

## Perturbation of Gamma-Ray Angular Correlations Following Continual Random Reorientation of the Axis of Interaction

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*Abstract.* Attenuation coefficients of perturbed gamma-ray angular correlations are calculated for classical extranuclear fields which fluctuate at random both in direction and in magnitude. Explicit expressions are given for time-integral attenuation coefficients. Time-differential attenuation coefficients have been calculated numerically. The theory applies to any correlation time.

### 1. Introduction

In a recent paper [1] we have described the exactly soluble FOGA model for the perturbation of angular correlations due to randomly-fluctuating extranuclear magnetic or electric fields. The model assumes that the strength of the perturbing field can change at random according to a Gaussian probability distribution, while the direction of the field has to remain fixed with time. For an isotropic interaction one must average over the direction as, for example, in magnetic domains. The same assumption of a “fixed orientation” of the axis of interaction is inherent in the Blume model [2] in which the perturbing magnetic field is allowed to jump between two possible states. Similarly, the attenuation coefficients for a magnetic field of the fixed orientation type jumping between three possible states have been given by Spanjaard and Hartmann-Boutron [3].

It has been argued [4] that the fluctuating isotropic hyperfine interaction in highly-ionized and -excited free atoms can be simulated by such stochastic “fixed orientation” models, provided that the average lifetime of the electronic states, described by the correlation time  $\tau_c$  of the model, is not much smaller than the typical time of observation of the perturbation process. In this case the dipole rule  $\Delta m_J = 0, \pm 1$  for the free decay of the electronic states favours the approximate preservation of the initial orientation of the hyperfine field if  $J > 1$ . Fixed orientation models become meaningless for free ions, however, if the correlation time is small compared to the observation time. In this case one must average over the direction of the perturbing field within the observation time in each individual nucleus. Such situations may conveniently be simulated by stochastic models using the simplifying assumption that the *direction* of the perturbing field fluctuates completely at random, i.e. that after each “jump” the orientation of the axis of interaction is equally likely in any direction. For free ions this assumption obviously contradicts the dipole rule. Nevertheless, such models are of interest because the actual behaviour of perturbing systems can usually be approximated by one or other of the limiting cases of “fixed orientation” or “fluctuating orientation”. Throughout this paper the latter term will be understood to signify random changes of the axis of interaction without memory of the past.

A stochastic model of the fluctuating orientation type has been given by Scherer [5]. This model applies to the isotropic magnetic hyperfine interaction  $a \mathbf{I} \cdot \mathbf{J}$  for free atoms in a gas. The orientation of the atomic angular momentum  $\mathbf{J}$  is assumed to be completely random after each collision with a neighbouring gas atom. The size of  $\mathbf{J}$  is assumed to be preserved. In the following a model of the fluctuating orientation type will be described which uses the classical interaction Hamiltonian  $-\boldsymbol{\mu} \cdot \mathbf{H}$ . Some of the stochastic details of this model are similar

to those of the Scherer model. It will be allowed, however, that the orientation as well as the size of the perturbing interaction fluctuates at random. In Section 2 a short description of the statistics of our model will be given. In Section 3 the intermixing of the quantum mechanical and probabilistic aspects of the model will be formulated and in Section 4 explicit results for time-integral attenuation coefficients are given and compared with the perturbation theoretic results of Abragam and Pound [6]. In Section 5 time-differential attenuation coefficients will be discussed.

## 2. Statistics of the Fluctuating Orientation Model

We start by selecting a sample of random points  $t_1, t_2, \dots, t_n, \dots$  having uniform probability distribution in the time interval  $(0, t)$  and assume ordering according to  $0 < t_1 < t_2 < \dots < t_n < \dots < t$ . The model implies that the probability of having exactly  $n$  points in the time interval of length  $t$  is given by the Poisson distribution

$$P\{n \text{ in } (0, t)\} = \frac{(\lambda t)^n}{n!} e^{-\lambda t}, \quad (1)$$

where  $\lambda$  is the mean number of points per unit time and  $\tau_c = 1/\lambda$  will be called the correlation time. We assume that in each time interval  $(t_n, t_{n+1})$  the size as well as the direction of the perturbing magnetic field  $\mathbf{H}$  is fixed. Exactly at the points  $t_n$  the direction  $\Omega_{n-1}$  of the field is abruptly changed and it will be allowed that also the size of the field  $H_{n-1}$  is changed, i.e. the initial magnetic field of strength  $H_0$ , pointing in the direction  $\Omega_0$ , abruptly jumps to  $H_1$  in the direction  $\Omega_1$  at the random point  $t_1$ . The angle of Larmor-precession around the direction  $\Omega_0$  is given by

