

UNIVERSITÀ DEGLI STUDI DI MESSINA

DIPARTIMENTO DI SCIENZE MATEMATICHE E

INFORMATICHE, SCIENZE FISICHE E SCIENZE DELLA TERRA

Dottorato di Ricerca in Fisica

XXXIV CICLO

ELECTROMAGNETIC SCATTERING CALCULATIONS FOR OPTICAL TRAPPING AND SPACE APPLICATIONS

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SSD: MATTER PHYSICS - FIS/03

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To my wife, Mariasole: my new Family ...

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Introduction

The aim of this thesis is to discuss theoretical approaches and numerical results suitable for describing some phenomena that characterize the interaction between electromagnetic radiation and micro-nanostructured matter, going from nano-plasmonic particles and nanowires in optical traps, to interstellar and interplanetary dust particles.

We know that the electromagnetic scattering theory is accurately described by the transition matrix formalism (T-matrix) that allows to calculate the optical properties of spherical, composite, aggregated, and multistratified model particles. The T-matrix method, initially introduced by Waterman [1], is among the most efficient tools for the accurate calculation of light scattering from non-spherical particles, both isolated and interacting in composite structures [2]. It is based on the calculation of a linear operator (Transition operator) which, acting on the multipole amplitudes of the incident fields, gives as a result the multipole amplitudes of the scattered field. For a homogeneous spherical particle under plane wave illumination this coincides with the Mie theory [3]. The T-matrix approach is particularly advantageous when we deal with particles that can be modeled as cluster or aggregates of spheres, spheres with spherical (eccentric) inclusions, and multilayered spheres [2]. This technique takes into proper account the multiple scattering processes occurring among the spherical sub-units composing the aggregate and the contribution of all the details of the model structure. Optical properties of composite scatterers can be exactly calculated without introducing any approximation except the truncation of the expansion of the fields, being able to check the convergence of the results at every step. The elements of the T-matrix contain all the information on the particle nature (refractive index, size relative to the wavelength, and shape) and on the orientation of the scatterer with respect to the incident field. A fundamental feature is that the T-matrix is independent of the propagation direction and polarization states of the incident and of the scattered fields. This means that, once we compute the T-matrix elements, we do not need to compute them again if the incident field direction and polarization state changes [4]. This is a key property when dealing with optical trapping calculations since the particle has to be placed in different positions to reconstruct the optical force within the focal spot [5]. Thanks to the flexibility and the accuracy of the T-matrix technique, we have the possibility to explore several systems and configurations in a broad range of applications, going from optical force theory, *i.e.* Optical Tweezers [6–8], through fundamental physics problems such as optical forces related to non-local spin-dependent components [9], interstellar dust modelling [10–12], to the study of the behaviour of plasmonic systems [13, 14].

This thesis is structured as follows. In the first chapter, we describe the electromagnetic theory of light scattering in the T-matrix formalism. After introducing different scattering regimes, useful for justifying approximations under certain conditions, we introduce the scattering problem. Then, we present the T-matrix formalism discussing the special cases of Mie theory (scattering by a spherical particle) and light scattering by aggregates of spheres.

The second chapter deals with the theory of optical forces, focusing in particular on optical tweezers. After an historical overview, we introduce the Maxwell stress tensor and describe the general equations from which optical forces and torques can be calculated within the electromagnetic scattering theory. We focus on two different particle size regimes, much larger (ray optics) and much smaller (dipole approximation) than the light wavelength, where simple expressions of the force are obtained thanks to specific approximations. We then describe how to calculate optical forces within the T-Matrix formalism both in a plane wave configuration and in optical tweezers. We conclude by discussing a key example of optical trapping calculations on non-spherical particles, the size scaling of optical trapping of nanowires.

In the third chapter, we present the behaviour of resonant gain assisted metallic/dielectric nanoshells in optical tweezers. We investigate gain-assisted optical forces on dye-enriched silver nanoshells in the quasi-static limit by means of a theoretical/numerical approach. We demonstrate the onset of nonlinear optical trapping of these resonant nanostructures in a counterpropagating Gaussian beam configuration. We study the optical forces and trapping behaviour as a function of wavelength, particle gain level, and laser power. We support the theoretical analysis with Brownian dynamics simulations that show how particle position locking is achieved at high gains in extended optical trapping potentials. Finally, for wavelengths blue-detuned with respect to the plasmon-enhanced resonance, we observe particle channeling by the standing wave antinodes due to gradient force reversal.

In the fourth chapter, spin-dependent optical forces in optically trapped nanowires are studied. We present computational results associated with the onset of a spin-dependent optical force component occurring on zinc oxide nanowires trapped in optical tweezers with circularly polarized light. This type of non-conservative force appears directed perpendicularly with respect to the propagation direction of the incident light on the nanowires both for plane wave illumination and for optical tweezers. We show how this transverse optical force component is also shape dependent and connected with the imaginary part of the local Poynting vector and the local spin density.

In the fifth chapter the polarization constraints on core-mantle interstellar dust grain models are discussed. The morphology, structure, and composition of interstellar dust grains is still a much debated issue in the literature. Strong evidences point toward the possibility that dust grains are characterized by a stratified structure, made by a silicate core covered with a carbon shell. However, such model has been challenged by the spectropolarimetric observations of the carbon absorption feature at 3.4 μ m in the diffuse interstellar medium. The negligible polarization of this feature, together with the observation of a strong polarization in the silicate feature at 9.7 μ m along the same sightline, seems to suggest that carbon and silicate are separate components in grains. We numerically investigate this problem, comparing the computed polarization feature with the observational evidences and trying to understand if a model in which carbon and silicate co-exist in the same core-mantle structure is still plausible.

In the last chapter, we present calculations of optical forces for extraterrestrial applications, space tweezers. We describe how optical tweezers can be used to trap and characterize extraterrestrial particulate matter. In particular, we calculate radiation pressure and optical trapping properties of a variety of complex particles of astrophysical interest. Our results open perspectives in the investigation of extraterrestrial particles on our planet, in controlled laboratory experiments, aiming for space tweezers applications where optical tweezers are used to trap and characterize dust particles directly in space or on extraterrestrial bodies during exploratory missions.

Chapter 1

Light scattering and T-matrix formalism

1.1 Introduction

By electromagnetic scattering we mean a wide class of radiation-matter interaction phenomena in which electromagnetic fields are deflected due to collision with material particles. We can distinguish between elastic scattering when no energy transfers are involved in the process, and inelastic, in the otherwise. For example, Rayleigh scattering or Mie scattering belongs to the first category while a typical example of inelastic scattering is Raman scattering. In this thesis, we will consider only the case of elastic scattering because we assume that, in the considered matter-radiation interactions, the energy is conserved. A complete study of the problem should be dealt by treating both radiation and matter using the Quantum Electrodynamics (QED). However, computing with wavelengths greater than those of De Broglie, we can neglect eventually quantum contributions. In such manner, we can take advantage of appropriate approximations to treat the radiation-matter interaction in Classical Electrodynamics (CED). Historically, the light scattering classical theoretical understanding has been investigated through the use of suitable approximations that depend on the size of the particle [2, 8, 15]. For homogeneous spherical particles, accurate evaluation of the radiation fields can be obtained by Mie theory [3]. In general, when we study the scattering process involving non-spherical or non-homogeneous particles, we must use a full electromagnetic theory based on the Maxwell's equations [4] and the integration of the Maxwell stress tensor [15]. However, such calculations can be extremely complex through computational intensive procedures. For this reason different methods, such as the transition matrix (T-matrix) approach [1, 2], have been developed to solve the scattering problem and calculate optical forces more efficiently. The T-matrix is the matrix representation of a linear operator which, acting on the multipole amplitudes of the incident field, gives as a result the multipole amplitudes of the scattered field. When the scatterer is a homogeneous spherical particle, *i.e.*, in the highly i = 1symmetric case, the scattered field does not depend on the orientation of the particle and is exactly described by the Mie theory [3]. However, the spherical model is not able to describe a great number of real scatterers, which, in general, may exhibit an asymmetric shape or may result from the aggregation of several constituent monomers. The introduction of asymmetry is immediately reflected in the polarization of the scattered field as well as in its dependence on the position and orientation of the particle. In such case, a model scatterer must be used which can simulate as accurately as possible the details of the structure of such composite particles. The T-matrix approach is particularly advantageous when we deal with particles composed by spherical constituents, *i.e.*, cluster or aggregates of spheres, spheres with spherical (eccentric) inclusions, and multilayered spheres [2]. By varying the number of the constituent spheres (as well as of the layers of the inclusions), their refractive index and their mutual position, structures can be obtained which better approximate the shape and the composition of the scatterers in the analysed system. This technique takes into proper account the multiple scattering processes occurring among the spherical sub-units composing the aggregate and the contribution of all the details of the model structure. Optical properties of composite scatterers can be exactly calculated without introducing any approximation except the truncation of the expansion of the fields, being able to check the convergence of the results at every step.

The elements of the T-matrix contain all the information on the particle nature (refractive index, size relative to the wavelength, and shape) and on the orientation of the scatterer with respect to the incident field. A fundamental feature is that the T-matrix is independent of the propagation direction and polarization states of the incident and of the scattered fields. This means that, once we compute the T-matrix elements, we do not need to compute them again if the incident field direction and polarization state changes [4]. Borghese et al. [16] succeeded in showing that the transformation properties of the multipole fields under rotation of the coordinate frame imply corresponding transformation properties of the T-matrix elements under rotation of the scattering particle. Such transformation properties enable us to calculate orientational averages of the optical quantities of interest with a reasonable computational effort [2]. This is one of the greatest advantages offered by the T-matrix approach respect to other computational techniques like, for example, the discrete dipole approximation [17, 18]. Thanks to the flexibility and the accuracy of the T-matrix technique, we have the opportunity to explore several systems and configurations in a broad range of fields of applications, going from interstellar dust modelling [10, 11] to the study of the behaviour of plasmon systems and finally to the optical trapping theory that we discuss in this thesis. We will formally discuss this method in details afterwards, while in the next section we give an overview of the approximated approaches that often can grant a fast and simple way to obtain reasonable results in specific regimes.

1.2 Scattering regimes

For calculating the light fields acting on spherical or quasi-spherical particles, it is customary to identify several regimes which depend on the particle size [15]. For each regime, simplifications and approximations have been made for a better and more qualitative understanding and calculation of the light scattering. The size parameter x is crucially used to determine the range of validity of these approximations:

$$x = k_{\rm m}a = \frac{2\pi n_{\rm m}}{\lambda_0}a \tag{1.1}$$

where $k_{\rm m} = 2\pi n_{\rm m}/\lambda_0$ is the light wavenumber in the medium surrounding the particle, a is the particle radius, λ_0 is the incoming monochromatic wavelength in vacuum and $n_{\rm m}$ is the refractive index of the surrounding non-magnetic medium. When the particle radius is larger than the incident wavelength, *i.e.* $k_{\rm m}a \gg 1$, we can use the geometrical optics (GO) approximation. In some of the problems addressed in this thesis, we can perform the computations within the ray optics regime, as we will see more systematically in the Chap. 2. The accuracy of this approximation increases with the size parameter, whereas the exact theories become unpractical due to the increasing computational complexity. This makes ray optics very useful when dealing with large particles. In the opposite case, if the radius of the particle is much smaller than λ_0 ($k_{\rm m}a \ll 1$), we can use the Rayleigh approximation and consider the particle as a dipole [19] or a collection of dipoles [17]. This means we are considering the electromagnetic fields homogeneous inside the particle under the condition $\left|\frac{n_{\rm p}}{n_{\rm m}}\right| ka \ll 1$ where $n_{\rm p}$ is the refractive index of the particle. This particular relation has to be considered when we deal with high refractive index dielectric particles (e.g., silicon) or noble metal (e.g., silicon)gold, silver) nanoparticles, where the presence of plasmonic resonances dominate the optical response [13]. In the intermediate regime, where we work for most of this work, the particle size is comparable with the light wavelength



Figure 1.1: Pictorial view of a scattering process. Scattering theory studies how an incoming electromagnetic wave is scattered by a particle. In general, when light impinges on an object, the latter emits a scattered electromagnetic field, which in the far-field is a spherical wave. In particular, given an incoming linearly polarised plane electromagnetic wave (\mathbf{E}_i) in a medium of refractive index n_m impinging on a particle of homogeneous refractive index n_p , one wants to determine the electromagnetic field inside the particle (\mathbf{E}_p) and the scattered electromagnetic field (\mathbf{E}_s), both in the near-field and in the far-field.

 $(k_{\rm m}a \simeq 1)$. In this way, we need a complete wave-optical modeling of the particle-light interaction to calculate the trapping forces. In such case, the T-matrix approach proves to be a very convenient choice.

1.3 The scattering problem

The scattering problem aims at describing the electromagnetic fields scattered by a particle when it is illuminated by an incoming electromagnetic wave. In Fig. (1.1) we represent a pictorial view of a scattering process. For a homogeneous particle of refractive index n_p in a medium of refractive index n_m , the three-dimensional homogeneous Helmholtz equations describe the scattering process:

$$\begin{cases} (\nabla^2 + k_{\rm m}^2) \mathbf{E}_{\rm i}(\mathbf{r}) &= \mathbf{0} \\ (\nabla^2 + k_{\rm m}^2) \mathbf{E}_{\rm s}(\mathbf{r}) &= \mathbf{0} \\ (\nabla^2 + k_{\rm p}^2) \mathbf{E}_{\rm p}(\mathbf{r}) &= \mathbf{0} \end{cases},$$
(1.2)

where $\mathbf{E}_{i}(\mathbf{r})$ is the incident electric field, $\mathbf{E}_{s}(\mathbf{r})$ is the scattered electric field, so that the total electric field outside the particle is $\mathbf{E}_{t}(\mathbf{r}) = \mathbf{E}_{i}(\mathbf{r}) + \mathbf{E}_{s}(\mathbf{r})$, $\mathbf{E}_{p}(\mathbf{r})$ is the total electric field inside the particle, $k_{m} = n_{m}k_{0}$, $k_{p} = n_{p}k_{0}$ and k_{0} is the vacuum wavenumber. To derive the vector solutions of the previous equations, it is advisable to introduce a spherical coordinate system (r, ϑ, φ) and a scalar function $F(r, \vartheta, \varphi)$, solution of the scalar Helmholtz equation:

$$(\nabla^2 + k^2)F(r,\vartheta,\varphi) = 0. \tag{1.3}$$

The most general solution of the Helmholtz equation that is regular at the origin is:

$$F_{j}(\mathbf{r}) = \sum_{l=0}^{+\infty} \sum_{m=-l}^{+l} B_{lm} j_{l}(kr) Y_{lm}(\hat{\mathbf{r}})$$
(1.4)

and the general solution that satisfies the radiation condition at infinity is

$$F_h(\mathbf{r}) = \sum_{l=0}^{+\infty} \sum_{m=-l}^{+l} C_{lm} h_l(kr) Y_{lm}(\hat{\mathbf{r}}) , \qquad (1.5)$$

where B_{lm} and C_{lm} are the amplitudes corresponding to each mode, $j_l(kr)$ and $h_l(kr)$ are respectively the Bessel and the Hankel functions, $Y_{lm}(\hat{\mathbf{r}})$ are the spherical harmonics and l is a natural number. The electromagnetic fields outside and inside the particle are related by the boundary conditions across the surface of the particle.

1.3.1 Cross sections

Since any electromagnetic field can be described as a superpositions of plane waves and Maxwell's equations are linear, it suffices to consider the scattering produced by a single linearly polarised incoming homogeneous plane wave [15]:

$$\mathbf{E}_{\mathbf{i}}(\mathbf{r}) = E_0 e^{i\mathbf{k}_{\mathbf{i}}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\mathbf{i}},\tag{1.6}$$

where E_0 is the modulo of a vector specifying the amplitude, $\hat{\mathbf{e}}_i$ is the unit vector indicating the polarisation direction and $\mathbf{k}_i = k_m \hat{\mathbf{k}}_i$ is the real wavevector along the incidence propagation direction. $\mathbf{E}_s(\mathbf{r}) = [E_{s,x}(\mathbf{r}), E_{s,y}(\mathbf{r}), E_{s,z}(\mathbf{r})]$ satisfies the vector Helmholtz equation and its Cartesian components must satisfy the scalar Helmholtz equation. The solution to this equation satisfies the radiation condition at infinity [20, 21], *i.e.*, for the x-component:

$$E_{\mathbf{s},x}(\mathbf{r}) = E_{\mathbf{s},x}(r, \hat{\mathbf{k}}_{\mathbf{s}}) = \sum_{lm} h_l(k_m r) C_{lm,x}(\hat{\mathbf{k}}_{\mathbf{i}}) Y_{lm}(\hat{\mathbf{k}}_{\mathbf{s}}), \qquad (1.7)$$

where $\hat{\mathbf{k}}_{s}$ is the radial unit vector indicating the direction of the scattered wave, $\mathbf{r} = r\hat{\mathbf{k}}_{s}$ and the amplitudes $C_{lm,x}(\hat{\mathbf{k}}_{i})$, which depend on the direction of the incident wave, are determined by the boundary conditions at the surface of the particle.

Using the asymptotic properties of $h_l(k_{\rm m}r)$ for $k_{\rm m}r \to \infty$, the asymptotic form of $E_{{\rm s},x}({\bf r})$ can be written as a spherical wave

$$E_{\mathbf{s},x}(\mathbf{r}) = E_0 \; \frac{e^{ik_{\mathrm{m}}r}}{r} \; f_x(\hat{\mathbf{k}}_{\mathrm{s}}, \hat{\mathbf{k}}_{\mathrm{i}}) \;, \tag{1.8}$$

where the x-component of the normalised scattering amplitude has been introduced as

$$f_x(\hat{\mathbf{k}}_{\rm s}, \hat{\mathbf{k}}_{\rm i}) = k_{\rm m}^{-1} \sum_{lm} (-i)^{l+1} C_{lm,x}(\hat{\mathbf{k}}_{\rm i}) Y_{lm}(\hat{\mathbf{k}}_{\rm s}) .$$
(1.9)

Repeating the same procedure on $E_{s,y}(\mathbf{r})$ and $E_{s,z}(\mathbf{r})$, the normalised scattering amplitude $\mathbf{f}(\hat{\mathbf{k}}_{s}, \hat{\mathbf{k}}_{i}) = \left[f_{x}(\hat{\mathbf{k}}_{s}, \hat{\mathbf{k}}_{i}), f_{y}(\hat{\mathbf{k}}_{s}, \hat{\mathbf{k}}_{i}), f_{z}(\hat{\mathbf{k}}_{s}, \hat{\mathbf{k}}_{i})\right]$ and the asymptotic form of the scattered field can be obtained and

$$\mathbf{E}_{s}(\mathbf{r}) = \mathbf{E}_{s}(r, \hat{\mathbf{k}}_{s}) = E_{0} \mathbf{f}(\hat{\mathbf{k}}_{s}, \hat{\mathbf{k}}_{i}) \frac{e^{ik_{m}r}}{r} .$$
(1.10)

As known, from the scattering amplitude $\mathbf{f}(\hat{\mathbf{k}}_{s}, \hat{\mathbf{k}}_{i})$ it is possible to derive all the other optical properties such as the scattering cross-section σ_{scat} (taking the square modulus of the scattering amplitude and integrating over the solid angle), the extinction cross-section σ_{ext} thanks to the optical theorem, and finally the absorption cross-section σ_{abs} . The asymmetry of the scattering with respect to the incoming wave direction and polarisation can be quantified by the asymmetry parameters and, in particular, by the asymmetry parameter in the direction of the incoming wave defined as

$$g_{i} = \frac{1}{\sigma_{\text{scat}}} \oint_{\Omega} \frac{\mathrm{d}\sigma_{\text{scat}}}{\mathrm{d}\Omega} \hat{\mathbf{r}} \cdot \hat{\mathbf{k}}_{i} \,\mathrm{d}\Omega \tag{1.11}$$

and the transverse asymmetry parameters defined as

$$g_1 = \frac{1}{\sigma_{\text{scat}}} \oint_{\Omega} \frac{\mathrm{d}\sigma_{\text{scat}}}{\mathrm{d}\Omega} \hat{\mathbf{r}} \cdot \hat{\mathbf{u}}_1 \,\mathrm{d}\Omega \tag{1.12}$$

and

$$g_2 = \frac{1}{\sigma_{\text{scat}}} \oint_{\Omega} \frac{\mathrm{d}\sigma_{\text{scat}}}{\mathrm{d}\Omega} \hat{\mathbf{r}} \cdot \hat{\mathbf{u}}_2 \,\mathrm{d}\Omega \,, \qquad (1.13)$$

where $\hat{\mathbf{u}}_1 = \hat{\mathbf{e}}_i$ and $\hat{\mathbf{u}}_2 = \hat{\mathbf{k}}_i \times \hat{\mathbf{e}}_i$. It is often convenient to characterize a scattering particle through efficiencies that are defined by the ratios:

$$Q_{\text{scat}} = \frac{\sigma_{\text{scat}}}{\sigma_{\text{geom}}}, Q_{\text{ext}} = \frac{\sigma_{\text{ext}}}{\sigma_{\text{geom}}}, Q_{\text{abs}} = \frac{\sigma_{\text{abs}}}{\sigma_{\text{geom}}}.$$
 (1.14)

in which, σ_{geom} is the geometrical cross section of the particle in a plane orthogonal to the direction of incidence. Another quantity that characterizes the optical properties of a particle is the albedo that is defined as

$$\bar{\omega} = \frac{\sigma_{\text{scat}}}{\sigma_{\text{ext}}} = \frac{Q_{\text{scat}}}{Q_{\text{geom}}}.$$
(1.15)

The albedo, when considered in terms of photons impinging on a particle, gives the probability that a photon be scattered rather than absorbed [22].

1.3.2 Multipole expansion of a plane wave

Considering a homogeneous electromagnetic plane wave where planes of equal phase and of equal amplitude are mutually parallel to each other (Eq. 1.6). Since it is finite at the origin, it can be decomposed in **J**-multipoles whose radial function is a spherical Bessel function $j_l(kr)$ to ensure the finiteness at the origin:

$$\mathbf{E}_{\mathbf{i}}(r,\hat{\mathbf{r}}) = E_0 \sum_{p=1,2} \sum_{lm} W_{\mathbf{i},lm}^{(p)}(\hat{\mathbf{e}}_{\mathbf{i}},\hat{\mathbf{k}}_{\mathbf{i}}) \mathbf{J}_{lm}^{(p)}(kr,\hat{\mathbf{r}}).$$
(1.16)

In this equation the scalar spherical harmonics $Y_{lm}(\hat{\mathbf{r}})$ are been extended in the radial vector orthonormal spherical harmonic $\mathbf{Y}_{lm}(\hat{\mathbf{r}})$ and in the transversal vector orthonormal spherical harmonic $\mathbf{Z}_{lm}^{(1)}(\hat{\mathbf{r}})$ and $\mathbf{Z}_{lm}^{(2)}(\hat{\mathbf{r}})$ to deal with vector fields. So they are defined as [2]:

$$\begin{cases} \mathbf{Y}_{lm}(\hat{\mathbf{r}}) &= Y_{lm}(\hat{\mathbf{r}})\hat{\mathbf{r}} \\ \mathbf{Z}_{lm}^{(1)}(\hat{\mathbf{r}}) &= -\frac{i}{\sqrt{l(l+1)}} \hat{\mathbf{r}} \times \nabla Y_{lm}(\hat{\mathbf{r}}) \\ \mathbf{Z}_{lm}^{(2)}(\hat{\mathbf{r}}) &= \mathbf{Z}_{lm}^{(1)}(\hat{\mathbf{r}}) \times \hat{\mathbf{r}} \end{cases}$$
(1.17)

Then:

$$W_{i,lm}^{(1)} = 4\pi i^{l} \hat{\mathbf{e}}_{i} \cdot \mathbf{Z}_{lm}^{(1)*}(\hat{\mathbf{k}}_{i})$$

$$W_{i,lm}^{(2)} = 4\pi i^{l+1} \hat{\mathbf{e}}_{i} \cdot \mathbf{Z}_{lm}^{(2)*}(\hat{\mathbf{k}}_{i})$$
(1.18)

are numerical coefficients referred to the multipolar components of magnetic (p = 1) and electric (p = 2) fields [23]. In analogy to incoming field, the scattered wave is expanded in **H**-multipoles, whose radial function is a spherical Hankel function $h_{(1),l}(kr)$ of the first kind because the scattered field has to satisfy the radiation condition at infinity [21]:

$$\mathbf{E}_{s}(r,\hat{\mathbf{r}}) = E_{0} \sum_{p=1,2} \sum_{lm} A_{s,lm}^{(p)}(\hat{\mathbf{e}}_{i},\hat{\mathbf{k}}_{i}) \mathbf{H}_{lm}^{(p)}(k_{m}r,\hat{\mathbf{r}}), \qquad (1.19)$$

where $A_{s,lm}^{(1)}$ and $A_{s,lm}^{(2)}$ are the amplitudes of the magnetic and electrical scattered fields respectively which are determined by the boundary conditions across the surface of the particle. In general, they depend on the orientation of the scattering particle with respect to the incident field. The multipole expansion of the normalized scattering amplitude is easily obtained by taking the limit of the **H**-multipole fields for $kr \to \infty$ [2]. Then, the asymptotic form of the scattered field is:

$$\mathbf{E}_{s}(r, \hat{\mathbf{k}}_{s}) = E_{0} \frac{e^{ik_{m}r}}{k_{m}r} \sum_{plm} (-i)^{l+p} A_{s,lm}^{(p)}(\hat{\mathbf{e}}_{i}, \hat{\mathbf{k}}_{i}) \mathbf{Z}_{lm}^{(p)}(\hat{\mathbf{k}}_{s}).$$
(1.20)

The comparison with Eq. 1.10 yields

$$\mathbf{f}(\hat{\mathbf{k}}_{\rm s}, \hat{\mathbf{k}}_{\rm i}) = \frac{1}{k_{\rm m}} \sum_{plm} (-i)^{l+p} A_{{\rm s},lm}^{(p)}(\hat{\mathbf{u}}_{{\rm i},\eta}, \hat{\mathbf{k}}_{\rm i}) \mathbf{Z}_{lm}^{(p)}(\hat{\mathbf{k}}_{\rm s}), \qquad (1.21)$$

in which the temporary argument $\hat{\mathbf{e}}_i$ has been substituted by the label η that recalls the polarization of the incident field. Therefore the normalized scattering amplitude matrix elements on the basis $\hat{\mathbf{u}}_{s,\eta'}$ of the scattered field is:

$$f_{\eta'\eta} = -\frac{i}{4\pi k_{\rm m}} \sum_{plm} W_{\rm i,lm}^{(p)*}(\hat{\mathbf{u}}_{{\rm s},\eta'}, \hat{\mathbf{k}}_{\rm s}) A_{{\rm s},lm}^{(p)}(\hat{\mathbf{u}}_{{\rm i},\eta}, \hat{\mathbf{k}}_{\rm i}).$$
(1.22)

At this stage, the explicit expressions of the scattering and the extinction cross section of a particle can be written in terms of the amplitudes of the fields. Taking the square modulus of Eq. 1.21 and operating the integral over the entire solid angle, the scattering cross section is:

$$\sigma_{\rm scat} = \frac{1}{k_{\rm m}^2} \sum_{plm} A_{\rm s,lm}^{(p)*}(\hat{\mathbf{e}}_{\rm i}, \hat{\mathbf{k}}_{\rm i}) A_{\rm s,lm}^{(p)}(\hat{\mathbf{e}}_{\rm i}, \hat{\mathbf{k}}_{\rm i}).$$
(1.23)

The extinction cross section is related to the scattering amplitude through the optical theorem, then:

$$\sigma_{\text{ext}} = -\frac{1}{k_{\text{m}}^2} \Re \left\{ \sum_{plm} W_{\text{i},lm}^{(p)*}(\hat{\mathbf{e}}_{\text{i}}, \hat{\mathbf{k}}_{\text{i}}) A_{\text{s},lm}^{(p)}(\hat{\mathbf{e}}_{\text{i}}, \hat{\mathbf{k}}_{\text{i}}) \right\}$$
(1.24)

1.4 T-matrix

The transition matrix, or T-matrix, formalism was derived by Waterman [1] starting from the integral equation formulation of electromagnetic scattering to solve the scattering problem. The T-matrix was calculated by expanding the field into a series of spherical multipole fields and by imposing boundary conditions across the surface of the particles. This formulation of the T-matrix method, which is known as extended boundary condition method (EBCM), can then be regarded as a generalisation of Mie theory which is the paradigmatic solution of the light scattering problem obtained by Gustav Mie for a homogeneous sphere of arbitrary size. While, as we have seen, the analytical approach of the multipole expansion is relatively simple and general, the computational methods needed to calculate the expansion coefficients from the imposition of the boundary conditions can be quite complex [4]. The formulation used hereinafter is provided by Ferdinando Borghese and coworkers and shows the advantage that most calculations are carried out analytically and the resulting algorithms are, therefore, computationally efficient and accurate.

Because of the linearity of Maxwell's equations and of the boundary conditions, the scattering process can be considered as a linear operator \mathbb{T} (transition operator) so that

$$\mathbf{E}_{s} = \mathbb{T}\mathbf{E}_{i} , \qquad (1.25)$$

with \mathbf{E}_{i} the incoming electric field and \mathbf{E}_{s} the scattered electric field. Therefore, if both \mathbf{E}_{i} and \mathbf{E}_{s} are expanded on suitable bases (not necessarily the same), it is possible to find a transition matrix \mathbb{T} that relates the coefficients of such expansions, encompassing all the information on the morphology and orientation of the particle with respect to the incident field [1]. Since \mathbf{E}_{i} is in general finite at the origin, its expansion is conveniently given in terms of **J**-multipoles (Eq. 1.16) with amplitudes $W_{i,lm}^{(p)}$ (Eq. 1.18). Since \mathbf{E}_{s} must satisfy the radiation condition at infinity, it is convenient to expand it in terms of **H**-multipoles (Eq. 1.19) with amplitudes $A_{s,lm}^{(p)}$. These amplitudes are determined by imposing the boundary conditions across the surface of the scattering particle. The transition matrix $\mathbb{T} = \{T_{l'm'lm}^{(p'p)}\}$ of the scattering particle acts on the known multipole amplitudes of the incident field $W_{i,lm}^{(p)}$ to give the unknown amplitudes of the scattered field $A_{s,l'm'}^{(p')}$, *i.e.*,

$$A_{s,l'm'}^{(p')}(\hat{\mathbf{e}}_{i},\hat{\mathbf{k}}_{i}) = \sum_{plm} T_{l'm'lm}^{(p'p)} W_{i,lm}^{(p)}(\hat{\mathbf{e}}_{i},\hat{\mathbf{k}}_{i}) .$$
(1.26)

The quantities $T_{l'm'lm}^{(p'p)}$ take into account the morphology of the particle as well as the boundary conditions, but are independent of the state of polarization of the incident field. Therefore, Eq. 1.26 holds true whatever the polarization is. For instance, the equation

$$A_{s,\eta l'm'}^{(p')} = \sum_{plm} T_{l'm'lm}^{(p'p)} W_{i,\eta lm}^{(p)}$$
(1.27)

relates the basis-polarized amplitudes of the incident and of the scattered field. Then, substituting Eq. 1.27 into Eq. 1.22, the explicit relation between the scattering amplitude and the T-matrix can be obtained:

$$f_{\eta'\eta} = -\frac{i}{4\pi k_{\rm m}} \sum_{plm} \sum_{p'l'm'} W^{(p)*}_{{\rm s},\eta'lm} T^{(p'p)}_{l'm'lm} W^{(p')}_{{\rm i},\eta l'm'}.$$
 (1.28)

This equation, giving the explicit relation between the scattering amplitude and the Transition matrix, is perhaps the most important equation in the theory of light scattering. In fact, the observable quantities, such as the optical cross sections of the particles, are given in terms of the scattering amplitude matrix elements, and so can be easily computed once the T-matrix elements are known.

1.5 Applications to model particles

The T-matrix method can be used to rigorously describe light scattering by many particle models, specifically, homogeneous spheres, first described by Gustav Mie [3], radially non-homogeneous spheres, which is an extension of Mie theory to spheres where the refractive index is a regular function of the distance from the center [24], and aggregates or cluster of spheres, where the T-matrix approach proves to be a very powerful approach.

