Stochastic perturbation of the γ-ray angular correlation in the case of a quadrupole interaction

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The angular correlation of γ rays emitted from a quadrupole nucleus perturbed stochastically by an axially asymmetric electric field gradient has been derived explicitly. The average attenuation coefficients for the specific cases of the stochastic environment have been deduced from the general expression. Numerical results of the derived expressions are represented graphically.

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I. INTRODUCTION

The joint probability of emission of two γ rays in cascade is a measure of the correlation function. After emission of the first γ ray and before emission of the second, the nucleus remains in its intermediate state for some time. During the lifetime of the intermediate state, if the nucleus is perturbed by the external environment, then the angular correlation between the two γ rays will consequently be affected. This is called perturbed angular correlation (PAC). The environment being large, its detailed interaction with the system (the nucleus) is impossible to handle. One thus resorts to modeling of the environment by a random process. Matching of the experimental results with the theoretically calculated values of PAC provides the value of the characteristic feature of the random process or environment.

Time-dependent perturbation may be visualized as follows: The radioactive probe nucleus immersed in a liquid acts as a Brownian particle and undergoes a large number of collisions with the bath constituting the surrounding molecules. During the lifetime of the intermediate state, it does not traverse much. What is important is that the orientation of the nucleus changes at each molecular collision. The effect of such perturbation, which is time dependent, could thus provide information about the rotational diffusion constant. It is also observed that transport properties (viscosity and diffusion coefficient) of a superviscous liquid differ strongly from the properties of "normal liquids" [1–3]. Therefore, PAC measurements at different temperatures could also provide information about the variation of the diffusion constant with temperature at different viscosity regions.

The theory of time-dependent perturbed angular correlation is succinctly described and the final expression of the attenuation coefficient is written formally for arbitrary interaction in Ref. [4]. As the number of collisions taking place within a given time is a random phenomenon, the attenuation coefficient should be properly averaged out with respect to the number of collisions. That is, one must consider an ensemble of nuclei having faced a different number of collisions within a given time. Next, one focuses one's attention on the nature of the interaction Hamiltonian. If the nucleus is considered a dipole, it must couple with the perturbing field vector characterized solely by its magnitude and direction. While in the quadrupole approximation of the nucleus, it should couple with the field gradient in the interaction Hamiltonian. In the coordinate system where the electric field gradient (EFG) tensor becomes diagonal, the interaction Hamiltonian is characterized by two parameters, namely, the quadrupole frequency and the asymmetry parameter. After each collision, the orientation of the EFG is also changed; thus it necessitates different coordinate systems in which the EFG becomes diagonal. As the extranuclear perturbing field is random, apart from the collisional average mentioned before, the interaction parameters are random in nature at each time. Therefore, the attenuation coefficient should properly be averaged out with respect to the direction of the field and the magnitude of the strength of the field in the case of dipole approximation or with respect to the direction of the field gradient and the magnitude of the quadrupole frequency and asymmetry parameter in the case of the quadrupole approximation.

Different models have been proposed by different authors in order to carry out the averaging process. A stochastic model of the fluctuating orientation type has been given by Scherer and Blume [5]. Some authors do not consider the flips of the orientation axis; obviously, this model will be true when the direction of the perturbing field does not change appreciably within the lifetime of the intermediate state. The PAC with the "fixed orientation type" but with a Gaussian distribution of the strength of the extranuclear electric or magnetic field has been calculated [6]. This model is popularly known as the fixed orientation Gaussian approximation (FOGA) model. The assumption of fixed orientation of the axis of interaction is inherent in the Blume model [7] in which the perturbing field is allowed to jump between two possible states. The attenuation coefficient of similar type for a magnetic field jumping between three possible states has been given by Spanjaard and Hartmann-Boutron [8]. Subsequently, Bosch and Spehl [9] have generalized their model to include fluctuation of the orientation axis. In all these models no consideration of correlation time of the stochastic processes has been made. However, in reality no process is free from their autocorrelation time. The present authors [4] thus have calculated the expression of PAC when the environment is of fluctuating orientation type and the strength of the field is modified as an Ornstein-Uhlenbeck process. With the quadrupole approximation of the nucleus, Fraunfelder and Steffen [10] have given the perturbation function for a nonaxially symmetric interaction in a polycrystalline source. In their derivation they [10] have assumed the fixed orientation type model. Later, Dattagupta and Blume [11] have considered a model where the EFG does not change its magnitude but can be oriented in any direction. Recently, Martinez, Sanchez, and Vasquez [12] have considered the case of an axially symmetric quadrupole interaction. However, no general expression of time-dependent perturbed angular correlation is available for an axially asymmetric quadrupole interaction where both quadrupole frequency and the asymmetry factor are varying randomly.

In what follows, we extend all these models to include the cases where orientation and both parameters, namely, quadrupole frequency and asymmetry factor, vary randomly. In Sec. II the actual derivation of the attenuation coefficient has been given. Next, we take two specific cases of the random EFG and calculate numerically the derived expressions of averaged attenuation factors. Finally, we offer a few concluding remarks in Sec. III.