$$\omega_0 t_1 = -g_I \frac{\mu_N}{\hbar} H_0 t_1,$$

the precession angle around  $\Omega_1$  is  $\omega_1(t_2 - t_1)$  and  $\omega_1$  is allowed to be different from  $\omega_0$ , and so on. The events  $\{\Omega_0 \text{ in } (0, t_1)\}, \{\Omega_1 \text{ in } (t_1, t_2)\}, \dots, \{\Omega_n \text{ in } (t_n, t_{n+1})\}$  are assumed to be independent and uniformly distributed in the solid angle  $4\pi$ , i.e. the  $n$ -th order probability distribution [7] has the form

$$W(\Omega_0 0; \Omega_1 t_1; \dots \Omega_n t_n) = W(\Omega_0 0) \cdot W(\Omega_1 t_1) \dots W(\Omega_n t_n) = \left(\frac{1}{4\pi}\right)^n. \quad (2)$$

Similarly, the events  $\{\omega_0 \text{ in } (0, t_1)\}; \{\omega_1 \text{ in } (t_1, t_2)\} \dots$  are assumed to be independent:

$$W(\omega_0 0; \omega_1 t_1; \dots \omega_n t_n) = W(\omega_0 0) \cdot W(\omega_1 t_1) \dots W(\omega_n t_n) \quad (3)$$

and the first order distribution  $W(\omega)$  has to be specified. Finally, we assume that the strength of the interacting field and its direction are uncorrelated, i.e. the joint distribution of  $\omega$  and  $\Omega$  is given by

$$W(\omega, \Omega) = W(\omega) \cdot W(\Omega). \quad (4)$$

## 3. Calculation of the Attenuation Factors

### 3.1. General Expressions

The general form of the perturbed angular correlation function is given by [8]

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = \sum_{\substack{k_1, k_2 \\ N_1, N_2}} A_{k_1}(1) \cdot A_{k_2}(2) \cdot G_{k_1 k_2}^{N_1 N_2}(t) [(2k_1 + 1)(2k_2 + 1)]^{-\frac{1}{2}} Y_{k_1}^{N_1}(\Theta_1, \Phi_1) \cdot Y_{k_2}^{N_2}(\Theta_2, \Phi_2) \quad (5)$$

with the perturbation factor

$$G_{k_1 k_2}^{N_1 N_2}(t) = \sum_{m_a, m_b} (-1)^{2I + m_a + m_b} [(2k_1 + 1)(2k_2 + 1)]^{\frac{1}{2}} \begin{pmatrix} I & I & k_1 \\ m'_a - m_a & N_1 & \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b - m_b & N_2 & \end{pmatrix} \langle m_b | \Lambda(t) | m_a \rangle \langle m'_b | \Lambda(t) | m'_a \rangle^*. \quad (6)$$

The time evolution operator  $\Lambda(t)$  describes the change in population of the substates  $|m\rangle$  with time and is given by the interaction Hamiltonian  $\mathcal{H} = -\boldsymbol{\mu} \cdot \mathbf{H}$  according to

$$\Lambda(t' - t'') = \exp \left[ -\frac{i}{\hbar} \mathcal{H} \cdot (t' - t'') \right] \quad (7)$$

if  $\mathcal{H}$  does not depend on time. In fact, in our model, Eq. (7) represents the correct operator within each time interval  $(t_n, t_{n+1})$ . Since the perturbing magnetic field  $H$  changes at the random points  $t_1, t_2, \dots, t_n, \dots$ , the mean value (the mathematical expectation) of the perturbation factor (6) has to be calculated. This can be done in a very convenient way if the ensemble of nuclei is divided into classes according to the number of “flips” of the magnetic field in the time interval  $(0, t)$ . Since the events {no flip in  $(0, t)$ }, {one flip in  $(0, t)$ }, ... { $n$  flips in  $(0, t)$ } ... are mutually exclusive and their sum equals the certain event, the mean of the perturbation factor is given by the following rule of probability theory:

$$\begin{aligned} \langle G_{k_1 k_2}^{N_1 N_2}(t) \rangle_{t_1, t_2, \dots, t_n, \dots} &\equiv E \{ G_{k_1 k_2}^{N_1 N_2} \}_{t_1, t_2, \dots, t_n, \dots} \\ &= E \{ G_{k_1 k_2}^{N_1 N_2} | n=0 \} \cdot P(n=0) + E \{ G_{k_1 k_2}^{N_1 N_2} | n=1 \}_{t_1, t_2, \dots, t_n} \cdot P(n=1) + \dots + E \{ G_{k_1 k_2}^{N_1 N_2} | n \}_{t_1, t_2, \dots, t_n} \cdot P(n) + \dots \end{aligned} \quad (8)$$