1.5.1 Mie theory

A milestone result in electromagnetic scattering theory is the complete solution to the problem of light scattering of a linearly polarised plane wave by a homogeneous sphere of arbitrary radius a and refractive index n_p surrounded by a medium of refractive index n_m [3]. This result was obtained by Gustav Mie in 1908 and is therefore known as Mie theory. If the material of the sphere and that of the surrounding medium are nonmagnetic, the boundary conditions reduce to the requirement of continuity of the tangential components of both the electric and magnetic fields, where the latter is related to the electric components by the rotor operator. The relations between the amplitudes of the scattered and of the incident fields represent the Mie coefficients, which are defined as:

$$a_{l} = -\frac{A_{s,lm}^{(2)}}{W_{i,lm}^{(2)}}$$

$$b_{l} = -\frac{A_{s,lm}^{(1)}}{W_{i,lm}^{(1)}}.$$
(1.29)

Mie coefficients are used to calculate the exact expressions of the scattered electrical and magnetic fields and, imposing the appropriate boundary conditions, they are:

$$a_{l} = \frac{n_{\rm m} u_{l}'(\rho_{\rm p}) u_{l}(\rho_{\rm m}) - n_{\rm p} u_{l}(\rho_{\rm p}) u_{l}'(\rho_{\rm m})}{n_{\rm m} u_{l}'(\rho_{\rm p}) w_{l}(\rho_{\rm m}) - n_{\rm p} u_{l}(\rho_{\rm p}) w_{l}'(\rho_{\rm m})}$$
(1.30)

$$b_{l} = \frac{n_{\rm p} u_{l}'(\rho_{\rm p}) u_{l}(\rho_{\rm m}) - n_{\rm m} u_{l}(\rho_{\rm p}) u_{l}'(\rho_{\rm m})}{n_{\rm p} u_{l}'(\rho_{\rm p}) w_{l}(\rho_{\rm m}) - n_{\rm m} u_{l}(\rho_{\rm p}) w_{l}'(\rho_{\rm m})}$$
(1.31)

where $u_l(\rho) = \rho j_l(\rho)$ and $w_l(\rho) = \rho h_l(\rho)$ are Riccati-Bessel and Riccati-Hankel functions, respectively [25], while the prime denotes derivation with respect to the argument, $\rho_m = n_m k_0 a$ and $\rho_p = n_p k_0 a$. Thus, the scattering problem is reduced to the calculations of these coefficients through, *e.g.*, the imposition of the boundary conditions across the particle surface or by point matching numerically the fields at the surface [2]. The T-matrix formalism for a spherical scatterer provides the Mie coefficients. The T-matrix for a homogenous spherical particle is diagonal, independent of m and connected to the Mie coefficients a_l and b_l , *i.e.*,

$$\mathbf{A}_{\mathrm{s}} = -\mathbb{R}\mathbf{W}_{\mathrm{i}} , \qquad (1.32)$$

where $\mathbb{R} = \{-T_{l'm'lm}^{(p'p)}\} = \{R_{l'm'lm}^{(p'p)}\}$ [22] and

$$R_{l'm'lm}^{(p'p)} = \begin{cases} b_l & p = p' = 1 \text{ and } l = l' \text{ and } m = m' \\ a_l & p = p' = 2 \text{ and } l = l' \text{ and } m = m' \\ 0 & \text{otherwise} \end{cases}$$
(1.33)

Moreover, according to Eq. 1.28 and Eq. 1.32 the scattering amplitude has the form:

$$f_{\eta'\eta} = \frac{i}{4\pi k_{\rm m}} \sum_{plm} W_{{\rm s},\eta lm}^{(p)*} R_l^{(p)} W_{{\rm i},\eta' lm}^{(p)}.$$
 (1.34)

It is actually diagonal in η on account of the reciprocity theorem and the diagonal elements $f_{\eta'\eta}$ are complex numbers with a different phase, so that the scattered wave may turn out to be elliptically polarized even when the incident wave is linearly polarized. To get an accurate representation of the scattered field, the sum in Eq. 1.34 must be extended to a sufficiently high value of l, say l_M . In other words, the convergence of the calculation must always be checked [2]. To get a fair convergence for a sphere of size parameter x, it is necessary to include into Eq. 1.34 terms up to $l_M > x$ [20]. In practice, when $x \leq 0.1$ one needs to include terms up to $l_M = 1$ or, at most, up to $l_M = 2$ only. For smaller values of x, one can expand the elements of the Tmatrix in powers of x, thus obtaining the Rayleigh scattering approximation that assumes the scattered field from a small sphere is well approximated by the field of the dipole moment induced by the incident electromagnetic wave [22]. However, the elements of the T-matrix as well as the convergence of the scattered field depend not only on the size parameter but also on the refractive index $n_{\rm p}$ (that is contained in $\rho_{\rm p}$). Therefore, as long as the refractive indexes are frequency independent, the response of a spherical scatterer does not depend separately on a and λ , but rather on their ratio. This is the principle of optical scaling that allows the people to test the reliability of the theoretical predictions using microwave devices and large scale scatterers [26].

1.5.2 Aggregates of spheres

The spherical scatterer model, on account of the ease of computation, has been widely used in the scientific literature and in many fields of application. However, the particles that are most commonly met in actual observations are non-spherical and the effects that stem from the lack of sphericity may be attenuated but never cancelled, not even by the use of an averaging procedure. Several attempts were made to devise model non-spherical particles such that the optical properties could be calculated as exactly as possible, *i.e.*, without resorting to any approximation. The first real progress was marked by Bruning and Lo [27], who devised a technique to calculate the optical properties of linear chains of identical spherical scatterers. The properties of this model were investigated by Peterson and Ström [28] for general geometry of the aggregation, whereas, the first application of the cluster model to the description of real particles is due to Gérardy and Ausloos [29]. In this subsection, we present the procedure devised by Borghese et al. for the calculation of the T-matrix for a group of N, not necessarily equal, spheres whose mutual distances are so small that they must be dealt with as one object [16]. The geometry of such kind of scatterer is arbitrary to a large extent, so that aggregates can be built to model particles of various shapes. The emphasis is on the T-matrix on account of the usefulness of the latter for performing orientational averages. The surrounding medium is assumed to be a homogeneous dielectric so that the incident field still has the form of a polarized plane wave whose multipole expansion is given by Eq. 1.16. The spheres are numbered by an index α while \mathbf{R}_{α} is the vector position of the center of the α th sphere of radius a_{α} and refractive index n_{α} . Furthermore, the following theory refers to aggregates of spheres that, if isolated, could be described by Mie Theory. The field scattered by the whole aggregate as the superposition of the fields scattered by each of the spheres is

$$\mathbf{E}_{\mathrm{s},\eta} = E_0 \sum_{\alpha=1}^{N} \sum_{plm} \mathcal{A}_{\eta\alpha lm}^{(p)} \mathbf{H}_{lm}^{(p)}(k_{\mathrm{m}}, \mathbf{r}_{\alpha})$$
(1.35)

where the amplitudes $\mathcal{A}_{\eta\alpha lm}^{(p)}$ should be calculated so that $\mathbf{E}_{s\eta}$ satisfy the appropriate boundary conditions at the surface of each of the spheres. The radiation condition at infinity is automatically satisfied because the expansion includes **H**-multipole fields only. The field within each sphere is taken in the form

$$\mathbf{E}_{\mathbf{p},\eta\alpha} = E_0 \sum_{plm} \mathcal{C}_{\eta\alpha lm}^{(p)} \mathbf{J}_{lm}^{(p)}(k_\alpha, \mathbf{r}_\alpha)$$
(1.36)

where k_{α} is the wavenumber for each sphere. Due to the presence of the **J**-multipole fields, the field is regular everywhere within the sphere. While the scattered field is given by a linear combination of multipole fields that have different origins, the incident field is given by a combination of multipole

fields centered at the origin of the coordinates. Since the boundary conditions must be imposed at the surface of each of the spheres, *e.g.*, of the α th sphere, the whole field can be rewritten in terms of multipole fields centered at \mathbf{R}_{α} , resorting to the addition theorem [30]. The scattered field at the surface of the α th sphere turns out to be

$$\mathbf{E}_{\mathrm{s},\eta\alpha} = E_0 \sum_{plm} \left[\mathcal{A}_{\eta\alpha lm}^{(p)} \mathbf{H}_{lm}^{(p)}(k_{\mathrm{m}},\mathbf{r}_{\alpha}) + \sum_{\alpha'} \sum_{p'l'm'} \mathcal{A}_{\eta\alpha'l'm'}^{(p')} \mathcal{H}_{\alpha lm\alpha'l'm'}^{(pp')} \mathbf{J}_{lm}^{(p)}(k_{\mathrm{m}},\mathbf{r}_{\alpha}) \right].$$
(1.37)

Analogously, the incident field at the surface of the α th sphere is

$$\mathbf{E}_{\mathbf{i},\eta\alpha} = E_0 \sum_{plm} \left[\sum_{p'l'm'} W_{\mathbf{i},\eta l'm'}^{(p')} \mathcal{J}_{\alpha lm0l'm'}^{(pp')} \right] \mathbf{J}_{lm}^{(p)}(k_{\mathrm{m}},\mathbf{r}_{\alpha})$$
(1.38)

where $\mathbf{R}_0 = 0$ is the vector position of the origin and the quantities \mathcal{H} and \mathcal{J} respectively in Eqs. 1.37, 1.38 are obtained applying the addition theorem to the vector Helmholtz harmonics \mathbf{H} and \mathbf{J} together with the properties of the Clebsch-Gordan coefficients. At this stage, using the same technique for homogeneous spheres, the boundary conditions can be imposed. Once this elimination is done, a system of linear nonhomogeneous equations is obtained such as

$$\sum_{\alpha'} \sum_{p'l'm'} \mathcal{M}^{(pp')}_{\alpha l m \alpha' l'm'} \mathcal{A}^{(p)}_{\eta \alpha l m} = -\mathcal{W}^{(p)}_{\mathbf{i},\eta \alpha l m}, \qquad (1.39)$$

where new coefficients have been defined as

$$\mathcal{W}_{\mathbf{i},\eta\alpha lm}^{(p)} = \sum_{p'l'm'} W_{\mathbf{i},\eta l'm'}^{(p')} \mathcal{J}_{\alpha lm0l'm'}^{(pp')}$$
(1.40)

$$\mathcal{M}^{(p,p')}_{\alpha l m \alpha' l' m'} = (R^{(p)}_{\alpha l})^{-1} \delta_{\alpha \alpha'} \delta_{pp'} \delta_{ll'} \delta_{mm'} + \mathcal{H}^{(pp')}_{\alpha l m \alpha' l' m'}.$$
 (1.41)

In the last equation, the quantities $R_{\alpha l}^{(p)}$ are the Mie coefficients (Eq. 1.33) for the scattering from the α th sphere. The matrix \mathcal{H} describes the multiple scattering processes that, in view of the small mutual distance, occur with noticeable strength among the spheres of the aggregate. The amplitudes of the scattered field are calculated by solving the system of Eq. 1.39. Furthermore, the elements $\mathcal{H}_{\alpha lm\alpha' l'm'}^{(p,p')}$ of the transfer matrix couple multipole fields both of the same and of different parity with origin on different spheres. Then, the formal solution to the system of Eq. 1.39 is

$$\mathcal{A}_{\eta\alpha lm}^{(p)} = -\sum_{p'l'm'} \left[M^{-1} \right]_{\alpha lm\alpha' l'm'}^{(pp')} \mathcal{W}_{\mathbf{i},\eta\alpha' l'm'}^{(p')}.$$
 (1.42)

This equation may lead to the conclusion that matrix \mathbb{M}^{-1} be the Tmatrix of the aggregate. This conclusion is incorrect however, because, according to Eq. 1.27, the T-matrix relates the multipole amplitudes of the incident field to those of the field scattered by the whole object. On the contrary, Eq. 1.42 relates the amplitudes of the incident field to those of the fields scattered by each sphere in the aggregate. In order to define the Tmatrix for the whole aggregate it is necessary to express the scattered field in terms of multipole fields with the same origin. Actually, with the help of the addition theorem, the scattered field (Eq. 1.37) can be cast into the form

$$\mathbf{E}_{\mathbf{s},\eta} = E_0 \sum_{plm} \left[\sum_{\alpha'} \sum_{p'l'm'} \mathcal{A}_{\eta\alpha'l'm'}^{(p')} \mathcal{J}_{0lm\alpha'l'm'}^{(pp')} \right] \mathbf{H}_{lm}^{(p)}(k_{\mathrm{m}}, \mathbf{r}_{\alpha}) \\
= E_0 \sum_{plm} \mathcal{A}_{\eta lm}^{(p)} \mathbf{H}_{lm}^{(p)}(k_{\mathrm{m}}, \mathbf{r}_{\alpha}),$$
(1.43)

which is valid at a large distance from the aggregate or, at least, outside the smallest sphere with center at \mathbf{R}_0 that includes the whole aggregate. The preceding equation shows that the field scattered by the whole cluster can be expanded as a series of vector multipole fields with a single origin provided that the amplitudes are

$$A_{\eta lm}^{(p)} = \sum_{\alpha'} \sum_{p'l'm'} \mathcal{A}_{\eta\alpha'l'm'}^{(p')} \mathcal{J}_{0lm\alpha'l'm'}^{(pp')}.$$
 (1.44)

Then, the T-matrix of the aggregate can be defined in a compact form as

$$T_{lml'm'}^{(pp')} = -\sum_{\alpha\alpha'} \sum_{qLM} \sum_{q'L'M'} \mathcal{J}_{0lm\alpha LM}^{(pq)} \left[M^{-1} \right]_{\alpha LM\alpha'L'M'}^{(qq')} \mathcal{J}_{\alpha L'M'0l'm'}^{(q'p')}.$$
 (1.45)

The T-matrix defined in the preceding equation has the correct transformation properties under rotation, although it is non diagonal as a consequence of the lack of spherical symmetry of the aggregate.

1.5.3 Convergence

A crucial aspect concerning the computation is the convergence of the results. The calculation of the T-matrix of an aggregate requires inverting the

matrix \mathbb{M} whose order is, in principle, infinite. Of course the system (1.39) is truncated to some finite order by including in Eq. 1.45 terms up to order L_M , the maximum value both for L and L' in Eq. 1.45. The maximum value is chosen to ensure the required accuracy of the transition matrix elements. For a cluster of N spheres this implies the solution of a system of order $D_M = 2NL_M(L_M + 2)$, which may become too large. Actually, the inversion of the matrix \mathbb{M} is responsible for most of the time required for the calculation and this time scales, in fact, as D_M^3 . Thus, on account of the definition of D_M , the computation time scales as L_M^6 whereas the storage requirements scale as L_M^4 , so that it pays, in terms of both CPU time and storage requirements, to keep L_M as low as practicable. The choice of the appropriate value of L_M has been the concern of several researchers. For instance Quinten et al. |31| used the criterion suggested by Wiscombe [32]. The choice of L_M has been guided by the considerations provided by Iati et al [11]. For an isolated dielectric sphere with radius a_s we have to choose $L_M > ka_s + m_s$ [20], where $k = nk_{\nu}, k_{\nu} = \omega/c$ is the wavenumber of the particle and m_s is an integer that depends on the refractive index. Now, by looking to Eqs. 1.39, 1.41, we see that what distinguishes a set of independent spheres from a true aggregate of spheres is the presence of the elements $\mathcal{H}^{(pp')}_{\alpha lm\alpha' l'm'}$ that describe the multiple scattering processes occurring among the spheres. Of course, these processes become less and less effective the more the spheres are separated. By looking to the explicit expression of the \mathcal{H} -elements [2], one sees that their magnitudes actually become smaller and smaller for well separated spheres. As a result, we can assume that, choosing L_M a little larger than necessary to ensure the convergence of the component spheres as if they were isolated, we should also get fairly convergent values for the transition matrix elements. Thus we make the ansatz that we can choose $L_M > k_{\nu}a_c + m_c$, where a_c is the radius of the smallest sphere including the entire aggregate. Even in this case m_c is an integer that depends on the refractive index.

Chapter 2

Theory of optical forces and optical tweezers

2.1 Historical perspective

Historically, the idea that light exerts a mechanical action on matter has been known since the explanation of comet tails given by Kepler [15, 33]. In fact, comets generally show two tails while approaching the Sun. Α first tail, more diffused, composed by dust and ice particles formed by the radiation pressure of the solar light; and a second one composed of ions and charged particles due to the solar wind. It was only in 1873 that James C. Maxwell gave the first theoretical explanation of electromagnetic phenomena[34]. Within his electromagnetic theory the identification of light momentum associated to the Poynting vector yields that light can exert a force on matter due to the momentum exchanged between light and matter upon scattering [35]. The Italian physicist Bartoli independently predicted the pressure of light on the basis of thermodynamics [36]. Since this momentum exchange is extremely small, only in 1901 Lebedev [37], Nichols and Hull [38] provided a first experimental evidence of the radiation pressure due to an arc or electric lamps on a mirror fixed on a torsion balance. Other experiments were carried out during the next decades but, because of the non coherent nature of the light sources, the results were small and hard to be detected. Only from the 1960s, radiation pressure and its applicability have been better understood thanks to the invention and availability of laser sources [39] that, compared to standard lamps, have increased drastically the intensity of the electromagnetic fields provided. In the early 1970s, Arthur Ashkin, at Bell laboratories, while trying to reproduce the effects of the solar wind, demonstrated that the motion of microscopic particles [40] and neutral atoms [41] could be altered by laser-induced optical forces. In particular, he found out an unexpected effect: the micro-spherical particles suspended in water were attracted perpendicular to the propagation axis and pushed in the propagation direction [40]. This attraction is due to the gradient force perpendicular to the propagation axis and it is caused by the focusing of the laser beam [42]. After the discovery of the optical gradient force component, Ashkin built a first optical trap made of two focused counter-propagating laser beams in order to balance the detrimental effects of radiation pressure and get a stable optical trap [42]. This kind of trap is a static trap since the two lasers fix, on average, the particle's position along the propagation axis, unless the power of one of the two beams is changed. In this situation the particle can shift along the propagation direction. Soon after, in 1971 Ashkin demonstrated the first levitation trap. In this case the radiation pressure is balanced by gravity and the gradient force keeps the sample trapped [43]. The restriction of this method is that the maximum radiation pressure applicable is equal to the gravity. This kind of trap is generally not very stable and the sample can be moved only along the propagation direction. A real breakthrough occurred in 1986, when all the problems encountered before were solved by using a highly focused laser beam [44]. Ashkin and his collaborators demonstrated that by using a high numerical aperture objective the focal spot is so tight that it is possible to obtain a gradient force also along the propagation direction. This force is directed towards the focal spot and can be used to trap and manipulate dielectric micro-spheres and atoms, this technique is called Optical Tweezers (OT) [15]. A scheme of a typical OT setup in shown in Fig. 2.1 The applications of optical forces and optical tweezers in atomic physics with the development of techniques for atom trapping and cooling lead to the Nobel prize in Physics in 1997 for Steven Chu, Claude Cohen-Tannoudji and William D. Phillips [7, 15, 42]. Since its first demonstration, optical tweezers are commonly used tools for the manipulation of micro- [45, 46] and nanostructures [47–49] and as a force transducer with resolution at the femtonewton [50]. As shown in Fig. 2.2, optical tweezers find applications in many fields of physics, biology, chemistry and material sciences. They are useful tools to sort and organize cells, control bacterial motion, measure linear and torsional forces, alter biological structures via modification of cellular membranes, cellular fusion, or the interaction between red blood cells and viruses [51-54] with the possibility to apply and measure forces with femtonewton sensitivity on micro- and nanometer-sized particles [50, 55–57].



Figure 2.1: Sketch of a typical experimental OT setup. Trapping is achieved by a tightly focused laser beam using a high-numerical-aperture objective lens (OBJ), which is also used to image the sample on a camera. The beam produced by the laser source (LS) is enlarged through a telescope lens system (lenses L1 and L2) to overfill the back aperture of the objective. A dichroic mirror (DM1) is used to reflect the laser light to the objective and to transmit the visible light to the camera. The sample is held and moved by a sample holder (SH). Inset (a) represents the real image of a SiNW optically trapped and aligned along the propagating axis of the laser beam [57],[58]. When the laser is switched off the SiNW performs Brownian motion and its alignment is randomized. Scattered and transmitted lights are collected and overlapped by a condenser lens (C) and projected on a QPD by a second dichroic mirror (DM2) and a lens (L4). Signals from the QPD are analyzed by a PC in order to calculate the calibration factors and to reconstruct the 3D Brownian motion of the trapped sample. From Ref. [7].



Figure 2.2: Optical trapping of particles in different regimes. Optical tweezers are able to confine atoms, viruses, small nanostructures, for which a dipole approximation can often give a reasonable account of optical forces (Rayleigh regime). In the opposite size regime optical trapping of bacteria, algae, cells, and microparticles can be studied using geometrical optics. The intermediate mesoscale is instead the most interesting and complex and a full electromagnetic theory must be used. From Ref. [47]

2.2 Introduction to the optical trapping theory

Optical trapping of particles is a consequence of the radiation force that stems from the conservation of electromagnetic momentum upon scattering [7, 15]. The theoretical understanding of this physical process can been investigated through the use of suitable approximations that depend on the size of the particle x, as discussed in the Sect. 1.2. For homogeneous spherical particles, accurate evaluation of optical forces can be obtained by Mie theory [3] (Subsect. 1.5.1). Generally, when we study the optical trapping involving non-spherical or non-homogeneous particles, we must use a full electromagnetic theory based on the Maxwell's equations [4] and the integration of the Maxwell stress tensor [15]. Since such calculations can be extremely complex, the Transition Matrix approach, treated in the Sect. 1.4, appears quite suitable [1, 2].

2.2.1 Maxwell stress tensor, radiation force, and torque

As mentioned, particles illuminated by a radiation field experience a radiation force \mathbf{F}_{rad} and torque \mathbf{T}_{rad} , which contribute to determining their dynamical behaviour. Since the interaction between radiation and matter is regulated by conservation laws, it is possible to derive \mathbf{F}_{rad} and \mathbf{T}_{rad} using the conservation of linear and angular momentum. So, the time-averaging optical force exerted by a monochromatic light on a particle is given by [2, 59–61]:

$$\mathbf{F}_{\rm rad} = \oint_{S} \hat{\mathbf{n}} \cdot \langle \mathsf{T}_{\rm M} \rangle \, \mathrm{d}S \tag{2.1}$$

where the integration is carried out over the surface S surrounding the scattering particle, $\hat{\mathbf{n}}$ is the outward normal unit vector, and $\langle \mathsf{T}_{\mathrm{M}} \rangle$ is the averaged Maxwell stress tensor which describes the mechanical interaction of light with matter. The general expression of the Maxwell stress tensor in a medium in the Minkowski form is:

$$\mathsf{T}_{\mathrm{M}} = \boldsymbol{\mathcal{E}} \otimes \boldsymbol{\mathcal{D}} + \boldsymbol{\mathcal{H}} \otimes \boldsymbol{\mathcal{B}} - \frac{1}{2} \left(\boldsymbol{\mathcal{E}} \cdot \boldsymbol{\mathcal{D}} + \boldsymbol{\mathcal{H}} \cdot \boldsymbol{\mathcal{B}} \right) \mathsf{I} . \tag{2.2}$$

where \mathcal{E} is the electric field, \mathcal{D} is the electric displacement, \mathcal{H} is the magnetic field, \mathcal{B} is the magnetic induction, \otimes represents the dyadic product and \mathbf{I} is the dyadic unit. Since we consider always harmonic fields, at angular frequency ω in a homogeneous, linear, and non-dispersive medium, we can simplify this expression by using the complex amplitudes of the fields, $\mathbf{E} = \mathbf{E}(\mathbf{r})$ and $\mathbf{B} = \mathbf{B}(\mathbf{r})$, so that, *i.e.*, the real physical electric field is written as

 $\mathcal{E}(\mathbf{r},t) = \Re \{ \mathbf{E}(\mathbf{r})e^{-i\omega t} \}$ and in the same manner the real physical magnetic field [15]. Thus, the averaged Maxwell stress tensor simplifies as:

$$\langle \mathsf{T}_{\mathrm{M}} \rangle = \frac{\varepsilon_{\mathrm{m}}}{2} \Re \left\{ \mathbf{E} \otimes \mathbf{E}^{*} + \frac{c^{2}}{n_{\mathrm{m}}^{2}} \mathbf{B} \otimes \mathbf{B}^{*} - \frac{1}{2} \left(|\mathbf{E}|^{2} + \frac{c^{2}}{n_{\mathrm{m}}^{2}} |\mathbf{B}|^{2} \right) \mathsf{I} \right\}$$
(2.3)

with $\varepsilon_{\rm m}$ dielectric permittivity of the medium, the fields $\mathbf{E} = \mathbf{E}_{\rm i} + \mathbf{E}_{\rm s}$ and $\mathbf{B} = \mathbf{B}_{\rm i} + \mathbf{B}_{\rm s}$ are the total electric and magnetic fields, superposition of the incident $(\mathbf{E}_{\rm i}, \mathbf{B}_{\rm i})$ and scattered $(\mathbf{E}_{\rm s}, \mathbf{B}_{\rm s})$ fields.

In a similar way, considering the conservation of the angular momentum, the time averaged radiation torque is expressed as [2]:

$$\mathbf{\Gamma}_{\rm rad} = -\oint_{S} (\langle \mathsf{T}_{\rm M} \rangle \times \mathbf{r}) \cdot \hat{\mathbf{n}} \, \mathrm{d}S \tag{2.4}$$

where \mathbf{r} is the vector position.

2.2.2 Angular momentum of light

In the previous part of this chapter we mentioned the conservation of angular momentum which leads to the definition of torque, given by Eq. 2.4. The total angular momentum \mathbf{J} can be split in two contributions [62], in which \mathbf{J}_{mech} takes into account the mechanical effects on the scatterer while $\mathbf{J}_{\text{field}}$ is the angular momentum of the field, that is [63]:

$$\mathbf{J}_{\text{field}} = \frac{n_{\text{m}}^2}{c^2} \int_V \mathbf{r} \times \boldsymbol{\mathcal{S}} \, \mathrm{d}V, \qquad (2.5)$$

where the term on the right-hand-side represents the flux of angular momentum that enters the surface S and $\frac{n_m^2}{c^2} S = \varepsilon_m \mathcal{E} \times \mathcal{B}$ is the Poynting's vector that is the energy flux density associated with a propagating wave. Using the Helmholtz decomposition of the electromagnetic fields and introducing respectively the scalar and the vector potentials ϕ and \mathcal{A} in the Coulomb gauge, the Eq. 2.5 can be rewritten as:

$$\mathbf{J}_{\text{field}} = \varepsilon_{\text{m}} \int_{V} \mathbf{r} \times \left[\boldsymbol{\mathcal{E}}_{\parallel} \times (\nabla \times \boldsymbol{\mathcal{A}}) + \boldsymbol{\mathcal{E}}_{\perp} \times (\nabla \times \boldsymbol{\mathcal{A}}) \right] \, \mathrm{d}V, \qquad (2.6)$$

in which \mathcal{E}_{\parallel} is the irrotational component and \mathcal{E}_{\perp} is the rotational one of the electric field. The first term on the right-hand-side of Eq. 2.6 is related to the canonical angular momentum and is associated with the source term ρ [15]:

$$\mathbf{J}_{\text{field,canonical}} = \varepsilon_{\text{m}} \int_{V} \mathbf{r} \times \left[\boldsymbol{\mathcal{E}}_{\parallel} \times (\nabla \times \boldsymbol{\mathcal{A}}) \right] \, \mathrm{d}V = \rho \int_{V} \mathbf{r} \times \boldsymbol{\mathcal{A}} \, \mathrm{d}V.$$
(2.7)

Therefore, in a source-free space or in the case of radiating fields rapidly vanishing at infinity, the canonical part of the electromagnetic angular momentum is negligible. The second term on the right-had-side of Eq. 2.6 is related to the rotational fields and can be further split into orbital and spin components:

$$\varepsilon_{\rm m} \int_{V} \mathbf{r} \times [\boldsymbol{\mathcal{E}}_{\perp} \times (\nabla \times \boldsymbol{\mathcal{A}})] \, \mathrm{d}V = \varepsilon_{\rm m} \int_{V} \left[\sum_{j=x,y,z} \boldsymbol{\mathcal{E}}_{\perp,j} (\mathbf{r} \times \nabla) \boldsymbol{\mathcal{A}}_{j} \right] - \mathbf{r} \times (\boldsymbol{\mathcal{E}}_{\perp} \cdot \nabla) \boldsymbol{\mathcal{A}} \, \mathrm{d}V$$
(2.8)

After other manipulations, the following expressions for the orbital, \mathbf{L} , and spin, \mathbf{s} , components of the light angular momentum can be obtained [62]:

$$\mathbf{L} = \varepsilon_{\mathrm{m}} \int_{V} \sum_{j=x,y,z} \mathcal{E}_{\perp,j}(\mathbf{r} \times \nabla) \mathcal{A}_{j} \, \mathrm{d}V \,, \qquad (2.9)$$

$$\mathbf{s} = \varepsilon_{\mathrm{m}} \int_{V} \boldsymbol{\mathcal{E}}_{\perp} \times \boldsymbol{\mathcal{A}} \, \mathrm{d}V \,. \tag{2.10}$$

For a monochromatic transverse electromagnetic field, we can use the complex amplitudes of the field to express the electric field in terms of the vector potential, *i.e.*, $\mathbf{E} = \mathbf{E}_{\perp} = i\omega \mathbf{A}$. Thus, we can write the orbital, $\langle \mathbf{L}_{d} \rangle$, and spin, $\langle \mathbf{s}_{d} \rangle$, averaged angular momentum densities in a form that is useful in many practical cases:

$$\langle \mathbf{L}_{\mathrm{d}} \rangle = i \frac{\varepsilon_{\mathrm{m}}}{2\omega} \sum_{j=x,y,z} E_j(\mathbf{r} \times \nabla) E_j^*$$
 (2.11)

$$\langle \mathbf{s}_{\mathrm{d}} \rangle = i \frac{\varepsilon_{\mathrm{m}}}{2\omega} \mathbf{E} \times \mathbf{E}^* .$$
 (2.12)

2.3 Ray optics regime $(x \gg 1)$

We start by considering a particle with refractive index $n_{\rm p}$, immersed in a medium with refractive index $n_{\rm m} < n_{\rm p}$. When we can use the geometrical optics, also known as the ray optics regime, the optical field is described by considering it as a collection of N light rays and employing the tools of geometrical optics [15]. Each ray carries with it an portion of the incident power P_i so that the total power is $P = N \sum_i P_i$ and a linear momentum per second $n_m P_i/c$. When a ray impinges on a surface with an incident angle θ_i , it will be partly reflected and partly transmitted with a transmitted angle θ_t , according to the well-known Snell's law [64]. As a consequence of energy conservation, the power is split between the reflected and transmitted part of the ray following Fresnel coefficients [65]. Moreover, at each scattering event, the ray changes its direction and hence its momentum causing a reaction force on the center-of-mass of the particle. Thus, the force associated with the reflection and transmission of a ray \mathbf{r}_i and power P_i in the direction $\hat{\mathbf{r}}_i$ is written as:

$$\mathbf{F}_{\mathrm{ray},0} = \frac{n_{\mathrm{m}} P_{\mathrm{i}}}{c} \hat{\mathbf{r}}_{\mathrm{i}} - \frac{n_{\mathrm{m}} P_{\mathrm{r}}}{c} \hat{\mathbf{r}}_{\mathrm{r}} - \frac{n_{\mathrm{p}} P_{\mathrm{t}}}{c} \hat{\mathbf{r}}_{\mathrm{t}}.$$
 (2.13)

where $\hat{\mathbf{r}}_{r}$, P_{r} and $\hat{\mathbf{r}}_{t}$, P_{t} are the unit vector and power in the direction of the reflected and transmitted rays, respectively. Most of the power carried by the incident ray is delivered to the transmitted ray that travels inside the particle until it impinges on the opposite surface. Here, it will be reflected and transmitted again and a large portion of the power will be transmitted outside the sphere. The process will continue until all light escapes from the sphere. By considering these reflection and refraction events the optical force can be calculated directly as [65, 66]:

$$\mathbf{F}_{\rm ray} = \frac{n_{\rm m} P_{\rm i}}{c} \hat{\mathbf{r}}_{\rm i} - \frac{n_{\rm m} P_{\rm r}}{c} \hat{\mathbf{r}}_{\rm r}^{(0)} - \sum_{j=1}^{+\infty} \frac{n_{\rm p} P_{\rm t}^{(j)}}{c} \hat{\mathbf{r}}_{\rm t}^{(j)}.$$
 (2.14)

The optical force \mathbf{F}_{ray} has components only in the incidence plane and can be split in two perpendicular components. The component in the direction of the incoming ray $\hat{\mathbf{r}}_i$ represents the scattering force $\mathbf{F}_{ray,scat}$, that pushes the particle away from the center of the trap. The component perpendicular to the incoming ray is the gradient force $\mathbf{F}_{ray,grad}$, that pulls the particle towards the optical axis when $n_m < n_p$. Instead, if $n_m > n_p$ the particle is pushed away from the high intensity focal region.

$$\mathbf{F}_{ray} = \mathbf{F}_{ray,scat} + \mathbf{F}_{ray,grad} \tag{2.15}$$

It is often useful to define the dimensionless quantities (trapping efficiencies) obtained dividing the force components $F_{\rm ray,scat}$ and $F_{\rm ray,grad}$ by $n_{\rm m}P_{\rm i}/c$, that quantify how efficiently the momentum is transferred from the ray to the particle.

For a circularly polarized ray on a sphere Ashkin derived the following theoretical expression for the scattering and gradient efficiencies [66]:

$$Q_{\text{scat}} = 1 + R \cos 2\theta_{\text{i}} - T^{2} \frac{\cos(2\theta_{\text{i}} - 2\theta_{\text{r}}) + R \cos 2\theta_{\text{i}}}{1 + R^{2} + 2R \cos 2\theta_{\text{r}}},$$

$$Q_{\text{grad}} = R \sin 2\theta_{\text{i}} - T^{2} \frac{\sin(2\theta_{\text{i}} - 2\theta_{\text{r}}) + R \sin 2\theta_{\text{i}}}{1 + R^{2} + 2R \cos 2\theta_{\text{r}}}$$
(2.16)

where R and T are the (intensity) Fresnel reflection and transmission coefficients and θ_i and θ_r are the incidence and transmission angle relative to the scattering of the incident beam.

In general, if more than one ray interacts with a particle, the total force is given by the sum of the forces generated by the reflection and refraction of each ray. To model an optical trap we need to model a highly focused laser beam that means a set of many rays that converge at a very large angle in the focal spot, and hence sum up all contributions from each ray forming the beam. This means that the total force acting on the particle is the sum of all the contributions from each ray forming the beam. Considering Eq. 2.13, the force acting on the centre of mass of the sphere is

$$\mathbf{F}_{\rm GO} = \sum_{m} \mathbf{F}_{\rm ray}^{(m)} = \sum_{m} \left[\frac{n_{\rm m} P_{\rm i}^{(m)}}{c} \hat{\mathbf{r}}_{\rm i}^{(m)} - \frac{n_{\rm m} P_{\rm r}^{(m)}}{c} \hat{\mathbf{r}}_{\rm r,0}^{(m)} - \sum_{j=1}^{+\infty} \frac{n_{\rm p} P_{{\rm t},j}^{(m)}}{c} \hat{\mathbf{r}}_{{\rm t},j}^{(m)} \right].$$
(2.17)

For a single-beam optical tweezers, the focused rays will generate a restoring force proportional to the particle's displacement from an equilibrium point, that is for small displacements optical trapping can be modeled as an harmonic response. Due to the scattering force, the particle's is displaced from the nominal focus to an equilibrium position $\mathbf{C}_{eq} = [x_{eq}, y_{eq}, z_{eq}]$. Thus, for small displacements optical trapping forces are modeled as:

$$F_x \approx -\kappa_x (x - x_{eq})$$

$$F_y \approx -\kappa_y (y - y_{eq})$$

$$F_z \approx -\kappa_z (z - z_{eq})$$
(2.18)

where κ_x , κ_y and κ_z are the trap stiffnesses or spring constants of the trap. Calculating or measuring the spring constants, we can obtain a calibration of the optical trap.