II. DETERMINATION OF THE ATTENUATION COEFFICIENT

When two successive γ rays are emitted in the $\mathbf{k}_1$ and $\mathbf{k}_2$ directions, the general form of the angular correlation function [13] is

$$W(\mathbf{k}_1,\mathbf{k}_2,t)=\sum_{\substack{\mathbf{k}_1,\mathbf{k}_2\\ N_1,N_2}} A_{k_1}(1)A_{k_2}(2)G_{k_1k_2}^{N_1N_2}(t)\times[(2k_1+1)(2k_2+1)]^{-1/2}\times Y_{k_1}^{N_1^*}(\vartheta_1,\phi_1)Y_{k_2}^{N_2}(\vartheta_2,\phi_2)\ , \quad (1)$$

where the perturbation factor is given by

$$G_{k_1k_2}^{N_1N_2}(t)=\sum_{m_a,m_b}(-1)^{2I+m_a+m_b}[(2k_1+1)(2k_2+1)]^{1/2}\begin{pmatrix} I & I & k_1\\ m_a' & -m_a & N_1\end{pmatrix}\times\begin{pmatrix} I & I & k_2\\ m_b' & -m_b & N_2\end{pmatrix}\langle m_b|\hat{\Lambda}(t)|m_a\rangle\langle m_a'|\hat{\Lambda}^{\dagger}(t)|m_b'\rangle\ . \quad (2)$$

The factors A_{k_1}, A_{k_2} are related to the matrix elements of the interaction Hamiltonian inducing a transition between the nuclear states and associated with the emission of the γ rays in the $\mathbf{k}_1$ and $\mathbf{k}_2$ directions, respectively. The angles θ_1,ϕ_1 define the direction $\mathbf{k}_1$ and, similarly, θ_2,ϕ_2 for $\mathbf{k}_2$. The central quantity of interest is $G_{k_1k_2}^{N_1N_2}(t)$, which is related to the matrix element of the operator $\hat{\Lambda}(t)$ between magnetic states $|m>s$. The quantity $\hat{\Lambda}(t)$ is the time development operator describing the development of the intermediate nuclear state under extranuclear perturbation through hyperfine interaction. Thus, $\hat{\Lambda}(t)$ is given by

$$\hat{\Lambda}(t)=\exp[-(i/\hbar)\hat{H}t]\ . \quad (3)$$

The operator $\hat{H}$ describes the interaction between the intermediate nuclear state and extranuclear perturbation. In this paper we assume the nucleus to be an electric quadrupole; hence, in the interaction Hamiltonian $\hat{H}$ it should couple with the EFG.

The detailed method of the averaging procedure mentioned in the Introduction and obtaining the expression of the average attenuation coefficient $\langle G_{k_1k_2}^{N_1N_2}(t)\rangle$ in the case of an arbitrary interaction Hamiltonian has been given formally in Ref. [4]. Here, we only quote the expressions

$$\langle G_{k_1k_2}^{N_1N_2}(t)\rangle=\sum_{n=0}^{\infty}\lambda^n e^{-\lambda t}E\{G_{k_1k_2}^{N_1N_2}|n\}\ , \quad (4)$$

where λ refers to the mean number of collisions per unit time and

$$E\{G_{k_1k_2}^{N_1N_2}|n\}=(1/4\pi)^{n+1}\int_{(0\le t_1\le t_2\le\cdots\le t_n\le t)}\cdots\int\prod_{i=1}^{n}dt_i\{G_{k_1k_2}^{N_1N_2}\}_{t_1,t_2,\ldots,t_n}\times\prod_{i=0}^{n}(d\omega_i d\eta_i d\Omega_i)P(\omega_0,\eta_0|0;\omega_1,\eta_1|t_1;\ldots;\omega_n,\eta_n|t_n)\ . \quad (5)$$

The quantity $\{G_{k_1k_2}^{N_1N_2}\}_{t_1,t_2,\ldots,t_n}$ corresponds to the evaluation of $G_{k_1k_2}^{N_1N_2}(t)$ at n time points $t_1,t_2,\ldots,t_n$ $(t\ge t_n\ge t_{n-1}\ge\cdots\ge t_1\ge 0)$. The variables ω,η refer to the characteristics of the interaction Hamiltonian (explained below) and Ω corresponds to the direction for which EFG is diagonal. The subscripts attached to them refer to the time points $\{t_i\}$ at which the collisions take place. The quantity

$$P(\omega_0,\eta_0|0;\omega_1,\eta_1|t_1;\ldots;\omega_n,\eta_n|t_n)$$

in Eq. (5) is the joint conditional probability of having the specified values of the characteristics of the interaction Hamiltonian after choosing n ordered time points. The hyperfine quadrupole interaction Hamiltonian $\hat{H}$ takes the following form in the present case:

