Effects of Instrumental Energy Resolution on the Measured MSD as Obtained by Elastic Incoherent Neutron Scattering Data

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Abstract

In the present paper we compare the mean square displacement (MSD) values obtained on dry and hydrated (H_2O andD_2O) lysozyme samples by using two spectrometers, IN13 and IN10 at the Institute Laue Langevin (Grenoble, France), working at the energy resolution value of $8\mu eV$, corresponding to an elastic time resolution of 516 ps, and at the energy resolution value of $1\mu eV$, corresponding to an elastic time resolution of 4136 ps. In particular the comparison is performed when only vibrational motions occur, i.e. when the system MSD can be considered almost constant$\langle r^2\rangle(t) \rightarrow \langle r^2\rangle^V$. Such a condition is approximately satisfied in the low temperature range up to T = 40 K. The analysis furnish values that agree very well with data reported in literature while comparison between the measured MSDs shows the role played by the instrumental energy resolution.

Keywords: Mean Square Displacement, Elastic Incoherent Neutron Scattering, Lysozime, Instrumental Energy Resolution.

Introduction

It is well-known that neutron scattering give the possibility to characterize the dynamical and structural properties of many material systems, such as polymers, proteins, glasses, and so forth. These information are expressed by the time-dependent spatial correlation functions $G(r,t)$ introduced by Van Hove whose spacetime Fourier transform corresponds to the scattering function $S(\vec{Q},\omega)$.
The system observables, for example, distribution functions and mean square displacement (MSD), are influenced by instrumental effects because the experimentally obtained neutron scattering data are also connected with the employed spectrometer instrumental features. In the present paper we use two spectrometers working, IN13 and IN10, with different instrumental resolutions, to apply to EINS data collected, on dry and hydrated (H_2O and D_2O) lysozyme samples. Lysozyme is an enzyme occurring naturally in egg white, human tears, saliva, and other body fluids, capable of destroying the cell walls of certain bacteria and there by acting as a mildantiseptic.

Experimental Section

Experimental data were obtained by the IN13 and IN10 spectrometers at the Institute Laue Langevin (Grenoble, France). In these spectrometers the incident neutron shave a relatively high energy ($16meV$) and two different energy resolutions. In particular:

	IN13 spectrometer	IN10 spectrometer
Incident wavelength	2.23 Å	6.27 Å
Q-range	0.28 ÷4.27 Å^{-1}	0.30 ÷ 2.00 Å^{-1}
Instrumental energy resolution (FWHM)	8 μeV	1 μeV
Instrumental time resolution	516 ps	4136 ps

Figure 1: *Instrumental characteristics*

The scattering particles which move in a time scale much slower than the characteristic time corresponding to the energy resolution are seen as elastic scatterers, whereas a decrease of the elastic intensity is observed for scattering particles which move faster. This implies that a scattering particle which moves in a time scale between there solution time of IN13 and IN10 contributes as an elastic process in the IN13 spectra and as a non elastic process in the IN10 spectra. Raw data were corrected for cell scattering and detector response [1]. Partially deuterated lysozyme, in a dry state, in D_2O, and in H_2O environments at a hydration value of h =0.4 (h =water/protein weight fraction) have been employed. Data were collected by the IN13 spectrometer in the temperature range

of 20÷310 K and by the IN10 spectrometer in the 20÷320 K temperature range.

Theoretical Approach

It is well known that experimental neutron scattering data are connected both with the structure and dynamical properties of the investigated sample, represented with the instrumental apparatus and also by the scattering law $S(\vec{Q},\omega)$ and by the intermediate scattering function $I(\vec{Q},t)$,that are connected by a direct and an inverse time Fourier transform:

$$S(\vec{Q},\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} I(\vec{Q},t)e^{-i\omega t}\,dt \tag{1}$$

$$I(\vec{Q},t) = \int_{-\infty}^{\infty} S(\vec{Q},\omega)e^{i\omega t}\,d\omega \tag{2}$$