where the expectation assuming  $n$  flips at exactly the points  $t_1, t_2, \dots, t_n$  is defined by

$$E \{ G_{k_1 k_2}^{N_1 N_2} | n \}_{t_1, t_2, \dots, t_n} = \iint \dots \int G_{k_1 k_2}^{N_1 N_2} \cdot W(\omega_0, \Omega_0, 0; \omega_1, \Omega_1, t_1; \dots) d\omega_0 \cdot d\omega_1 \dots d\omega_n \cdot d\Omega_0 \dots d\Omega_n. \quad (9)$$

$P(n)$  is the probability that exactly  $n$  flips occur in  $(0, t)$ , as given by the Poisson distribution Eq. (1).

### 3.2. Class with Static Interaction

The first term in Eq. (8) describes the class of nuclei with a completely static interaction and can easily be evaluated following the well-known procedure [8]. The interaction Hamiltonian is diagonal in a system having the  $z'$  axis parallel to  $\Omega_0$ , the direction of the magnetic field  $H_0$ . The direction  $\Omega_0$  will be specified by the Euler angles  $(\varphi_0, \vartheta_0, 0)$  with respect to the  $z$  axis. If  $|p\rangle$  denotes the eigenstates in the  $z'$  system,  $|m\rangle$  being those in the  $z$  system, we have

$$|m_a\rangle = \sum_p |p\rangle \langle p | m_a \rangle, \quad \langle m_b | = \sum_{p'} \langle m_b | p' \rangle \langle p' |,$$

where the matrix elements are given by the rotation matrix

$$\langle p | m_a \rangle = D_{p m_a}^{(I)}(\varphi_0, \vartheta_0, 0), \quad \langle m_b | p' \rangle = D_{m_b p'}^{(I)}(0, -\vartheta_0, -\varphi_0).$$

The matrix elements in Eq. (6), assuming no flip in the interval  $(0, t)$ , are therefore given by

$$\begin{aligned} \langle m_b | A(t) | m_a \rangle &= \sum_{p, p'} D_{m_b p'}^{(I)}(0, -\vartheta_0, -\varphi_0) \cdot D_{p m_a}^{(I)}(\varphi_0, \vartheta_0, 0) \cdot e^{-ip\omega_0 t} \langle p' | p \rangle \\ &= \sum_p D_{m_b p}^{(I)}(0, -\vartheta_0, -\varphi_0) \cdot D_{p m_a}^{(I)}(\varphi_0, \vartheta_0, 0) \cdot e^{-ip\omega_0 t}, \end{aligned}$$

where the energy eigenvalues  $E_p$  of the interaction Hamiltonian are written in terms of the Larmor frequency  $\omega_0$

$$E_p = -p \cdot \hbar \omega_0 = -p \cdot g_I \mu_N H_0.$$

According to (6) and (9) the first term in the series (8) is now given by

$$\begin{aligned} E \{ G_{k_1 k_2}^{N_1 N_2} | n=0 \} &= \int d\omega_0 W(\omega_0) \int d\Omega_0 W(\Omega_0) \cdot \sum_{m_a, m_b} \sum_{p, p'} (-1)^{2I+m_a+m_b} \\ &\cdot D_{m_b p}^{(I)}(0, -\vartheta_0, -\varphi_0) \cdot D_{p m_a}^{(I)}(\varphi_0, \vartheta_0, 0) \cdot D_{m_b p'}^{(I)*}(0, -\vartheta_0, -\varphi_0) \cdot D_{p' m_a}^{(I)*}(\varphi_0, \vartheta_0, 0) \\ &\cdot [(2k_1+1)(2k_2+1)]^{\frac{1}{2}} \begin{pmatrix} I & I & k_1 \\ m'_a - m_a & N_1 & \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b - m_b & N_2 & \end{pmatrix} \cdot e^{-i(p-p')\omega_0 t}. \end{aligned}$$

Inserting for  $W(\Omega_0)$  the uniform distribution of Eq. (2), integration over the Euler angles simply yields

$$E \{ G_{kk} | n=0 \} = \int d\omega_0 \cdot W(\omega_0) \sum_{p, p'} \begin{pmatrix} I & I & k \\ p' & -p & N \end{pmatrix}^2 e^{-i(p-p')\omega_0 t}, \quad (10)$$

where by virtue of the orthogonality of the  $D$ -functions  $k_1 = k_2 = k$  results and the perturbation factor becomes independent of  $N_1$  and  $N_2$ . To evaluate the last expression further, the probability distribution  $W(\omega_0)$  has to be

specified. We arbitrarily choose a Gaussian distribution,

$$W(\omega_0) = \frac{1}{\sqrt{2\pi\langle\omega^2\rangle}} \exp\left[-\frac{\omega_0^2}{2\langle\omega^2\rangle}\right], \quad (11)$$