The geometrical optics approach can be also used when we deal with non spherical particles, such as cylindrical objects. The basic interaction of the ray with this kind of particles is the same introduced in Eq. 2.13 but now two new aspects must be considered: induced torque and transverse radiation force. Induced torque is calculated from the difference of the angular momentum associated with the incoming and outgoing ray with respect to a pole [15]. Also in this case, the total torque on the object can be obtained as the sum of the torque produced by each ray, $\Gamma = \sum_m \Gamma_{ray}^{(m)}$. For example, the effect of the torque due to the rays is to align a cylindrical particle along the optical axis. The second aspect, the transverse radiation force, yields the optical lift effect [67]. This component arises from the anisotropic shape of non-spherical particles and generates a motion transversely to the incident light propagation direction. We can note that the accuracy of ray optics approximation increases with the size of the particle, whereas exact electromagnetic theories become unpractical due to the increasing computational complexity. Thus, ray optics has not only a pedagogical value but represents a key technique for modeling optical trapping of large particles [68].

2.4 Focusing

The easiest and most used configuration of optical tweezers is the singlebeam optical trap as devised by Ashkin in 1986 [44]. This is obtained by a single highly-focused light beam. In fact, rays originating from diametrically opposite points of a high numerical aperture (NA) focusing lens produce in practise a set of rays that converge at very large angle. While the ray optics view is extremely useful for rapid calculations, it misses out many aspects of the focusing process that have a crucial importance when performing accurate modelling. Thus, here we give a brief description on the focusing of a paraxial optical beam by an ideal aplanatic optical lens [69, 70]. An exemplary figure is shown in Fig. 2.3.

Using the Abbe's sine condition, the deflection angle θ at position **R** (the intersection point of a ray with the aplanatic lens p_2) is:

$$\theta = \arcsin\left(\frac{\rho}{f}\right) = \arcsin\left(\rho \frac{\mathrm{NA}}{n_{\mathrm{m}}R}\right)$$
(2.19)

where ρ is the radial coordinate of the incident wave, R is the radius of the iris, $n_{\rm m}$ is the index of refraction for the medium beyond p₂ and NA is the numerical aperture of the objective lens:

$$NA = n_{\rm m} \sin(\theta_{\rm max}) = n_{\rm m} \frac{R}{f}$$
(2.20)

with θ_{max} angle over which the rays are focused and which determines the trapping characteristics of the focus. The complex focused field $\mathbf{E}_{\mathbf{f}}(x, y, z)$


Figure 2.3: An optical beam crosses an aperture stop, or iris, with radius R and then propagates toward the principal plane p_1 of the lens and is transferred to the principal plane p_2 , which is a spherical surface with centre at the focal point **O** and with radius equal to the focal length f. The diffraction that occurs inside the objective is modelled by propagating the electromagnetic wave from the aperture stop to the principal plane p_1 . The aperture stop is often placed in the back focal plane, *i.e.*, at a distance f from p_1 , which results in a telecentric imaging system. At p_2 , the beam is refracted and focused towards **O**. Taken from Ref. [15].

near ${\bf O}$ is then:

$$\mathbf{E}_{\mathrm{f}}(x,y,z) = \frac{ik_{\mathrm{m}}fe^{-ik_{\mathrm{m}}f}}{2\pi} \int_{0}^{\theta_{\mathrm{max}}} \sin\theta \int_{0}^{2\pi} \mathbf{E}_{\mathrm{ff},\mathrm{m}}(\theta,\varphi)e^{i[k_{\mathrm{m},x}x+k_{\mathrm{m},y}y]}e^{ik_{\mathrm{m},z}z} \,\mathrm{d}\varphi \,\mathrm{d}\theta \,.$$
(2.21)

This is the integral representation of the focused field [71] where the plane wave angular spectrum representation is used such as $\mathbf{E}_{\mathrm{ff},\mathrm{m}}(\theta,\varphi)$ is the far field.

2.5 Dipole approximation regime $(x \ll 1)$

When the particle size parameter is small, $x \ll 1$, optical trapping forces can be calculated exploiting a dipole approximation. When placed in an external electric field, the negative electron cloud surrounding the positive nucleus will be displaced, leading to a separation between the centre-of-mass of the positive and negative charge distributions. An induced dipole is thus generated that experiences electrostatic forces arising from its interaction with the inducing electric field. Consequently, an oscillating electromagnetic field, such as that of the laser beam used for an optical tweezers, induces an oscillating dipole, which also experiences forces arising from its interaction with the inducing electromagnetic field. Furthermore, an oscillating dipole radiates an electromagnetic field that can produce a mechanical effect on other induced dipoles leading to, in some cases, optical binding [15, 72].

This picture can be extended to a small particle, so that if the external field is not too large, the induced dipole moment, $\mathbf{p}(\mathbf{r}, t)$, can be expressed in terms of a linear polarisability:

$$\mathbf{p}(\mathbf{r},t) = \alpha_{\mathrm{p}} \mathbf{E}(\mathbf{r},t) \tag{2.22}$$

where $\alpha_{\rm p}$ is the complex polarisability of the particle with respect to the surrounding medium and it is given by [73]:

$$\alpha_{\rm p} = \alpha_0 \left(1 - i \frac{k_{\rm m}^3 \alpha_0}{6\pi\varepsilon_{\rm m}} \right)^{-1} \tag{2.23}$$

 α_0 being the static Clausius-Mossotti polarisability:

$$\alpha_0 = 3V\varepsilon_{\rm m} \left(\frac{\varepsilon_{\rm p} - \varepsilon_{\rm m}}{\varepsilon_{\rm p} + 2\varepsilon_{\rm m}}\right) \tag{2.24}$$

where V is the particle volume and $\varepsilon_{\rm p}$ dielectric permittivity of the particle. Therefore, for an electric dipole of polarisability $\alpha_{\rm p}$, we can write the cross-sections in accordance with Poynting's theorem [15]:

$$\sigma_{\text{ext,d}} = \frac{k_{\text{m}}}{\varepsilon_{\text{m}}} \Im\{\alpha_{\text{p}}\}$$
(2.25)

$$\sigma_{\rm scat,d} = \frac{k_{\rm m}^4}{6\pi\varepsilon_{\rm m}^2} |\alpha_{\rm p}|^2 \tag{2.26}$$

$$\sigma_{\rm abs,d} = \sigma_{\rm ext,d} - \sigma_{\rm scat,d} = \frac{k_{\rm m}}{\varepsilon_{\rm m}} \Im\{\alpha_{\rm p}\} - \frac{k_{\rm m}^4}{6\pi\varepsilon_{\rm m}^2} |\alpha_{\rm p}|^2 \qquad (2.27)$$

2.5.1 Optical forces

The time-averaged optical force experienced by a small particle when illuminated by time-varying electromagnetic field can be also expressed in terms of its polarisability [74]:

$$\langle \mathbf{F} \rangle_{\mathrm{DA}} = \frac{1}{2} \Re \left\{ \sum_{i} \alpha_{\mathrm{p}} E_{i} \nabla E_{i}^{*} \right\}.$$
 (2.28)

where E_i are the electric field components. Starting from this expression, one can explicitly write the optical force in terms of extinction cross-section and particle's polarisability [75–77]:

$$\langle \mathbf{F} \rangle_{\mathrm{DA}} = \frac{1}{4} \Re \left\{ \alpha_{\mathrm{p}} \right\} \nabla |\mathbf{E}|^{2} + \frac{n_{\mathrm{m}}}{c} \sigma_{\mathrm{ext}} \langle \mathbf{S} \rangle - \frac{1}{2} c n_{\mathrm{m}} \sigma_{\mathrm{ext}} \nabla \times \langle \mathbf{s} \rangle$$
(2.29)

where $\langle \mathbf{S} \rangle = \frac{1}{2} \Re \{ \mathbf{E} \times \mathbf{H}^* \}$ is the time-averaged Poynting vector of the incoming wave and $\langle \mathbf{s} \rangle = i \frac{\varepsilon_m}{2\omega} \mathbf{E} \times \mathbf{E}^*$ is the time-averaged spin angular momentum density [76, 77].

The first term in Eq. 2.29 represents the gradient force and is responsible for particle confinement in optical tweezers:

$$\mathbf{F}_{\mathrm{DA,grad}}(\mathbf{r}) = \frac{1}{2} \frac{n_{\mathrm{m}}}{c\varepsilon_{\mathrm{m}}} \Re \{\alpha_{\mathrm{p}}\} \nabla I(\mathbf{r})$$
(2.30)

where $I(\mathbf{r}) = \frac{1}{2}n_{\rm m}c|\mathbf{E}(\mathbf{r})|^2$ is the intensity of the electric field and $\mathbf{r}_{\rm p}$ is the position of the center of the dipole. The gradient force, arising from the potential energy of a dipole immersed in the electric field, is conservative and its work does not depend on the path taken. Particles with refractive index higher than that of the surrounding medium $(n_{\rm p} > n_{\rm m})$ have a positive $\Re \{\alpha_{\rm p}\}$, and will be attracted toward the high intensity region of the optical field [44]. Conversely, when $n_{\rm p} < n_{\rm m}$ the polarisability is negative and the particles are repelled by the high intensity region.

The second term in Eq. 2.29 is the scattering force:

$$\mathbf{F}_{\mathrm{DA,scat}}(\mathbf{r}) = \frac{n_{\mathrm{m}}}{c} \sigma_{\mathrm{ext}} \langle \mathbf{S} \rangle = \frac{n_{\mathrm{m}}}{c} \sigma_{\mathrm{ext}} I(\mathbf{r}).$$
(2.31)

This term is responsible for the radiation pressure and is non-conservative. We can note the dependence on the extinction cross-section because momentum transfer from the electromagnetic field to the particle is a result of both scattering and absorption processes. This force is directed along the propagation direction of the laser beam [40].

The last term in Eq. 2.29 is a spin-dependent force [76]:

$$\mathbf{F}_{\mathrm{DA,spin}}(\mathbf{r}) = -\frac{1}{2}cn_{\mathrm{m}}\sigma_{\mathrm{ext}}\nabla\times\langle\mathbf{s}\rangle.$$
(2.32)

This term is also non-conservative and dependent on the extinction crosssection. It can be generated by polarisation gradients in the electromagnetic



Figure 2.4: The intensity distribution of a Gaussian beam is (a) Gaussian in the transverse x, y plane and (b) cylindrically symmetric around the propagation z-axis. (c) As the beam propagates along the z-axis, its phase deviates (top) with respect to the one of a reference plane wave (bottom) leading to a phase shift of exactly π as z goes from $-\infty$ to $+\infty$ (Gouy phase shift). The shades of grey represent the phase of the beam from 0 to 2π . From Ref. [15].

field, but usually does not play a major role in optical trapping because it is zero or very small compared to the other contributions. However, it may play a more significant role when considering optical trapping with optical beams of higher order with inhomogeneous polarization patterns such as cylindrical vector beams [78, 79] or superpositions of circularly polarized Hermite-Gauss beams [80].

2.5.2 Gradient force with Gaussian beam

We now consider some examples that show the usefulness of the dipole approximation. Being a simple analytical approach it permits to obtain quantitative information on optical trapping (force components, trap stiffness) of small particles in many different beam configuration.

The first case is the single-beam trap configuration. We calculate the gradient force and the related trap stiffness of an incident laser beam with a typical Gaussian intensity profile which propagates along z axis [15] as in Fig. 2.4.

The complex electric field of a Gaussian beam $\mathbf{E}^{\mathrm{G}}(\rho, z)$ is [15]:

$$\mathbf{E}^{G}(\rho, z) = \mathbf{E}_{0} \frac{w_{0}}{w(z)} e^{-\frac{\rho^{2}}{w(z)^{2}}} e^{i\Phi(z)} .$$
(2.33)

where ρ is the radial coordinate, \mathbf{E}_0 is a vector in xy plan specifying the amplitude, phase and polarisation of the beam, w_0 is the waist radius, w(z) is the beam width such that $w(z) = w_0 \sqrt{1 + \frac{z^2}{z_0^2}}, \ \Phi(z) = k_{\rm m}z - \zeta(z) + \frac{z^2}{z_0^2}$

 $k_m \rho^2/2R(z)$, R(z) is the wave-front radius $R(z) = z \left(1 + \frac{z_0^2}{z^2}\right)$, $\zeta(z)$ is phase correction $\zeta(z) = \operatorname{atan}\left(\frac{z}{z_0}\right)$ and z_0 is the Rayleigh range which denotes the distance from the beam waist at z = 0 to where the beam width has increased by a factor $\sqrt{2}$, $z_0 = \frac{k_m w_0^2}{2}$. In the last equation the coordinate φ does not appear due to the cylindrical symmetry and the paraxial approximation has been used because the electromagnetic fields of a laser beam propagate mostly along a certain direction, which in this case along the z axis is assumed, spreading out only slowly in the transverse direction. Then the wave number along z axis can be approximate as follows:

$$k_{\mathrm{m},z} = k_{\mathrm{m}} \sqrt{1 - \frac{k_{\mathrm{m},x}^2 + k_{\mathrm{m},y}^2}{k_{\mathrm{m}}^2}} \approx k_{\mathrm{m}} - \frac{k_{\mathrm{m},x}^2 + k_{\mathrm{m},y}^2}{2k_{\mathrm{m}}} .$$
(2.34)

Thus, we can write the expression for the intensity of the beam:

$$I^{G}(\rho, z) = \frac{1}{2} c n_{m} |\mathbf{E}^{G}(\rho, z)|^{2}$$

$$= \frac{1}{2} c n_{m} |\mathbf{E}_{0}|^{2} \frac{w_{0}^{2}}{w(z)^{2}} e^{-\frac{2\rho^{2}}{w(z)^{2}}}$$

$$= I_{0} \frac{w_{0}^{2}}{w(z)^{2}} e^{-\frac{2\rho^{2}}{w(z)^{2}}}$$
(2.35)

where $I_0 = \frac{1}{2} c n_{\rm m} |\mathbf{E}_0|^2$ is the maximum intensity at the center of the beam.

In the transverse plane (z = 0), for small displacements from axis, we can approximate the intensity profile:

$$I^{\rm G}(\rho) \approx I_0 \left(1 - 2\frac{\rho^2}{w_0^2} \right)$$
 (2.36)

so that the radial component of the gradient force (Eq. 2.30) can be approximated by an elastic restoring force proportional and opposite to the displacement from the origin:

$$F_{\rm DA,grad,\rho}^{\rm G} = -\kappa_{\rho}^{\rm G}\rho \tag{2.37}$$

in which the trap stiffness is:

$$\kappa_{\rho}^{\rm G} = 2 \frac{\Re \left\{ \alpha_{\rm p} \right\}}{c \varepsilon_0 n_{\rm m}} \frac{I_0}{w_0^2}.$$
(2.38)

Similarly, we can calculate the force along z axis ($\rho = 0$). Then for small displacements in the transverse plane:

$$I^{\rm G}(z) \approx I_0 \left(1 - \frac{z^2}{z_0^2}\right)$$
 (2.39)

so that the z component of the gradient force (Eq. 2.30)

$$F_{\mathrm{DA,grad},z}^{\mathrm{G}} = -\kappa_{z}^{\mathrm{G}}z \tag{2.40}$$

has spring constant:

$$\kappa_z^{\rm G} = \frac{\Re \left\{ \alpha_{\rm p} \right\}}{c \varepsilon_0 n_{\rm m}} \frac{I_0}{z_0^2}.$$
(2.41)

Eqs. 2.38, 2.41 reveals that the spring constants are proportional to the electric field intensity maximum and the real part of the polarisability. Furthermore, these trap stiffness are inversely proportional to the beam area so, as may be expected, tighter focusing leads to stronger confinement.

2.5.3 Gradient force with counterpropagating Gaussian beams

A second important example is the situation of two counterpropagating gaussian beams. This is not only one of the first configuration used by Ashkin in his pioneering work [42], but a configuration often used in vacuum for the optical trapping and laser cooling of atoms and particles [81]. In liquid, this permits the trapping and optical binding of spherical and non-spherical particles with a wide tunability [82, 83]. Recent experiments in optomechanics with levitated particles also exploit this counterpropagating configuration [47, 84, 85]. However, the most important feature of OT in counterpropagating configuration is that permits a systematic analysis without the scattering forces contribution which could push away the scatterer for a certain range in wavelength. The calculation of the gradient force follows the scheme of optical trap composed by two incident Gaussian beam which travel one opposite to the other along the z axis and the two waste coincide with the origin of the laboratory reference system [86]. Moreover, the polarization directions of the electro-magnetic field for the two beams lie on the xy plane and they are co-linear. In this manner, taking the expression of the Gaussian electric field for this counterpropagating configuration, the intensity of the laser beam, depending by radial ρ and axial z directions, assumes a stationary profile and [6, 87]:

$$I^{\text{c.p.}}(\rho, z) = 4I_0 \frac{w_0^2}{w(z)^2} e^{-\frac{2\rho^2}{w(z)^2}} \cos^2 \Phi(z).$$
(2.42)

In Eq. 2.42, the interference between the two Gaussian beams generates a standing wave with a modulation of intensity along the z-axis that results in a strong wavelength-dependent modulation of the axial optical force. Thus, using Eq. 2.29 (without the scattering part) and Eq. 2.42, we get the expression of the gradient force components [14, 86]:

$$\begin{split} \langle F \rangle_{\mathrm{DA},\rho}^{\mathrm{c.p.}}(\rho,z) &= -\frac{4\Re\left\{\alpha\right\}I_{0}w_{0}^{2}\rho e^{\frac{-2\rho^{2}}{w^{2}(z)}}}{c\varepsilon_{0}n_{\mathrm{m}}w^{4}(z)} \left(\cos^{2}\Phi(z) + \frac{z\sin2\Phi(z)}{k_{\mathrm{m}}w^{2}(z)}\right) \\ \langle F \rangle_{\mathrm{DA},z}^{\mathrm{c.p.}}(\rho,z) &= -\frac{4\Re\left\{\alpha\right\}I_{0}w_{0}^{2}e^{\frac{-2\rho^{2}}{w^{2}(z)}}}{c\varepsilon_{0}n_{\mathrm{m}}k_{\mathrm{m}}^{2}w^{2}(z)} \\ &\cdot \left[\left(1 - \frac{2\rho^{2}}{w^{2}(z)}\right)\frac{2z\cos^{2}\Phi(z)}{w_{0}^{2}w^{2}(z)} + \left(\frac{k_{\mathrm{m}}^{2}}{4} - \frac{1}{2w^{2}(z)} - \rho^{2}\frac{w^{2}(z) - 2w_{0}^{2}}{w_{0}^{2}w^{4}(z)}\right)k_{\mathrm{m}}\sin2\Phi(z)\right] \\ \end{split}$$

In the transverse plane (z = 0), for small displacements from axis:

$$I^{\text{c.p.}}(\rho) \approx 4I_0 \left(1 - 2\frac{\rho^2}{w_0^2}\right)$$
 (2.44)

so that the radial component of the gradient force of the Eq. 2.43 can be simplify:

$$F_{\mathrm{DA,grad},\rho}^{\mathrm{c.p.}}(\rho) = -\kappa_{\rho}^{\mathrm{c.p.}}\rho \qquad (2.45)$$

in which the trap stiffness is:

$$\kappa_{\rho}^{\text{c.p.}} = 8 \frac{\Re \left\{ \alpha_{\text{p}} \right\}}{c \varepsilon_0 n_{\text{m}}} \frac{I_0}{w_0^2}.$$
(2.46)

We note that this spring constant is four times greater than that associated with the Gaussian beam (Eq. 2.38), demonstrating that stationary setup traps better than one single laser beam. Similarly, we can calculate the force along z axis ($\rho = 0$). Then for small displacements in the transverse plane we have:

$$I^{\text{c.p.}}(z) \approx 4I_0 \left[1 - \left(2 - 2k_{\text{m}}z_0 + k_{\text{m}}^2 z_0^2 \right) \frac{z^2}{z_0^2} \right]$$
 (2.47)

so that the z component of the gradient force (Eq. 2.43)

$$F_{\mathrm{DA,grad},z}^{\mathrm{c.p.}}(z) = -\kappa_z^{\mathrm{c.p.}} z \tag{2.48}$$

with a spring constant:

$$\kappa_z^{\text{c.p.}} = 4 \frac{\Re \left\{ \alpha_p \right\}}{c\varepsilon_0 n_m} \left(2 - 2k_m z_0 + k_m^2 z_0^2 \right) \frac{I_0}{z_0^2}.$$
 (2.49)

This result is more complex than the trap stiffness for a single Gaussian beam (Eq. 2.41) and it depends also on the wavenumber $k_{\rm m}$. Indeed, the generation of a stationary wave yield a modulation of intensity along the *z*-axis that result in a strong wavelength-dependent modulation of the axial optical force.

2.6 Radiation force and torque in T-matrix formalism

In the final section we introduce the optical force and torque using the asymptotic properties of the vector Helmholtz harmonics and the T-matrix formalism. In fact, the expressions for the radiative force (Eq. 2.1) and torque (Eq. 2.4) can be significantly simplified in the far-field region $(r \to \infty)$ and using the T-matrix formalism. Here, the incident \mathbf{E}_{i} , scattered \mathbf{E}_{s} and internal \mathbf{E}_{p} fields are expanded in terms of vector spherical harmonics \mathbf{Z}_{lm}^{*} . Moreover the integration can be performed over a spherical surface of radius r. large enough so that only transverse fields are taken in the integration since vanishing terms at infinity are neglected in the integration. A basic requirement for the correct integration of the Eqs. 2.1, 2.4 is that the particle has to be contained inside the sphere and it is centered at the origin of the reference system. Due to the orthogonality between \mathbf{Z}_{lm}^* and the radial unit vector $\hat{\mathbf{r}}$, the integrals of the first and second term of the Eq. 2.3 are identically zero. Therefore the optical force is due only to the contribution of the integral of the third term of Maxwell stress tensor. For a non-magnetic medium the optical force is:

$$\mathbf{F}_{\rm rad} = -\frac{\varepsilon_{\rm m} r^2}{4} \int_{\Omega} \left[|\mathbf{E}_{\rm s}|^2 + \frac{c^2}{n_{\rm m}^2} |\mathbf{B}_{\rm s}|^2 + 2\Re \left\{ \mathbf{E}_{\rm i} \cdot \mathbf{E}_{\rm s}^* + \frac{c^2}{n_{\rm m}^2} \mathbf{B}_{\rm i} \cdot \mathbf{B}_{\rm s}^* \right\} \right] \hat{\mathbf{r}} \, \mathrm{d}\Omega$$
(2.50)

where the integration is now carried out over the full solid angle $\Omega = 4\pi$ that represents the full solid angle describing the spherical surface. In the same way also the torque can be integrated over a spherical surface of radius rwhich contains the particle. Here it is convenient to set the center-of-mass of the particle as origin of the reference system. Considering the Eq. 2.3, the integrals of the last two terms do not give any contribution to the torque, therefore it is given by:

$$\Gamma_{\rm rad} = -\frac{\varepsilon_{\rm m} r^3}{2} \Re \left\{ \int_{\Omega} \left[(\hat{\mathbf{r}} \cdot \mathbf{E}) (\mathbf{E}^* \times \hat{\mathbf{r}}) + \frac{c^2}{n_{\rm m}^2} (\hat{\mathbf{r}} \cdot \mathbf{B}) (\mathbf{B}^* \times \hat{\mathbf{r}}) \right] \, \mathrm{d}\Omega \right\}. \quad (2.51)$$

The latter two expressions are the starting point for the electromagnetic calculations of optical forces and torque in optical trapping. We recall that the key point is to solve the scattering problem by calculating the scattered fields and consequently the Maxwell stress tensor. However, the calculation of forces an torques in this regime is usually a complicate procedure [2]. Thus, various algorithms have been developed to handle this problem [88, 89]. Among the different approaches, a successful method is based on the calculation of the transition matrix [2]. This is particularly useful and computationally effective because it is possible to exploit the rotation and translation properties of the T-matrix to obtain at once optical forces and torques for different positions and orientations of the trapped particles [5, 60, 61, 90–95]. For these reasons, we have used this approach to compute the quantities of interest (forces, cross sections, maps) on the model particles studied in this thesis.

2.6.1 Optical forces due to a plane wave

An important case is the calculation of the optical force exerted by a linearly polarized plane wave on a particle. The incoming electric field associated to the linear polarized plane wave is expressed by Eq. 1.6. Starting from Eq. (2.50), after some substitutions and mathematical steps [15], the force originated from the scattering process of a linear polarized plane wave by a spherical homogeneous particle is written as [59]:

$$\mathbf{F}_{\rm rad} = \frac{n_{\rm m}}{c} I_0 \left[\sigma_{\rm scat} \mathbf{\hat{k}}_{\rm i} - \int_{\Omega} \frac{\mathrm{d}\sigma_{\rm scat}}{\mathrm{d}\Omega} \mathbf{\hat{r}} \,\mathrm{d}\Omega \right]$$
(2.52)

where $I_0 = \frac{n_{\rm mc}}{2} E_0^2$ is the intensity of the incident plane wave and $d\sigma_{\rm scat}/d\Omega = |\mathbf{f}(\hat{\mathbf{r}}, \hat{\mathbf{k}}_i)|^2$ is the differential scattering cross-section. The right side of this equation is composed by a first term that represents a force in $\hat{\mathbf{k}}_i$ direction and a second term which can present also a force component perpendicular to $\hat{\mathbf{k}}_i$ [59]. Therefore the component of the force along the propagation direction represents the radiation pressure:

$$\mathbf{F}_{\mathrm{rad}}^{\parallel} = \frac{n_{\mathrm{m}}}{c} I_0 \left[\sigma_{\mathrm{ext}} - g_{\mathrm{i}} \sigma_{\mathrm{scat}} \right] \hat{\mathbf{k}}_{\mathrm{i}}$$
(2.53)

while the transverse component of the force is:

$$\mathbf{F}_{\mathrm{rad}}^{\perp} = -\frac{n_{\mathrm{m}}}{c} I_0 \ \sigma_{\mathrm{scat}} \left[g_1 \hat{\mathbf{e}}_{\mathrm{i}} + g_2 (\hat{\mathbf{k}}_{\mathrm{i}} \times \hat{\mathbf{e}}_{\mathrm{i}}) \right]$$
(2.54)

where g_1 is the asymmetry parameter in the incoming wave direction (Eq. 1.11), while g_1 and g_2 are transverse asymmetry parameters (Eqs. 1.12 - 1.13). For a spherical particle only g_1 is different from zero, while for small dipolar particles all the parameters g_i , g_1 and g_2 are zero. The optical torque exerted by a linear polarized plane wave on a spherical particle is zero. However, torque is not zero in presence of elliptically polarized light and when the beads are made of an absorbing material [96].

On the other hand, when we deal with scatterers more complex than the single homogeneous sphere, such as radially symmetric non-homogeneous scatters or cluster, the expression of the radiation force (Eq. 2.50) should be rewritten in terms of T-matrix formalism. By substituting the expansions of the incident (Eq. 1.16) and scattered waves (Eq. 1.19) in terms of multipoles taken in the asymptotic limit $(r \to \infty)$ into Eq. 2.50 [15], the expression for the radiation force along the direction of a unit vector $\hat{\mathbf{u}}$, *i.e.*, $F_{\rm rad}(\hat{\mathbf{u}}) = \mathbf{F}_{\rm rad} \cdot \hat{\mathbf{u}}$ can be obtained, such as:

$$F_{\rm rad}(\hat{\mathbf{u}}) = -\frac{\varepsilon_{\rm m} E_0^2}{2k_{\rm m}^2} \Re \left\{ \sum_{plm} \sum_{p'l'm'} i^{l-l'} I_{lml'm'}^{(pp')}(\hat{\mathbf{u}}) \left[A_{{\rm s},lm}^{(p)*} A_{{\rm s},l'm'}^{(p')} + W_{{\rm i},lm}^{(p)*} A_{{\rm s},l'm'}^{(p')} \right] \right\},$$
(2.55)

where the amplitudes $W_{i,lm}^{(p)*}$ of the incident field and the amplitudes $A_{s,l'm'}^{(p)}$ of the scattered field are given by Eq. 1.27 or Eq. 1.42 in terms of the elements of the T-matrix, depending on if we deal with a single sphere or a cluster. Furthermore, in this equation,

$$I_{lml'm'}^{(pp')}(\hat{\mathbf{u}}) = \oint_{\Omega} (\hat{\mathbf{r}} \cdot \hat{\mathbf{u}}) \ i^{p-p'} \ \mathbf{Z}_{lm}^{(p)*}(\hat{\mathbf{r}}) \cdot \mathbf{Z}_{l'm'}^{(p')}(\hat{\mathbf{r}}) \,\mathrm{d}\Omega \ .$$
(2.56)

The integrals $I_{lml'm'}^{(pp')}(\hat{\mathbf{u}})$ can be expressed in closed form [5] as

$$I_{lml'm'}^{(pp')}(\hat{\mathbf{u}}) = \frac{4\pi}{3} \sum_{\mu=-1,0,1} Y_{1\mu}^*(\hat{\mathbf{u}}) K_{\mu;lml'm'}^{(pp')} , \qquad (2.57)$$

where the unit vectors are expressed in terms of spherical harmonics through:

$$K_{\mu;lml'm'}^{(pp')} = \oint_{\Omega} Y_{1\mu}(\hat{\mathbf{r}}) i^{p-p'} \mathbf{Z}_{lm}^{(p)*}(\hat{\mathbf{r}}) \cdot \mathbf{Z}_{l'm'}^{(p')}(\hat{\mathbf{r}}) d\Omega
 = \sqrt{\frac{3}{4\pi}} C_1(l',l;\mu,m-\mu) O_{ll'}^{(pp')},$$
(2.58)

in which in turn

$$O_{ll'}^{(pp')} = \begin{cases} \sqrt{\frac{(l-1)(l+1)}{l(2l+1)}} & l' = l-1 \text{ and } p = p' \\ -\frac{1}{\sqrt{l(l+1)}} & l' = l \text{ and } p \neq p' \\ -\sqrt{\frac{l(l+2)}{(l+1)(2l+1)}} & l' = l+1 \text{ and } p = p' \\ 0 & \text{otherwise} \end{cases}$$

and $C_1(l', l; \mu, m - \mu)$ are Clebsch-Gordan coefficients. The force expressed by Eq. 2.55 can be separated into two parts, *i.e.*,

$$F_{\rm rad}(\hat{\mathbf{u}}) = -F_{\rm scat}(\hat{\mathbf{u}}) + F_{\rm ext}(\hat{\mathbf{u}}) , \qquad (2.59)$$

where

$$F_{\rm scat}(\hat{\mathbf{u}}) = \frac{\varepsilon_{\rm m} E_0^2}{2k_{\rm m}^2} \Re \left\{ \sum_{plm} \sum_{p'l'm'} A_{{\rm s},lm}^{(p)*} A_{{\rm s},l'm'}^{(p')} i^{l-l'} I_{lml'm'}^{(pp')}(\hat{\mathbf{u}}) \right\}$$
(2.60)

and

$$F_{\text{ext}}(\hat{\mathbf{u}}) = -\frac{\varepsilon_{\text{m}} E_0^2}{2k_{\text{m}}^2} \Re \left\{ \sum_{plm} \sum_{p'l'm'} W_{\mathbf{i},lm}^{(p)*} A_{\mathbf{s},l'm'}^{(p')} i^{l-l'} I_{lml'm'}^{(pp')}(\hat{\mathbf{u}}) \right\} .$$
(2.61)

 $F_{\text{scat}}(\hat{\mathbf{u}})$ depends on the amplitudes $A_{\text{s},lm}^{(p)}$ of the scattered field only, while $F_{\text{ext}}(\hat{\mathbf{u}})$ depends both on $A_{\text{s},lm}^{(p)}$ and on the amplitudes $W_{\text{i},lm}^{(p)}$ of the incident field. This dependence is analogous to that on the scattering and extinction cross-sections for the force exerted by a plane wave.

