Nuclear Resonant scattering of Synchrotron radiation from nuclei in the Brownian motion

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Abstract:
The time evolution of the coherent forward scattering of the synchrotron radiation for resonant nuclei in Brownian motion is studied. Apart from target thickness, the appearance of the dynamical beats also depends on \( \alpha \) which is the ratio of the harmonic force constant to the damping force constant of harmonic oscillator undergoing Brownian motion.
Introduction:

In recent years, synchrotron radiation (SR) has been used to study Mössbauer spectroscopy in the time domain. By measuring the time dependence of the intensity of nuclear de-excitation [1 and references therein] various solid state and nuclear parameters are being studied. Synchrotron radiation is produced when ultra-relativistic electrons or positrons accelerate while transgressing bending magnets or periodic magnetic structures and emit intense radiation. SR is emitted in short repetitive pulses whereas emission of photons from a radioactive nucleus are random in time.

The Mössbauer spectra of iron-protein crystals have special shape which provide a unique signature, indicating that Mössbauer atom undergoes bounded diffusive motions in the biological systems. It is observed for biological systems that above a particular temperature, which is the characteristic of the system (i) the Mössbauer lineshape becomes non-Lorentzian, (ii) the mean square displacement (msd) shows temperature dependence which is not typical of either the Einstein or the Debye model and (iii) the width of the Mössbauer lineshape increases with the temperature. The non-Lorentzian lineshape was observed by Parak et. al. [2], Cohen et. al.[3] in myoglobin. Similar phenomena was seen in haemoglobin by Mayo,Parak and Mössbauer [4]. Pioneering studies on the dynamics of proteins using Mössbauer Spectroscopy were done by Keller and Debrunner [5], Parak et. al. [6,7]. These unusual results observed for biological systems, have been explained by using a dynamical model of the bounded diffusion. These models are based on the concept of Brownian motion of an overdamped harmonic oscillator [2,3,8,9]. Recently synchrotron radiation has been used to study dynamics
of Mössbauer atom in the biological systems [10,11,12,13]. The goal of the present paper is to study the time evolution of resonant scattering of SR from nuclei undergoing Brownian motion in harmonic potential.

**Methodology:**

In this study we use mathematical treatment developed in reference [8,9] as a starting point. A short pulse of SR can be decomposed into a continuum set of monochromatic spectral components within a frequency interval. Smirnov and Kohn [14,15,20] calculated the forward transmitted wave packet by integrating over all spectral components of the transmitted radiation to obtain

\[ E(t, Z) = \frac{E_{\omega \theta}(Z)}{2\pi} \int d\omega \exp(-i\omega t) \exp\left(\frac{iKC(k, \omega)Z}{2}\right) \]  

(1)

\( E(t, Z) \) is the time dependent electric field amplitude of the synchrotron radiation transmitted through a nuclear target of thickness \( Z \). The function \( E_{\omega \theta}(Z) \) has the modulus \( (\frac{I_0}{\Delta \omega})^{1/2} \exp(-\frac{\mu e z^2}{2}) \). The electronic absorption coefficient is given as \( \mu_e = K \chi \) where \( \chi = \frac{\sigma_e V_0 K}{\hbar} \), \( \sigma_e \) being the total cross section of electronic inelastic scattering and \( V_0 \) is the target volume corresponding to one nucleus. \( K \) is the wave number defined as \( K = \frac{\omega}{c} \) and \( \omega \) is the frequency having wave vector \( k \). \( I_0 \) is the intensity of the SR within the frequency band \( \Delta \omega \), as determined by the monochromator system. \( C(k, \omega) \) is the nuclear part of the susceptibility of the target and is related to the scattering amplitude and can be written as

\[ C(k, \omega) = \frac{i\Gamma_0}{2\hbar} \sum_{ge} B_{ge} \phi(k, \omega - \omega_{eg}) \]  

(2)

where

\[ B_{ge} = \frac{8\pi f_{LM}(k)}{\omega^2 V_0(2I_g + 1)\Gamma_0} |< g|j(k)|e >|^2 \]  

(3)
characterizes the strength of the nuclear response at the resonant frequency \( \omega_{eg} \) and \( \phi(k, \omega - \omega_{eg}) \) is the universal resonance function. Here, \( g \) and \( e \) define sublevels of the ground and the excited states of nucleus respectively and \( \Gamma_0 \) is the natural linewidth of the excited levels. \( f_{LM}(k) \) is the Lamb Mössbauer factor, \( I_g \) is the nuclear spin in the ground state while \( < g | j(\mathbf{k}) | e > \) represents the matrix element of the scaler component of the nuclear current density vector.