$$\hat{H}=\omega\{3\hat{I}_z^2-\hat{I}^2-(\eta/2)(\hat{I}_+^2+\hat{I}_-^2)\}\ , \quad (6)$$

where the quadrupole frequency ω is defined by

$$\omega = eQV_{zz}/4I(2I-1) \ . \tag{7}$$

In expression (7), I, Q, and e are the nuclear spin, the electric quadrupole moment, and the electronic charge, respectively. The quantity η refers to the asymmetry factor and is given by

$$\eta = (V_{xx} - V_{yy})/V_{zz} \ . \tag{8}$$

The quantity V_{ii} is the double derivative of the potential in the suitably chosen axes for which the EFG tensor becomes diagonal and the axes are chosen such that

$$|V_{xx}| \le |V_{yy}| \le |V_{zz}| \ .$$

This restriction implies that the asymmetry factor η would always lie between 0 and 1. In this frame of axes where EFG is diagonal, if the $|p\rangle$ refer to magnetic states, then the nontrivial matrix elements of $\hat{H}$ are given by

$$\langle Ip|\hat{H}|Ip\rangle = \hbar\omega[3p^2 - I(I+1)] \ , \tag{9}$$

$$\langle Ip'|\hat{H}|Ip\rangle = \hbar\omega(\eta/2)[(I\mp p-1)(I\mp p) \times (I\pm p+1)(I\pm p+2)]^{1/2} \times \delta_{p',p'\pm 2} \ . \tag{10}$$

We now evaluate Eq. (4) term by term with the interaction Hamiltonian given by Eq. (6).

A. Class with static interaction

The first term in Eq. (4), corresponding to $n=0$, represents the class of nuclei having no flip of the orientation axis. The quantity that is to be evaluated is

$$(1/4\pi)\int\int\int d\omega_0 d\eta_0 d\Omega_0 P(\omega_0,\eta_0) G_{k_1k_2}^{N_1N_2}(t) \ , \tag{11}$$

where $G_{k_1k_2}^{N_1N_2}(t)$ is given by Eq. (2). We note that $|m\rangle, |m'\rangle$ are magnetic states corresponding to our fixed z axis in laboratory frame although the matrix elements of the interaction Hamiltonian are known [Eqs. (9) and (10)] in the frame, say, z', where the EFG tensor is diagonal. Let $|p\rangle$ be the eigenstates corresponding to the frame where EFG is diagonal. Thus we must express the matrix elements in terms of $|p\rangle$. We write

$$|m\rangle = \sum_p |p\rangle\langle p|m\rangle \ , \tag{12}$$

where $\langle p|m\rangle$ are known to be Wigner coefficients $D_{pm}^{(I)}(\phi_0,\vartheta_0,0)$, with $(\phi_0,\vartheta_0,0)$ being the Eulerian angles of the direction Ω_0 with respect to the z axis of the laboratory frame. Inclusion of the identity

$$I = \sum_p |p\rangle\langle p| \tag{13}$$

in matrix elements of $\hat{\Lambda}, \hat{\Lambda}^\dagger$ in Eq. (2) implies that we would get the product of four Wigner coefficients. Then we use the integration formula

$$\int D_{aa'}^{A}(\Omega)D_{bb'}^{B}(\Omega)D_{cc'}^{C}(\Omega)D_{dd'}^{D}(\Omega)d\Omega = (8\pi^2)\sum_K(-1)^{m-1}(2K+1)\begin{bmatrix} A & B & K \\ a & b & l \end{bmatrix}\begin{bmatrix} A & B & K \\ a' & b' & m \end{bmatrix}\begin{bmatrix} C & D & K \\ c & d & -1 \end{bmatrix}\begin{bmatrix} C & D & K \\ c' & d' & -m \end{bmatrix} . \tag{14}$$

This formula is obtained using

$$D_{aa'}^{A}(\Omega)D_{bb'}^{B}(\Omega) = \sum_C (2C+1)\begin{bmatrix} A & B & C \\ a & b & c \end{bmatrix}\begin{bmatrix} A & B & C \\ a' & b' & c' \end{bmatrix} D_{cc'}^{C}(\Omega) \ , \tag{15}$$

the orthogonality of the Wigner coefficients, namely,

$$\int D_{mm'}^{j*}(\Omega)D_{MM'}^{J}(\Omega)d\Omega = (8\pi^2/2j+1)\delta_{jJ}\delta_{mM}\delta_{m'M'} \ , \tag{16}$$

and noting that

$$D_{mm'}^{j}(\alpha\beta\gamma) = (-1)^{m-m'}D_{-m-m'}^{j}(\alpha\beta\gamma) \ . \tag{17}$$