The experimentally accessible quantity in the $\omega-$*space*, due to the finite energy instrumental resolution , is the convolution of the scattering law $S(\vec{Q},\omega)$ with the instrumental resolution function $R(\omega,\Delta\omega)$, i.e. the measured scattering law $S_R(\vec{Q},\omega,\Delta\omega)$:

$$\begin{aligned} &S_R(\vec{Q},\omega,\Delta\omega) = S(\vec{Q},\omega)\otimes R(\omega,\Delta\omega),= \\ &\int_{-\infty}^{\infty} S(\vec{Q},\omega-\omega')R(\omega',\Delta\omega)\,d\omega' \end{aligned} \tag{3}$$

that, taking into account eq. 1, yields:

$$\begin{aligned} &S_R(\vec{Q},\omega,\Delta\omega) = \left[\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} I(\vec{Q},t)e^{-i\omega t}\,dt\right]\otimes R(\omega,\Delta\omega) = \\ &\int_{-\infty}^{+\infty}\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} I(\vec{Q},t)e^{-i(\omega-\omega')t}\,dtR(\omega',\Delta\omega)\,d\omega' = \\ &\int_{-\infty}^{\infty} I(\vec{Q},t)e^{-i\omega t}\,dt\left[\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} e^{-i\omega' t}R(\omega',\Delta\omega)\,d\omega'\right] = \\ &\int_{-\infty}^{\infty} I(\vec{Q},t)R(t)e^{-i\omega t}\,dt \end{aligned} \tag{4}$$

We highlight that the sub-index R indicates that the relative function is affected by the instrumental resolution,instead, the absence of this sub-index indicates that the relative function is connected only to the sample. In the ideal elastic case in which the resolution is a delta function in the ω-space, we obtain from eq. 4 that the measured scattering law coincides with the scattering law evaluated at $\omega = 0$:

$$S_R(\vec{Q},\omega=0,\Delta\omega) = \int_{-\infty}^{\infty} I(\vec{Q},t)R(t)\,dt \tag{5}$$

$$\begin{aligned} &S_R(\vec{Q},\omega=0,\Delta\omega) = \\ &\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} I(\vec{Q},t)\,dt = S(\vec{Q},\omega=0) \end{aligned} \tag{6}$$

In the opposite limit in which the instrumental resolution is a very broad function in the ω-space (in respect to the time behaviour of $I(\vec{Q},t)$), under the assumption that it can be treated as a constant, the measured scattering law corresponds to the intermediate scattering function evaluated at $t = 0$:

$$\begin{aligned} S_R(\vec{Q},\omega,\Delta\omega) = &\int_{-\infty}^{\infty} I(\vec{Q},t)R(t)\,dt = \\ &\int_{-\infty}^{\infty} I(\vec{Q},t)\delta(t)\,dt = \\ &I(\vec{Q},t=0) \end{aligned} \tag{7}$$

Now, analyzing eq. 3 and eq. 5, we evaluate the effect of the instrumental resolution on the MSD. We begin introducing a general expression for the measured MSD

$$\langle\vec{r^2}\rangle_R = \int_{-\infty}^{\infty} G_R^{self}(r)r^2\,dr \tag{8}$$

So, we can consider two procedures for MSD evaluation:

- the first one is based on the following equation:

$$= \int_{-\infty}^{\infty} FT_r\big\{I_R(Q,t)\big\}r^2\,dr \tag{9}$$

and leads to the following conclusion:

$$\langle\vec{r^2}\rangle_R(t) = \sqrt{2\pi}R(t)\langle r^2\rangle(t) \tag{10}$$

The MSD obtained by this procedure is a function of time.

- The second one is based on the following equation:

$$= \int_{-\infty}^{\infty} FT_r\big\{S_R(\vec{Q},\omega=0)\big\}r^2\,dr \tag{11}$$

and leads to the following conclusion:

$$\langle\vec{r^2}\rangle_R = \int_{-\infty}^{\infty}\langle r^2\rangle(t)R(t)\,dt \tag{12}$$

The MSD obtained by this procedure is a number The connection between the two MSD is:

$$\langle\vec{r^2}\rangle_R = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}\langle r^2\rangle_R(t)\,dt \tag{13}$$

Moreover, it depends on the employed instrumental resolution.