At resonance, nuclear part of the susceptibility of the target has a frequency dependence, which is determined by the nature of universal resonance function, obtained (in the present case) by taking into account the diffusive motion. It is described by the following

\[
\phi(k, \omega) = \int_0^\infty dt \exp(-i\omega t - \frac{\Gamma_0 t}{2\hbar})F_s(k, t)
\]  

where

\[
F_s(k, t) = \int dr \exp(-ik\.r)G(r, t)
\]  

Here \( G(r,t) \) is the correlation function. The derivation of above equations is given in references [14,15] in detail. We consider only that case where polarization is absent in the coherent forward scattering. The most frequent application of SR in the nuclear resonance spectroscopy is the measurement of the time-dependence of the forward scattering intensity, given by

\[
I_f(t, z) = |E(t, Z)|^2
\]  

It is interesting to note that Mössbauer absorption spectrum is dependent on the real part of the universal resonance function \( \phi(k, \omega) \). On the other hand, the time dependence is determined by the entire universal function.
Harmonically bound nuclei in Brownian motion:

The time evolution of nuclear resonant scattering of SR pulse by nuclei undergoing various types of diffusion (like free diffusion, continuous localised and jump diffusion) has been studied earlier [14,15]. Here we consider an important case of diffusion in the form of Brownian motion. Gunther et. al. [16,17] were the first to predict that Brownian motion of a Mössbauer atom results in the non-Lorentzian nature of Mössbauer line shape. The theory for the cases of motion of harmonically bound Mössbauer atom in the Brownian motion has been extensively developed both for the classical [2,3,8,9] and quantum cases [18]. In the present paper we will confine to the classical case and use the theory as developed by Nowik et. al. [8,9]. For one-dimensional harmonic oscillator in the Brownian motion, Uhlenbeck and Ornstein [19] derived a general formula for self correlation function $G(x, x_0, t)$ which is the probability that at time $t$ the nucleus will be at position $x$ if at time $t=0$ it was at position $x_0$. This self correlation is given as [8]

$$G(x, x_0, t) = \left(\frac{\alpha}{2\pi D(1 - \exp(-2\alpha t))}\right)^{1/2} \exp\left(\frac{-\alpha(x - x_0 \exp(-\alpha t))^2}{2D(1 - \exp(-2\alpha t))}\right)$$

where

$$\alpha = \frac{w^2}{\beta}$$

is the ratio of the harmonic force constant $w$ and damping force constant $\beta$. The diffusion constant $D$ is given by

$$D = \frac{k_B T}{m\beta}$$

Nowik et. al.[8,9] generalised this self-correlation function to three dimensions for the over-damped case and obtained a simple formula for the line
Mössbauer spectra were computed for a range of α values and fixed diffusion constant D. The parameter α [8,9] decides the nature of the Mössbauer lineshape. The Mossbauer line-shape is non-Lorentzian for smaller values of α [8,9]. Since Mössbauer lineshape for biological systems is non-Lorentzian in nature, we will focus on smaller values of α in our calculations.