2.6.2 Optical forces due to a focused beam

The most important case for the scope of this thesis is the computation of the radiation forces in a optical trap composed by focused laser beam with high-NA. In order to calculate the multipole amplitudes $\tilde{W}_{i,lm}^{(p)}$ of a focused beam, the expansion of the incoming beam into plane waves and its focusing can be exploited as described in the Sect. 2.4. The expansion of the focused beam around the focal point is given by the Eq. 2.21, *i.e.*,

$$\mathbf{E}_{\mathbf{f}}(x,y,z) = \frac{ik_{\mathbf{t}}fe^{-ik_{\mathbf{t}}f}}{2\pi} \int_{0}^{\theta_{\max}} \sin\theta \int_{0}^{2\pi} \mathbf{E}_{\mathbf{f},t}(\theta,\varphi)e^{i[k_{\mathbf{t},x}x+k_{\mathbf{t},y}y]}e^{ik_{\mathbf{t},z}z} \,\mathrm{d}\varphi \,\mathrm{d}\theta ,$$

where each plane wave transmitted through the objective lens $\mathbf{E}_{\mathrm{ff},t}(\theta,\varphi)$ can be expanded into multipoles according to the Eq. 1.16, *i.e.*,

$$\mathbf{E}_{\mathrm{ff},t}(\theta,\varphi) \equiv \mathbf{E}_{\mathrm{i}}(r,\hat{\mathbf{r}}) = E_0 \sum_{plm} W_{\mathrm{i},lm}^{(p)}(\hat{\mathbf{k}}_{\mathrm{i}},\hat{\mathbf{e}}_{\mathrm{i}}) \mathbf{J}_{lm}^{(p)}(k_{\mathrm{m}}r,\hat{\mathbf{r}}) ,$$

with the amplitudes given by Eqs. 1.18. Therefore, the amplitudes of the focused field are:

$$\tilde{W}_{i,lm}^{(p)}(\mathbf{P}) = \frac{ik_{\rm t}fe^{-ik_{\rm t}f}}{2\pi} \int_{0}^{\theta_{\rm max}} \sin\theta \int_{0}^{2\pi} E_{\rm i}(\theta,\varphi) \ W_{i,lm}^{(p)}(\hat{\mathbf{k}}_{\rm i},\hat{\mathbf{e}}_{\rm i}) \ e^{i\mathbf{k}_{\rm t}\cdot\mathbf{P}} \ \mathrm{d}\varphi \ \mathrm{d}\theta, \quad (2.62)$$

where the centre around which the expansion is performed is considered displaced by **P** with respect to the focal point **O** and the amplitudes $\mathcal{W}_{lm}^{(p)}(\mathbf{P})$ define the focal field and can be numerically calculated once the characteristics of the optical system are known. The radiation force are calculated through the knowledge of the scattered amplitudes $\tilde{A}_{s,lm}^{(p)}$, *e.g.*, by using the T-matrix (Eqs. (1.27, 1.42) and:

$$F_{\rm rad}(\hat{\mathbf{u}}) = -\frac{\varepsilon_{\rm m}}{2k_{\rm m}^2} \Re \left\{ \sum_{plm} \sum_{p'l'm'} i^{l-l'} I^{(pp')}_{lml'm'}(\hat{\mathbf{u}}) \left[\tilde{A}^{(p)*}_{{\rm s},lm} \tilde{A}^{(p')}_{{\rm s},l'm'} + \tilde{W}^{(p)*}_{{\rm i},lm} \tilde{A}^{(p')}_{{\rm s},l'm'} \right] \right\} .$$
(2.63)

In practice, the expression of the force in this equation is obtained from the correspondent one for the plane wave (Eq. 2.55) mutatis mutandis by changing $E_0 W_{i,lm}^{(p)} \to \tilde{W}_{i,lm}^{(p)}(\mathbf{P})$ and $E_i A_{s,lm}^{(p)} \to \tilde{A}_{s,lm}^{(p)}$ [5].

2.7 Size scaling in optical trapping of nanowires

The formalism described in this chapter has crucial applications for the calculation of optical forces on non-spherical particles. Here, we discuss its application for the study of the size scaling behaviour of optical trapping forces on nanowires. Size scaling is crucial in the study of nanoscience [97]. It characterises solid state systems for many applications in the most different

research fields [98]. Many particularly interesting properties of materials and interactions change dramatically with size [99]. Much progress has already been done in the synthesis, assembly, and fabrication of nanomaterials, and, equally important, toward a wide variety of technological applications [100]. The properties of materials with nanometric dimensions are significantly different from those of atoms or bulk materials, and the appropriate control of such properties have led to new science as well as new products, devices, and technologies [97]. The size scaling properties of optical forces help us to understand the important features of optical trapping in a wide size range and their comparison with experiments.

Nanowires have attracted considerable interest within the scientific community as an innovative material with applications in nanotechnology [101]. They are defined as structures with a high aspect ratio, being characterised by two spatial dimensions in the range of tens of nanometers and the third one on a much longer scale, typically micrometers. Moreover, due to their very large surface-to-volume ratio, nanowires can lead to strongly enhanced surface effects as compared to bulk materials. Their extreme geometry, combined, in the case of semi-conducting nanowires, with important intrinsic physical properties, leads to a wide range of novel physical applications. Because of their potential technological importance, the ability to manipulate, characterise and integrate nanowires on an individual basis is highly desirable, and optical trapping provides an ideal platform to achieve these aims [57, 102]. The possibility of investigating the structural [103], optical [104, 105], and thermal [106, 107] properties of individual nanowires in optical tweezers has been investigated in recent years. In this context, the role of their elongated shape on optical forces [90, 108], and dynamical stability [109] has been studied. From the experimental point of view, the dynamics of non-spherical particles in optical traps can be quite complex [56-58, 78, 88, 110, 110, 111]. For instance, elongated particles align, on average, with the optical (z) axis of the trap due to the optical torque and show small angular thermal fluctuations about their stability axis that can be measured together with optical aligning torques [56, 57]. Moreover, the occurrence of transverse optical forces [61, 88] yielding a translation-rotation coupling in non-spherical particles can result in a regular biased orbital motion that has been the subject of intense research [58, 112–115]. Thus, it is of crucial importance to have a better theoretical understanding of the optical forces and torques acting on these nanosystems in optical tweezers and their scaling with size.

Here, we consider model nanowires structured as linear chains of homogeneous non-absorbing latex nanospheres [90]. Despite their simplicity these model linear dielectric particles can grasp most of the optical trapping fea-



Figure 2.5: Trapping efficiencies (Q_x, Q_y, Q_z) for a nanowire composed by latex $(n_p = 1.57)$ spheres immersed in water $(n_m = 1.33)$ in the transverse, x - y, and longitudinal z, directions, as function of displacement in the same directions from the location of the paraxial focus. The considered linear clusters have half-length L/2: 50 nm (a), 300 nm (b), 600 nm (c), 1400 nm (d). The focal spot is obtained overfilling an objective lens with NA= 1.30.



Figure 2.6: Optical trap stiffnesses, κ_x (a), κ_y (b), κ_z (c), for a nanowire composed of latex ($n_p = 1.57$) spheres immersed in water ($n_m = 1.33$) in the transverse, x (a) - y (b), and longitudinal z (c), directions, as a function of the half-length L/2 of the linear cluster. The dimension of the considered aggregate spans in the interval [50 nm - 1500 nm] or, in other words, between number of spheres N [1 - 30].



Figure 2.7: Comparison of the optical trap stiffness, in logarithmic scale, κ_z , between a nanowire, composed by latex ($n_p = 1.57$) spheres, and a single sphere, immersed in water ($n_m = 1.33$), as a function of the dimension, d, corresponding to the half-length, L/2, for the nanowire and the radius, a, for the sphere. The dimension spans in the interval [50 nm - 1400 nm]. The two curves have in common a maximum at about 300 nm, corresponding to the maximum overlap of particle volume with the diffraction limited laser spot. The main difference is in the size scaling. In fact, for small size the stiffness of the sphere scales as d^3 while for the nanowire as d. For large size the stiffness of the sphere scales as d^{-1} , while for the nanowire decreases as d^{-3} .

tures related to the shape and geometry of the problem. Moreover, latex particles are a standard example of polymer particles that are commonly used in optical tweezers experiments [7, 15, 113]. We focus our attention only on nanowires aligned along the direction of propagation of the incident field (z) with the aim to investigate their length scale behaviour. This is justified by the fact that the optical torque aligns the nanowires along the axial propagation direction [90] as also experimentally observed on average [57, 102]. We model the trapping of a linear particle cluster by a laser beam with fixed wavelength $\lambda_0 = 830$ nm and numerical aperture NA = 1.3. We are interested, once again, in the calculation of the optical forces and in particular on how the trap stiffnesses scale as a function of the nanowire length, L. Each sphere composing the linear cluster has refractive index $n_{\rm p}(\lambda_0 = 830 \text{ nm}) = 1.57$ [116] and is immersed in water $(n_{\rm m} = 1.33)$. The radius of the single sphere is fixed at 50 nm because the idea is to model a linear structure that grows along the longitudinal direction, z, up to the microscale and remains fixed at the nanoscale in the xy transverse plane. The calculation is conducted for different lengths and, by adding the spheres, we work in a range in which the half-length of the cluster, L/2, spans from 50 nm (a single sphere) to 1500 nm (corresponding to 30 spheres in the cluster).

Given the complexity of the scatterer, which has only cylindrical symmetry with respect to the longitudinal direction of the incident beam, the T-matrix method performs very well thanks to its high precision and calculation speed. Thus, the incident and scattered fields are expanded in a series of vector spherical harmonics with amplitudes $W_{i,lm}^{(p)}$ and $\mathcal{A}_{s,lm}^{(p)}$, respectively given by Eq. 1.42. Then, as seen in Subsect. 1.5.2, the elements of the transition matrix $T_{l'm'lm}^{(p'p)}$ are calculated by the inversion of the matrix of the linear system, obtained by imposing the boundary conditions to the fields across the surface of the scatterer (see Eq. 1.42). The incident fields of the scattering problem are the focal fields calculated in the angular spectrum representation (Eq. 2.21). Finally, optical forces and trapping properties are obtained through the Maxwell tensor as described in the Subsect. 2.3. Convergence has been carefully checked and we adopted a truncation of the multipole expansion index $l_{\rm M} = 8$ for nanowire half-lengths L/2 between 50 nm and 500 nm (N = [1 - 10]) while $l_{\rm M} = 15$ for L/2 between 600 nm and 1.5 μ m (N = [12 - 30]).

2.7.1 Optical force components, trap stiffnesses, and size scaling

We have computed the (x, y, z) components of the optical force on the points of a grid employed with specified resolution. These components are calculated in a micron-sized range, $[-1.2 \ \mu m, 1.2 \ \mu m]$, around the paraxial nominal focus of the beam. So, we can plot the force as a function of particle displacement in each spatial direction, x, y, z. The trapping position of the particle in the longitudinal (z) direction is typically offset from the centre of the grid because of the 'pushing' effect of the optical scattering force. To calculate the force on the particle at the equilibrium position, the z (longitudinal) coordinate at which the axial force vanishes must first be found. The force plots in the transverse directions (x, y) can then be calculated. It is often convenient to calculate the dimensionless force (trapping) efficiencies along the three Cartesian directions, $Q_i = cF_i/n_{\rm m}P$ with i = x, y, z [7]. We present in Fig. 2.5 the results of the computation of the cartesian components of the optical force efficiencies for nanowires with half-length L/2 = 50 nm, 300 nm, 600 nm, 1400 nm]. Consequently, in Fig. 2.6, we have shown the optical trap stiffnesses, κ_x , κ_y , κ_z , as a function of the half-length of the linear cluster.

In Fig. 2.5 we investigate an interval of length spanning from the nanoto the microscale. We can immediately notice how in Figs. 2.5a, 2.5b, 2.5c, for short length of the scatterer, the graphs present the typical maximum and minimum that is maintained at the same positions for the transverse, x, y, directions, while it moves approximately with the edges of the nanowire for the axial, z, direction. Consequentially, the linear cluster is trapped at an equilibrium position in proximity to the nominal focus with a small dissipating scattering force. It is at the ends of the nanowire that a greater trap efficiency is developed in z and, given the particular cylindrical symmetry of the aggregate with respect to the direction of incidence of the beam, the equilibrium point is set at the midpoint of the nanowire. In this regard, heuristic considerations are reported by Simpson and Hanna [109], while experimental demonstrations were obtained by Irrera et al. [57]. As can be expected, when the length of the nanowire grows to such an extent that it is no longer completely contained in the high intensity region of the laser spot, the trap efficiencies collapse towards zero showing the flat pattern of Fig. 2.5d.

Now let us analyze the trap constant size scaling behaviour as the length of the nanowire varies. We plot these trends in Figs. 2.6a, 2.6b, 2.6c. We observe two different trends for κ_x , Fig. 2.6a, and κ_y , Fig. 2.6b, in the transverse plane, and κ_z , Fig. 2.6c, along the longitudinal direction. The transverse stiff-

nesses show a linear growth at short length that saturates when the length reaches the axial spot size optimizing the optomechanical interaction when the nanowire overlaps the laser spot high intensity region in the axial direction. In other words, the contribution to the transverse spring constants of the spheres composing the linear cluster outside the interaction region, determined by the diffraction limited laser spot, is negligible.

Instead, the size scaling behaviour of the axial stiffness, κ_z , can appear with a similar trend to the one for a single sphere. As we can see in Fig. 2.7, the two axial graphs have in common the occurrence of the maximum around 300 nm because, as in the case of a single sphere, the linear aggregate at this particular value has a length comparable with the axial size of the high intensity spot. On the other hand, the main difference between optical trapping of the two model particles lies in the size scaling. Here we directly compare the size scaling for the two model systems by plotting the axial spring constants for the nanowire as a function of the half-length, d = L/2, and for the sphere as a function of its radius, d = a. The stiffness of the nanowire grows linearly for short length, $\kappa_z \propto d$, while for the spheres we recall the cubic growth, $\kappa_z \propto d^3$. This is justified in dipole approximation by the one-dimensional geometry of the nanowire growth in contrast to the three-dimensional geometry of the sphere growth. For large length, instead, the axial spring constant decreases in a hyperbolic cubic manner, $\kappa_z \propto d^{-3}$, in contrast to the hyperbolic scaling for the sphere, $\kappa_z \propto d^{-1}$. This asymptotic scaling behaviour for nanowires is in agreement with analytical calculations provided by Simpson et al. [109], while experimental evidence of the size scaling in the optical trapping of silicon nanowires has been studied by Irrera et al. [57].

Chapter 3

Gain-assisted optomechanical position locking of metal/dielectric nanoshells in optical potentials

3.1 Introduction

As just discussed in the Chap. 2, OT [15, 117] are crucial tools for the manipulation and study of micro- and nanoscopic particles of different nature without mechanical contact [7, 118]. In recent years, a tremendous effort has been devoted to the optical trapping and optical manipulation of nanoparticles in liquid, air or vacuum [119, 120]. The difficulties in optical trapping nano-sized matter are mainly related to the fact that optical forces decrease with the particle volume for small particles [7], which yields trapping potentials lower than the energy of thermal fluctuations for reasonable incident laser [119]. Standard OT, *i.e.*, single-beam optical traps, are also affected by the unavoidable light scattering forces which tend to push the particle along the light propagation direction, and might have a particularly destabilizing effect for highly absorbing, resonant or plasmonic nanoparticles [119, 120]. Morphology [121, 122], material composition [123–125], material hybridization [126, 127], and resonant opto-plasmonic response [128–130] can increase optical trapping at the nanoscale. Scattering forces can be balanced at equilibrium in dual-beam optical traps based on the use of low numerical aperture lenses in a counterpropagating beam geometry [131]. For laser beams with the same polarization, a standing wave is formed with an intensity modulation along the beam axis that generate an optical potential with

many equilibrium position[132–136].

Among the various applications of optical forces at the nanoscale, the study of optical forces in optically trapped gain-enriched plasmonic nanostructures appears to be of particular interest. In fact, a plethora of remarkable phenomena occurs in these systems [137, 138] due to the resonant interplay between plasmonic structures and gain media (*e.g.* dye molecules or quantum dots). In particular, the coupling with a gain medium located in the core of a metallic nanoshell, when excited by means of an external pump, produces intense changes of the electromagnetic fields around the structure, thus producing novel features which can be useful for a variety of applications, such as photothermal therapy, enhanced spectroscopy, and spasing [139-141].

In this chapter, we present a study of the optical forces acting on a gainenriched silver nanoshell in the quasi-static limit. Specifically, we analyze the optomechanical response of this nanostructure in a counterpropagating Gaussian beam optical trap, where a systematic analysis can be performed without any detrimental effect of the scattering force component. In particular, we study the behaviour of the optical force constants as a function of wavelength and for different gain levels, which can be achieved by fixing the molecular density of the gain medium and varying the power of the external pump. We show that optical trapping strongly depends both on the wavelength and on the gain level. Moreover, we investigate the stable configurations and particle dynamics in the trap by means of Brownian dynamics simulation. Interesting localization effects appear for wavelengths red-detuned with respect to the gain-enhanced resonance, while for bluedetuned wavelengths, we observe particle channeling by the standing wave antinodes due to the reversal of the gradient force.

3.2 Metal/dielectric nanoshells

The system under study (Fig. 3.1) is a metal nanoshell embedding a gain enriched dielectric core, studied in the quasi-static limit and below the emission threshold [140] (*i.e.*, when the gain is not enough to completely overcome the metal losses). In this regime, the nanoshell geometry ensures that the plasmonic field can be described as dipolar without any approximation. Thus, we can use a steady state model for the particle polarizability α :

$$\alpha = \frac{(\varepsilon_{\rm sh} - \varepsilon_{\rm m})(\varepsilon_{\rm h} + 2\varepsilon_{\rm sh}) + f^3(\varepsilon_{\rm h} - \varepsilon_{\rm sh})(\varepsilon_{\rm m} + 2\varepsilon_{\rm sh})}{(\varepsilon_{\rm h} + 2\varepsilon_{\rm sh})(\varepsilon_{\rm sh} + 2\varepsilon_{\rm m}) + 2f^3(\varepsilon_{\rm h} - \varepsilon_{\rm sh})(\varepsilon_{\rm sh} - \varepsilon_{\rm m})},\tag{3.1}$$



Figure 3.1: In (a), a schematic of the system under study is represented. A core-shell particle is composed by an external silver shell and a silica core, doped with rhodamine dye molecules which act like gain material under the action of a pumped field (purple) considered in a counterpropagating configuration to balance radiation pressure. This system is placed in a doublebeam optical tweezers configured with co-linearly polarized counterpropagating Gaussian beams (green). The double beam configuration ensures the balance of scattering forces in the trap region. In (b), the scheme of the energy levels is presented. System pumping inverts the population \tilde{N} of the dye using a pump frequency higher than the frequency of the nanoshell plasmonic resonance. In this way, an emission of the dye at the wavelength of the silver plasmon resonance is induced. In (c), counter-propagating laser beam intensity is represented and projected on the yz plane. The probe wavelength is $\lambda = 531.9$ nm, its single-beam power is 50 mW, and NA= 0.6. Nanoparticle trajectories is plotted for G = 0 (blue), G = -0.132 (green), and G = -0.22 (magenta). Note how the highest gain (magenta) shows the tightest confinement. Brownian dynamics simulations are performed for all cases with a time step of $\Delta t = 10^{-7}$ s and a sampling time of $t_{\text{samp}} = 0.1$ s.

where $\varepsilon_{\rm sh}(\omega)$ is the complex steady state metal shell permittivity, $\varepsilon_{\rm h}(\omega)$ is the complex steady state permittivity of the gain enriched core, $\varepsilon_{\rm m}$ is a real number representing the permittivity of the external medium (solvent) hosting the nanoparticle, and $f = a_0/a_1$ is the ratio between the internal and the external radius of the nanoshell (Fig. 3.1a). In all of the calculations and simulations presented in this work the nanoparticle has an external radius of $a_1 = 20$ nm and a ratio between the internal and the external radius of f = 0.8 (meaning that the internal radius is $a_0 = 16$ nm).

For the metal permittivity we used a Drude-Lorentz model in which the losses due to the interband transitions $\delta \varepsilon_{\rm sh}(\omega)$ have been added heuristically by interpolating them from the Johnson and Christie data set [142]:

$$\varepsilon_{\rm sh}(\omega) = \varepsilon_{\infty} + \delta \varepsilon_{\rm sh}(\omega) - \frac{\omega_{\rm pl}^2}{\omega(\omega + 2i\Gamma)},\tag{3.2}$$

where ε_{∞} is a constant offset for the real part of the dielectric function, accounting for the high frequency contributions and the vacuum contribution, $\omega_{\rm pl}$ is the plasma frequency and Γ is the ionic collisions friction coefficient. In all of our calculations we will use $\varepsilon_{\infty} = 5.3$, $\hbar \omega_{\rm pl} = 9.6$ eV and $\hbar \Gamma = 0.0228$ eV which are compatible with the complex dielectric function of silver. As one can see in Fig. 3.1b, the gain elements (*e.g.*, molecule/quantum dot) have been chosen in order to resonate with the plasmon. Being below the emission threshold we can model that using a Lorentzian shape:

$$\varepsilon_{\rm h}(\omega) = \varepsilon_{\rm b} - \frac{G\Delta}{2(\omega - \omega_{\rm g}) + i\Delta},$$
(3.3)

where $\varepsilon_{\rm b}$ is the permittivity of the dielectric core in which the gain elements are embedded, $\omega_{\rm g}$ is the emission centerline of the gain elements, $\Delta = 2/\tau$ is the width of the Lorentzian shape where τ is the time constants associated with energy (spontaneous emission) relaxation processes of the gain element, and G is a dimensionless parameter equal to the maximum in absolute value of the imaginary part of $\varepsilon_{\rm h}(\omega)$ and measuring the amount of gain present in the system; it can be shown to be equal to[140]:

$$G = \Im[\varepsilon_{\rm h}(\omega_g)] = -\frac{n\mu^2\tau}{3\hbar\varepsilon_0}\tilde{N}, \qquad (3.4)$$

where *n* is the element density of the gain medium, μ is the amplitude of the transition dipole moment of the gain element and \tilde{N} represents the population inversion produced through the action of the external pump (*e.g.*, when $\tilde{N} = 0$ all of the gain molecules are in the ground state, when $\tilde{N} = 1$ they are in the excited state), \hbar is the reduced Planck constant and ε_0 is the vacuum permittivity. For a given gain molecule (where μ , ω_q and τ are set), the molecule density n describes the maximum possible value for G although, even once the molecule density n of the nanoparticle s core is fixed, G can be experimentally modulated by means of the external pump power, from zero (corresponding to $\tilde{N} = 0$ or pump off) to its maximum value (corresponding to N = 1 which is when the system is completely pumped). In all of our simulations, G ranged between zero and G = -0.22; this last minimum value, when using conservative estimates for the physical quantities in Eq. 3.4 (such as a transition dipole moment of $\mu = 10$ D and a relaxation time $\tau = 10^{-14}$ s) corresponds to a element density of n = 0.35 nm⁻³. It is worth noting that this estimation has been done for the *minimum value* we used for G (corresponding to the highest gain), while the effect on the optical forces begin to appear for much lower gain levels. This means that, even if we are neglecting efficiency-reducing quenching effects due to the proximity of the gain element to the metal in the nanoparticle, we are working in a realistic range of gain elements density.

3.3 Gain-assisted nonlinear optical trapping

We consider a silver nanoshell with an external radius of 20 nm and trapping light with a wavelength in the visible range. Therefore, the particle size is small enough that optical forces can be calculated within a dipole approximation [74–76] and expressed in terms of the linear effective complex polarizability, α , as already discussed in the Subsect. 2.5.1. It is worth noting that the trapping light provides also the probe field exciting the plasmonic resonance of the nanoshell, meaning that the dipole moment **p** of the nanoparticle can be written as the Eq. 2.23. Considering the double-beam OT in their standing wave configuration, we fix a low numerical aperture objective that at a typical value [129, 136] of NA= 0.6. In this manner, without the detrimental scattering contribution, a gradient force unset whose analytical expression is provided by the Eqs. 2.43.

We start our analysis of the nanoshell trapping behaviour by studying the spring constants associated to small displacements from the equilibrium trapping point, where the gradient force can be approximated by a harmonic force. In particular, the radial component trap stiffness is provided by the Eq. 2.46 while the axial component trap stiffness is given by the Eq. 2.49. In Fig. 3.2, we compare the particle polarizability for three different gain values (G = [0; -0.132; -0.22]) with the wavelength dependence of the trap constants normalized to the power, κ_{ρ}/P and κ_z/P . We can note that $\Im \{\alpha\}$ (red lines in Fig. 3.2a, 3.2d, 3.2g) increases for higher gain around the plasmon



Figure 3.2: Trap stiffnesses normalized to the beam power in counterpropagating configuration are shown for three different gains. (a-c) G = 0, (d-e) G = -0.132, and (g-i) G = -0.22. The first column represent the real (black line) and imaginary (red line) parts of the polarizability normalized by ε_0 . The second and third column represent the radial and axial trap stiffnesses, κ_{ρ} and κ_z , normalized by the optical power, respectively.



Figure 3.3: In (a), the maximum value of the trapping constants κ_{ρ}^{\max} (black line) and κ_z^{\max} (red line) in the counterpropagating configuration is plotted as a function of the gain in semi-logarithmic scale. κ_{ρ}^{\max} and κ_z^{\max} are intended as the highest positive values that κ_{ρ} and κ_z , respectively, take spanning through the wavelengths according to the trends pictured in Fig. 3.2. Intensity maps of the gradient force in yz plane are shown for G = 0 and G = -0.22, respectively in (b) and (c). The wavelength $\lambda = 531.9$ nm is chosen so that the stiffness obtained in the harmonic approximation assumes the highest values for each of the two gains. The power of the single Gaussian beam laser is 50 mW.

resonance, and its profile is mostly peaked and narrow. As the gain increases the enhanced emission of the pumped dye, which is suitably tuned to the frequency $\omega_{\rm pl}$, dominates the optical response in intensity with respect to that of the plasmon mode. Here we have that $\Im \{\alpha\} (G = -0.22) / \Im \{\alpha\} (G =$ $0 \simeq 50$ and the absorption appears increasingly spectrally confined around the resonance. On the other hand, $\Re\{\alpha\}$ changes sign in correspondence to the plasmonic resonance and for increasing gain shows a sharpening of the dispersive curve. Thus, from Eqs. 2.46, 2.49, trap stiffnesses have a similar trend as shown in Figs. 3.2b, 3.2c, 3.2e, 3.2f, 3.2h, 3.2i. In all cases we have that: i) for wavelengths lower than the resonance, the force constants are negative, optical forces are repulsive, and the nanoshell is pushed away from the high intensity region; ii) for wavelengths higher than the resonance, the force constants are positive, and the nanoshell is attracted to the high intensity region of the standing wave. As expected for a standing wave configuration, for a fixed gain, the axial force constants, κ_z , are greater, in modulo, than the transverse ones, κ_{ρ} .

Fig. 3.3a shows the maximum value of the trap stiffness as a function of gain. The onset of a nonlinear behaviour is clearly visible. Both κ_{ρ}^{\max} (black line) and κ_{z}^{\max} (red line) increase similarly in the logarithmic scale for increasing gain. While the spring constants give us an idea of the strength of gradient forces for small displacements around the equilibrium point,

to understand how the optical force is spatially distributed for the whole counter-propagating pattern, Eqs. 2.43 must be considered. This is shown in Figs. 3.3b, 3.3c that represent such distributions in yz plane, respectively at G = 0 and G = -0.22. The wavelength $\lambda = 531.9$ nm has been chosen so that the force constant calculated for a small displacements (harmonic) approximation is the highest for each of the two gains we have considered. The standing wave modulation of the light intensity (Fig. 3.3c) yields the modulated spatial profile of the force. Since we considered a wavelength on the red-side of the resonance, a nanoshell will be attracted towards the maximum intensity points, represented with the hottest colors in Fig. 3.3c and corresponding to the minima, blue regions, of the gradient force in Figs. 3.3b, 3.3c. We note how the gradient force for G = -0.22 (Fig. 3.3c) is strikingly higher than the force for G = 0 (Fig. 3.3b), (*i.e.* $\langle F \rangle_{\text{DA}}^{\text{max}}(G = -0.22) / \langle F \rangle_{\text{DA}}^{\text{max}}(G = 0) \sim 2 \cdot 10^3$, meaning that, by diverting energy from the pump, the interplay between the plasmonic resonance and the gain elements is able to produce a trapping force about three orders of magnitude stronger than the one found in the unpumped system.

3.4 Optomechanical gain-assisted position locking and channeling

To realistically simulate the dynamics of the silver-silica dye-doped nanoshell in the double-beam OT, we performed Brownian dynamics simulations[143] in water for different gain values. Interesting behaviours can arise for the motion of the nanoshells, including optomechanical position locking and channeling. The Brownian dynamics takes into account the thermal noise contribution, which tends to jiggle the nanoshell in the optical trap. The motion of the particle is, therefore, the result of the interplay between this random motion and the deterministic optical forces. The time scale on which the gradient force acts is given by the ratio $\tau_{\rm ot} = \gamma/\kappa$, where γ is the particle friction coefficient in the surrounding fluid, determined by Stokes' law [15, 143]. In our case, $\tau_{\rm ot}$ is always significantly greater than the momentum relaxation time $\tau_{\rm in} = m/\gamma$, so that inertial effects can be safely neglected. Thus, we can describe the particle Brownian dynamics through three independent overdamped Langevin equations [15, 144]:

$$\begin{cases} \frac{dx(t)}{dt} = \frac{F_{\mathrm{DA},x}(\mathbf{r},t)}{\gamma} + \sqrt{2D} W_x(t) \\ \frac{dy(t)}{dt} = \frac{F_{\mathrm{DA},y}(\mathbf{r},t)}{\gamma} + \sqrt{2D} W_y(t) \\ \frac{dz(t)}{dt} = \frac{F_{\mathrm{DA},z}(\mathbf{r},t)}{\gamma} + \sqrt{2D} W_z(t) \end{cases}$$
(3.5)

where $F_{\text{DA},i}$, with i = x, y, z, is the *i*-th Cartesian component of the time dependent optical force in dipole approximation, $D = k_{\text{B}}T/\gamma$ is the diffusion coefficient according to fluctuation-dissipation theorem with T the temperature and k_{B} the Boltzmann's constant, $W_x(t)$, $W_y(t)$ and $W_z(t)$ are the independent white noises related to the thermal fluctuations. Approximating this ordinary differential equations with finite difference equations [143, 145, 146], the corresponding system to Eq. 3.5 is:

$$\begin{cases} x_n = x_{n-1} - \frac{\langle F \rangle_{\mathrm{DA},x,n}}{\gamma} \Delta t + \sqrt{2D\Delta t} w_{x,n} \\ y_n = y_{n-1} - \frac{\langle F \rangle_{\mathrm{DA},y,n}}{\gamma} \Delta t + \sqrt{2D\Delta t} w_{y,n} \\ z_n = z_{n-1} - \frac{\langle F \rangle_{\mathrm{DA},z,n}}{\gamma} \Delta t + \sqrt{2D\Delta t} w_{z,n} \end{cases}$$
(3.6)

where $[x_n, y_n, z_n]$ represent the position of the particle at time t_n , $w_{n,i}$ are the independent Gaussian random numbers with zero mean and unitary variance that emulate the white noise, and n = 1, ..., N is an index where $N = 10^6$ is the number of computation steps. Moreover, $\langle F \rangle_{\text{DA},i,n}$ is the *i*-th component of the optical force in dipole approximation (Eqs. 2.43) calculated at the *n*-th position. In our simulation, we consider T = 300 K and different time steps, Δt , depending on the laser power, *e.g.*, in Fig. 3.1c a typical value of $\Delta t = 10^{-7}$ s has been used for P = 50 mW. The sampling time, t_{samp} , is chosen so that $t_{\text{samp}} \gg \tau_{\text{ot}}$. In fact, because of the trap asymmetry we have two timescales, $\tau_{\text{ot},z}$ and $\tau_{\text{ot},\rho}$, associated to the axial and radial stiffnesses, respectively, and, *e.g.*, at P = 50 mW we obtain $\tau_{\text{ott},z}(G = 0) \simeq 4 \cdot 10^{-6}$ s and $\tau_{\text{ot},\rho}(G = 0) \simeq 10^{-4}$ s. Thus, both timescales are much smaller than the sampling time of $t_{\text{samp}} = 0.1$ s.

With these tools at hand, it is possible to investigate the dynamics of the dye-doped silver-silica nanoshell. First of all, we consider three different gain G = 0, G = -0.132 and G = -0.22. The power in each laser beam is fixed at 50 mW and the trapping wavelength λ is chosen so that the



Figure 3.4: (a-c) Brownian dynamics simulation of the dye-doped nanoshell trajectory in water for gains, G = 0 (a), G = -0.132 (b) and G = -0.22 (c) and laser power 50 mW. (d-f) Histograms of the nanoshell Brownian motion for the position tracks considered in (a-c). In (g), mean square displacement (MSD) along the z direction for various gains (solid lines). These trends are compared respectively with the analytic limit of the MSD_z to infinity, such as $MSD_z(\infty) \rightarrow 2k_BT/\kappa_z^{max}$ (dashed lines). In (h), the statistical MSD_z computed at infinity vs gain is shown for different power values in solid lines. These trends are compared respectively with the analytic limit of the MSD_z to infinity, shown in dashed lines. The simulation time is 0.1 s and the trapping wavelength has been chosen so that optical trapping forces are maximized for each gain considered.

trap stiffnesses are at their maxima for each gain. In Figs. 3.4a and 3.4c, we show the simulation of the particle trajectories along the axial direction z that are the starting point of the analysis. When G = 0, Fig. 3.4a, the particle is not trapped, the nanoshell jumps in the standing wave maxima and therefore its position is never locked in any of the optical potential minima exploring more than 2 μ m within 0.1 s. When G = -0.132, Fig. 3.4b, the particle trajectory fluctuates around its equilibrium position (z = 0) and the nanoshell is trapped in the central intensity maximum of the standing wave. For the highest gain G = -0.22, (Fig. 3.4c), the tracking profile is tightly locked in the z = 0 position with much smaller fluctuations. We can further quantify our analysis by calculating the position histograms and the mean square displacements of these trajectories. Figs. 3.4d and 3.4f show the position histograms corresponding to the three considered gain. For the case of G = 0, Fig. 3.4d, the nanoshell interacts very weakly with the trapping beam and the thermal noise contribution to the dynamics is much larger than the trapping potential depth. The particle explores several standing wave intensity maxima that show up in the structure of the histogram as different peaks with widths of the order of ~ 200 nm, of the same order of the standing wave periodicity. Instead, increasing gain (G = -0.132, Fig. 3.4e), the particle position appears locked to the central intensity maximum of the double-beam OT. The thermal noise contribution is smaller than the trapping potential depth and the nanoparticle explores only a 19 nm region around the central high intensity spot. For the highest gain G = -0.22, (Fig.3.4f), the position distribution is extremely narrow, and the gain-enhanced gradient force tightly confines the particle within only 2 nm.