Results and Discussion

The relation between the measured MSD, $\langle \vec{r^2} \rangle_R$ and the system MSD, $\langle \vec{r^2} \rangle_R(t)$, in experiments which give a direct connection with the scattering law evaluated at $= 0$, $S_R(\vec{Q}, \omega = 0, \Delta\omega)$, is given by equation:

$$\langle \vec{r^2} \rangle_R = \int_{-\infty}^{\infty} \langle \vec{r^2} \rangle(t) R(t)\, dt \tag{14}$$

We can say that:

- For two different systems with the same instrumental resolution yields, the comparison between the MSDs measured is:

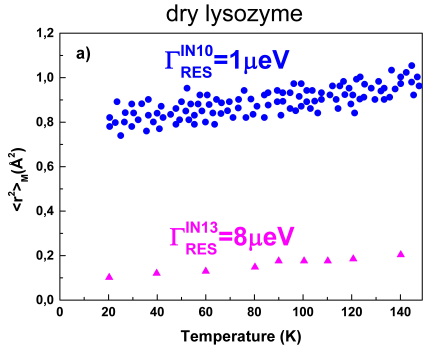
$$\langle r^2 \rangle_{1,R} - \langle r^2 \rangle_{2,R} = \int_{-\infty}^{\infty} \Big[\langle r^2 \rangle_1(t) - \langle r^2 \rangle_2(t) \Big] R(t)\, dt \tag{15}$$

  So, using the same instrument working at the same resolution, on two different systems,the difference between the measured MSD does not correspond to the difference between their MSDs [2].

- For the same system evaluated at different instrumental resolutions yields, the comparison between the MSDs measured is:

$$\langle r^2 \rangle_{R1} - \langle r^2 \rangle_{R2} = \int_{-\infty}^{\infty} \langle r^2 \rangle(t) \Big[R_1(t) - R_2(t) \Big]\, dt \tag{16}$$

  Below is shown the comparison between the measured MSDs obtained for the same systems, that is, dry and hydrated (H_2O and D_2O with $h = 0.4$) lysozyme, respectively, by the IN13 spectrometer working at the energy resolution value of $8\mu eV$, corresponding to an elastic time resolution of $516ps$, and by the IN10 spectrometer working at the energy resolution value of The MSD values have been obtained by employing the following common Q-range: $0.30 \div 2.16 \mathring{A}^{-1}$



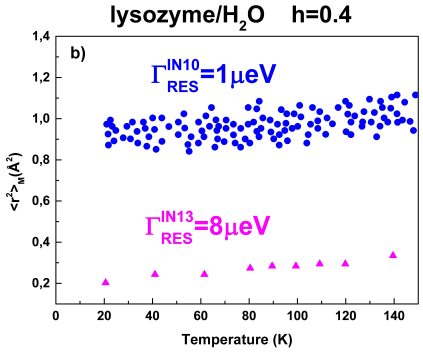


Figure 2: *Comparison between the measured MSDs temperature behavior obtained from data collected by the IN13 and IN10 spectrometers on (a) dry and (b) hydrated (H_2O) lysozyme samples.*

LYSOZYME DRY					
MSD_M^{IN10} (Å²)	MSD_M^{IN13} (Å²)	MSD^{IN10} (Å²)	MSD^{IN13} (Å²)	MD^{IN10} (Å)	MD^{IN13} (Å)
1,241	0,172	0,00029	0,00034	0,017	0,018
1,228	0,199	0,00030	0,00036	0,017	0,019
1,251	0,219	0,00030	0,00042	0,017	0,020

LYSOZYME/H_2O					
MSD_M^{IN10} (Å²)	MSD_M^{IN13} (Å²)	MSD^{IN10} (Å²)	MSD^{IN13} (Å²)	MD^{IN10} (Å)	MD^{IN13} (Å)
2,165	0,253	0,00052	0,00049	0,023	0,022
2,471	0,460	0,00060	0,00089	0,024	0,029
2,853	0,462	0,00069	0,00090	0,026	0,030