When the integrated result is plugged into Eq. (2), the consistency criterion demands that $N_1 = N_2$. This condition is a characteristic of the isotropy of the model. We could sum the expression over m_a, m_b and dummy index K using the property

$$\sum_{\alpha,\beta}(2C+1)\begin{bmatrix} a & b & c \\ \alpha & \beta & \gamma \end{bmatrix}\begin{bmatrix} a & b & c' \\ \alpha & \beta & \gamma' \end{bmatrix} = \delta_{cc'}\delta_{\gamma\gamma'} \ , \tag{18}$$

and get the closed expression for the attenuation coefficient, when there is no flipping, as

$$E\{G_{KK}|n=0\} = \int\int d\omega_0 d\eta_0 P(\omega_0,\eta_0) G_{KK}^{(0)} \ , \tag{19}$$

where $G_{KK}^{(0)}$ is given by

$$G_{KK}^{(0)}=\sum_{\{p^{(i)}\}_{i=1,\ldots,4}}(-1)^{[p^{(1)}-p^{(2)}]}\begin{bmatrix} I & I & K \\ p^{(4)} & -p^{(1)} & l \end{bmatrix}\begin{bmatrix} I & I & K \\ p^{(3)} & -p^{(2)} & l \end{bmatrix}\langle p^{(1)}|\hat{\Lambda}|p^{(2)}\rangle\langle p^{(3)}|\hat{\Lambda}^{\dagger}|p^{(4)}\rangle \ . \quad (20)$$

The matrix elements of $\hat{\Lambda}$ and $\hat{\Lambda}^{\dagger}$ involve interaction parameters ω_0,η_0. In Eq. (20) we use the symbol K as a subscript in G. In fact, the summation formula (18) results in $K_1=K_2=K$ (say).

B. Class with one flip

The $n=1$ term in Eq. (4) is the following:

$$\lambda e^{-\lambda t}/(1/4\pi)^2\int_0^t dt_1\prod_{i=0}^{1}(d\omega_i d\eta_i d\Omega_i)\times P(\omega_0,\eta_0|0;\omega_1,\eta_1|t_1)\times\{G_{k_1k_2}^{N_1N_2}(t)\}_{t_1} \ , \quad (21)$$

where $\{G_{k_1k_2}^{N_1N_2}(t)\}_{t_1}$ is given by Eq. (2), with

$$\hat{\Lambda}(t)\to\hat{\Lambda}(t-t_1)\hat{\Lambda}(t_1) \ . \quad (22)$$

During time 0 to t_1, the EFG tensor is diagonal in one frame and, after collision at t_1, as the orientation of the EFG flips, we need another frame to calculate the matrix element of $\hat{\Lambda}(t-t_1)$. This indicates that we need two subscripts of $|p\rangle$, while dummy indices that appear in the superscript of $|p\rangle$ always run from 1 to 4 as in Eq. (20). Since we are using Euler angles with respect to the laboratory frame as arguments in the Wigner coefficients, we need to insert identity operators of the type

$$\sum_{m_1}|m_1\rangle\langle m_1|=I \ , \quad \sum_{m_1'}|m_1'\rangle\langle m_1'|=I \ , \quad (23)$$

apart from the type (13). Thus here we will get eight D functions, four of which have the arguments Ω_0 and the other four have the arguments Ω_1. Integrations with respect to Ω_0 and Ω_1 are performed first. Then summations over m_a,m_b,m_1,m_1' are carried out to obtain

$$E\{G_{KK}|n=1\}=\int_0^t dt_1\prod_{i=0}^{1}(d\omega_i d\eta_i)P(\omega_0,\eta_0|0;\omega_1,\eta_1|t_1)\times G_{KK}^{(0)}(t-t_1)G_{KK}^{(0)}(t_1) \quad (24)$$

where $G_{KK}^{(0)}$ is given by Eq. (20).

C. Class with n flips

Noting the above systematics, one readily writes the central quantity for general n, and the attenuation coefficient can be written more neatly as

$$\langle G_{KK}(t)\rangle=\sum_{n=0}^{\infty}\lambda^n e^{-\lambda t}\int_0^t dt_n\int_0^{t_n}dt_{n-1}\cdots\int_0^{t_2}dt_1\prod_{i=0}^{n}(d\omega_i d\eta_i)P(\omega_0,\eta_0|0;\omega_1,\eta_1|t_1;\ldots;\omega_n,\eta_n|t_n)\times G_{KK}^{(0)}(t-t_n)G_{KK}^{(0)}(t_n-t_{n-1})\cdots G_{KK}^{(0)}(t_1) \ . \quad (25)$$