We now consider the behaviour of the particle mean square displacement, MSD_z , along axial z direction as a function of gain. This quantity computes the deviation of the particle position (Eqs. 3.6), z_n , from its preceding position for each time interval Δt , $MSD_z(t_n) = \langle |z_n - z_{n-m}|^2 \rangle_m =$ $\sum_{m=1}^{n} |z_n - z_{n-m}|^2/n$. Therefore, in Fig. 3.4g, we have considered the calculated statistical MSD_z versus time for increasing the gain (solid lines) and compared them to the MSD analytically calculated considering the harmonic approximation for small displacements around z = 0 (dashed lines) where MSD_z can be calculated analytically in terms of trap stiffnesses [7, 15], $MSD_z(t) = (2k_BT/\kappa_z^{max}) [1 - \exp(-t/\tau_{to})]$. Hence, at long times we have that $MSD_z(\infty) \rightarrow 2k_BT/\kappa_z^{max}$. We note that in Fig. 3.4g the value of MSD_z approaches to $MSD_z(\infty)$ only for the highest gain (G = -0.22). This occurs when the potential energy barrier is so high that the thermal fluctuations do not have enough energy to drive the hopping between the different potential wells in the standing wave. Consequently, the small displacements approximation gives a reliable description of the dynamics and the theoretical trapping constants (Fig. 3.2i) can be safely used to predict the MSD values. On the other hand, for the lower gains the MSD_z from the simulation is larger than the values predicted by the small displacements approximation with increasing discrepancies as the gain decreases. At zero gain optical potential barriers are much lower than the energy of the thermal fluctuations. Thus, the particle hops between several standing wave maxima and the small displacement approximation does not give a reliable description of the simulated particle dynamics.

Another parameter of the double-beam OT is the laser beam power that will crucially determine the optical trapping dynamics of the dye-doped nanoshell. In Fig. 3.4h (solid lines) we study the MSD at infinity, $MSD_z(\infty)$, as a function of gain parameterized for different power P. Since optical forces increase with laser power strengthening the trap, for each gain value $MSD_z(\infty)$ decreases with increasing power and the tightest confinement occurs at high gain values and high power. Also in this case, we compare the analysis of the Brownian dynamics results with the analytical values of $MSD_z(\infty)$ obtained using the harmonic approximation for the standing wave trapping (dashed lines). For each power, the simulation results tend to overlap with the harmonic approximation as gain increases. The trapping of the nanoshell is so efficient at high gain that the dynamics can be faithfully studied in the single-trap harmonic approximation.

Finally, we describe simulations of a possible practical situation when dye-doped nanoshell particles are immersed in a microfluidic flow and laser light is selectively tuned across the gain-enhanced resonance for position locking, channeling or sorting in a fashion similar to what has been developed for cold atoms [147, 148]. We consider nanoshell particles in water with a high gain G = -0.22 and tune the double-beam OT wavelength within a fraction of nanometer across the sharp resonance peak (see Fig. 3.2g) so that the sign of the gradient force is switched (see Figs. 3.2h,i). In the simulations the power of the single Gaussian beam is fixed at 50 mW and the trajectory is simulated for 0.1 s. In order to emulate a generic particle flux, we have considered three identical nanoshells whose dynamics evolve from three different starting positions $\mathbf{r}_{01} = (0, -400, -250) \text{ nm}, \mathbf{r}_{02} = (0, -400, 0)$ nm, $\mathbf{r}_{03} = (0, -400, 250)$ nm with constant flow velocity $\mathbf{v}_0 = (0, 1, 0)$ mm/s. The reference system origin is taken in correspondence of the center of the double-beam OT. First, we simulate the particle motion with a double-beam OT wavelength of $\lambda_{\rm max} \simeq 531.9$ nm, corresponding to the maximum trapping force. Due to the positive sign of the optical force components, the flowing nanoshells are *high-field seekers* and their position evolve towards specific equilibrium points within the standing wave maxima. Fig. 3.5a show the trajectories, projected on the yz plane, of the three particles superposed



Figure 3.5: Nanoparticles trajectory projections on yz plane are plotted for G = -0.22 both for $\lambda \simeq 531.9$ nm (a), when $\operatorname{Re}\{\alpha\}/\varepsilon_0 \simeq 5.02 \cdot 10^6$ nm³, and for $\lambda \simeq 531.5$ nm (b), when $\operatorname{Re}\{\alpha\}/\varepsilon_0 \simeq -5.88 \cdot 10^6$ nm³. In (a), the radiation intensity maxima correspond to the minima of the optical potential. Therefore, these are attraction points for nanoshells that flow into the standing wave region from three different points $\mathbf{r}_{01} = (0, -400, -250)$ nm, $\mathbf{r}_{02} = (0, -400, 0)$ nm, $\mathbf{r}_{03} = (0, -400, 250)$ nm with constant flow velocity $\mathbf{v}_0 = (0, -400, 0)$ nm/s. Instead, in (b), the intensity maxima correspond to the maximum of the optical potential and the gradient force is maximally repulsive. In this way, the motion of the three nanoshells (with the same initial conditions of the previous situation) is channeled. Each trajectory is simulated for 0.1 s and the power of the single Gaussian beam is fixed at 50 mW.

with the standing wave intensity pattern. Conversely, when the wavelength is tuned to $\lambda_{\min} \simeq 531.5$ nm, the optical force is reversed and the flowing nanoshells are *low-field seekers*. Fig. 3.5b shows that the standing wave maxima act as repulsive spots and the particles are efficiently channelled through the standing wave intensity minima. We note that the role of radiation pressure by the pump field has not been considered in our analysis. Although it can be a source of instability for optical trapping, a double beam configuration can be always applied so that also for the pump field scattering forces are balanced in the trap region.

Chapter 4

Spin-dependent optical forces in optically trapped nanowires

4.1 Introduction

In classical electrodynamics, the linear momentum of light \mathbf{p} , related to the Poynting vector \mathbf{S} , is not trivially associated with, not directed parallel to, the local wavevector \mathbf{k} [149]. In recent years, there has been an increasing interest in investigating, both theoretically and experimentally, configurations and effects connected with the transverse (with respect to the wavevector) components of the Poynting vector, where the interaction of light with particles can lead to extraordinary spin-dependent optical forces [77, 80, 150–160].

The origin of this extraordinary component can be understood in the framework of relativistic field theory [161, 162] applied to the free electromagnetic field Lagrangian, in vacuum. Nöether's theorem produces a current for the canonical non-symmetric gauge dependent energy-momentum tensor [158, 161–163]. In 1940, Belinfante suggested a symmetrization procedure starting from the spin tensor, in such manner to make it gauge-invariant and symmetric [164]. However, the Belinfante spin-correction term has been usually regarded as 'virtual' because it does not contribute to the energy-momentum conservation law, energy transport, and integral momentum of a localized field [150, 165]. Despite this, recently it has been shown that simple optical fields offer an opportunity to investigate, simultaneously and independently, effects connected to the canonical and spin momenta of light in experiments with optical forces [156, 159]. In particular, Bekshaev et al. [155] analyzed the interference field formed by two electromagnetic plane waves (with the same frequency but different wave vectors) and they found that such a field reveals a rich and highly non-trivial structure of the local momentum and spin densities. Antognozzi et al. [158] measured for the first time the extraordinary optical momentum and transverse spin-dependent force using a femtoNewton-resolution nano-cantilever immersed in an evanescent optical field above a total internal reflecting glass surface. Similarly, Liu et al. [166] reported the simultaneous measurement on Mie particle of all components of the polarization-dependent optical force by using a 3D force spectroscopy technique with femto-Newton sensitivity. Ginis et al. argue that the Belinfante momentum transfer to particles in the evanescent field of waveguides depends locally on the light helicity [167]. In brief, the idea behind these theoretical configurations and experiments is the use of nonhomogeneous incident fields to bring out measurable mechanical effects of the 'Belinfante' contribution.

In this chapter, we present the role of the shape of the scatterer on the spin-dependent components of the optical force. We first consider a plane wave illumination, hence we investigate the configuration of focused fields occurring in optical tweezers [15, 44], where a laser beam is tightly focused by a high numerical aperture lens. OT has been extensively used in recent years to trap non-spherical particles and to detect forces and torques in the femtonewton range [47, 58, 110, 168]. Alternatively, a striking range of non-equilibrium phenomena have been revealed in levitated particles caused by the transverse spin-dependent forces combined with thermal fluctuations [169] and birefringence [170]. More specifically, we show the occurrence of spin-dependent optical forces on an optically trapped tilted zinc oxide (ZnO) nanowire illuminated by circularly polarized light. In fact, through light scattering calculations in the T-matrix formalism [2, 90], that the non-spherical shape of the scatterer and its angular tilting yield non-conservative components of the optical force perpendicular to the circularly polarized light propagation direction. We connect these non-conservative components to the symmetry breaking of local spin density maps and to the occurrence of an optical force vorticity [9].

4.2 Extraordinary momentum component and spin density on ZnO nanowires

In our calculations we consider a parameters close to typical system used in real experiments. In particular, we model the ZnO nanowire by a linear cluster of N = 10 homogeneous spherical particles, each with a radius of 50 nm, so that the nanowire length is equal to $L_{\rm NW} = 1 \ \mu {\rm m}$. The particle


Figure 4.1: (a) The model nanowire has 1 μ m length and it is tilted by an angle θ with respect to the incident light wavevector, $\mathbf{k}_{i}||_{z}$, in the x - zplane. It is composed by N = 10 spheres of radius a = 50 nm. A circularly polarized plane wave impinging along z on the tilted nanowire yields a shape-dependent optical force component along x and a spin-dependent optical force component along y. (b) When placed in a circularly polarized optical tweezers (NA = 1.3) contributions to the optical force from the extraordinary momentum are enhanced by the elongated shape. The directions of the canonical momentum \mathbf{P}_{can} (parallel to the local wavevector \mathbf{k}) and the spin momentum \mathbf{P}_{spin} (along y-direction) are sketched. In (c), the intensity xy map of the incident circularly polarized focused field square module $|\mathbf{E}_i|^2$, normalized to the field square module entering the objective lens $|\mathbf{E}_0|^2$, is shown. Its spin-density maps, $\tilde{\mathbf{s}}_{z,i}$, are shown in (d) and (e) for left and right polarization, respectively.

refractive index at the exciting wavelength ($\lambda_0 = 830$ nm) is $n_p = 1.96$ and the surrounding medium is water, $n_m = 1.33$. The nanowire is tilted in the *xz*-plane by an angle θ (see geometry in Figs. 4.1a, 4.1b). By irradiating the nanowire, the time-averaged canonical, spin, and Poynting momentum densities can be defined. Therefore, for a generic harmonic field in a dielectric medium, assuming the Coulomb gauge, [150, 160, 171–173]:

$$\mathbf{P}_{can} = \frac{\varepsilon_{m}}{4\omega} \Im \left\{ (\mathbf{E}^{*} \cdot \nabla) \mathbf{E} + \frac{c^{2}}{n_{m}^{2}} (\mathbf{B}^{*} \cdot \nabla) \mathbf{B} \right\}, \mathbf{P}_{spin} = \frac{\varepsilon_{m}}{8\omega} \nabla \times \Im \left\{ \mathbf{E}^{*} \times \mathbf{E} + \frac{c^{2}}{n_{m}^{2}} \mathbf{B}^{*} \times \mathbf{B} \right\}, \mathbf{S} = \mathbf{P}_{can} + \mathbf{P}_{spin} = \frac{\varepsilon_{0}c^{2}}{2} \Re \left\{ \mathbf{E}^{*} \times \mathbf{B} \right\}.$$
(4.1)

Here, \mathbf{E} and \mathbf{B} are the total electric and magnetic field, obtained summing the incident and the scattering contributions. The spin momentum in the Eqs. 4.1 represents a solenoidal edge current, which is generated by the intrinsic spin angular momentum of the field:

$$\mathbf{P}_{\rm spin} = \frac{1}{2} \nabla \times \mathbf{s}, \\ \mathbf{s} = \frac{\varepsilon_{\rm m}}{2\omega} \Im \left\{ \mathbf{E}^* \times \mathbf{E} \right\} = \frac{\varepsilon_{\rm m}}{2\omega} |\mathbf{E}_0|^2 \mathbf{\tilde{s}}, \tag{4.2}$$

where $\tilde{\mathbf{s}}$ is the vector of spin-1 matrices and produced by the spin angular momentum density \mathbf{s} of the field in the case of non-magnetic medium [150, 160, 171, 173, 174]. The canonical momentum in Eqs. 4.2 is independent of polarization and referred to as the orbital component of the Poynting linear momentum. This component is responsible for the scattering force on a point particle [169]. In contrast to the orbital component, the Belinfante spin momentum, \mathbf{P}_{spin} , is determined by inhomogeneous circular polarization and phase inhomogeneity of the field rather than by its wavevector and it is responsible for the difference between the local propagation and Poynting-vector directions in structured light [151, 155, 164, 165]. The origin of this component can be understood in terms of spin momentum loops [151] that are balanced in homogeneous fields (such as a circularly polarized plane wave), while whose balance is broken when spatial inhomogeneity, such as intensity gradients, are present. In this latter case, spin-dependent force components can arise in transverse directions with respect to the field propagation. For example, for a tightly focused circularly polarized optical beam (see Figs. 4.1b, 4.1c) propagating along the z-axis, we have that the averaged scattering force component related to the canonical momentum is directed along the z direction. While we expect that the transverse component of the optical scattering force related to the linear spin momentum



Figure 4.2: Cartesian components of the radiation force normalized to power, F_x , F_y , F_z , as a function of the nanowire tilting polar angle θ . In (a), (b), (c) the incoming field is a plane-wave, in which the flat/top approximation is used, and in (d), (e), (f) the incoming field is a the focused field where NA= 1.3. The considered laser power is P = 10 mW.

can be revealed along the y-axis. In Figs. 4.1d, 4.1e, we have also plotted $\tilde{\mathbf{s}}_{z,i} = \Im \{ \mathbf{E}_i^* \times \mathbf{E}_i \} / |\mathbf{E}_0|^2|_z$ for circularly left- and right-polarized incident focused field \mathbf{E}_i respectively. This is proportional (Eq. 4.2) to the spin density $\mathbf{s}_{z,i}$. As expected, in both cases the spin-density sign is well defined with $\mathbf{s}_{z,i}^{\sigma_+} = -\mathbf{s}_{z,i}^{\sigma_-}$, since they correspond to two opposite values of the helicity of the incident radiation. We note that for non-paraxial focusing small transverse orbital components are produced by spin-orbit coupling [175] that might contribute to the spin-dependent transverse force. However, even for the tight focusing of optical tweezers we expect these orbital contributions to be at least six times smaller than spin-dependent ones [169].

4.3 Spin-dependent optical force components

A first indication of the occurrence of the spin-dependent force components on the tilted nanowire is given by calculating optical forces for plane wave illumination (Fig. 4.1a) and at the nominal focus of an OT focused field (Fig. 4.1b). In Fig. 4.2 we show the optical force dependence on the nanowire tilting polar angle θ and field polarization for plane wave illumination, Figs. 4.2a, 4.2b, 4.2c, and in the optical tweezers, Figs. 4.2d, 4.2e, 4.2f. Thus, we can appreciate quantitatively how the OT enhances the effect under study. Concerning the plane wave case, we calculated F_x, F_y, F_z starting from the Eq. 2.55, and we consider $|\mathbf{E}_0|^2 = 2\varepsilon_{\rm m}I_0/c$ with I_0 is the intensity of the incoming field \mathbf{E}_0 [15]. We consider a realistic value of intensity $I_0 = 10 \text{ mW}/\mu\text{m}^2$ that can be easily compared to the case of the tightly focused beam of an optical tweezers. In Figs. 4.2a, 4.2b, 4.2c we represent the graphs through different coloured lines comparing the forces for linear x-polarized (blue) and circularly polarized (red and magenta) plane waves. The first plot (Fig. 4.2a) shows the behaviour of the in-plane transverse x-component of the radiation force. Both linear and circular polarization result in a similar trend with tilting angle, showing a negative force pushing the nanowire opposite to its tilting. Most importantly, both the circular polarizations, left and right, yield the same transverse radiation pressure cross section (shown in red). Hence, the in-plane force x-component is the results of the non-spherical shape, but it is not dependent on spin. Similar considerations hold for Fig. 4.2c, that represents the longitudinal radiation pressure, *i.e.* $F_z \sim \sigma_{\rm rad}$. Also in this case, both the x-linear and circular polarizations yield a similar angular dependence for a positive force, and no dependence of the force on light helicity. We note that the quantitative differences in the optical forces for linear and circular polarization shown in Figs. 4.2a, 4.2c are related to the anisotropic polarizability of the nanowire. This is more polarizable parallel to its axis than in perpendicular directions. Thus, the polarization field within the nanowire depends on its orientation relative to the polarization of the external optical field. In turn, the Lorentz force acting on an infinitesimal element of the nanowire depends on its polarization. The total force on the wire is the sum of such contributions and depends, therefore, on the orientation of the wire relative to the polarization of the external field. Alternatively, optical forces can be considered in terms of the T-matrix formulation described in Sect. 2.6. The incident Gaussian beam can be written in terms of a helicity basis of left- and right-hand circularly polarized beams. In this notation, a linearly polarized beam is a sum of a right circularly polarized beam and a left circular polarized beam with a phase shift that determines the direction of linear polarization. Similarly, the

scattered field contains two components, one associated with each circular polarization state. In the context of T-matrix theory the force is a quadratic function of the total (incident plus scattered) field. For a linearly polarized beam, this quadratic term contains cross terms involving, for example, the incident right circular polarized beam, and the field scattered by the left circular polarized beam. These cross terms are completely absent from the expression for the force produced by a right circular polarized or left circular polarized beam independently.

Instead, a striking difference occurs for the out-of-plane force component, F_{y} , shown in Fig. 4.2b. This is zero for x-linear polarization, while for circular polarization a non-zero transverse force component occurs as the nanowire is tilted. Moreover, it changes its sign for opposite circular polarized light. For left-handed light, σ_+ , the spin-dependent force onsets from 0 at $\theta = 0^\circ$, it becomes negative up to $\theta = 45^{\circ}$, then changes sign up to a maximum at about $\theta = 70^{\circ}$, and finally goes back to zero, as expected, when the nanowire is at $\theta = 90^{\circ}$. For right-handed light, σ_{-} , the dependence is opposite. This already clearly shows that the onset of a spin-component of the optical force is crucially related to the shape and symmetry of the scatterer. Indeed, optical force components on a scattering body will generally be non-zero, unless prohibited by symmetry. For instance, an optical force cannot act normally to a symmetry plane. Thus the forces shown in Fig. 4.2b are consistent with symmetry. However, they are unusual in that the y component of the incident momentum is identically zero. These forces arise due to a compensating momentum appearing in the total field as a consequence of scattering.

Figures 4.2d, 4.2e, 4.2f show different coloured lines comparing the forces for linear (blue) and circularly polarized (red and magenta) focused fields. In this case, the (x, y, z) components of the optical trapping force are computed on the points of a grid employed with specified resolution (51 points per axis). These components are calculated over a range of 1 μ m around the nominal focus of the beam. Analytically, we use the Eqs. 2.62, 2.63, in which a laser power P = 10 mW is considered. Even in this case, Figs. 4.2d, 4.2f show that for the components x and z of the radiation force $F_{x,z}^l \neq F_{x,z}^{\sigma}$. The two curves have a minimum in proximity of $\theta \approx 50^{\circ}$, which shows that the module of the transverse optical force is maximum when the symmetry breaking is maximum. We expect this angle to depend on the numerical aperture of the system because of the geometrical interplay between the focal spot (axial and transverse) size and the length of the nanowire. For a nanowire longer than the axial spot size the numerical aperture has an influence in the shape of the spot. For a focused Gaussian beam, considering the axial to transverse size ratio, $z_{\rm R}/w_0$ and $w_0 = 0.5\lambda/{\rm NA}$ being the diffraction limited transverse size, and by geometrical consideration, we expect a maximum at

around $\arctan(z_{\rm R}/w_0)$ that for NA = 1.3 is about $\theta \approx 40^{\circ}$. For a nanowire much shorter than the transverse spot size, the nanowire is contained within the high intensity region of the focal spot and we expect a behavior similar to the plane wave case with differences controlled by the beam polarization and a shift of the maximum toward longer tilt angles ($\theta \approx 55^{\circ}$). Thus, for intermediate situations, such as the one considered in Fig. 2, we expect the angle for which the maximum transverse force occurs to lie between these two situations, as observed. In analogy with the phenomenon of optical lift [67, 93], the nanowire behaves like 'a sail in the wind' that can turn left or right depending on its orientation. This 'sailing effect' is also evident in the longitudinal component, F_z , as visualized in Fig. 4.2f. For both linear and circular polarization $F_z^{l,\sigma}$ at 0° are small and, by tilting the nanowire, they increase until a maximum is reached at $\theta = 90^{\circ}$ when the nanowire is perpendicular to the laser beam propagation direction maximizing its cross section. Both x and z force components are not dependent on the light helicity and the calculations for left-handed σ_+ and right-handed σ_- yield exactly the same results. Instead, Fig. 4.2e shows a breaking of chiral symmetry since $F_y^{\sigma_+}$ is exactly opposite to $F_y^{\sigma_-}$ while for each polar angle F_y^l is null. Thus, the optical force y-component is spin-dependent and occurs only when the light is circularly polarized. Both circular polarizations show a maximum modulus of the force at around $\theta \approx 45^{\circ}$, when the symmetry breaking is maximum. Comparing the curves in the plane-waves (Figs. 4.2a, 4.2b, 4.2c) and those in the focused field (Figs. 4.2d, 4.2e, 4.2f), it is evident how, when the focused laser beams are used, the component of the non-conservative force F_y is greater than in the case of a plane wave, *i.e.* $|F_{y,\text{foc.}}^{\sigma}|/|F_{y,\text{p.w.}}^{\sigma}| \sim 10^2$. Moreover, $|F_{y,\text{foc.}}^{\sigma}|$ has the same order of magnitude with respect to the other components and, therefore, it is in principle detectable.

The strong dependence between the optical force y-component and the spin is deducible by analyzing the same spin density behaviour (Eq. 4.2). Therefore, in Fig. 4.3 we present how the z-component of the total field spindensity $\tilde{\mathbf{s}}_z$ is distributed in the focal region xy-plane when the nanowire is tilted at $\theta = (0^{\circ}, 30^{\circ}, 45^{\circ}, 60^{\circ})$. In particular, we pay attention only on the $\mathbf{s}_z^{\sigma_+}$ because $|\mathbf{s}_z^{\sigma_+}| = |\mathbf{s}_z^{\sigma_-}|$, as we can predict by observing the z-component of the incident field spin intensity (Figs. 4.3c, 4.3d). Starting from the symmetrical configuration at $\theta = 0^{\circ}$ (Fig. 4.3e), and tilting the nanowire at $\theta = 30^{\circ}$ (Figs. 4.3f), 45° (Figs. 4.3g), and at $\theta = 45^{\circ}$ (Fig. 4.3h), $\tilde{\mathbf{s}}_z^{\sigma_+}$ appears with an increasing number of hot-spots. In the origin, the spin density is strongly non-zero just when the cylindrical symmetry is broken. To emphasize the relation between the nanovic shape and the inhomogeneities of the spin distribution, we place the nanowire center of mass is at a generic point of the grid that, for instance, is chosen at (0.1,0,0) μ m (Figs. 4.3i, 4.3j, 4.3k,



Figure 4.3: Maps in the *xy*-plane near the focus of the total field spindensity along z-direction in left circular polarization $\tilde{\mathbf{s}}_z^{\sigma_+}$. The nanowire is at $\theta = (0^\circ, 30^\circ, 45^\circ 60^\circ)$, sketched respectively in (a), (b), (c), (d). In (e), (f), (g), (h) for these three configurations the nanowire is placed in such a manner its center of mass is on the reference system center. Instead, in (i), (j), (k), (l) respectively the nanowire is placed in such a manner its center of mass is on the coordinate point $(0.1,0,0)\mu$ m.

4.31). In addition to the features shown in the case of nanowire with center of mass placed on the reference system center, the $\tilde{\mathbf{s}}_{z}^{\sigma_{+}}$ distributions appears less and less circular as the tilting angle increases.

4.4 Non-conservative force components and optical force vorticity

To better appreciate how the left and right-handed circular polarization forces behave in the focal region, in Fig. 4.4 we highlight the differences between the in-plane non-conservative components of force. In particular, we plot the local difference $F_i^{\sigma_+} - F_i^{\sigma_-}$, with i = (x, y), for four different nanowire orientations, $\theta = (0^{\circ}, 30^{\circ}, 45^{\circ}, 60^{\circ})$, to also evidence any chiral effect. In Figs. 4.4a, 4.4b we have this comparison for $\theta = 0$. In this configuration, the transverse components, zero at the origin, are weakly non-zero and opposite in sign away from the optical axis. Figs. 4.4c, 4.4d, 4.4e, 4.4f, 4.4g, 4.4h show the differential maps for $\theta = 30^{\circ}$, $\theta = 45^{\circ}$, and $\theta = 60^{\circ}$, respectively. Here, the symmetry of the differential maps is fully broken. In particular, for the spin-dependent component, F_y , the map shows three quasi-oblate regions standing out: one around the origin (red) where there is a maximum, and the other two (blue), on the opposite sides of the origin where there is a minimum. This shows clearly as the transverse optical force depends on the helicity of the incident radiation, that, in turn, transfers its own chiral behaviour to the nanowire dynamics. Concerning F_x , the difference is much smaller because this component is less dependent on the light helicity and it is not zero only when the nanowire is shifted away from the origin.

To highlight the effects of the transverse force on the nanorod dynamics, we show in Fig. 4.5 the vectorial force distribution $\mathbf{F}_{\rho}^{\sigma} \equiv F_x^{\sigma} \hat{\mathbf{x}} + F_y^{\sigma} \hat{\mathbf{y}}$ calculated in the xy focal plane. We notice that in the focal region the interaction between the incident radiation and the nanowire causes the trapping to take place so to brings the nanostructure to its maximum stability configuration, that is when the wire is aligned with the optical axes ($\theta = 0^{\circ}$).

According to what we have just shown, it is evident how the chiral effects are significant in the tilted configuration. In fact, in this case the force distribution has appeared cylindrically unsymmetrical. Therefore, we have analyzed the trend of the force curl calculating the quantity $\Omega_c \equiv (\nabla \times \mathbf{F}_{\rho}^{\sigma_+})|_z - (\nabla \times \mathbf{F}_{\rho}^{\sigma_-})|_z$. We refer it as *optical force vorticity* being the local difference between the z-component of the force curl for the two different helicities. In fact, the force curl z-component is related to the non-conservative y-component of radiation force. In Figs. 4.6a, we have the vorticity for $\theta = 0$.



Figure 4.4: Maps in the *xy*-plane near the focus of the difference between radiation forces in left circular polarization and right one. The nanowire is presented in three configurations: when it is at polar angle $\theta = 0^{\circ}$, the local differences $F_x^{\sigma_+} - F_x^{\sigma_-}$ and $F_y^{\sigma_+} - F_y^{\sigma_-}$ are respectively shown in (a) and (b), when it is at polar angle $\theta = 30^{\circ}$, $F_x^{\sigma_+} - F_x^{\sigma_-}$ and $F_y^{\sigma_+} - F_x^{\sigma_-}$ are respectively shown in (c) and (d), for $\theta = 45^{\circ}$ they are, respectively, shown in (e) and (f); and, when it is at polar angle $\theta = 60^{\circ}$, $F_x^{\sigma_+} - F_x^{\sigma_-}$ and $F_y^{\sigma_+} - F_y^{\sigma_-}$ are respectively shown in (g) and (h). The considered laser power is P = 10 mW.



Figure 4.5: Maps in the *xy*-plane strongly near the focus of radiation force vector in left circular polarization and right one. The nanowire is presented in three configurations. When it is at polar angle $\theta = 0^{\circ}$, $\mathbf{F}_{\rho}^{\sigma_{+}}$ and $\mathbf{F}_{\rho}^{\sigma_{-}}$ are respectively shown in (a) and (b), when it is at polar angle $\theta = 30^{\circ}$, $\mathbf{F}_{\rho}^{\sigma_{+}}$ and $\mathbf{F}_{\rho}^{\sigma_{-}}$ are respectively shown in (c) and (d), for $\theta = 45^{\circ}$ they are, respectively, shown in (e) and (f); and, when it is at polar angle $\theta = 60^{\circ}$, $\mathbf{F}_{\rho}^{\sigma_{+}}$ and $\mathbf{F}_{\rho}^{\sigma_{-}}$ are respectively shown in (g) and (h). The considered laser power is P = 10 mW.



Figure 4.6: Maps in the *xy*-plane near the focus of the 'Vorticity' Ω_c , understood as the difference between the curl of the radiation force in left circular polarization and right one. The nanowire is presented in the four configurations when it is at polar angle $\theta = (0^{\circ}, 30^{\circ}, 45^{\circ}, 60^{\circ})$. For each angle, Ω_c are respectively shown in (a), (b), (c), (d). The considered laser power is P = 10 mW.

In this configuration, as the local distributions $\mathbf{F}_{\rho}^{\sigma_+}$ and $\mathbf{F}_{\rho}^{\sigma_-}$ are similar, the map shows a circular symmetry around z-axis. Figs. 4.6b, 4.6c, and 4.6d show the differential maps for $\theta = 30^{\circ}$, $\theta = 45^{\circ}$ and $\theta = 60^{\circ}$, respectively. Here, the symmetry of the differential maps is fully broken, not only because of the different orientation of the nanostructure, but also because of the different helicity transferred from the incident field to the non-conservative force components.

Chapter 5

Spectropolarimetric constraints on interstellar dust modelling

5.1 A dusty universe

We live in a dusty galaxy, one of billions of galaxies in the Universe. Almost all of them are dusty. Up to the mid-20th century, this dust was considered only as an unfortunate impediment to precise observations of stars and galaxies, regarded as the most important items in the Universe. The modern view of dust as a cosmic component has completely reversed the earlier view. We now know that many aspects of the formation of planets, stars, and galaxies are influenced in some way by interstellar dust [176]. The involvement of dust grains in providing molecules important for the origin of life, and in the safe transmission of those species to newly-forming planets orbiting Sun-like stars [177], has been one of the greatest surprises of all. Therefore, it is no wonder that considerable efforts are currently placed in trying to understand nature, composition, and evolution of dust in the interstellar medium.

There is now a robust evidence that dust grains condense in circumstellar environments, mainly AGB (Asymptotic Giant Branch) stars [178] and supernovae [179, 180], from which they are ejected in the interstellar medium. There, dust is tightly mixed with the gas, with the dust representing only a minor fraction of the total mass. The gas-to-dust mass ratio locally assumes the fairly constant value of 100 [181]. The dependence of this ratio on the metal content among galaxies and within a galaxy is an important issue from a cosmic perspective for a number of reasons [182], including the appearance of the first solids in the early Universe [183]. In late-type galaxies, like our own, this ratio scales with the metallicity, decreasing with the galactocentric radius, a clear indication that dust growth in the interstellar medium dominates over destruction [184]. As a consequence, stardust, literally dust formed around stars, is not the same as interstellar dust, and must be regarded as the raw material from which true interstellar grains are formed. Such a material is modified and destructured by violent interstellar processes [185], before being reformed and reassembled in denser interstellar regions [186].

Modifying processes continue to act throughout the lifetime of a grain, some hundreds of millions of years in the Milky Way. In some cases, such modifications may be catastrophic, as during the formation of a planetary system. In the Solar System, the patterns of isotopic abundances that are found in survived presolar grains identify their origins in the cool envelopes of evolved low mass stars and in supernovae ejecta. These particles have passed through many destruction processes, including their ejection from the stellar envelopes, their passage through the interstellar medium where they have been subjected to intense radiation fields and dynamical shocks, their incorporation into the molecular cloud that formed the Solar System, and through all the violent processes involved in the formation of the Sun and its planets. Near-stellar dust is modified in the interstellar medium and becomes interstellar dust which, in turn, may be severely modified or destroyed when it is incorporated in the gas that forms newly-born planetary systems.

5.1.1 Interstellar dust

Most of the information that we have about interstellar dust is obtained remotely, by the influence of dust on various kinds of astronomical observations. These observations may be carried out at a very wide range of wavelengths, from X-rays to radio waves, but traditionally the most important and defining data have come from the infrared, visible, and ultraviolet parts of the spectrum, summarized through a wavelength-dependent extinction curve along the lines of sight to individual stars [12]. There are detailed extinction measurements along hundreds of lines of sight that present similarities in their shapes. The extinction typically increases from low values in the infrared to high values in the far ultraviolet, a near-linear portion in the optical region, a pronounced and broad "bump" near 217.5 nm, and a final rise of varying slope into the far ultraviolet. Both the general aspect of the extinction and the details of specific curves along particular lines of sight provide useful information, and generally indicate that grains of a wide range of sizes (roughly nanometers to micrometers) are required [187]. The dominant feature in the extinction curve is the prominent bump in the near ultraviolet. Its central position is fixed, although the width of the feature can vary significantly from

one line of sight to another. It is widely attributed to $\pi^* \leftarrow \pi$ transitions in aromatic carbon solids [188] or polycyclic aromatic hydrocarbons [189]. The far-ultraviolet rise may be decomposed in a linear contribution, due to nano-sized particles, and a non-linear component belonging to the partially invisible (because located beyond the Lyman continuum) $\sigma^* \leftarrow \sigma$ resonance in aromatic carbon. In the near-infrared, there is a weak absorption feature at a wavelength of 3.4 μ m, detectable on long paths through diffuse gas. It is characteristic of absorption in the sp^3 (aliphatic) C-H stretching bond [190]. Further into the infrared are two stronger absorption features, at 9.7 and 18 μ m, ascribable to silicate materials, from Si-O stretching and O-Si-O bending modes, respectively. Taken together, these features strongly support the presence of some form of carbon/hydrocarbon and silicate in the dust. X-ray scattering and absorption edges provide constraints on grain size and composition, specifically O, Mg, Si, Fe, and C atoms [191]. There is also a set of detected infrared emission features occurring at 3.3, 6.2, 7.7, 8.6, and 11.3 μ m, indicative of aromatic CH groups. The mechanism responsible for the excitation of such infrared emission may involve non-equilibrium emission from polycyclic aromatic hydrocarbons stochastically heated to high temperatures by the absorption of individual photons from the interstellar radiation field [192]. However, the requirement of microscopic sizes can be relaxed if the emitters of the 3.3 μ m and other infrared bands are heated by the chemical energy released from reactions within larger carbon interstellar grains of mixed sp^2/sp^3 carbon composition [193]. Such structures have been in fact observed in some extragalactic objects [194]. Exploiting a ternary phase diagram where the hydrogen content and the two main bonding types (sp^2) and sp^3) for carbon constitute the poles, Dartois et al. (2007) [194] were able to identify the carrier of the spectral features as an interstellar hydrocarbon belonging to the class of polymeric-like hydrogenated amorphous carbon (a-C:H), dominated by an aliphatic/olefinic backbone structure. The change from aliphatic to aromatic structures may occur in environments that selectively dehydrogenate the a-C:H, providing an opportunity for aromatic molecules to form. These observations, together with observations of very evolved stars (protoplanetary and planetary nebulae), suggest an evolution in which aliphatics are converted into aromatic structures [195, 196].