(results for other distributions will be given in Section 4 whereas the more general aspects are discussed in Section 5) and obtain

$$E\{G_{kk}|n=0\} \equiv G_{kk}^{(0)}(t) = \frac{1}{2k+1} \sum_{N=-k}^{+k} \exp\left[-\frac{N^2\langle\omega^2\rangle t^2}{2}\right]. \quad (12)$$

### 3.3. Class with one Discontinuity Point

The second term in Eq. (8) describes the class of nuclei which witness at exactly the point  $t_1$  one flip of the perturbing field. The interaction Hamiltonian  $\mathcal{H}$  is diagonal in the system  $z'$  parallel to  $\Omega_0$  within the time interval  $(0, t_1)$ , whereas in the subsequent interval  $(t_1, t)$  a new system with  $z''$  axis parallel to  $\Omega_1$  must be chosen in order to diagonalize  $\mathcal{H}$ . Denoting the Euler angles of the direction  $\Omega_1$  by  $(\varphi_1, \vartheta_1, 0)$  and the eigenstates in the system  $z''$  by  $|q\rangle$ , we have

$$\begin{aligned} A_1(t-t_1)A_0(t_1)|m_a\rangle &= A_1(t-t_1) \sum_p |p\rangle \langle p|m_a\rangle e^{-ip\omega_0 t_1} = \sum_{p,q} A_1(t-t_1)|q\rangle \langle q|p\rangle \langle p|m_a\rangle \cdot e^{-ip\omega_0 t_1} \\ &= \sum_{p,q} |q\rangle \langle q|p\rangle \langle p|m_a\rangle \cdot e^{-iq\omega_1(t-t_1)} \cdot e^{-ip\omega_0 t_1}. \end{aligned}$$

If the matrix element  $\langle q|p\rangle$  which couples the  $z'$  and  $z''$  systems is written in the form

$$\langle q|p\rangle = \sum_m \langle q|m\rangle \langle m|p\rangle = \sum_m D_{qm}^{(l)}(\varphi_1, \vartheta_1, 0) \cdot D_{mp}^{(l)}(0, -\vartheta_0, -\varphi_0),$$

we obtain for the matrix elements in Eq. (6)

$$\begin{aligned} \langle m_b|A_1(t-t_1)A_0(t_1)|m_a\rangle \cdot \langle m'_b|A_1(t-t_1)A_0(t_1)|m'_a\rangle^* &= \sum_{\substack{p,q,m \\ p',q',m'}} D_{m_b q}^{(l)}(0, -\vartheta_1, -\varphi_1) \cdot D_{q m'}^{(l)}(\varphi_1, \vartheta_1, 0) \\ &\cdot D_{m p}^{(l)}(0, -\vartheta_0, -\varphi_0) \cdot D_{p m_a}^{(l)}(\varphi_0, \vartheta_0, 0) \cdot D_{m'_b q'}^{(l)*}(0, -\vartheta_1, -\varphi_1) \cdot D_{q' m'}^{(l)*}(\varphi_1, \vartheta_1, 0) \cdot D_{m'_p}^{(l)*}(0, -\vartheta_0, -\varphi_0) \\ &\cdot D_{p' m'_a}^{(l)*}(\varphi_0, \vartheta_0, 0) \cdot \exp[-i(q-q')\omega_1 \cdot (t-t_1)] \cdot \exp[-i(p-p')\omega_0 \cdot t_1]. \end{aligned}$$

If this expression is used in (6), multiplied according to (9) by

$$W(\omega_0 \Omega_0 0; \omega_1 \Omega_1 t_1) = \frac{1}{(4\pi)^2} \cdot W(\omega_0) \cdot W(\omega_1),$$

and integrated over the Euler angles and the interaction frequencies  $\omega_0$  and  $\omega_1$ , one obtains

$$E\{G_{kk}|n=1\}_{t_1} = \sum_{p,p',q,q'} \begin{pmatrix} I & I & k \\ p'-p & s_1 & \end{pmatrix}^2 \begin{pmatrix} I & I & k \\ q'-q & s_2 & \end{pmatrix}^2 \cdot \exp\left[-\frac{(q-q')^2\langle\omega^2\rangle(t-t_1)^2}{2}\right] \exp\left[-\frac{(p-p')^2\langle\omega^2\rangle t_1^2}{2}\right], \quad (13)$$