The universal resonance function characteristic of the Brownian motion can be written as [9]

\[
\phi(k, \omega) = \exp\left(-\frac{k^2D}{\alpha}\right) \sum_{m=0}^{\infty} \frac{1}{m!} \left(\frac{k^2D}{\alpha}\right)^m \frac{it_0}{(\omega + it_0 + im\alpha)}
\]  

(10)

where

\[t_0 = \frac{\Gamma_0}{2\hbar}\]  

(11)

Thus, in the present case, for equation (1), \(C(k, \omega)\) is given by

\[C(k, \omega) = \sum_{ge} \sum_m \frac{B_{ge}t_0a_k(m)}{(\omega - \omega_0) + i(t_0 + m\alpha)}\]  

(12)

where

\[a_k(m) = \frac{1}{m!} \left(\frac{k^2D}{\alpha}\right)^m \exp\left(-\frac{k^2D}{\alpha}\right) \exp\left(-t_0 - m\alpha\right)\]  

(13)

Thus \(E(Z, t)\) is given as

\[E(Z, t) = \frac{E_{\omega_0}(Z)}{2\pi} \int d\omega \exp(-i\omega t) \exp\left(-\frac{\mu_nZt_0}{(\omega - \omega_0) + i(t_0 + m\alpha)}\right)\]  

(14)

where

\[\mu_n = K \sum_{ge} B_{ge}\]  

(15)

The summation over ‘ge’ in equation (15) is related to the summing over the transition between the ground and the excited states of nucleus [14,15]. It is important to note that for any particular value of \(\left(\frac{k^2D}{\alpha}\right)\), only a few Lorentzians contribute to the universal line shape [9].
Results:

To illustrate the role of Brownian motion in the time dependence of forward scattering, we first consider a case of a thin target (single line sample) having an effective resonance thickness $\mu_n Z=1$. The results of the numerical calculation for various values of $\alpha$ are shown in the Figure 1. It is evident from this figure, that the natural decay appears (manifested by straight segment of time response curve) faster for smaller values of $\alpha$ and slower for larger values of $\alpha$. The time response in the case of single-line target, having an effective resonant thickness $\mu_n Z=10$ (thick samples) for various values of $\alpha$, are shown in the Figure 2 where dynamical beats appear. It is also clear from this figure that smaller the value of $\alpha$, earlier the dynamical beat occurs and vice versa. The appearance of the dynamical beats is also dependent on the value of $\mu_n Z$. Comparing Figures 1 and 2 it is clear that $\alpha$ and $\mu_n Z$ share a definite relationship, with each other so far as the appearance of dynamical beats are concerned. In order to further investigate this relationship we fix the value of $\alpha$ and see how the variation of $\mu_n Z$ affects the appearance of dynamical beats in the time response behaviour. The results are shown in the Figure 3 which indicates that larger the value of $\mu_n Z$, earlier does the dynamical beat appear and vice versa.

Comparison with diffusion results:

In general the Mössbauer lineshape has a Lorentzian character. The diffusion of the Mössbauer atom causes broadening in this Lorentzian lineshape. For the case of free diffusion, the increase in the Mössbauer linewidth is proportional to the diffusion constant. The presence of the this diffusion constant in the exponential factor of the time response causes an accelerated
decay of the coherent signal. However, the position of the appearance of the 'dynamical beat minimum' is independent of the value of the diffusion constant. All dynamical beats appear at the same time with variable minimum values for different values of the diffusion constants. But in the cases of bounded diffusion inside a potential well and jump diffusion, the universal resonance functions have complicated shapes represented, in general, by the coherent superposition of the Lorentzian lineshapes. The width and weight of each Lorentzian is determined by the specific nature of the diffusion process. Thus, the appearance of the dynamical beats in the time response is sensitive to the diffusion rate and cage-size for the cases of the jump diffusion and bounded diffusion, respectively.

The universal resonance function for the case of Brownian motion in general (and for smaller values of $\alpha$ in particular) is non-Lorentzian. This is because the universal resonance function is the result of superposition of various Lorentzian line shapes. Each Lorentzian line shape has its characteristic weight and width as determined by the value of $\alpha$. This type of the nature of universal resonance function results in the complicated behaviour in the time response. As in the cases of continuous diffusion and jump diffusion, the physical reason for the complicated behaviour in the time response for the case of Brownian motion, is the split of the universal resonance function into several terms.