The interpretation of dust observations must take also account of the available abundances along the line of sight. Astronomical dust is likely to be an amalgam of a number of different materials, very chaotic in composition and structure, with different individual substances dominating at different wavelengths. These materials are thus fundamentally different from terrestrial materials. Nuth & Hecht (1990) introduced the concept of chaotic silicates in which the level of disorder is even greater than for glasses, that

are characterized by the absence of long-range order in the atomic arrangement beyond nearest neighbours [197]. Since materials may be assembled in the agglomerate, an astronomical silicate cannot be considered a solid with a definite stoichiometric composition. They may also occur in groups that recall solid solutions, in which one or more types of atoms or molecules of the solid may be partly substituted for the original atoms and molecules without changing the structure. Olivine and enstatite are excellent examples of solid solutions. Forsterite, Mg₂SiO₄, and fayalite, Fe₂SiO₄, have identical structures because the ions Mg^{2+} and Fe^{2+} are very nearly the same size and are chemically similar. Very frequently amorphous silicates in space are misleadingly described in terms of the optical properties of these materials. Indeed, as pointed out by Rietmeijer and Nuth (2013) [198], there are no amorphous silicates, as the word "silicate" already implies that the material is crystalline and could be a mineral. Moreover, astronomical solids may be porous and therefore of much lower density than a glass. Ultimately, the nature of an astronomical silicate is rather loosely constrained, to same extent just limited to a material whose infrared spectrum is dominated by Si-O stretching and bending vibrations. Thermal annealing (e.g. in shocks) or intense X-ray irradiation [199] of precursor materials, that were probably amorphous, may explain the presence of crystalline silicates (see however Ritmejer and Nuth 2013 [198]) in circumstellar regions and protoplanetary disks [200].

Even carbonaceous materials in space are difficult to constrain. A striking example is given by the nature of the carrier of the interstellar ultraviolet extinction bump at 217.5 nm, that was originally attributed to small crystalline graphite particles [201], followed by a plethora of proposals including mixture of spheres composed of graphite, amorphous carbon, and silicate [202], irregular or fractal arrangement of graphite and amorphous carbon [203], polycyclic aromatic hydrocarbons [204], natural coal [205], and even electronic transitions of OH⁻ ions in sites of low coordination in silicates [206]. In general, carbonaceous materials contain greater or lesser hydrogen fractions, varying proportions of different chemical bonding, and different degrees of long-range order. All these forms of carbon can, under suitable conditions, be readily converted from one to another. The manifold of possible bonding arrangements produces several allotropes of carbon. The best known are graphite, diamond and amorphous carbon. The physical properties of carbon vary widely with the allotropic form. Amorphous carbonaceous materials cover a wide range of compositions, from wide band gap, H-rich, aliphatic-rich a-C:H to narrow band gap, H-poor, aromatic-rich a-C materials. The properties of a-C:H materials are determined by the sp^3/sp^2 ratio for the carbon atoms and the hydrogen concentration. A C-H bond contributes to the formation of sp^3 bonding and the reduction of the defects in the amorphous carbon network. In general, it is found that the optical energy gap increases with hydrogen concentration [207].

5.1.2 Core-mantle model and spectropolarimetric constraints

Despite the good amount of reliable information regarding interstellar dust, its nature, morphology, and composition are still much debated and there are still many open questions. There is not a unique model capable of justifying the observational evidences described in Subsect. 5.1.1. In this context, the spectropolarimetric investigation has been used to distinguish between dust models and gather more information on the grain structure and composition [208]. In 1949 Hall and Hiltner were the first to realize that starlight is often slightly linearly polarized, at a level of a few percent [209]. The interpretation was that the interstellar material exhibits differential extinction, that favours the extinction of starlight in one plane of polarization over another. Evidently, non-spherical dust grains are aligned. The knowledge of the polarization state of radiation provides far more astrophysical information than intensity alone, including clues on the optical properties of grains [210], the conditions under which grains can be aligned [211], and their morphology [212].

Basically, the carbon and the silicate components play the main role in current dust models, although they appear in different forms. Draine & Lee consider that graphite is the major grain carbon sink and that the silicate and graphite components are bare and physically separated in dust [213]. Mathis & Whiffen assume the dust to be low-density aggregates of small silicates and carbonaceous particles [214]. Jones, Duley & Williams assume that silicate grains are coated with a carbonaceous mantle made of either HAC or organic refractory [215]. Such model has proven to be successful in reproducing the interstellar extinction curve also respecting the element abundance constraints [215–217]. In this model carbon from the gas phase is deposited on silicate grain cores as H-rich sp^3 carbon, then processed by the ultraviolet field and aromatized, progressively becoming sp^2 carbon, and ultimately removed in shocks [195, 217]. The process can be conversed under the grain exposure to hot H atoms. Within this picture, we have a silicate core covered by two distinct carbon mantles, an inner 'old' processed sp^2 carbon mantle and an outer 'young' mantle of freshly deposited sp^3 carbon. So, we have two competitive processes characterized by two different timescales, the rate of carbon deposition from the gas phase and the photodarkening rate, giving the conversion rate from sp^3 to sp^2 carbon [195, 217].

The core-mantle model has been seriously challenged by the spectropolarimetric observations of the carbon 3.4 μ m feature [212]. Such absorption feature, observed in the diffuse interstellar medium, is commonly attributed to the C-H stretching mode in aliphatic hydrocarbons residing in some components of interstellar dust. Chiar et al. found that this feature is negligibly polarized for a line of sight toward the Galactic Center [208]. Such evidence, together with the observation of a strong polarization in the silicate feature at 9.7 μ m along the same sightline, suggests that carbon and silicate grains are separate components. In this picture the silicate grains appear to be non-spherical and aligned so as to originate the observed strong polarization feature at 9.7 μ m, while the carbon grains should not be aligned to explain the non-detection of polarization in the 3.4 μ m feature [212].

5.2 Polarization results

Interstellar polarization is caused by the differential extinction of the two perpendicular electric vectors of starlight by aligned, non-spherical grains. A lower polarization degree of the 3.4 μ m feature substantially corresponds to a lower elongation of the 3.4 μ m carrier. Chiar et al. places an upper observational limit on the 3.4 μ m relative polarization of the Galactic Centre quintuplet object GCS 3-II [208]:

$$\frac{P_{3.4}/A_{3.4}}{P_{9.7}/A_{9.7}} \approx 0.13 \tag{5.1}$$

where $P_{3.4}$ and $P_{9.7}$ are the 'excess' polarization referred respectively to the 3.4 μ m and 9.7 μ m features, while $A_{3.4}$ and $A_{9.7}$ are the excess average extinctions referred respectively to the 3.4 μ m and 9.7 μ m features. By 'excess', we mean the extinction and the absorption feature polarization in excess of the continuum extinction and polarization underneath the feature. To alleviate the challenge on the core-mantle model, Li et al. considered an extreme case, proposing a model made by elongated (spheroidal) silicate cores coated with spherical layers of aliphatic hydrocarbons [212]. The basic idea behind this model is that, since the carbon mantle is much less elongated than the silicate core, the 3.4 μ m feature would result polarized to a much smaller degree than the 9.7 μ m silicate feature. However, the computed 3.4 μ m feature polarization still exceeds the observational upper limit placed by Chiar [212].

We follow a different approach and try to understand if the challenge may be alleviated modeling the dust as aggregates of core-shell grains. To this aim, we consider many different cluster models and calculate the resulting extinction and polarization through the T-Matrix method (Subsect. 1.5.2).

5.2.1 Results for core-mantle grain aggregates and composite dust models

In our numerical investigation, first we present the extinction and polarization results for some cluster configurations inspired by the core-mantle model provided by Jones et. al [215, 217]. In agreement with the abundance constraints and with the evidence that interstellar dust grains are rather elongated, we have generated aggregates whose related inertia ellipsoid has an eccentricity lower than unity. Each structure presents nine spheric monomers which have different radii. In Figs. 5.1a, 5.1b, 5.1c three aggregates of silicate core-carbon mantle spheres are shown. The silicate core is made of olivine $(Fe, Mg)_2SiO_4$ while the outer shell is sp^3 amorphous carbon [195]. The olivine refractive index is provided by Draine & Li while the carbon optical constants are given by Ashok et al. [213, 218]. The three models have different morphologies, going from the most elongated structure in Fig. 5.1a (very fluffy aggregate) to the most compact one in Fig. 5.1c. The major semiaxis of the equivalent inertia ellipsoid is respectively $r = 0.2 \ \mu m$ (Fig. 5.1a), $r = 0.16 \ \mu \text{m}$ (Fig. 5.1b), $r = 0.11 \ \mu \text{m}$ (Fig. 5.1c). The polarization to extinction ratio corresponding to these three structures is shown in Fig. 5.1h as a function of the carbon-silicate volume fraction (dashed line: fluffy cluster, dotted line: very fluffy, and dash-dotted line: compact cluster).

In Fig. 5.1d, we model a composite interstellar grain (mixed aggregate) made by six stratified spheres, with a silicate core and a sp^2/sp^3 carbon coating, and three homogeneous spheres of amorphous carbon. The geometric configuration of the cluster is the same as in Fig. 5.1b. In the numerical simulations we increased gradually the percentage composition of sp^2 carbon in the mantle, going from zero (corresponding to a completely sp^3 carbon mantle) to a completely sp^2 carbon mantle (see Fig. 5.1h) corresponding to a situation where all the carbon mantle has been processed by the ultraviolet radiation. In Fig. 5.1h the polarization to extinction ratios are shown corresponding to the different carbon mantle composition and assuming $V_{\text{Carb}}/V_{\text{Sil}} = 1$.

In Fig. 5.1e (big fluffy model) we show again a core-mantle cluster model, with dimensions ten times larger than the cluster in Fig. 5.1b. The aim is to explore the same configuration but on a larger scale. In Fig. 5.1f we model a composite aggregate, following the Mathis model [214], made by homogeneous spheres. Specifically, we have four silicate spheres and five amorphous



Figure 5.1: Polarization to extinction ratios for different dust grain structures. (a), (b), (c) Aggregates of core-mantle grains with a silicate core and a carbon shell. (d) Composite core-shell and carbon sphere cluster. Six stratified spheres present a silicate core and a sp^2/sp^3 carbon external coating while three spheres are entirely composed of amorphous carbon. (e) Aggregate of core-mantle grains, with dimensions ten times larger than the cluster in (b). (f) Composite model made by four homogeneous silicate spheres and five homogeneous amorphous carbon spheres. (g) Stratified cluster models like the cluster in (b) with the addition of an ice external shell. (h) 3.4 μ m polarization to extinction ratios for the different models.



f _{sp2} tail (%)												
			50			75		100				
f _{sp2} head (%)		$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	P _{9,7} /A _{9,7}	$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	$P_{9,7}/A_{9,7}$	$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	P _{9,7} /A _{9,7}		
	0	0.728	0.26	0.35	0.564	0.21	0.37	0.312	0.12	0.38		
	50	0.882	0.32	0.37	0.696	0.27	0.38	0.256	0.10	0.40		
	75	1.039	0.39	0.37	0.871	0.34	0.39	0.211	0.09	0.41		
	95	1.242	0.47	0.37	1.173	0.47	0.40	0.071	0.03	0.42		

Table 5.1: Polarization to extinction ratios for different configurations of the head-tail model. The head and the tail are characterized by different percentages f_{sp2} of processed carbon in the shells. The cluster major axis is oriented perpendicularly to the incident field direction.

carbon spheres. In this way, we completely separate the silicate components from the carbon ones, making in principle the polarization of the 3.4 μ m feature independent from that of the silicate feature at 9.7 μ m. Finally, in Fig. 5.1g we show a core-mantle cluster model, similar to the one in Fig. 5.1b, with the addition of an ice external shell. As can be seen from Fig. 5.1h, none of the aforementioned models presents satisfactory polarimetric results. All the results are above the threshold value of 0.13 (solid line) indicated by Chiar et al. [208]. These results show that all the explored models so far do not respect the polarimetric observational constraints and seem to push towards the failure of the core-mantle model.

5.2.2 Head-tail cluster model

We try to alleviate the polarization challenge exploring different grain structures. We model the core-mantle grain aggregate with a head-tail con-

		$\left. \frac{r}{R} \right _{\text{head}} < \frac{r}{R} \right _{\text{tail}}$			^r / _R	head = r/F	al tail	$\left. \frac{r_{R}}{r_{R}} \right _{\text{head}} > \frac{r_{R}}{r_{R}}$			
		P _{3,4} /A _{3,4} P _{9,7} /A _{9,7}	$P_{3,4}/A_{3,4}$	P _{9,7} /A _{9,7}	$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	P _{9,7} / A _{9,7}	$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	$P_{9,7}/A_{9,7}$	
V _{Sil}	1.25	0.105	0.05	0.47	0.194	0.08	0.4	0.948	0.33	0.34	
$V_{\rm Carb}/$.	0.071	0.03	0.419	0.236	0.09	0.37	0.893	0.3	0.33	
	0.75	0.019	0.008	0.39	0.274	0.01	0.35	1.6	0.5	0.31	

Table 5.2: Polarization to extinction ratios corresponding to different volume fractions of the silicate and carbon components in the core-mantle spherical sub-units. Also the ratio between the core radius (r) and the external radius (R) in the core-mantle units is changed independently in the head and in the tail. The cluster major axis is oriented perpendicularly to the incident field direction. f_{sp2} is 95% for the head and 100% for the tail.

figuration, kind of a 'mouse' like configuration (see the top of Tab. 5.1 for the model). This new structure is made by a compact 'head' and a quite elongated 'tail', both made by core-mantle spherical grains. The calculations are performed by independently varying the percentage of processed carbon f_{sp2} for the tail and the head, and assuming different sizes for the spherical sub-units. The cluster is oriented so that the major axis is perpendicular to the direction of the incident radiation. As we can see from Tab. 5.1, the polarization to extinction ratio settles above the upper observational limit of 0.13 in most of the cases under investigation. However, when the carbon is highly aromatized (95%) in the head and completely aromatized (100%) in the tail, the computed polarization to extinction ratio finally goes below the observational limit, giving a value of 0.071.

			(;	а)	9	P	(b)	(c)			
	PRINCIPAL						r	STRETCHED			
z		$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	P _{3,4} /A _{3,4}	P _{9,7} /A _{9,7}	$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	$P_{9,7}/A_{9,7}$	$\frac{P_{3,4}/A_{3,4}}{P_{9,7}/A_{9,7}}$	$P_{3,4}/A_{3,4}$	P _{9,7} / A _{9,7}	
	×	0.587	0.189	0.321	2.815	0.433	0.154	0.213	0.09	0.424	
N AXIS OR	~	0.071	0.03	0.419	0.887	0.186	0.21	0.457	0.24	0.524	
MAI		2.222	0.218	0.01	4.39	0.247	0.056	3.299	0.33	0.1	

(c)

Table 5.3: Polarization to extinction ratios for different configurations of the head-tail model (a) principal, (b) compact, (c) stretched. The results are shown changing the main axis orientation of the corresponding rotation ellipsoid. $V_{\text{Carb.}}/V_{\text{Sil.}} = 1$. f_{sp2} for the head is 95% while f_{sp2} for the tail is 100% for all the models.

So, choosing this particular configuration, we run the T-matrix computations using as free parameters in the model:

- the ratio between the overall carbon volume and the silicate volume,
- the ratio between the internal spherical core radius r and the radius of the external sphere R, both for the core-mantle spherical particles in the head $(\frac{r}{R}|_{\text{head}})$ and for those in the tail $(\frac{r}{R}|_{\text{tail}})$. We consider three cases: $\frac{r}{R}|_{\text{head}} < \frac{r}{R}|_{\text{tail}}, \frac{r}{R}|_{\text{head}} = \frac{r}{R}|_{\text{tail}}, \text{ and } \frac{r}{R}|_{\text{head}} > \frac{r}{R}|_{\text{tail}}.$

As Tab. 5.2 shows, the results are in agreement with the observational limit only when $\frac{r}{R}|_{\text{head}} < \frac{r}{R}|_{\text{tail}}$. In such case, the polarization to extinction ratio is below 0.13 for all the considered carbon/silicate volume fractions.

Then, we change the cluster structures. In particular, setting the carbon/silicate volume ratio at unity, we consider different orientations for the aggregates and three different configurations. In Tab. 5.2 the three cluster models are shown: the 'Principal' cluster (a) is the same used for the previous computations, the 'Compact' (b) and the 'Stretched' (c). Analyzing the results in Tab. 5.2, the 'principal' model is the only one giving values below the upper observational limit. Moreover, this works well only for a specific (*i.e. y*-axis) cluster orientation. All the models give the worst results (that is the highest values compared with the observational limit) when oriented along the z-axis, that is along the direction of propagation of the incident field.

5.3 An evolutionary scenario

Analyzing the results of our computations, we observe that the silicate core-carbon mantle dust grains meet the polarization constraints only in few of the cases under investigation. From an evolutionary point of view, we find that the best configuration corresponds to aggregates of core-mantle particles made by a compact head, with traces of aliphatic carbon, and an old carbon elongated tail where carbon is totally processed and aromatized showing a $100\% sp^2$ composition. Moreover, the structure and orientation of the cluster strongly affects the results. These observations pose reasonable doubts on the validity of the core-mantle model due to the limited range of structures satisfying the polarization constraints. However, there is an interesting point that still deserves to be discussed and that could open new perspectives. The demand for fully aromatized carbon mantles, emerging from our investigation, is actually reasonable in an evolutionary scenario related to the carbon cycle in the interstellar medium [219]. It is known that the interstellar radiation field progressively processes the sp^3 hydrogenated amorphous carbon in the grain mantles into an H-poor sp^2 carbon on a relatively short timescale of the order of a million years [220]. Therefore, it is to be expected that small grains and thin mantles of amorphous hydrocarbon material should be maximally aromatized in the diffuse ISM. The outer layers of freshly deposited, still unprocessed, aliphatic carbon would be very thin and negligible [219]. Within this evolutionary history picture, the carbon mantle would not be responsible for the 3.4 μ m feature that would be rather due to small, non aligned, aliphatic hydrocarbon grains. Then, silicate and carbon could still co-exist in a core-mantle structure without being in conflict with the polarization constraints.

Chapter 6

Modeling optical forces for space tweezers applications

6.1 Space tweezers

Optical trapping and optical manipulation methods have seen a tremendous advances in many research fields from physics to chemistry and biology. However, their application to planetary exploration is still to be developed, even though already conceived by, e.g., NASA [221]. The development of the optical trapping technique to collect and analyze in situ or return to Earth a variety of extraterrestrial particles will open doors to information on space materials that is currently unreachable, e.q., the dust volatile component not detectable in situ by the Rosetta/ESA dust instruments (GIADA, MIDAS, COSIMA) and not retrievable by samples return missions, or have biases due to collection media contamination, e.g. cometary dust samples trapped in aerogel and returned by the Stardust/NASA space probe [222, 223]. In this context, the project Space Tweezers (https://www.spacetweezers.org/home) has taken shape and its purpose is to develop methods for using OT to trap and spectroscopically characterize (Raman Tweezers) extraterrestrial dust particles and/or their analogs. This study would provide solid ground for the application of OT techniques in the near future to solar system study, e.g., cometary particles analyses including the volatile component, dust particles in the Martian atmosphere and/or on the Martian, Lunar surfaces. Such application will also be strategic for the "clean" handling/preliminary characterization of restricted and unrestricted samples of planetary bodies returned from space missions in curatorial facilities.

Therefore, in this chapter we show the first theoretical results that serve as a starting point for space tweezers applications [12]. After a review on the role of dust in the solar system and in the extra-planetary environment, we describe the models and methods we use to calculate light pressure and optical trapping properties on a variety of realistic dust particle models. Then, we show results on solar radiation pressure calculations that can help to a greater understanding of micro-particle dynamics as well as to estimate its detrimental role in optical trapping in space. Finally, we consider and compare results of calculations for optical trapping of dust particles in standard OT in water (typical laboratory conditions) with those calculated for OT in air or in space.

6.2 The Solar System dust complex

The interplanetary space of the solar system is very dusty. This conspicuous feature of the solar system is actually appreciable to the naked eye through the faint solar colour cone of light above the western horizon after sunset, or above the eastern horizon just before sunrise, called the Zodiacal Light [12]. Just like the Milky Way reveals the billions of stars forming our galaxy, so the Zodiacal Light reveals the presence of a huge amount of fine dust particles that scatter solar radiation in the interplanetary space of the solar system. The brightness of the Zodiacal Light provides information on the overall space density of the dust known as the interplanetary dust complex. The interplanetary dust complex consists of microscopic (mainly rocky) particles, typically less than a few millimetres in size, called micrometeoroids, moving in the interplanetary space of the solar system [228]. Dust is produced by collisions among solid bodies, by disruptions of icy bodies [229, 230] and by cometary activity [231, 232]. As such the interplanetary dust complex is an inventory of the constituent materials of a large variety of solid bodies in the solar system. Most of the compositional properties of the interplanetary dust complex derives from the cosmochemical analyses of samples recovered from the Earth's: 1) surface, e.g., micrometeorites collected in Antarctica [233–235]; 2) stratosphere, by balloon born instruments as, e.g., DUSTER, designed for non-destructive and uncontaminated collection of solid particles from tens of microns down to 200 nm in size [236] and by stratospheric NASA/aircraft passive sticking on silicon oil coated plates [237]. A critical contribution is also given by laboratory analyses of samples, *i.e.*, collected, and brought back to Earth, from asteroid surfaces [238] and in a cometary coma [222, 223]. In addition, cometary dust was studied in situ from the onset of cometary activity to its cessation after perihelion by the Rosetta/ESA space mission [239–241].



Figure 6.1: Scattering models with shape and composition inspired by interstellar, interplanetary (DUSTER mission [224]), and planetary [225] particles. On the top, the models emulate hypothetical interstellar dust grains whose constituents are olivine and aliphatic carbon. In (a) and (b), the constituents refractive indexes are mixed according to the Bruggeman criterion. Instead, in (c) and (d), the olivine is considered covered by a carbon layer. In (e), the model of a silica particle arranged in quenched melt spheres shown in the TEM image [224] (f). In (g), the model of condensed Ca[O] nanograins that are accreted onto a larger melted aggregate of tiny carbonate grains shown in the TEM image [224] (h). The larger sphere is calcite and the other spheres are CaO. In (i), a spherical model of the particle Fe,Mg-rich 'TP2' [223], in which we consider an effective refractive index constructed mixing iron (67%) and magnesium (33%), according to the Bruggeman criterion. In (j), a spherical model of Martian hematite [225]. In (k), a spherical model of Lunar regolith [226]. In (1), a model of an ellipsoidal fassaite shown in the TEM image (m). Here, we consider an effective refractive index constructed mixing silica (53%), CaO (27%), FeO (10%), Al₂O₃ (10%), according to the Bruggeman criterion [227].



Figure 6.2: Optical forces distributions exerted by the Sun on dust particles modeled as shown in Fig. 6.1. The optical force parallel, $f_{\parallel}(\lambda)$ (blue line), and perpendicular, $f_{\perp}(\lambda)$ (red line), are intended respect to the light propagation direction $\hat{\mathbf{k}}$. In (a-d) we show calculations on olivine-aliphatic carbon structures. In (a) and (b), the constituents refractive indexes are mixed according to the Bruggeman criterion [227]. While in (c) and (d) the olivine and the carbon are distributed in a core-shell structure. In (e) we show results for the quenched melt silica particle, in (f) for the bunch-of-grape carbon Ca-rich, in (g) for the 'TP2' sphere, in (h) for the martian hematite sphere, and in (i) for the fassaite ellipsoid.

Planetary dust. Many terrestrial planets and satellites of both terrestrial and giant planets in our Solar System show dusty environmental conditions. In particular, the mostly explored dusty bodies are the Moon and Mars.

The Moon is the only Earth's natural satellite. Since the Moon has neither a magnetic field nor a significant atmosphere, the lunar regolith and the near-surface environment are mainly affected by space weathering processes such as meteoroid impacts, solar ultraviolet radiation, solar wind, galactic cosmic rays and plasma processes in the tail of the Earth's magnetosphere. On the Moon, all the locations explored so far have cratered surfaces covered with loose regolith of several meters. We can therefore assume that the entire surface is covered by regolith although the thickness may vary. The optical parameters of the dust particles depend on the composition and can vary significantly, not only over a wide region, but even locally in a microscopic scale. For the glassy component, the real part values range from 1.570 to 1.749 and they vary directly with the total Fe and Ti contents and inversely with the Al content [242]. The imaginary part of the complex refractive index is more variable and can span from 0.0005 up to 0.15 according to the composition and wavelength of interest.

Mars, the fourth planet of our Solar System, has many features in common with the Earth. Much of the Martian surface is covered by unconsolidated soils (dust) derived from impact, aeolian and other sedimentary processes. Such dust is likely produced by impacts early in Mars' history and subsequently recycled at its surface. Sometimes the wind in Mars' thin atmosphere blows the dust on the surface into dust storms, carrying dust particles up to altitudes of about 50 km. The composition of the Martian dust grains was obtained from several space missions, starting from the mission Mariner 9 in 1971 [243]. Data from recent rovers (e.g. MSL Curiosity at Rocknest, Oct 2012) and orbital spacecraft show that Martian surface is dominated by a soil (dust grain dimension less than 150 μ m) of basaltic composition with primarily pyroxene, plagioclase feldspar, and olivine, as well as minor amounts of Fe and Ti oxides (e.g., magnetite, ilmenite, and hematite) and alteration minerals (e.g., sulfates, phyllosilicates, and carbonates) [244–247]. In general, dust particle sizes range from 1.2 to about 4.1 μ m with a mean dust radius of about 1.6-1.8 μ m [248, 249] and a positive correlation between dust opacity and particle sizes. Phase function results show asymmetry parameter values of $q = 0.601 \pm 0.108$ for high atmospheric dust loading scenarios and $g = 0.710 \pm 0.065$ for non-dusty periods. Regarding the shape of the particles, considering a modified log-normal aspect ratio distribution for a mixture of spheroids, data suggest more elongated particles are present during dust storms, with aspect ratios of 2.8 ± 0.9 for high-opacity days, in contrast to values of 1.8 measured during post-storm period. The particles single scattering albedo is found ranging in the solar band between 0.89-0.90 (dark regions) and 0.92-0.94 (bright regions) [250].

6.3 Solar radiation pressure and optical trapping of dust particles

6.3.1 Models

The diversity of dust particles in an astrophysical context implies a richness of models that we need to build in order to calculate realistic optical forces for space tweezers applications. Here, we consider several models of extra-planetary dusts with shape and composition inspired by interstellar, interplanetary, and planetary particles. Some of them have been collected in the DUSTER mission [224, 251], others are inspired by the particles that could be collected on the Moon or on Mars. We show these models in Fig. 6.1. On the top row, the homogeneous and stratified single/aggregated spheres emulate hypothetical interstellar dust grains whose constituents are olivine and aliphatic carbon [195]. The olivine refractive index is provided by Draine & Li while the carbon one by Ashok et al. [213, 218]. In Figs. 6.1a, 6.1b, the constituents refractive indexes are mixed in such a way as to treat the particle homogeneously with a single effective refractive index according to the Bruggeman criterion [227]. On the other hand, in Figs. 6.1c, 6.1d, an olivine core is considered covered by a carbon layer [195]. The spheres radius of Figs. 6.1a, 6.1c is $r = 0.15 \ \mu m$. On the other hand, the clusters of Figs. 6.1b, 6.1d are composed by 9 spheres of different sizes with the major semi-axis $r = 0.16 \ \mu m$. In Fig. 6.1e, we present a model according to a Field Emission Scanning Electron Microscope (FESEM) image of a silica particle clustering arranged in quenched melt spheres, shown in Fig. 6.1f, and collected in the DUSTER mission [224]. The refractive index is provided by Malitson [252]. The model is composed by 4 spheres of different radius with the major semi-axis $r = 0.23 \ \mu m$. Fig. 6.1g represents the model of condensed Ca[O] nanograins that are accreted onto a larger melted aggregate of tiny carbonate grains, shown in the TEM image of Fig. 6.1h, and collected in the DUSTER mission [224, 251]. The larger sphere is calcite and the other spheres are CaO. The cluster model is composed by 30 spheres with the calcite refractive index provided by Ghosh while the calcium oxide one is provided by Liu & Sieckmann [253, 254]. Moreover, its major semi-axis has $r = 0.25 \ \mu m$. In Fig. 6.1i, a spherical model ($r = 1.25 \ \mu m$) of the particle Fe,Mg-rich TP2, collected during DUSTER mission, is shown [223]. We consider an effective refractive index obtained mixing iron (67%) and magnesium (33%), according to the



Figure 6.3: Extinction cross-sections, $\langle \sigma_{\text{ext}} \rangle$, and optical trapping force, F, components along x (blue line), y (red line), and z (yellow dots) when the particle is trapped in water (center column) or in air (right column). Results in (a-c) are related to the olivine-aliphatic carbon core-shell sphere. (d-f) show calculations for the olivine-aliphatic carbon core-shell cluster. (g-i) concern the quenched melt silica. (j-l) show the results for the bunch-of-grape carbon Ca-rich. (m-o) are the results for the 'TP2' sphere. For the optical trapping calculations the laser power is fixed at 50 mW and the wavelength at 0.83 μ m.

Bruggeman criterion whose the two refractive indexes are respectively provided by Johnson & Christy and Hagemann [142, 227, 255]. In Fig. 6.1j, a spherical model of Martian hematite $(r = 1 \ \mu m)$ [225]. In Fig. 6.1k, a spherical model of Lunar regolith $(r = 1 \ \mu m)$ [226]. Fig. 6.1l shows the model of a microscale fassaite ellipsoidal, collected during DUSTER mission, and shown in a TEM image of Fig. 6.1m [224]. We consider an effective refractive index constructed mixing silica (65 %), and CaO (35 %) according to the Bruggeman criterion [227], the major semi-axis is 2 μm . All non-spherical models are oriented in such a way that their major axis is aligned with the incident light propagation direction.

The calculation of the solar radiation pressure and optical trapping forces on model particles of Figs. 6.1a - 6.1k is carried out with the T-matrix formalism because their size parameters fall within the range $x \approx [0.1 - 6]$. On the other hand, when the particle size parameter is too high (e.g. $x \ge 10$) such as in the fassaite micro ellipsoid of Fig. 6.1l, the calculation is carried out in the ray optics approximation. For this case we exploited the optimized computational MATLAB codes for dielectric particles provided by Callegari et al. [65] (http://opticaltweezers.org/software/otgo-optical-tweezersgeometrical-optics/).

6.3.2 Results

Solar radiation pressure calculations are important to understand its relevance in optical trapping applications in space. In general external forces, such as solar radiation pressure, can have detrimental effects on optical trapping of particles in space or the high atmosphere. Here we aim to show that T-matrix methods can be used to give accurate estimates of these effects on individual dust particles. The total radiation force, $\bar{\mathbf{F}}$, and force spectrum, $f_s(\lambda)$, that the Sun exerts on particles are calculated as:

$$\bar{F}_s = \int_{\lambda} \mathrm{d}\lambda \ f_s(\lambda) \ , \tag{6.1}$$

$$f_s(\lambda) = \frac{i_{\odot}(\lambda)}{I_{\odot}} F_{\mathrm{rad},s}(\lambda) , \qquad (6.2)$$

where the index $s = (\parallel, \perp)$ specifies respectively the parallel and the orthogonal component of $F_{\mathrm{rad},s}(\lambda)$ respect to the radiation incident direction, $\hat{\mathbf{k}}$. The expressions of $F_{\mathrm{rad},s}(\lambda)$ are the wavelength dependent optical forces calculated in the ray optics approximation [15] or in the T-matrix approach [2] according to the different models under investigation. The term $f_s(\lambda)$ specifies the spectral force distribution obtained scaling the computation outputs

	r	$\sigma_{ m geom}$	\overline{F}_{\parallel}	\overline{F}_{\perp}	σ_{ext}	n	k	κz	ñ	κz	ñ	
MODELS	(µm)	(µm²)	(pN)		λ=0.83 μm (μm²)	λ=	<i>λ</i> =0.83 μm		WATER (pN·µ		AIR/VACU UM µm-1)	
	0.15	7.06·10 ⁻²	55.45	0	3.79·10 ⁻²	2.59	2.59 4.73.10-2		No trapping			
3 0%	0.16	1.98∙10 ⁻²	0.93	3.09∙10 ⁻³	2.4·10 ⁻⁴			6	25	2	14.5	
	0.15	7.06·10 ⁻²	60.74	0	4.11·10 ⁻²	(2.94	ORE 0.1	No trapping			J	
÷.	0.16	1.98∙10 ⁻²	0.95	3.08∙10 ⁻³	2.42·10 ⁻⁴	SHELL 2.28 8.83·10 ⁻⁵		5.5	24.5	2	14.5	
6	0.23	7.85∙10 ⁻²	15.78	0.2	7.96∙10 ⁻³	2.11	0	42.5	185.5	25.5	158.5	
	0.25	1.77·10 ⁻¹	18.2	1.46	1.15∙10 ⁻²	2.72 1.94	CaCO ₃ 0 CaO 0	63	248.5	31	171	
	1.25	4.91	7.61·10 ³	0	12.17		20.0		No tra	pping	I	
	0.05	7.85·10 ⁻³	18.06	0	4.74·10 ⁻³	- 35	20.9	27	62	2	13.5	
	1	3.14	5.31·10 ³	0	6.84	8.02	0.14	No trapping			I	
	0.1	3.14·10 ⁻²	70.66	0	2.19·10 ⁻²	8.03	0.14	148.5	433.5	33	151	
	1	3.14	2,49	0	10.85	1.69	6 72 10-2		No tra	pping	I	
	0.1	3.14·10 ⁻²	2.49∙10 ³	0	2.2·10 ⁻³	1.00	0.73.10*	19	89	9	57	
	2	12.57	2.56·10 ³	0	-	2.42	0.11	No trap		pping	I	

Table 6.1: Summary table of radiation pressure and optical trapping properties. For each model particle we show: the radius, r, or the radius of the smallest sphere enclosing the cluster; the geometric cross-section, σ_{geom} ; the integrated optical force on the solar intensity spectrum along the parallel, \bar{F}_{\parallel} , and perpendicular direction, \bar{F}_{\perp} , with respect to the light propagation; the extinction cross-section σ_{ext} at the trapping wavelength $\lambda = 0.83 \ \mu\text{m}$; the real, n, and the imaginary part, k, of the refractive index at the trapping wavelength $\lambda = 0.83 \ \mu\text{m}$; and the optical trap stiffnesses along the optical axis, κ_z , and perpendicular to it, $\tilde{\kappa}$, in water and in air.