We note that the attenuation coefficient can be expressed in terms of $G_{KK}^{(0)}$, whose expression is given by Eq. (20) when we consider the case of no flipping, namely, the case described in Sec. II A. This fact simplifies enormously the algebra of evaluating the attenuation coefficient. In order to evaluate $G_{KK}^{(0)}$, we need to see more closely the matrix elements of $\hat{\Lambda}$. If we define

$$\hat{\alpha}=\omega(-it/\hbar)[3\hat{I}_z^2-\hat{I}^2] \ ,$$
$$\hat{\beta}=\omega(-it/\hbar)[\hat{I}_+^2+\hat{I}_-^2] \ , \quad \varepsilon=(\eta/2) \ , \quad (26)$$

then $\hat{\Lambda}(t)$ in Eq. (3) can be written as

$$\hat{\Lambda}(t)=\exp[\hat{\alpha}+\varepsilon\hat{\beta}] \ . \quad (27)$$

Since $0\le\varepsilon\le 0.5$, for small ε we can write

$$\hat{\Lambda}(t)=e^{\hat{\alpha}}+\varepsilon\int_0^1 e^{(1-\lambda)\hat{\alpha}}\hat{\beta}e^{\lambda\hat{\alpha}}d\lambda+O(\varepsilon^2) \ . \quad (28)$$

In the representation $|Ip\rangle$, $\hat{\alpha}$ is diagonal, as can be seen from Eq. (9). Therefore,

$$\langle p|\hat{\Lambda}|p'\rangle=\langle p|e^{\hat{\alpha}}|p'\rangle+\varepsilon\sum_{p'',p'''}\int_0^1\langle p|e^{(1-\lambda)\hat{\alpha}}|p''\rangle\langle p''|\hat{\beta}|p'''\rangle\times\langle p'''|e^{\lambda\hat{\alpha}}|p'\rangle d\lambda \ , \quad (29)$$

which is simplified to

$$\langle p|\hat{\Lambda}|p'\rangle=e^{\alpha_p}\delta_{pp'}+\varepsilon\int_0^1 e^{(1-\lambda)\alpha_p}\langle p|\hat{\beta}|p'\rangle e^{\lambda\alpha_{p'}}d\lambda \ . \quad (30)$$

From Eqs. (9) and (10) it is clear that

$$\alpha_p=(it)\omega[3p^2-I(I+1)] \ ,$$
$$\langle p|\hat{\beta}|p'\rangle=(-it)\omega\gamma_{p,p\pm 2}\delta_{p',p\pm 2} \ , \quad (31)$$

where $\gamma_{p,p\pm 2}$ is defined by

$$\gamma_{p,p\pm 2}=[(I\mp p-1)(I\mp p)(I\pm p+1)(I\pm p+2)]^{1/2} \ . \quad (32)$$

Thus we get the following nontrivial matrix elements of $\hat{\Lambda}$ up to order ε:

$$\langle p|\hat{\Lambda}|p\rangle = e^{\alpha_p} , \quad (33)$$

$$\langle p|\hat{\Lambda}|p\pm 2\rangle = \varepsilon\gamma_{p,p\pm 2}\{\pm[\tfrac{1}{12}(p\pm 1)](e^{\alpha_{p\pm 2}}-e^{\alpha_p})(1-\delta_{p,\mp 1}) \times(-it)\omega e^{\alpha_p}\delta_{p,\mp 1}\} . \quad (34)$$

Again, from the definition of the $3j$ symbol in Eq. (20), one immediately gets

$$p^{(4)}-p^{(1)}=p^{(3)}-p^{(2)} , \quad (35)$$

and we have already seen that the nontrivial matrix elements for $\langle p^{(1)}|\hat{\Lambda}|p^{(2)}\rangle$ exist for

$$p^{(2)}=p^{(1)} , \quad p^{(2)}=p^{(1)}+2 , \quad p^{(2)}=p^{(1)}-2 . \quad (36)$$

Combining (35) with (36), we obtain the following:

$$\begin{aligned} &p^{(2)}=p^{(1)} \text{ must be associated with } p^{(3)}=p^{(4)} , \\ &p^{(2)}=p^{(1)}+2 \text{ must be associated with } p^{(3)}=p^{(4)}+2 , \\ &p^{(2)}=p^{(1)}-2 \text{ must be associated with } p^{(3)}=p^{(4)}-2 . \end{aligned} \quad (37)$$

Thus, nonzero components that would appear in the summation of $G_{KK}^{(0)}$ will be

$$\langle p^{(1)}|\hat{\Lambda}|p^{(2)}\rangle\langle p^{(4)}|\hat{\Lambda}|p^{(3)}\rangle^* , \quad (38a)$$

$$\langle p^{(1)}|\hat{\Lambda}|p^{(2)}+2\rangle\langle p^{(4)}|\hat{\Lambda}|p^{(3)}+2\rangle^* , \quad (38b)$$

$$\langle p^{(1)}|\hat{\Lambda}|p^{(2)}-2\rangle\langle p^{(4)}|\hat{\Lambda}|p^{(3)}-2\rangle^* , \quad (38c)$$

where the respective factors are given explicitly in Eqs. (33) and (34). From condition (36), we can see that the factor $(-1)^{[p^{(1)}-p^{(2)}]}$ will be absent from Eq. (20) and the summation is over variables $p^{(1)}$ and $p^{(4)}$ only; the variables $p^{(2)}$ and $p^{(3)}$ will be constrained by the condition (37). Hence, the final expression of the average attenuation factor is given by Eq. (25) where $G_{KK}^{(0)}$ takes the following form:

$$G_{KK}^{(0)} = \sum_{p^{(1)},p^{(4)}} \begin{bmatrix} I & I & K \\ p^{(4)} & -p^{(1)} & l \end{bmatrix} \times \begin{bmatrix} I & I & K \\ p^{(3)} & -p^{(2)} & l \end{bmatrix} \times\langle p^{(1)}|\hat{\Lambda}|p^{(2)}\rangle\langle p^{(3)}|\hat{\Lambda}^{\dagger}|p^{(4)}\rangle . \quad (39)$$