LYSOZYME/D_2O					
MSD_M^{IN10} ($Å^2$)	MSD_M^{IN13} ($Å^2$)	MSD^{IN10} ($Å^2$)	MSD^{IN13} ($Å^2$)	MD^{IN10} (Å)	MD^{IN13} (Å)
0,897	0,079	0,00022	0,00015	0,015	0,012
0,748	0,114	0,00018	0,00022	0,013	0,015
0,832	0,079	0,00020	0,00015	0,014	0,012

Figure 3: *vibrational MSDs values for dry, hydrated (H_2O and D_2O) lysozyme.*

In agreement with eq. 12 , the MSD evaluated by IN10, in all the temperature ranges, is higher in respect to that evaluated by IN13. This is because that even at the lowest temperature values, where only vibrational contributions are expected to contribute, the measured MSD is the integral of the product between the resolution function and the system MSD [3].

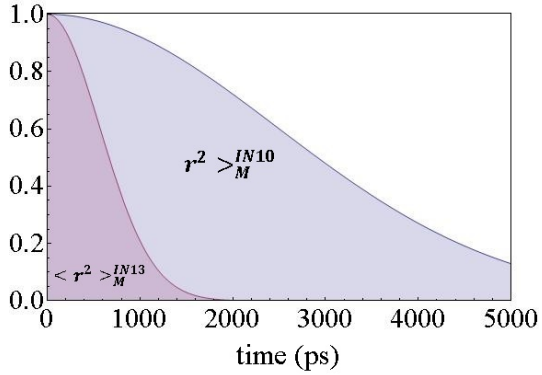


Figure 4: *Effect on the measured MSD because of the different energy resolution of the two spectrometers, IN13 and IN10,when there are only vibrational motions; inparticular different measured MSDs correspondat the same system MSD.*

- In particular, when only vibrational motions occur, the system MSD can be considered almost constant $\langle r^2\rangle(t) \to \langle r^2\rangle^V$ (such a condition is approximately satisfied in the low temperature range up to $T = 40K$); in such a case, the measured MSD from eq. 12 results

$$\begin{aligned}\langle r^2\rangle_R &= \int_{-\infty}^{\infty} \langle r^2\rangle(t) R(t)\, dt \\ &= \int_{-\infty}^{\infty} \langle r^2\rangle^V(t) R(t)\, dt\end{aligned} \tag{17}$$

$$\langle r^2\rangle_R = \langle r^2\rangle^V \int_{-\infty}^{\infty} R(t)\, dt \tag{18}$$

Then, starting from the last equation, its possible to determine the system MSD at the lowest temperature values

$$\langle r^2\rangle^V = \frac{\langle r^2\rangle_R}{\int_{-\infty}^{\infty} R(t)\, dt} \tag{19}$$

Figure 2 shows the effect on the measured MSD because of the employment of a different energy resolution, when there are onlyvibrational motions; as it can be seen, at the same vibrational system MSD, different measured MSDs correspond. It notes that if we apply this procedure to the data collected on the same systems by the two spectrometers IN13 and IN10, which operate at a different energy resolution, we obtain at the lowest temperature values the same system MSD value. This gives confirmation for the validity of the procedure.

Conclusions

MSD values obtained with different instrumental energy resolutions furnish different values. In this work the MSDs obtained on dry and hydrated (H_2O and D_2O) lysozyme samples by using two spectrometers, IN13 and IN10 at ILL, working with energy resolution values of $8\mu eV$, and $1\mu eV$are analysed. In particular the comparison is performed when only vibrational motions occur, i.e. when the system MSD can be considered almost constant $\langle r^2\rangle(t) \to \langle r^2\rangle^V$, i.e. in the low temperature range up to $T = 40K$. The obtained MSD values agree with findings reported in literature highlighting the role played by the instrumental energy resolution

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