**Conclusion:**

The method of coherent forward scattering of the SR reveals complex amplitudes of the oscillation of the electromagnetic field in the presence of the Brownian motion.
Figure captions:

Figure 1: The time dependence of nuclear forward scattering of the synchrotron radiation in the presence of Brownian motion for different values for a thin sample of effective thickness $\mu_n Z=1$ corresponding to $\alpha = 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 2.0, 4.0$. The lowermost curve corresponds to $\alpha=0.5$ and uppermost curve corresponds to $\alpha=4.0$. $\alpha$ is units of sec$^{-1}$.

Figure 2: The time dependence of nuclear forward scattering of the synchrotron radiation in the presence of Brownian motion for different values of $\alpha$ for a thick sample of effective thickness $\mu_n Z=10$. $\alpha$ is units of sec$^{-1}$. (1) $\alpha = 0.5$; (2) $\alpha=0.6$; (3) $\alpha=0.7$; (4) $\alpha=0.8$; (5) $\alpha=0.9$; (6) $\alpha=1.0$; (7) $\alpha=2.0$; (8) $\alpha=4.0$.

Figure 3: The time dependence of nuclear forward scattering of the synchrotron radiation with $\alpha=1$ for different values of $\mu_n Z$. $\alpha$ is in units of sec$^{-1}$. (1) $\mu_n Z = 50.0$; (2) $\mu_n Z=40.0$; (3) $\mu_n Z=30.0$; (4) $\mu_n Z=20.0$; (5) $\mu_n Z=10.0$; (6) $\mu_n Z=5.0$; (7) $\mu_n Z=1.0$. 
References:

1. W. Sturhahn, E. E. Alp, T. S. Toellner, P. Hession, M. Hu and J. Sutter, Hyperfine Interactions 113(1998)47

2. F. Parak, E. N. Frolov, R. L. Mossbauer and V. I. Goldanskii, J of Mol. Biology, 145(1981)825

3. S. G. Cohen, E. R. Bauminger, I. Nowik, S. Ofer, J. Yariv, Phys. Rev. Lett. 46(1981)1244

4. K. H. Mayo, F. Parak, R. L. Mossbauer, Phys. Lett. A 82(1981)468

5. H. Keller and P. G. Debrunner, Phys. Rev. Lett., 45(1980)68

6. F. Parak, E. W. Knapp, D. Kucheida, J. Mol. Biol. 161(1982)177

7. F. Parak et. al. Hyperfine Interactions, 58(1990)2381

8. I. Nowik, E. R. Bauminger, S. G. Cohen, S. Ofer, Phys. Rev. Lett. 50(1983)1528

9. I. Nowik, E. R. Bauminger, S. G. Cohen, S. Ofer, Phys. Rev. A 31(1985)2291

10. A. X. Trantwein and H. Winkler, Hyperfine Interactions 123/124(1999)561

11. H. Grunsteudel et. al. Inorganica Chimica Acta 275-276(1997)1334

12. C. Keppler et. al. European Biophys. J. 25(1997)221

13. F. Parak and K. Achterhold, Hyperfine Interactions 123/124(1999)825
14. G.V.Smirnov and V.G.Kohn, Phys. Rev. B 52(1995)3356
15. G.V.Smirnov and V.G.Kohn, Phys. Rev. B 57(1998)5788
16. L.Gunther, J.Zitkova-Wilcox, J.Phys. (France) 35(1974)6-519
17. L.Gunther, J.Zitkova-Wilcox, J.Stat.Phys. 12(1975)205
18. A.Razdan, Eur.Phys.J.B 8(1999)143
19. G.E.Uhlenbeck, L.S.Ornstein, Phys. Rev. 36(1930)823
20. V.G.Kohn, G.V.Smirnov, Hyperfine Interactions 123(1999/2000)327
Figure 1

\log\left(\frac{I(t)}{I(0)}\right) vs. time (ns)
Figure 2

\[ \log(I(t)/I(0)) \] vs. time (ns)
Figure 3

\[
\log\left(\frac{I(t)}{I(0)}\right)
\]

vs.

time (ns)