Equations (38a)–(38c) show that for static interaction where there is no flipping of the orientation of the EFG tensor, we have terms free from η and terms proportional to η^2. Considering the case where the EFG flips only once, we get terms, in the attenuation coefficient, free from η, proportional to η^2 and η^4. This implies that for n flips, we get terms proportional to

$$\eta^0, \eta^2, \eta^4, \ldots, (\eta^2)^{n+1} .$$

Noting that $0\le\eta\le 1$, we would expect the series to converge rapidly with an increase in the number of flips.

Until now, we have made no assumption about the conditional probability of the random variables ω and η. For the simplest case we assume that there is no correlation of strength parameters, namely, ω_i,η_i at different instants. This fact implies

$$P(\omega_0,\eta_0|0;\omega_1,\eta_1|t_1;\ldots;\omega_n,\eta_n|t_n)=\prod_{i=0}^{n} P(\omega_i,\eta_i|t_i) . \quad (40)$$

With this assumption, the evaluation of the average attenuation factor is considerably simplified and is given for $k=2$ as follows:

$$\langle G_{22}(t)\rangle = \sum_{n=0}^{\infty} \lambda^n e^{-\lambda t}\int_0^t dt_n \int_0^{t_n} dt_{n-1}\cdots\int_0^{t_2} dt_1 \bar{G}_{22}^{(0)}(t-t_n)\bar{G}_{22}^{(0)}(t_n-t_{n-1})\cdots\bar{G}_{22}^{(0)}(t_1) , \quad (41)$$

where

$$\bar{G}_{22}^{(0)} = \int\int G_{22}^{(0)}(t,\omega,\eta)P(\omega,\eta)d\omega\, d\eta . \quad (42)$$

We further assume that the correlation between these variables is also absent at every instant. That is,

$$P(\omega,\eta)=P(\omega)P(\eta) . \quad (43)$$

This decoupling greatly simplifies the evaluation of $\bar{G}_{22}^{(0)}$. Next, we take two specific models of the random environment and calculate numerically the attenuation coefficients.

D. δ distribution of the perturbing field

This model refers to the fact that although the direction of the interacting field is allowed to fluctuate at random with uniform distribution in $[0,4\pi]$, the size of the interaction Hamiltonian remains fixed in time. Hence, the quadrupole frequency as well as the asymmetry parameter take the fixed specific values

$$P(\omega_i|t_i)=\delta(\omega_i-\bar{\omega}) , \quad P(\eta_i|t_i)=\delta(\eta_i-\bar{\eta}) , \quad \forall i . \quad (44)$$

After considerable algebra, for $I=2$, the expression of $\bar{G}_{22}^{(0)}$ in Eq. (41) turns out to be

$$\bar{G}_{22}^{(0)}(t,\bar{\omega},\bar{\eta})=[a_0+a_1\cos\tau+a_3\cos 3\tau+a_4\cos 4\tau] +(\bar{\eta}^2/35)[\tau^2+\tau(\sin 3\tau-3\sin\tau)] , \quad (45)$$

with $\tau=\omega_0 t$ and $\omega_0=3\bar{\omega}$. The Alder coefficients [5(b)] a_0, a_1, a_3, and a_4 are obtained as

$$a_0=0.371\,428\,5 , \quad a_1=0.057\,142\,8 , \quad a_3=0.342\,857\,1 , \quad a_4=0.228\,571\,4 . \quad (46)$$

E. Gaussian distribution of quadrupole frequency

Next, we take another illustration where the probability distribution of the quadrupole frequency is assumed to be Gaussian at each time; however, the size of the asymmetry parameter assumes a fixed specified value:

$$P(\omega_i|t_i)=(2\pi\langle\omega_0^2\rangle)^{-1/2}\exp[-\omega_i^2/2\langle\omega_0^2\rangle]\ ,$$
$$P(\eta_i|t_i)=\delta(\eta_i-\bar{\eta})\ ,\quad \forall i \tag{47}$$

where $\langle\omega_0^2\rangle$ and $\bar{\eta}$ are some fixed specified value. The expression of $\bar{G}_{22}^{(0)}$ in this case is obtained with the help of Eq. (45) and it has the following form:

$$\bar{G}_{22}^{(0)}(t,\langle\omega_0^2\rangle,\bar{\eta})=[a_0+a_1\exp(-\tau^2/2)+a_3\exp(-9\tau^2/2)+a_4\exp(-8\tau^2)]$$
$$+(\bar{\eta}^2\tau^2/35)\{1+3[\exp(-9\tau^2/2)-\exp(-\tau^2/2)]\}\ , \tag{48}$$

with $\tau=\langle\omega_0^2\rangle^{1/2}t$, and the coefficients a_0, a_1, a_3, and a_4 are given by Eq. (46).