 $F_{\text{rad},s}(\lambda)$ upon the solar radiation intensity distribution $i_{\odot}(\lambda)$ normalized at the solar irradiance I_{\odot} , *i.e.*, $I_{\odot} = \int_{\lambda} d\lambda \ i_{\odot}(\lambda) = 1.34 \text{ kW/m}^2$ [256]. Therefore, the considered spectrum is chosen in the range $\lambda = [0.28 - 2] \ \mu\text{m}$ that is when the light emission by the Sun has the maximum intensity.

In the Fig. 6.2, we plot the solar radiation force distributions $f_s(\lambda)$ (Eq. 6.2) for all the models presented in Fig. 6.1. In particular, we show the force, $f_{\parallel}(\lambda)$, parallel to the propagation direction as a red line, and the perpendicular one, $f_{\perp}(\lambda)$, as a blue line. Figs. 6.2a - 6.2d concern the calculation on different models for the olivine-aliphatic carbon structures. Fig. 6.2e is referred to the quenched melt silica, Fig. 6.2f the bunch-of-grape carbon Carich, Fig. 6.2g the 'TP2' sphere, and Fig. 6.2h the martian hematite sphere. Finally, Fig. 6.2i shows the fassaite ellipsoid for which the radiation force is calculated with ray optics. We note how the parallel component, f_{\parallel} , is much larger than the perpendicular one, f_{\perp} . In fact, the parallel component is proportional to the particle extinction cross section, while the transverse one is related to the asymmetry parameters, g_i , quantifying the non-sphericity of the scatterer [2, 59, 61] (Eq. 1.11). Thus, for cylindrically symmetric particles the component f_{\perp} is zero (Figs. 6.2a, 6.2c, 6.2g, 6.2h, 6.2i). Figs. 6.2b, 6.2d, 6.2e show results for elongated nanoscale clusters that generally align with the incident electric field direction [90] yielding a low value of f_{\perp} . In Fig. 6.2f we report the highly non-symmetrical model of the bunch-of-grape carbon Ca-rich for which f_{\perp} is quite strong and $F_{\perp} \simeq 1.46$ pN, a value comparable to $\bar{F}_{\parallel} \simeq 18.2$ pN (Tab. 6.1). In summary, the parallel component of the solar radiation pressure, f_{\parallel} , describes an optomechanical interaction of the solar radiation pressure with the extra-planetary dust models in the tens of piconewton range. On the other hand, for non-spherical particles the perpendicular component of the solar radiation pressure can drive more complex transverse or rotational dynamics [257].

We now focus on the systematic characterization of optical trapping forces in OT, *i.e.*, a single Gaussian beam focused by a high-NA objective. In our calculations we fix the laser wavelength at 0.83 μ m, that is a typical wavelength for OT experiments in the near-infrared, and the power P = 50 mW. In Fig. 6.3, we show the three Cartesian components of the trapping force in the neighborhood of the OT paraxial nominal focus placed at the origin of the coordinate system (x = y = z = 0). The trapping position of the particle in the axial z-direction does not typically coincide with the origin because of the 'pushing' effect of the optical scattering force. To calculate the force on the particle at the equilibrium position $C_{eq} = (x_{eq}, y_{eq}, z_{eq})$, the z axial coordinate at which the axial force vanishes must firstly be found. Hence, the force plots in the transverse directions (x, y) can then be calculated. In the left column of the Fig. 6.3, we present the extinction cross-sections σ_{ext} for the visible and near-infrared wavelength spectrum. The extinction cross section, σ_{ext} , takes into account the rate at which the energy is removed from the electromagnetic wave through scattering and the absorption, allowing us to understand how effectively trapping takes place [15]. The trapping arises when the focused incoming field generates a restoring force proportional to the particle's displacement from an equilibrium point, and that, for small displacements, behaves harmonically [7]. Therefore, trap stiffnesses are defined as:

$$\kappa_i \equiv \frac{\mathrm{d}F_i}{\mathrm{d}x_i}\Big|_{x_{\mathrm{eq},i}} \,. \tag{6.3}$$

We calculate optical trapping in water $(n_{\rm m} = 1.33)$ with an objective NA = 1.3 (middle column of Fig. 6.3) and in air or vacuum $(n_{\rm m} = 1)$ with NA = 0.9 (right column of Fig. 6.3). We note that here we focus only on electromagnetic calculations, neglecting thermal fluctuations and hydrodynamics effects due to the surrounding medium [143]. Generally, for a given particle, optical trapping in water is stabilized by the overdamped viscous dynamics in the fluid, while in air or vacuum the underdamped dynamics might be more critical for stable optical trapping [169]. Moreover, the higher NA in water causes the equilibrium point C_{eq} to be closer to the nominal focus than in air. This is confirmed by comparing the graphs in Fig. 6.3, central column (in water), with those of the right column (in air). The dielectric particles, such as the quenched melt silica (Figs. 6.3h, 6.3i), and carbonate cluster (Figs. 6.3k, 6.3l) exhibit fairly stable trapping. Even the interstellar dust model of a sphere aggregate can be trapped thanks to the relatively low extinction (Figs. 6.3e, 6.3f). Indeed, for these model particles we are able to extrapolate the trapping constants along the axial direction, κ_z , and along the perpendicular direction, $\tilde{\kappa} \equiv (\kappa_x + \kappa_y)/2$ (Tab. 6.1). On the other hand, model particles, like the interstellar dust sphere (Figs. 6.3b, 6.3c), or the Fe-Mg sphere (Figs. 6.3n, 6.3o), can not be trapped neither in water nor in air. A similar behavior is also exhibited by the hematite sphere, regolith sphere, and the ellipsoidal fassaite as shown in Tab. 6.1 by observing the corresponding trapping constants. The reason is to be found in their large size (Fe-Mg sphere, hematite, regolith, and fassaite) and on their strong absorption (interstellar dust, Fe-Mg sphere, hematite, regolith and fassaite). To further confirm, we have calculated the trapping properties of the Fe-Mg, hematite spheres by reducing their size such that $r_{\rm Fe-Mg} = 0.05 \ \mu m$ and $r_{\rm hem} = 0.1$ μ m. In this way, the trap stiffnesses can be extrapolated (Tab. 6.1) and a quantitative indication of σ_{ext} is provided in Tab. 6.1. This behaviour is similar to the optical trapping of metal nanoparticles that can be efficiently trapped at small size where absorption and extinction cross sections are still small in the near-infrared [13].
Conclusions

In this thesis, different applications of electromagnetic scattering have been studied both in the transition matrix formalism and with appropriate approximations depending on the performed studies. Several systems have been analyzed ranging from optical trapping modeling, going to some specific plasmonic investigations, through fundamental physics problems to space applications. After describing in details the light scattering theory in the Tmatrix approach and optical tweezers, we analyzed four specific applications.

First, we studied optical trapping of hybrid core-shell nanoparticles with gain in a counter-propagating Gaussian beam configuration, highlighting the non-linear optical scaling of optical forces. We performed Brownian motion simulations in water, where we directly observed how the particle dynamics is more confined for increasing gain. We showed that by changing the light wavelength with respect to the nanoshell resonance it is possible to switch the sign of the optical forces and use the dual-beam configuration for position locking (red detuning) or channelling (blue detuning) of particles in a microfluidic flow.

Secondly, we investigated the onset of spin-dependent optical forces related to the Belinfante's linear spin momentum. In particular, we studied the role of shape in the occurrence of transverse spin-dependent optical force components in tilted ZnO nanowires illuminated by either a circularly polarized plane wave or a focused laser beam. In fact, because of the breaking of the cylindrical symmetry with respect to the optical axis z, a non-conservative spin-dependent component occurs when circularly polarized light is incident on the tilted nanowire. The strong connection between the non-local transverse y-component and the spin density has been demonstrated by analyzing the behavior of the spin distribution considering configurations in which the cylindrical symmetry is increasingly broken.

In the last part of the thesis, some applications of light scattering theory are exploited in an astrophysical context. More specifically, we focused on the modeling of interstellar dust properties by studying the polarization of the 3.4 μ m carbon absorption feature in the diffuse interstellar medium. The

non-detection of polarization in this feature, together with the observation of a strong polarization in the 9.7 μ m silicate feature, has seriously challenged the silicate core-carbon mantle dust model. We investigated the problem, trying to relax the core-mantle model crisis. Only for a particular model of aggregated core-mantle grains, which we have called the 'Head-tail cluster' model, the polarimetric results satisfy the observational constraints. Despite our results seem to confirm the doubts on the validity of the core-mantle model, there is an interesting point that still deserves to be discussed and that could open new perspectives. In fact, within an evolutionary history picture connected with the carbon cycle in the interstellar medium, the carbon mantle in stratified dust grains would not be responsible for the 3.4 μ m feature that would be rather due to small, non aligned, aliphatic hydrocarbon grains. Then, silicate and carbon could still co-exist in a core-mantle structure without being in conflict with the observational polarization constraints.

Finally, we studied computationally the solar radiation force and optical trapping properties for different cosmic dust particles. We showed that the radiation force exerted by the Sun is not trivially negligible and it can actively influence the dynamics of model dust particles at the nano- and microscale when compared to, e.q., optical trapping forces. Furthermore, we calculated single-beam optical trapping properties for cosmic dust model particles both in water and in air with parameters close to typical optical tweezers experiments in the near-infrared. We found that weakly absorbing and nanoscale particles can be captured, while particles with stronger absorption show a scattering component of the optical force that prevents optical trapping in a standard single-beam optical tweezers. This detrimental effects can be overcome through dual-beam traps made by stationary counter-propagating laser beams. This study opens perspectives for the application of optical tweezers techniques to solar system study, e.q., cometary particles analyses including the volatile component, dust particles in the Martian atmosphere and/or on the Martian and Lunar surfaces. Such applications will also be strategic for the "clean" handling/preliminary characterization of planetary particulate matter in curation facilities and can pave the way for future space applications and in situ analysis of planetary bodies.

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Bibliography

- Waterman P.C. Symmetry, unitarity, and geometry in electromagnetic scattering. *Physical Review D*, 3(4):825, 1971.
- [2] Fernando Borghese, Paolo Denti, and Rosalba Saija. Scattering from model nonspherical particles: Theory and applications to environmental physics. Springer Science & Business Media, 2007.
- [3] Mie G. Beiträge zur optik trüber medien, speziell kolloidaler metallösungen. Annalen der Physik, 330(3):377–445, 1908.
- [4] Mishchenko M.I., Travis L.D., and Lacis A.A. Scattering, absorption, and emission of light by small particles. Cambridge University Press, 2002.
- [5] Fernando Borghese, Denti P., Saija R., and Iatì M. A. Optical trapping of nonspherical particles in the T-matrix formalism. *Optics Express*, 15(19):11984–11998, 2007.
- [6] P. H. Jones, O. M. Maragò, and G. Volpe. Optical Tweezers: Principles and Applications. Cambridge University Press, Cambridge, UK, 2015.
- [7] Polimeno P., Magazzù A., Iatì M. A., Patti F., Saija R., Degli Esposti Boschi C., Donato M. G., Gucciardi P. G., Jones P. H., Volpe G., and Maragò O. M. Optical tweezers and their applications. *Journal of Quantitative Spectroscopy & Radiative Transfer*, 218:131–150, 2018.
- [8] Paolo Polimeno, Rosalba Saija, Cristian Degli Esposti Boschi, Onofrio M Maragò, and Maria Antonia Iatì. Optical forces in the t-matrix formalism. Atti della Accademia Peloritana dei Pericolanti-Classe di Scienze Fisiche, Matematiche e Naturali, 97(1):2, 2019.
- [9] P Polimeno, MA Iatì, C Degli Esposti Boschi, SH Simpson, V Svak, O Brzobohatỳ, P Zemánek, OM Maragò, and R Saija. T-matrix calculations of spin-dependent optical forces in optically trapped nanowires. *The European Physical Journal Plus*, 136(1):1–15, 2021.

- [10] Saija R., Iatì M.A, Borghese F., Denti P., Aiello S., and Cecchi-Pestellini C. Beyond Mie theory: The transition matrix approach in interstellar dust modeling. *The Astrophysical Journal*, 559(2):993, 2001.
- [11] Iatì M.A., Giusto A., Saija R., Borghese F., Denti P., Cecchi-Pestellini C., and Aiello S. Optical properties of composite interstellar grains: A morphological analysis. *The Astrophysical Journal*, 615(1):286, 2004.
- [12] P Polimeno, A Magazzù, MA Iatì, R Saija, L Folco, D Bronte Ciriza, MG Donato, A Foti, PG Gucciardi, A Saidi, et al. Optical tweezers in a dusty universe. *The European Physical Journal Plus*, 136(3):1–23, 2021.
- [13] Amendola V., Pilot R., Frasconi M., Maragò O.M., and Iatì M.A. Surface plasmon resonance in gold nanoparticles: A review. *Journal of Physics: Condensed Matter*, 29(20):203002, 2017.
- [14] Paolo Polimeno, Francesco Patti, Melissa Infusino, Jonathan Sánchez, Maria A Iatì, Rosalba Saija, Giovanni Volpe, Onofrio M Maragò, and Alessandro Veltri. Gain-assisted optomechanical position locking of metal/dielectric nanoshells in optical potentials. ACS Photonics, 7(5):1262–1270, 2020.
- [15] P. H. Jones, O. M. Marago, and G. Volpe. Optical tweezers: Principles and applications. Cambridge University Press, Cambridge, UK, 2015.
- [16] Fernando Borghese, Saija P., Denti R., Toscano G., and Sindoni O.I. Multiple electromagnetic scattering from a cluster of spheres. I. Theory. *Aerosol Science and Technology*, 3(2):227–235, 1984.
- [17] Purcell E.M. and Pennypacker C.R. Scattering and absorption of light by nonspherical dielectric grains. *The Astrophysical Journal*, 186:705– 714, 1973.
- [18] Draine B.T. The discrete-dipole approximation and its application to interstellar graphite grains. *The Astrophysical Journal*, 333:848–872, 1988.
- [19] Gordon J.P. Radiation forces and momenta in dielectric media. Physical Review A, 8(1):14, 1973.
- [20] Stratton J.A., Morse P.M, Chu L.J., and Hutner R.A. Elliptic cylinder and spheroidal wave functions: Including tables of separation constants and coefficients. J. Wiley & sons, inc., 1941.

- [21] Ishimaru A. Wave propagation and scattering in random media and rough surfaces. Proceedings of the IEEE, 79(10):1359–1366, 1991.
- [22] Van de Hulst H.C. Light scattering by small particles. Courier Corporation, 1957.
- [23] Fucile E., Denti P., Borghese F., Saija R., and Sindoni O.I. Optical properties of a sphere in the vicinity of a plane surface. *Journal of the Optical Society of America A*, 14(7):1505–1514, 1997.
- [24] Wyatt P.J. Scattering of electromagnetic plane waves from inhomogeneous spherically symmetric objects. *Physical Review*, 134(7AB):AB1, 1964.
- [25] Milton Abramowitz and Irene A Stegun. Handbook of mathematical functions with formulas, graphs, and mathematical table. In US Department of Commerce. National Bureau of Standards Applied Mathematics series 55, 1965.
- [26] Bo ÅS Gustafson, Ludmilla Kolokolova, Yu-lin Xu, J Mayo Greenberg, and Ralf Stognienko. Interactions with electromagnetic radiation: theory and laboratory simulations. In *Interplanetary dust*, pages 509–567. Springer, 2001.
- [27] Bruning J. and Lo Y. Multiple scattering of e.m. waves by spheres part I. Multipole expansion and ray-optical solutions. *IEEE Transactions* on Antennas and Propagation, 19(3):378–390, 1971.
- [28] Peterson B. and Ström S. T-matrix formulation of electromagnetic scattering from multilayered scatterers. *Physical Review D*, 10(8):2670, 1974.
- [29] Gerardy J.M. and Ausloos M. Absorption spectrum of clusters of spheres from the general solution of Maxwell's equations. The longwavelength limit. *Physical Review B*, 22(10):4950, 1980.
- [30] Borghese F., Denti P., Toscano G., and Sindoni O.I. An addition theorem for vector helmholtz harmonics. *Journal of Mathematical Physics*, 21(12):2754–2755, 1980.
- [31] Michael Quinten, Uwe Kreibig, Thomas Henning, and Harald Mutschke. Wavelength-dependent optical extinction of carbonaceous particles in atmospheric aerosols and interstellar dust. *Applied optics*, 41(33):7102–7113, 2002.

- [32] Warren J Wiscombe. Mie scattering calculations: Advances in technique and fast, vector-speed computer codes, volume 10. National Technical Information Service, US Department of Commerce, 1979.
- [33] Kepler J. De cometis libelli tres, 1619.
- [34] Maxwell J.C. A Treatise on electricity and magnetism. Clarendon Press, Oxford, 1873.
- [35] John Henry Poynting. Xv. on the transfer of energy in the electromagnetic field. *Philosophical Transactions of the Royal Society of London*, 175:343–361, 1884.
- [36] Adolfo Bartoli. Il calorico raggiante e il secondo principio di termodinamica. Il Nuovo Cimento (1877-1894), 15(1):193-202, 1884.
- [37] Lebedev P. Untersuchungen über die druckkräfte des lichtes. Annalen der Physik, 311(11):433–458, 1901.
- [38] Nichols E.F. and Hull G.F. A preliminary communication on the pressure of heat and light radiation. *Physics Review*, 13:307–320, 1901.
- [39] TH Maiman. Stimulated optical radiation in ruby. Nature, 187:493– 494, 1960.
- [40] Ashkin A. Acceleration and trapping of particles by radiation pressure. *Physical Review Letters*, 24(4):156, 1970.
- [41] Ashkin A. Atomic-beam deflection by resonance-radiation pressure. *Physical Review Letters*, 25(19):1321, 1970.
- [42] Ashkin A. History of optical trapping and manipulation of smallneutral particle, atoms, and molecules. *IEEE Journal of Selected Topics* in Quantum Electronics, 6(6):841–856, 2000.
- [43] Ashkin A. and Dziedzic J.M. Optical levitation by radiation pressure. Applied Physics Letters, 19(8):283–285, 1971.
- [44] Arthur Ashkin, Dziedzic J.M., Bjorkholm J.E., and Chu S. Observation of a single-beam gradient force optical trap for dielectric particles. *Optics Letters*, 11(5):288–290, 1986.
- [45] Dholakia K. and Čižmár T. Shaping the future of manipulation. Nature Photonics, 5(6):335–342, 2011.

- [46] Padgett M. and Bowman R. Tweezers with a twist. Nature Photonics, 5(6):343–348, 2011.
- [47] Maragò O.M., Jones P.H., Gucciardi P.G., Volpe G., and Ferrari A.C. Optical trapping and manipulation of nanostructures. *Nature Nan-otechnology*, 8(11):807–819, 2013.
- [48] Magazzú A. Optical trapping and thermal dynamics of Silicon nanowires. PhD thesis, Universitá degli studi di Messina, 2015.
- [49] Magazzú A., Spadaro D., Donato M.G., Sayed R., Messina E., D'Andrea C., Foti A., Fazio B., Iatí M.A., Irrera A., et al. Optical tweezers: A non-destructive tool for soft and biomaterial investigations. *Rendiconti Lincei*, 26(2):203–218, 2015.
- [50] Neuman K.C. and Nagy A. Single-molecule force spectroscopy: Optical tweezers, magnetic tweezers and atomic force microscopy. *Nature Methods*, 5(6):491–505, 2008.
- [51] Ashkin A., Dziedzic J.M., and Yamane T. Optical trapping and manipulation of single cells using infrared laser beams. *Nature*, 330(6150):769–771, 1987.
- [52] Ashkin A. and Dziedzic J.M. Optical trapping and manipulation of viruses and bacteria. *Science*, 235:1517–1521, 1987.
- [53] Fazal F.M. and Block S.M. Optical tweezers study life under tension. *Nature Photonics*, 5(6):318–321, 2011.
- [54] Svoboda K. and Block S.M. Biological applications of optical forces. Annual Review of Biophysics and Biomolecular Structure, 23(1):247– 285, 1994.
- [55] Maragò O.M., Gucciardi P.G., and Jones P.H. Photonic force microscopy: From femtonewton force sensing to ultra-sensitive spectroscopy. In Scanning Probe Microscopy in Nanoscience and Nanotechnology, pages 23–56. Springer, 2010.
- [56] Marago O.M., Jones P.H., Bonaccorso F., Scardaci V., Gucciardi P.G., Rozhin A.G., and Ferrari A.C. Femtonewton force sensing with optically trapped nanotubes. *Nano Letters*, 8(10):3211–3216, 2008.
- [57] Irrera A., Artoni P., Saija R., Gucciardi P.G., Iatì M.A., Borghese F., Denti P., Iacona F., Priolo F., and Marago O.M. Size-scaling in optical trapping of silicon nanowires. *Nano Letters*, 11(11):4879–4884, 2011.

- [58] Irrera A., Magazzú A., Artoni P., Simpson S.H., Hanna S., Jones P.H., Priolo F., Gucciardi P.G., and Maragó O.M. Photonic torque microscopy of the nonconservative force field for optically trapped silicon nanowires. *Nano Letters*, 16(7):4181–4188, 2016.
- [59] Mishchenko M.I. Radiation force caused by scattering, absorption, and emission of light by nonspherical particles. *Journal of Quantitative* Spectroscopy and Radiative Transfer, 70(4-6):811–816, 2001.
- [60] Timo A. Nieminen, Halina Rubinsztein-Dunlop, and Norman R. Heckenberg. Calculation and optical measurement of laser trapping forces on non-spherical particles. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 70(4-6):627–637, 2001.
- [61] Rosalba Saija, Iatì M.A., Giusto A., Denti P., and Borghese F. Transverse components of the radiation force on nonspherical particles in the T-matrix formalism. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 94(2):163–179, 2005.
- [62] B Piccirillo, S Slussarenko, L Marrucci, and E Santamato. The orbital angular momentum of light: genesis and evolution of the concept and of the associated photonic technology. La Rivista del Nuovo Cimento, 36(11):501–554, 2013.
- [63] Cohen-Tannoudji C., Dupont-Roc J., and Grynberg G. Atom-photon interactions: Basic processes and applications. John Wiley & Sons, Inc., New York, NY, 1992.
- [64] M. Born and E. Wolf. Principles of optics: Electromagnetic theory of propagation, interference and diffraction of light. Cambridge University Press, Cambridge, United Kingdom, 1999.
- [65] Callegari A., Mijalkov M., Gököz A.B., and Volpe G. Computational toolbox for optical tweezers in geometrical optics. JOSA B, 32(5):B11– B19, 2015.
- [66] Ashkin A. Forces of a single-beam gradient laser trap on a dielectric sphere in the ray optics regime. *Biophysical Journal*, 61(2):569–582, 1992.
- [67] Swartzlander jr G.A., Peterson T.J., Artusio-Glimpse A.B., and Raisanen A.D. Stable optical lift. *Nature Photonics*, 5(1):48–51, 2011.

- [68] Skelton S.E., Sergides M., Memoli G., Maragó O.M., and Jones P.H. Trapping and deformation of microbubbles in a dual-beam fibre-optic trap. *Journal of Optics*, 14(7):075706, 2012.
- [69] Wolf E. Electromagnetic diffraction in optical systems. I. An integral representation of the image field. In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, volume 253, pages 349–357. The Royal Society, 1959.
- [70] Bernard Richards and Emil Wolf. Electromagnetic diffraction in optical systems, ii. structure of the image field in an aplanatic system. *Proceedings of the Royal Society of London. Series A. Mathematical* and Physical Sciences, 253(1274):358–379, 1959.
- [71] Peter Debye. Das verhalten von lichtwellen in der nähe eines brennpunktes oder einer brennlinie. Annalen der Physik, 335(14):755–776, 1909.
- [72] Kishan Dholakia and Pavel Zemánek. Colloquium: Gripped by light: Optical binding. *Reviews of modern physics*, 82(2):1767, 2010.
- [73] Draine B.T. and Goodman J. Beyond clausius-mossotti-wave propagation on a polarizable point lattice and the discrete dipole approximation. *The Astrophysical Journal*, 405:685–697, 1993.
- [74] Chaumet P.C. and Nieto-Vesperinas M. Time-averaged total force on a dipolar sphere in an electromagnetic field. *Optics Letters*, 25(15):1065– 1067, 2000.
- [75] Arias-González J. R. and Nieto-Vesperinas M. Optical forces on small particles: attractive and repulsive nature and plasmon-resonance conditions. JOSA A, 20(7):1201–1209, 2003.
- [76] Albaladejo S., Laroche M.I., Marquésand M., and Sáenz J.J. Scattering forces from the curl of the spin angular momentum of a light field. *Physical Review Letters*, 102(11):113602, 2009.
- [77] Marqués M.I. and Sáenz J.J. Reply to comment on scattering forces from the curl of the spin angular momentum of a light field. *Physical Review Letters*, 111(5):059302, 2013.
- [78] Donato M.G., Vasi S., Sayed R., Jones P.H., Bonaccorso F., Ferrari A.C., Gucciardi P.G., and Maragò O.M. Optical trapping of nanotubes with cylindrical vector beams. *Optics Letters*, 37(16):3381–3383, 2012.

- [79] Skelton S.E., Sergides M., Saija R., Iatì M.A., Maragó O.M., and Jones P.H. Trapping volume control in optical tweezers using cylindrical vector beams. *Optics Letters*, 38(1):28–30, 2013.
- [80] Marqués M.I. Beam configuration proposal to verify that scattering forces come from the orbital part of the poynting vector. *Optics Letters*, 39(17):5122–5125, 2014.
- [81] Foot C.J. Atomic physics, volume 7. Oxford University Press, 2005.
- [82] Oto Brzobohatỳ, Martin Siler, Jan Trojek, Lukáš Chvátal, Vítězslav Karásek, Aleš Paták, Zuzana Pokorná, Filip Mika, and Pavel Zemánek. Three-dimensional optical trapping of a plasmonic nanoparticle using low numerical aperture optical tweezers. *Scientific reports*, 5(1):1–9, 2015.
- [83] Oto Brzobohatỳ, Alejandro V Arzola, Martin Šiler, Lukáš Chvátal, Petr Jákl, Stephen Simpson, and Pavel Zemánek. Complex rotational dynamics of multiple spheroidal particles in a circularly polarized, dual beam trap. *Optics express*, 23(6):7273–7287, 2015.
- [84] Stefan Kuhn, Peter Asenbaum, Alon Kosloff, Michele Sclafani, Benjamin A Stickler, Stefan Nimmrichter, Klaus Hornberger, Ori Cheshnovsky, Fernando Patolsky, and Markus Arndt. Cavity-assisted manipulation of freely rotating silicon nanorods in high vacuum. Nano letters, 15(8):5604–5608, 2015.
- [85] Stefan Kuhn, Alon Kosloff, Benjamin A Stickler, Fernando Patolsky, Klaus Hornberger, Markus Arndt, and James Millen. Full rotational control of levitated silicon nanorods. *Optica*, 4(3):356–360, 2017.
- [86] P Zemánek, A Jonáš, L Srámek, and M Liška. Optical trapping of rayleigh particles using a gaussian standing wave. Optics communications, 151(4-6):273–285, 1998.
- [87] Silvie Bernatová, Maria Grazia Donato, Jan Ježek, Zdeněk Pilát, Ota Samek, Alessandro Magazzù, Onofrio M Maragò, Pavel Zemánek, and Pietro G Gucciardi. Wavelength-dependent optical force aggregation of gold nanorods for sers in a microfluidic chip. J. Phys. Chem. C, 123(9):5608–5615, 2019.
- [88] Simpson S.H. Inhomogeneous and anisotropic particles in optical traps: Physical behaviour and applications. *Journal of Quantitative Spec*troscopy and Radiative Transfer, 146:81–99, 2014.

- [89] Timo A Nieminen, Nathaniel du Preez-Wilkinson, Alexander B Stilgoe, Vincent LY Loke, Ann AM Bui, and Halina Rubinsztein-Dunlop. Optical tweezers: Theory and modelling. *Journal of Quantitative Spec*troscopy and Radiative Transfer, 146:59–80, 2014.
- [90] Fernando Borghese, Denti P., Saija R., Iatì M.A., and Maragò O.M. Radiation torque and force on optically trapped linear nanostructures. *Physical Review Letters*, 100(16):163903, 2008.
- [91] Borghese F., Denti P., Saija R., and Iatì M.A. Radiation torque on nonspherical particles in the transition matrix formalism. *Optics Express*, 14(20):9508–9521, 2006.
- [92] Stephen H. Simpson and Simon Hanna. Numerical calculation of interparticle forces arising in association with holographic assembly. *Journal* of the Optical Society of America A, 23(6):1419–1431, 2006.
- [93] Stephen H. Simpson and Simon Hanna. Optical trapping of spheroidal particles in gaussian beams. Journal of the Optical Society of America A, 24(2):430–443, 2007.
- [94] Rosalba Saija, Denti P., Borghese F., Maragó O.M., and Iatì M.A. Optical trapping calculations for metal nanoparticles. Comparison with experimental data for Au and Ag spheres. *Optics Express*, 17(12):10231–10241, 2009.
- [95] Timo A. Nieminen, Loke V.L.Y., Stilgoe A.B., Heckenberg N.R., and Rubinsztein-Dunlop H. T-matrix method for modelling optical tweezers. *Journal of Modern Optics*, 58(5-6):528–544, 2011.
- [96] Marston P.L. and Crichton J.H. Radiation torque on a sphere caused by a circularly-polarized electromagnetic wave. *Physical Review A*, 30(5):2508, 1984.
- [97] Wolf E.L. Nanophysics and nanotechnology: An introduction to modern concepts in nanoscience. John Wiley & Sons, 2015.
- [98] Alivisatos A.P. Semiconductor clusters, nanocrystals and quantum dots. Science, 271(5251):933–937, 1996.
- [99] M.S. Gudiksen, Lauhon L-J, J. Wang, Smith. D. C., and C. M. Lieber. Growth of nanowire superlattice structures for nanoscale photonics and electronics. *Nature*, 415(6872):617, 2002.

- [100] K. A. Dick. A review of nanowire growth promoted by alloys and non-alloying elements with emphasis on Au-assisted III–V nanowires. *Progress in Crystal Growth and Characterization of Materials*, 54(3-4):138–173, 2008.
- [101] Huang Y., Duan X., Cui Y., Lauhon L.J., Kim K.H., and Lieber C.M. Logic gates and computation from assembled nanowire building blocks. *Science*, 294(5545):1313–1317, 2001.
- [102] Reece P.J., Toe W.J., F. Wang, Paiman S., Gao Q., Tan H.H., and Jagadish C. Characterization of semiconductor nanowires using optical tweezers. *Nano Letters*, 11(6):2375–2381, 2011.
- [103] Wang F., Toe W. J., Lee W. M., McGloin D., Gao Q., Tan H. H., Jagadish C., and Reece P. J. Resolving stable axial trapping points of nanowires in an optical tweezers using photoluminescence mapping. *Nano Letters*, 13(3):1185–1191, 2013.
- [104] Nakayama Y., Pauzauskie P. J., Radenovic A., Onorato R. M., Saykally R. J., Liphardt J., and Yang P. Tunable nanowire nonlinear optical probe. *Nature*, 447(7148):1098, 2007.
- [105] Dutto F., Raillon C., Schenk K., and Radenovic A. Nonlinear optical response in single alkaline niobate nanowires. *Nano Letters*, 11(6):2517– 2521, 2011.
- [106] Roder P. B., Smith B. E., Davis E. J., and Pauzauskie P. J. Photothermal heating of nanowires. *The Journal of Physical Chemistry C*, 118(3):1407–1416, 2014.
- [107] Smith B. E., Roder P. B., Zhou X., and Pauzauskie P. J. Nanoscale materials for hyperthermal theranostics. *Nanoscale*, 7(16):7115–7126, 2015.
- [108] Trojek J., Chvátal L., and Zemánek P. Optical alignment and confinement of an ellipsoidal nanorod in optical tweezers: a theoretical study. *JOSA A*, 29(7):1224–1236, 2012.
- [109] Simpson S.H. and Hanna S. Stability analysis and thermal motion of optically trapped nanowires. *Nanotechnology*, 23(20):205502, 2012.
- [110] Maria Grazia Donato, Oto Brzobohaty, Stephen H Simpson, Alessia Irrera, Antonio Alessio Leonardi, Maria Jose Lo Faro, Vojtech Svak,

Onofrio M Marago, and Pavel Zemanek. Optical trapping, optical binding, and rotational dynamics of silicon nanowires in counterpropagating beams. *Nano Letters*, 19:342–352, 2018.