Numerical calculations of the average attenuation factors have been carried out for some illustrative cases. Attenuation coefficients (41) with $\bar{G}_{22}^{(0)}$ given by Eq. (45) (in the case of a δ function distribution of the quadrupole frequency) for different values of (λ/ω_0), $\bar{\eta}$ as a function of $(\omega_0 t/2\pi)$ are shown graphically in Fig. 1. For the axially symmetric case, i.e., when $\bar{\eta}=0$, the results match those of Martinez, Sanchez, and Vasquez [12]. The attenuation coefficients obtained with the help of Eq. (48) (in the case of Gaussian distribution of the quadrupole frequency) are plotted for different values of $\lambda/\langle\omega_0^2\rangle^{1/2},\bar{\eta}$ as a function of $\langle\omega_0^2\rangle^{1/2}t$ in Fig. 2. Calculation shows that after some interval, the asymmetry parameter displays its contribution to the average attenuation coefficient. In the case of a Gaussian distribution of the quadrupole frequency, the attenuation coefficient always saturates to an asymptotic value for very large time, when exponential factors dominate over the multiplicative τ^2 factor. In Fig. 2 we display its behavior where $\langle\omega_0^2\rangle^{1/2}t$ takes values up to 10. For very small values of $\bar{\eta}$, $\langle G_{22}(t)\rangle$ does not differ much from the symmetric case. Increased values of the mean number of collisions per unit time blur the sensitive dependence of the attenuation factor on $\bar{\eta}$; however, expressions (45) and (48) suggest that its dependence would be recovered with large values of nuclear spin (in the case of a Gaussian distribution) or electric quadrupole moment in general.

III. CONCLUSIONS

We derived explicitly the expression for the time-differential attenuation coefficient when the nucleus is approximated as a quadrupole interacting with randomly fluctuating extranuclear EFG. We use the approximate formula (28) in order to obtain the closed form expression for the perturbation factor for arbitrary I. The first nontrivial term involving asymmetry parameter $\bar{\eta}$ appearing in the expression for the average attenuation factor is proportional to $\bar{\eta}^2/4$. Since $0\le\bar{\eta}\le 1$, this approximation therefore is not very severe. For static interaction, however, one can diagonalize the full Hamiltonian (6), and obtain the unitary matrix that diagonalizes the Hamiltonian. In Eq. (20), one should then sum over the eigenvector variables. But there is no systematic expression for eigenvalues as a function of I in the general asymmetry case. Hence, in this way one cannot obtain the expression for PAC function for arbitrary I. For $\bar{\eta}=0$, however, the expression (25) together with (39) is exact since in the representation $|p\rangle$, the Hamiltonian is diagonal and assumes simple form. One would then get back the result of Martinez, Sanchez, and Vasquez [12]. Expression (45), where the EFG does not change in magnitude but can be oriented in any direction, matches with the result obtained by Dattagupta and Blume [11] for the specific case. In the derivation we assume no correlation of the strength (ω_i,η_i) of the interaction Hamiltonian at different times [assumption (40)]. This implies that the time-differential attenuation coefficient $\langle G_{22}(t)\rangle$ may be

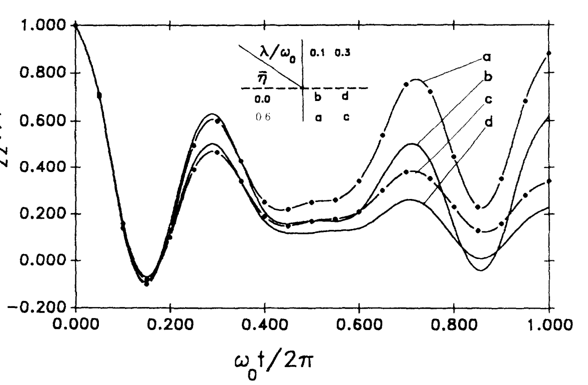

FIG. 1. Attenuation coefficients $\langle G_{22}(t)\rangle$ for $I=2$ are plotted against $(\omega_0 t/2\pi)$, in the case of a δ function distribution of quadrupole frequency for different values of (λ/ω_0) and $\bar{\eta}$.