- [111] Maragó O. M., Bonaccorso F., Saija R., Privitera G., Gucciardi P. G., Iati M. A., Calogero G., Jones P. H., Borghese F., Denti P., Nicolosi V., and Ferrari A. C. Brownian motion of graphene. ACS Nano, 4(12):7515–7523, 2010.
- [112] Simpson S. H. and Hanna S. First-order nonconservative motion of optically trapped nonspherical particles. *Physical Review E*, 82:031141, 2010.
- [113] Neves A. A. R., Camposeo A., Pagliara S., Saija R., Borghese F., Denti P., Iatì M. A., Cingolani R., Maragò O. M., and Pisignano D. Rotational dynamics of optically trapped nanofibers. *Optics Express*, 18(2):822–830, 2010.
- [114] Toe W. J., Ortega-Piwonka I., Angstmann C. N., Gao Q., Tan H. H., Jagadish C., Henry B. I., and Reece P. J. Nonconservative dynamics of optically trapped high-aspect-ratio nanowires. *Physical Review E*, 93(2):022137, 2016.
- [115] Mihiretie B. M., Snabre P., Loudet J.-C., and Pouligny B. Optically driven oscillations of ellipsoidal particles. part I: Experimental observations. *The European Physical Journal E*, 37(12):124, 2014.
- [116] Sultanova N., Kasarova S., and Nikolov I. Characteristics of optical polymers in the design of polymer and hybrid optical systems. *Bulgarian Journal of Physics*, 40(3):258–264, 2013.
- [117] A. Ashkin, J.M. Dziedzic, J.E. Bjorkholm, and S. Chu. Observation of a single-beam gradient optical trap for dielectric particles. *Opt. Lett.*, 11:288–290, 1986.
- [118] A. Jonáš and P. Zemánek. Light at Work: The Use of Optical Forces for Particle Manipulation, Sorting, and Analysis. *Electophoresis*, 29:4813– 4851, 2008.
- [119] O. M. Maragò, P. H. Jones, P. G. Gucciardi, G. Volpe, and A. C. Ferrari. Optical trapping and manipulation of nanostructures. *Nat. Nanotechnol.*, 8:807–819, 2013.

- [120] Susan E Skelton Spesyvtseva and Kishan Dholakia. Trapping in a material world. ACS Photonics, 3(5):719–736, 2016.
- [121] Z. Yan, M. Pelton, L. Vigderman, E.R. Zubarev, and N.F. Scherer. Why single-beam optical tweezers trap gold nanowires in three dimensions. ACS Nano, 7:8794–8800, 2013.
- [122] Alessia Irrera, Alessandro Magazzù, Pietro Artoni, Stephen H Simpson, Simon Hanna, Philip H Jones, Francesco Priolo, Pietro Giuseppe Gucciardi, and Onofrio M Maragò. Photonic torque microscopy of the nonconservative force field for optically trapped silicon nanowires. Nano Lett., 16(7):4181–4188, 2016.
- [123] Astrid Van der Horst, Peter DJ van Oostrum, Alexander Moroz, Alfons van Blaaderen, and Marileen Dogterom. High trapping forces for high-refractive index particles trapped in dynamic arrays of counterpropagating optical tweezers. Appl. Opt., 47(17):3196–3202, 2008.
- [124] O. M. Maragò, P. H. Jones, F. Bonaccorso, V. Scardaci, P. G. Gucciardi, A. G. Rozhin, and A. C. Ferrari. Femtonewton force sensing with optically trapped nanotubes. *Nano Lett.*, 8:3211–3216, 2008.
- [125] M. G. Donato, E. Messina, A. Foti, T. J. Smart, P. H. Jones, M. A. Iatì, R. Saija, P. G. Gucciardi, and O. M. Maragò. Optical trapping and optical force positioning of two-dimensional materials. *Nanoscale*, 10:1245–1255, 2018.
- [126] Donatella Spadaro, Maria A Iatì, Maria G Donato, Pietro G Gucciardi, Rosalba Saija, Anurag R Cherlakola, Stefano Scaramuzza, Vincenzo Amendola, and Onofrio M Maragò. Scaling of optical forces on au-peg core-shell nanoparticles. *RSC Adv.*, 5(113):93139–93146, 2015.
- [127] D Spadaro, Maria A. Iatì, J Pérez-Piñeiro, C Vázquez-Vázquez, Miguel A. Correa-Duarte, Maria G. Donato, Pietro G Gucciardi, R Saija, G Strangi, and Onofrio M Maragò. Optical trapping of plasmonic mesocapsules: Enhanced optical forces and sers. J. Phys. Chem. C, 121(1):691–700, 2016.
- [128] Anni Lehmuskero, Peter Johansson, Halina Rubinsztein-Dunlop, Lianming Tong, and Mikael Kall. Laser trapping of colloidal metal nanoparticles. ACS Nano, 9(4):3453–3469, 2015.
- [129] Oto Brzobohatý, Martin Šiler, Jan Trojek, Lukáš Chvátal, Vítězslav Karásek, Aleš Paták, Zuzana Pokorná, Filip Mika, and Pavel Zemánek.

Three-dimensional optical trapping of a plasmonic nanoparticle using low numerical aperture optical tweezers. *Sci. Rep.*, 5:8106, 2015.

- [130] E. Messina, M. G. Donato, M. Zimbone, R. Saija, M. A. Iatì, L. Calcagno, M. E. Fragala, G. Compagnini, C. D'Andrea, A. Foti, P. G. Gucciardi, and O. M. Maragò. Optical trapping of silver nanoplatelets. *Opt. Express*, 23(7):8720–8730, 2015.
- [131] A. Ashkin. Acceleration and trapping of particles by radiation pressure. *Phys. Rev. Lett.*, 24(4):156, 1970.
- [132] P. Zemánek, A. Jonáš, and M. Liška. Simplified Description of Optical Forces Acting on a Nanoparticle in the Gaussian Standing Wave. J. Opt. Soc. Am. A, 19:1025–1034, 2002.
- [133] T. Cižmár, M. Siler, and P. Zemánek. An Optical Nanotrap Array Movable over a Milimetre Range. Appl. Phys. B, 84:197–203, 2006.
- [134] D. M. Gherardi, A. E. Carruthers, T. Čižmár, E. M. Wright, and K. Dholakia. A Dual Beam Photonic Crystal Fibre Trap for Microscopic Particles. *Appl. Phys. Lett.*, 93:041110, 2008.
- [135] Wolfgang Singer, Manfred Frick, Stefan Bernet, and Monika Ritsch-Marte. Self-organized array of regularly spaced microbeads in a fiberoptical trap. J. Opt. Soc. Am. B, 20(7):1568–1574, 2003.
- [136] Maria G Donato, Oto Brzobohaty, Stephen H Simpson, Alessia Irrera, Antonio A Leonardi, Maria J Lo Faro, Vojtech Svak, Onofrio M Marago, and Pavel Zemánek. Optical trapping, optical binding, and rotational dynamics of silicon nanowires in counter-propagating beams. Nano Letters, 19(1):342–352, 2019.
- [137] Alessandro Veltri and Ashod Aradian. Optical response of a metallic nanoparticle immersed in a medium with optical gain. *Physical Review* B, 85(11):115429, 2012.
- [138] Melissa Infusino, Antonio De Luca, Alessandro Veltri, Carmen Vazquez-Vazquez, Miguel A Correa-Duarte, Rakesh Dhama, and Giuseppe Strangi. Loss-mitigated collective resonances in gain-assisted plasmonic mesocapsules. ACS Photonics, 1(4):371–376, 2014.
- [139] Vincenzo Caligiuri, Luigia Pezzi, Alessandro Veltri, and Antonio De Luca. Resonant gain singularities in 1d and 3d metal/dielectric multilayered nanostructures. ACS nano, 11(1):1012–1025, 2016.

- [140] Alessandro Veltri, Arkadi Chipouline, and Ashod Aradian. Multipolar, time-dynamical model for the loss compensation and lasing of a spherical plasmonic nanoparticle spaser immersed in an active gain medium. *Scientific Reports*, 6:33018, 2016.
- [141] Luigia Pezzi, Maria Antonia Iati, Rosalba Saija, Antonio De Luca, and Onofrio M Marago. Resonant coupling and gain singularities in metal/dielectric multishells: Quasi-static versus t-matrix calculations. *The Journal of Physical Chemistry C*, 123(48):29291–29297, 2019.
- [142] Peter B Johnson and R-WJPrB Christy. Optical constants of the noble metals. *Physical review B*, 6(12):4370, 1972.
- [143] Giorgio Volpe and Giovanni Volpe. Simulation of a brownian particle in an optical trap. *American Journal of Physics*, 81(3):224–230, 2013.
- [144] Paul Langevin. Sur la théorie du mouvement brownien. Comptes Rendus Acad. Sci., 146:530–533, 1908.
- [145] P. E. Kloeden and E. Platen. Numerical Solution of Stochastic Differential Equations. Springer Verlag, Heidelberg, Germany, 1999.
- [146] Agnese Callegari and Giovanni Volpe. Numerical simulations of active brownian particles. In *Flowing Matter*, pages 211–238. Springer, 2019.
- [147] C Salomon, J Dalibard, A Aspect, H Metcalf, and C Cohen-Tannoudji. Channeling atoms in a laser standing wave. *Physical Review Letters*, 59(15):1659, 1987.
- [148] D Meschede and H Metcalf. Atomic nanofabrication: Atomic deposition and lithography by laser and magnetic forces. *Journal of Physics* D: Applied Physics, 36(3):R17, 2003.
- [149] John David Jackson. Classical electrodynamics. John Wiley & Sons, 2012.
- [150] Konstantin Y Bliokh, Aleksandr Y Bekshaev, and Franco Nori. Dual electromagnetism: helicity, spin, momentum and angular momentum. *New Journal of Physics*, 15(3):033026, 2013.
- [151] K Y Bliokh, A Y Bekshaev, and F Nori. Extraordinary momentum and spin in evanescent waves. *Nature Communications*, 5:3300, 2014.
- [152] SB Wang and CT Chan. Lateral optical force on chiral particles near a surface. *Nature communications*, 5(1):1–8, 2014.

- [153] Sergey Sukhov, Veerachart Kajorndejnukul, John Broky, and Aristide Dogariu. Forces in aharonov-bohm optical setting. Optica, 1(6):383– 387, 2014.
- [154] K Yu Bliokh, FJ Rodríguez-Fortuño, Franco Nori, and Anatoly V Zayats. Spin-orbit interactions of light. *Nature Photonics*, 9(12):796, 2015.
- [155] Aleksandr Y Bekshaev, Konstantin Y Bliokh, and Franco Nori. Transverse spin and momentum in two-wave interference. *Physical Review* X, 5(1):011039, 2015.
- [156] Amaury Hayat, JP Balthasar Mueller, and Federico Capasso. Lateral chirality-sorting optical forces. *Proceedings of the National Academy of Sciences*, 112(43):13190–13194, 2015.
- [157] Martin Neugebauer, Thomas Bauer, Andrea Aiello, and Peter Banzer. Measuring the transverse spin density of light. *Physical review letters*, 114(6):063901, 2015.
- [158] Massimo Antognozzi, CR Bermingham, RL Harniman, S Simpson, J Senior, R Hayward, H Hoerber, MR Dennis, AY Bekshaev, KY Bliokh, et al. Direct measurements of the extraordinary optical momentum and transverse spin-dependent force using a nano-cantilever. *Nature Physics*, 12(8):731, 2016.
- [159] Shima Fardad, Alessandro Salandrino, Akbar Samadi, Matthias Heinrich, Zhigang Chen, and Demetrios N Christodoulides. Scattering detection of a solenoidal poynting vector field. Optics Letters, 41(15):3615–3618, 2016.
- [160] Giovanni Pellegrini, Marco Finazzi, Michele Celebrano, Lamberto Duo, Maria Antonia Iatì, Onofrio M Marago, and Paolo Biagioni. Superchiral surface waves for all-optical enantiomer separation. *The Journal of Physical Chemistry C*, 123(46):28336–28342, 2019.
- [161] N N Bogoliubov and D V Shirkov. Introduction to the Theory of quantized field. Interscience Publisher, New York, USA, 1959.
- [162] L D Landau and E M Lifsits. *Teoria dei campi*. Editori Riuniti, Roma, IT, 1976.
- [163] Emmy Noether. Invariant variation problems. Transport Theory and Statistical Physics, 1(3):186–207, 1971.

- [164] F J Belinfante. On the current and the density of the electric charge, the energy, the linear momentum and the angular momentum of arbitrary fields. *Physica*, 7(5):449–474, 1940.
- [165] H C Ohanian. What is spin? American Journal of Physics, 54(6):500– 505, 1986.
- [166] Lulu Liu, Andrea Di Donato, Vincent Ginis, Simon Kheifets, Arman Amirzhan, and Federico Capasso. Three-dimensional measurement of the helicity-dependent forces on a mie particle. *Physical review letters*, 120(22):223901, 2018.
- [167] Vincent Ginis, Lulu Liu, Alan She, and Federico Capasso. Using the belinfante momentum to retrieve the polarization state of light inside waveguides. *Scientific reports*, 9(1):1–6, 2019.
- [168] MG Donato, A Mazzulla, P Pagliusi, A Magazzù, RJ Hernandez, C Provenzano, PG Gucciardi, OM Maragò, and G Cipparrone. Lightinduced rotations of chiral birefringent microparticles in optical tweezers. *Scientific reports*, 6:31977, 2016.
- [169] V. Svak, O. Brzobohaty, M. Siler, P. Jákl, J. Kaňka, P. Zemánek, and S. H. Simpson. Transverse spin forces and non-equilibrium particle dynamics in a circularly polarized vacuum optical trap. *Nature communications*, 9(1):1–8, 2018.
- [170] Yoshihiko Arita, Stephen H Simpson, Pavel Zemánek, and Kishan Dholakia. Coherent oscillations of a levitated birefringent microsphere in vacuum driven by nonconservative rotation-translation coupling. *Sci*ence Advances, 6(23):eaaz9858, 2020.
- [171] M V Berry. Optical currents. Journal of Optics A: Pure and Applied Optics, 11(9):094001, 2009.
- [172] Konstantin Y Bliokh, Aleksandr Y Bekshaev, and Franco Nori. Optical momentum, spin, and angular momentum in dispersive media. *Physical review letters*, 119(7):073901, 2017.
- [173] Claudia Triolo, Adriano Cacciola, Salvatore Patanè, Rosalba Saija, Salvatore Savasta, and Franco Nori. Spin-momentum locking in the near field of metal nanoparticles. ACS Photonics, 4(9):2242–2249, 2017.
- [174] K Y Bliok and F Nori. Transverse and longitudinal angular momenta of light. *Physics Reports*, 592:1–38, 2015.

- [175] Timo A Nieminen, Alexander B Stilgoe, Norman R Heckenberg, and Halina Rubinsztein-Dunlop. Angular momentum of a strongly focused gaussian beam. Journal of Optics A: Pure and Applied Optics, 10(11):115005, 2008.
- [176] C. Cecchi-Pestellini, M. A. Iatì, and D. A. Williams. The nature of interstellar dust as revealed by light scattering. *Journal of Quantitative* Spectroscopy and Radiative Transfer, 113(18):2310–2320, 2012.
- [177] B. A. McGuire. 2018 Census of Interstellar, Circumstellar, Extragalactic, Protoplanetary Disk, and Exoplanetary Molecules. *The Astrophysical Journal Supplement*, 239(2):17, 2018.
- [178] H. P. Gail and E. Sedlmayr. Physics and Chemistry of Circumstellar Dust Shells. Cambridge Astrophysics. Cambridge University Press, 2014.
- [179] A. Sarangi and I. Cherchneff. Condensation of dust in the ejecta of Type II-P supernovae. Astronomy & Astrophysics, 575:A95, March 2015.
- [180] C. Gall, J. Hjorth, D. Watson, E. Dwek, J. R. Maund, O. Fox, G. Leloudas, D. Malesani, and A. C. Day-Jones. Rapid formation of large dust grains in the luminous supernova 2010jl. *Nature*, 511(7509):326–329, July 2014.
- [181] A. Giannetti, S. Leurini, C. König, J. S. Urquhart, T. Pillai, J. Brand, J. Kauffmann, F. Wyrowski, and K. M. Menten. Galactocentric variation of the gas-to-dust ratio and its relation with metallicity. *Astronomy* & Astrophysics, 606:L12, October 2017.
- [182] T. Zafar and D. Watson. The metals-to-dust ratio to very low metallicities using GRB and QSO absorbers; extremely rapid dust formation. *Astronomy & Astrophysics*, 560:A26, December 2013.
- [183] C. Gall, J. Hjorth, and A. C. Andersen. Production of dust by massive stars at high redshift. Astronomy & Astrophysics Review, 19:43, September 2011.
- [184] L. Mattsson, A. De Cia, A. C. Andersen, and T. Zafar. On the (in)variance of the dust-to-metals ratio in galaxies. *Monthly Notices* of the Royal Astronomical Society, 440(2):1562–1570, May 2014.

- [185] M. Bocchio, A. P. Jones, and J. D. Slavin. A re-evaluation of dust processing in supernova shock waves. Astronomy & Astrophysics, 570:A32, 2014.
- [186] B. T. Draine. Interstellar Dust Models and Evolutionary Implications. In T. Henning, E. Grün, and J. Steinacker, editors, Cosmic Dust - Near and Far, volume 414 of Astronomical Society of the Pacific Conference Series, page 453, December 2009.
- [187] D. A. Williams and C. Cecchi-Pestellini. The Chemistry of Cosmic Dust. Royal Society of Chemistry, 2015.
- [188] K. A. K. Gadallah, H. Mutschke, and C. Jäger. UV irradiated hydrogenated amorphous carbon (HAC) materials as a carrier candidate of the interstellar UV bump at 217.5 nm. Astronomy & Astrophysics, 528:A56, April 2011.
- [189] A.G.G.M. Tielens. The Physics and Chemistry of the Interstellar Medium. Cambridge University Press, 2005.
- [190] S. A. Sandford, L. J. Allamandola, A. G. G. M. Tielens, K. Sellgren, M. Tapia, and Y. Pendleton. The interstellar ch stretching band near 3.4 microns-constraints on the composition of organic material in the diffuse interstellar medium. *The Astrophysical Journal*, 371:607–620, 1991.
- [191] B. T. Draine. Scattering by interstellar dust grains. ii. x-rays. The Astrophysical Journal, 598(2):1026, 2003.
- [192] A. Leger and J. L. Puget. Identification of the "unidentified" IR emission features of interstellar dust? Astronomy & Astrophysics, 500:279– 282, August 1984.
- [193] W. W. Duley and D. A. Williams. Excitation of the Aromatic Infrared Emission Bands: Chemical Energy in Hydrogenated Amorphous Carbon Particles? *The Astrophysical Journal Lettersl*, 737(2):L44, August 2011.
- [194] E. Dartois, T. R. Geballe, T. Pino, A. T. Cao, A. Jones, D. Deboffle, V. Guerrini, Ph. Bréchignac, and L. D'Hendecourt. IRAS 08572+3915: constraining the aromatic versus aliphatic content of interstellar HACs. *Astronomy & Astrophysics*, 463(2):635–640, February 2007.

- [195] Iatì M. A., Saija R., Borghese F., Denti P., Cecchi-Pestellini C., and Williams D. A. Stratified dust grains in the interstellar medium–I. An accurate computational method for calculating their optical properties. *Monthly Notices of the Royal Astronomical Society*, 384(2):591–598, 2008.
- [196] C. Cecchi-Pestellini, A. Cacciola, M.A. Iatì, R. Saija, F. Borghese, P. Denti, A. Giusto, and D.A. Williams. Stratified dust grains in the interstellar medium-ii. time-dependent interstellar extinction. *Monthly Notices of the Royal Astronomical Society*, 408(1):535–541, 2010.
- [197] J. A. Nuth and J. H. Hecht. Signatures of aging silicate dust. Astrophysics and Space Science, 163(1):79–94, 1990.
- [198] F. J. M. Rietmeijer and J. A. Nuth. Petrologic Constraints on Amorphous and Crystalline Magnesium Silicates: Dust Formation and Evolution in Selected Herbig Ae/Be Systems. *The Astrophysical Journal*, 771(1):34, July 2013.
- [199] A. Ciaravella, C. Cecchi-Pestellini, Y. J. Chen, G. M. Muñoz Caro, C. H. Huang, A. Jiménez-Escobar, and A. M. Venezia. Soft X-Ray Irradiation of Silicates: Implications for Dust Evolution in Protoplanetary Disks. *The Astrophysical Journal*, 828(1):29, September 2016.
- [200] A. Juhász, J. Bouwman, Th. Henning, B. Acke, M. E. van den Ancker, G. Meeus, C. Dominik, M. Min, A. G. G. M. Tielens, and L. B. F. M. Waters. Dust Evolution in Protoplanetary Disks Around Herbig Ae/Be Stars—the Spitzer View. *The Astrophysical Journal*, 721(1):431–455, September 2010.
- [201] T. P. Stecher and B. Donn. On Graphite and Interstellar Extinction. *The Astrophysical Journal*, 142:1681, November 1965.
- [202] J. S. Mathis and G. Whiffen. Composite Interstellar Grains. *The Astrophysical Journal*, 341:808, June 1989.
- [203] E. L. Wright. Long-Wavelength Absorption by Fractal Dust Grains. The Astrophysical Journal, 320:818, September 1987.
- [204] Ch. Joblin, A. Léger, and P. Martin. Contribution of polycyclic aromatic hydrocarbon molecules to the interstellar extinction curve. *The Astrophysical Journal*, 393:L79–L82, 1992.

- [205] R. Papoular, J. Conard, O. Guillois, I. Nenner, C. Reynaud, and J.-N. Rouzaud. A comparison of solid-state carbonaceous models of cosmic dust. Astronomy and Astrophysics, 315:222–236, 1996.
- [206] T. M. Steel and W. W. Duley. A 217.5 Nanometer Absorption Feature in the Spectrum of Small Silicate Particles. *The Astrophysical Journal*, 315:337, April 1987.
- [207] A. Higa, T. Oshiro, Y. Saida, M. Yamazato, and M. Toguchi. Correlation between properties and hydrogen concentration of ac: H films prepared by rf magnetron sputtering. New Diamond and Frontier Carbon Technology, 16(5):245–253, 2006.
- [208] J. E. Chiar, A. J. Adamson, D. C. B. Whittet, A. Chrysostomou, J. H. Hough, T. H. Kerr, R. E. Mason, P. F. Roche, and G. Wright. Spectropolarimetry of the 3.4 μm feature in the diffuse ism toward the galactic center quintuplet cluster. *The Astrophysical Journal*, 651(1):268, 2006.
- [209] John S Hall and Alfred H Mikesell. Observations of polarized light from stars. The Astronomical Journal, 54:187–188, 1949.
- [210] NV Voshchinnikov, VB Il'in, and HK Das. Polarizing efficiency as a guide of grain growth and interstellar magnetic field properties. *Monthly Notices of the Royal Astronomical Society*, 462(3):2343–2354, 2016.
- [211] Bruce T Draine. *Physics of the interstellar and intergalactic medium*, volume 19. Princeton University Press, 2010.
- [212] Qi Li, SL Liang, and Aigen Li. Spectropolarimetric constraints on the nature of interstellar grains. *Monthly Notices of the Royal Astronomical Society: Letters*, 440(1):L56–L60, 2014.
- [213] BTf Draine and Hyung Mok Lee. Optical properties of interstellar graphite and silicate grains. *The Astrophysical Journal*, 285:89–108, 1984.
- [214] John S Mathis and G Whiffen. Composite interstellar grains. *The* Astrophysical Journal, 341:808–822, 1989.
- [215] AP Jones, WW Duley, and DA Williams. The structure and evolution of hydrogenated amorphous carbon grains and mantles in the interstellar medium. *Quarterly Journal of the Royal Astronomical Society*, 31:567–582, 1990.

- [216] Aigen Li and J Mayo Greenberg. A unified model of interstellar dust. Astronomy and Astrophysics, 323:566–584, 1997.
- [217] Cesare Cecchi-Pestellini and David A Williams. Evolving interstellar extinction. Monthly Notices of the Royal Astronomical Society, 296(2):414–418, 1998.
- [218] Edward D Palik. Handbook of optical constants of solids, volume 3. Academic press, 1998.
- [219] AP Jones, L Fanciullo, M Köhler, L Verstraete, V Guillet, M Bocchio, and N Ysard. The evolution of amorphous hydrocarbons in the ism: dust modelling from a new vantage point. Astronomy & Astrophysics, 558:A62, 2013.
- [220] Cesare Cecchi-Pestellini, Silvia Casu, Giacomo Mulas, and Alberto Zonca. Observational evidence of dust evolution in galactic extinction curves. *The Astrophysical Journal*, 785(1):41, 2014.
- [221] Paul Stysley. Laser-based optical trap for remote sampling of interplanetary and atmospheric particulate matter. Technical report, NASA, 2014. available online.
- [222] A Rotundi, GA Baratta, J Borg, JR Brucato, H Busemann, L Colangeli, L d'Hendecourt, Z Djouadi, G Ferrini, IA Franchi, et al. Combined micro-raman, micro-infrared, and field emission scanning electron microscope analyses of comet 81p/wild 2 particles collected by stardust. *Meteoritics & Planetary Science*, 43(1-2):367–397, 2008.
- [223] A Rotundi, FJM Rietmeijer, M Ferrari, V Della Corte, GA Baratta, R Brunetto, E Dartois, Z Djouadi, S Merouane, J Borg, et al. Two refractory wild 2 terminal particles from a carrot-shaped track characterized combining mir/fir/raman microspectroscopy and fe-sem/eds analyses. *Meteoritics & Planetary Science*, 49(4):550–575, 2014.
- [224] FJM Rietmeijer, VINCENZO Della Corte, MARCO Ferrari, Alessandra Rotundi, and R Brunetto. Laboratory analyses of meteoric debris in the upper stratosphere from settling bolide dust clouds. *Icarus*, 266:217–234, 2016.
- [225] F Oliva, E D'Aversa, GL Liberti, G Sindoni, F Altieri, and E Castelli. A database of aerosols and gases coefficients for vis-nir radiative transfer in the solar system planetary atmospheres. *Planetary and Space Science*, 166:101–109, 2019.

- [226] Walter G Egan and Theodore Hilgeman. Optical constants for terrestrial analogs of lunar materials. *The Astronomical Journal*, 78:799, 1973.
- [227] Craig F Bohren and Donald R Huffman. Absorption and scattering of light by small particles. John Wiley & Sons, 2008.
- [228] Alan E Rubin and Jeffrey N Grossman. Meteorite and meteoroid: New comprehensive definitions. *Meteoritics & Planetary Science*, 45(1):114– 122, 2010.
- [229] D. Nesvorny, D. Vokrouhlicky, W. F. Bottke, and M. Sykes. Physical properties of asteroid dust bands and their sources. *Icarus*, 181(1):107– 144, 2006.
- [230] G. J. Flynn, D. D. Durda, L. E. Sandel, J. W. Kreft, and M. M. Strait. Dust production from the hypervelocity impact disruption of the murchison hydrous cm2 meteorite: Implications for the disruption of hydrous asteroids and the production of interplanetary dust. *Planetary and Space Science*, 57(2):119–126, 2009.
- [231] M. Fulle, J. Blum, A. Rotundi, B. Gundlach, C. Güttler, and V. Zakharov. How comets work: nucleus erosion versus dehydration. *Monthly Notices of the Royal Astronomical Society*, 493(3):4039–4044, 2020.
- [232] C. Güttler, T. Mannel, A. Rotundi, S. Merouane, M. Fulle, D. Bockelée-Morvan, J. Lasue, A. C.l Levasseur-Regourd, J. Blum, G. Naletto, et al. Synthesis of the morphological description of cometary dust at comet 67p/churyumov-gerasimenko. Astronomy & Astrophysics, 630:A24, 2019.
- [233] C. Cordier and L. Folco. Oxygen isotopes in cosmic spherules and the composition of the near earth interplanetary dust complex. *Geochimica* et Cosmochimica Acta, 146:18–26, 2014.
- [234] Zelia Dionnet, Martin D Suttle, Andrea Longobardo, Alessandra Rotundi, Luigi Folco, Vincenzo Della Corte, and Andrew King. X-ray computed tomography: Morphological and porosity characterization of giant antarctic micrometeorites. *Meteoritics & Planetary Science*, 55(7):1581–1599, 2020.
- [235] M. D. Suttle and L. Folco. The extraterrestrial dust flux: Size distribution and mass contribution estimates inferred from the transantarctic

mountains (tam) micrometeorite collection. Journal of Geophysical Research: Planets, 125(2):e2019JE006241, 2020.

- [236] Vincenzo Della Corte, Pasquale Palumbo, Alessandra Rotundi, Simone De Angelis, Frans JM Rietmeijer, Ezio Bussoletti, Alessandra Ciucci, Marco Ferrari, Valentina Galluzzi, and Ernesto Zona. In situ collection of refractory dust in the upper stratosphere: the duster facility. *Space science reviews*, 169(1-4):159–180, 2012.
- [237] Frans JM Rietmeijer. The earliest chemical dust evolution in the solar nebula. *Geochemistry*, 62(1):1–45, 2002.
- [238] Zelia Dionnet, Rosario Brunetto, Alice Aléon-Toppani, Stefano Rubino, Donia Baklouti, Ferenc Borondics, Anne-Cécile Buellet, Zahia Djouadi, Andrew King, Tomoki Nakamura, et al. Combining ir and xray microtomography data sets: Application to itokawa particles and to paris meteorite. *Meteoritics & Planetary Science*, 55(7):1645–1664, 2020.
- [239] V. Della Corte, A. Rotundi, M. Fulle, S. Ivanovski, S. F. Green, F. J. M. Rietmeijer, L. Colangeli, P. Palumbo, R. Sordini, M. Ferrari, et al. 67p/cg inner coma dust properties from 2.2 au inbound to 2.0 au outbound to the sun. *Monthly Notices of the Royal Astronomical Society*, 462(Suppl_1):S210–S219, 2016.
- [240] Marco Fulle, VINCENZO Della Corte, Alessandra Rotundi, P Weissman, A Juhasz, K Szego, R Sordini, MARCO Ferrari, S Ivanovski, F Lucarelli, et al. Density and charge of pristine fluffy particles from comet 67p/churyumov–gerasimenko. *The Astrophysical journal letters*, 802(1):L12, 2015.
- [241] Alessandra Rotundi, Holger Sierks, Vincenzo Della Corte, Marco Fulle, Pedro J Gutierrez, Luisa Lara, Cesare Barbieri, Philippe L Lamy, Rafael Rodrigo, Detlef Koschny, et al. Dust measurements in the coma of comet 67p/churyumov-gerasimenko inbound to the sun. Science, 347(6220), 2015.
- [242] ECT Chao, Judith A Boreman, Jean A Minkin, Odette B James, and George A Desborough. Lunar glasses of impact origin: Physical and chemical characteristics and geologic implications. *journal of Geophysical Research*, 75(35):7445–7479, 1970.

- [243] Owen B Toon, James B Pollack, and Carl Sagan. Physical properties of the particles composing the martian dust storm of 1971–1972. *Icarus*, 30(4):663–696, 1977.
- [244] RV Morris, G Klingelhoefer, C Schroder, DS Rodionov, A Yen, DW Ming, PA De Souza, T Wdowiak, I Fleischer, R Gellert, and B. Bernhardt. Mossbauer mineralogy of rock, soil, and dust at meridiani planum, mars: Opportunity's journey across sulfate-rich outcrop, basaltic sand and dust, and hematite lag deposits. *Journal of Geophysical Research: Planets*, 111(E12), 2006.
- [245] Albert S Yen, Ralf Gellert, Christian Schröder, Richard V Morris, James F Bell, Amy T Knudson, Benton C Clark, Douglas W Ming, Joy A Crisp, Raymond E Arvidson, et al. An integrated view of the chemistry and mineralogy of martian soils. *Nature*, 436(7047):49–54, 2005.
- [246] Harry Y McSween Jr, Ian O McGlynn, and A Deane Rogers. Determining the modal mineralogy of martian soils. *Journal of Geophysical Research: Planets*, 115(E7), 2010.
- [247] David L Bish, DF Blake, DT Vaniman, SJ Chipera, RV Morris, DW Ming, AH Treiman, P Sarrazin, SM Morrison, Robert T Downs, et al. X-ray diffraction results from mars science laboratory: Mineralogy of rocknest at gale crater. *science*, 341(6153), 2013.
- [248] F Oliva, A Geminale, E D'Aversa, F Altieri, G Bellucci, FG Carrozzo, G Sindoni, and D Grassi. Properties of a martian local dust storm in atlantis chaos from omega/mex data. *Icarus*, 300:1–11, 2018.
- [249] MJ Wolff, MD Smith, RT Clancy, R Arvidson, M Kahre, F Seelos, S Murchie, and Hannu Savijärvi. Wavelength dependence of dust aerosol single scattering albedo as observed by the compact reconnaissance imaging spectrometer. *Journal of Geophysical Research: Planets*, 114(E2), 2009.
- [250] Michael J Wolff, R Todd Clancy, Jay D Goguen, Michael C Malin, and Bruce A Cantor. Ultraviolet dust aerosol properties as observed by marci. *Icarus*, 208(1):143–155, 2010.
- [251] Vincenzo Della Corte, Franciscus JM Rietmeijer, Alessandra Rotundi, Marco Ferrari, and Pasquale Palumbo. Meteoric cao and carbon smoke particles collected in the upper stratosphere from an unanticipated

source. Tellus B: Chemical and Physical Meteorology, 65(1):20174, 2013.

- [252] Ian H Malitson. Interspecimen comparison of the refractive index of fused silica. Josa, 55(10):1205–1209, 1965.
- [253] Gorachand Ghosh. Dispersion-equation coefficients for the refractive index and birefringence of calcite and quartz crystals. Optics communications, 163(1-3):95–102, 1999.
- [254] CJ Liu and EF Sieckmann. Refractive index of calcium oxide. Journal of Applied Physics, 37(6):2450–2452, 1966.
- [255] HJ Hagemann, W Gudat, and C Kunz. Desy report sr-74/7. Hamburg, Germany, 1974.
- [256] ASTM Standard. G173-03-standard tables for reference solar spectral irradiances: Direct normal and hemispherical on 37 tilted surface. Ann. Book of ASTM Standards 2003, 14, 2012.
- [257] Jozef Klačka and Miroslav Kocifaj. Motion of nonspherical dust particle under the action of electromagnetic radiation. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 70(4-6):595–610, 2001.