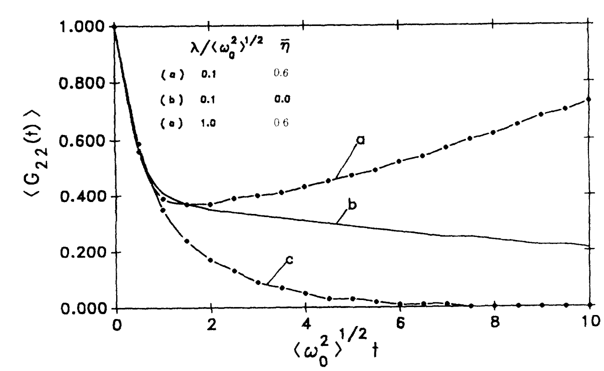

FIG. 2. Attenuation coefficients $\langle G_{22}(t)\rangle$ for $I=2$ are plotted against $\langle\omega_0^2\rangle^{1/2}t$, in the case of a Gaussian distribution of quadrupole frequency for different values of $(\lambda/\langle\omega_0^2\rangle^{1/2})$ and $\bar{\eta}$.

obtained by solving the following inhomogeneous Volterra integral equation of the second kind:

$$\langle G_{22}(t)\rangle=\bar{G}_{22}^{(0)}e^{-\lambda t}+\lambda\int_0^t\langle G_{22}(t')\rangle\bar{G}_{22}^{(0)}(t-t')\times e^{-\lambda(t-t')}dt' . \quad (49)$$

The structure of Eq. (49) suggests that the Laplace transform of $\langle G_{22}(t)\rangle$, namely, $\langle\tilde{G}_{22}(p)\rangle$, can be expressed more compactly in terms of the Laplace transform of $\bar{G}_{22}^{(0)}(t)$ or $\tilde{\bar{G}}_{22}^{(0)}(p)$ as

$$\langle\tilde{G}_{22}(p)\rangle=\tilde{\bar{G}}_{22}^{(0)}(\lambda+p)/[1-\lambda\tilde{\bar{G}}_{22}^{(0)}(\lambda+p)] . \quad (50)$$

For the two illustrative cases we give explicitly the Laplace transforms of the kernel $\bar{G}_{22}^{(0)}(t)$. For the case of the δ function distribution of the perturbing field, it is given by

$$\tilde{\bar{G}}_{22}^{(0)}(p)=(1/\omega_0)\tilde{\bar{G}}_{22}^{(0)}(p/\omega_0) , \quad (51)$$

where

$$\tilde{\bar{G}}_{22}^{(0)}(\bar{p})=[a_0/\bar{p}+a_1\bar{p}/(\bar{p}^2+1)+a_3\bar{p}/(\bar{p}^2+9)+a_4\bar{p}/(\bar{p}^2+16)]+(\bar{\eta}^2/35)[2/\bar{p}^3+6\bar{p}/(\bar{p}^2+9)^2-6\bar{p}/(\bar{p}^2+1)^2] , \quad (52)$$

with $\bar{p}=p/\omega_0$. For the case of a Gaussian distribution of quadrupole frequency, the corresponding expression would be

$$\tilde{\bar{G}}_{22}^{(0)}(p)=(1/\langle\omega_0^2\rangle^{1/2})\tilde{\bar{G}}_{22}^{(0)}(p/\langle\omega_0^2\rangle^{1/2}) , \quad (53)$$

where

$$\tilde{\bar{G}}_{22}^{(0)}(\bar{p})=a_0/\bar{p}+(\pi/2)^{1/2}[a_1\exp(\bar{p}^2)\mathrm{erfc}(\bar{p}/\sqrt{2})+a_3/3\exp(\bar{p}^2/9)\mathrm{erfc}(\bar{p}/3\sqrt{2})+a_4/4\exp(\bar{p}^2/16)\mathrm{erfc}(\bar{p}/4\sqrt{2})]+(\bar{\eta}^2/35)\{2/\bar{p}^3+3(\pi/2)^{1/2}[\tfrac{1}{27}(1+2\bar{p}^2/9)\exp(\bar{p}^2/9)\mathrm{erfc}(\bar{p}/3\sqrt{2})-(1+2\bar{p}^2)\exp(\bar{p}^2)\mathrm{erfc}(\bar{p}/\sqrt{2})]-3\bar{p}[\tfrac{1}{81}\exp(\bar{p}^2/18)-\exp(\bar{p}^2/2)]\} , \quad (54)$$

with $\bar{p}=p/\langle\omega_0^2\rangle^{1/2}$, and the symbol erfc denotes the complement of the error function. Substituting expressions (52) and (54) into Eq. (50), the Laplace inversion will directly yield the closed form expression of attenuation coefficient $\langle G_{22}(t)\rangle$ for the above two cases.

The incorporation of correlation of the strength variables at different times would necessitate the introduction of another parameter in the theory. Dependence of the PAC function on that parameter is measurable when the time-resolving power of the instrument will be very small. With these refined expressions for the PAC function, it would be interesting to study the transport properties of superviscous liquid and compare with the experimental results.

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