where for both  $W(\omega_0)$  and  $W(\omega_1)$  the same Gaussian distribution (11) has been used. As in Section 3.2 we have  $k_1 = k_2 = k$  and the dependence on  $N_1$  and  $N_2$  has disappeared, which is, of course, a consequence of the spatial isotropy of the model. On comparison of Eq. (13) with the corresponding expression (12), it is seen that (13) can be written

$$E\{G_{kk}(t)|n=1\}_{t_1} = E\{G_{kk}(t-t_1)|n=0\} \cdot E\{G_{kk}(t_1)|n=0\}. \quad (14)$$

The last two equations give the expectation of  $G_{kk}(t)$  assuming one flip at exactly the point  $t_1$ . Actually the discontinuity point is equally likely to occur anywhere in the time interval  $(0, t)$ . The average of Eq. (13) over  $t_1$  has, therefore, to be calculated with the uniform probability density  $W(t_1)dt_1 = dt_1/t$  that the discontinuity occurs at  $t_1$  in  $dt_1$ . The final result is

$$E\{G_{kk}(t)|n=1\} \cdot P(n=1) = \lambda \cdot e^{-\lambda t} \int_0^t G_{kk}^{(0)}(t-t_1) \cdot G_{kk}^{(0)}(t_1) dt_1, \quad (15)$$

where the shorthand notation of Eq. (12) has been used

$$G_{kk}^{(0)}(t) \equiv E \{G_{kk}(t) | n=0\} \quad (16)$$

for the attenuation factors of a completely static interaction. It should be noted that the property specified by Eq. (15) and the subsequent expressions (18) and (19) are, of course, quite independent of the probability distribution  $W(\omega)$ . The Gaussian distribution (11) has been used as an illustrative example only.

### 3.4. The Attenuation Coefficients

The procedure discussed in Section 3.3 can be generalized to the case of  $n$  discontinuities at exactly specified points in the time interval  $(0, t)$ . An expression similar to (13) can be derived which comprises now the product of  $n+1$  exponentials. As before, this expression has to be averaged over the random occurrence of the discontinuity points. The probability density of the ordered points  $t_1, t_2, \dots, t_n$ , which are independently and uniformly distributed in  $(0, t)$  is given by [9]

$$W(t_1, t_2, \dots, t_n) = \frac{n!}{t^n}.$$

The final result can be written as the  $n$ -th order convolution

$$E \{G_{kk}(t) | n\} \cdot P(n) = \lambda^n e^{-\lambda t} \int_0^t G_{kk}^{(0)}(t-t_n) dt_n \int_0^{t_n} G_{kk}^{(0)}(t_n-t_{n-1}) dt_{n-1} \dots \int_0^{t_2} G_{kk}^{(0)}(t_1) dt_1, \quad (17)$$

which is an obvious extension of Eq. (15).

From Eq. (8) we now obtain the attenuation coefficient

$$\langle G_{kk}(t) \rangle = e^{-\lambda t} \sum_{n=0}^{\infty} \lambda^n \int_0^t G_{kk}^{(0)}(t-t_n) dt_n \int_0^{t_n} G_{kk}^{(0)}(t_n-t_{n-1}) dt_{n-1} \dots \int_0^{t_2} G_{kk}^{(0)}(t_1) dt_1. \quad (18)$$

The mathematical structure of this expression is identical to the corresponding equation of the Scherer model [5]. To evaluate Eq. (18) we use the technique of the Laplace transform discussed by Blume [5]. The Laplace transform of the  $n$ -th order convolution reduces to the  $n$ -th order product and we obtain from Eq. (18)

$$\langle \tilde{G}_{kk}(p) \rangle = \tilde{G}_{kk}^{(0)}(p+\lambda) + \lambda [\tilde{G}_{kk}^{(0)}(p+\lambda)]^2 + \dots + \lambda^{n-1} [\tilde{G}_{kk}^{(0)}(p+\lambda)]^n + \dots$$

with the notation

$$\tilde{G}(p) \equiv \int_0^{\infty} e^{-pt} G(t) dt.$$

Summation of the geometric series gives

$$\langle \tilde{G}_{kk}(p) \rangle = \frac{\tilde{G}_{kk}^{(0)}(p+\lambda)}{1 - \lambda \tilde{G}_{kk}^{(0)}(p+\lambda)}. \quad (19)$$

This basic equation connects the attenuation factor for the fluctuating interaction with the attenuation factor for the class of nuclei with a static interaction.

If the Laplace transform is inverted, one obtains from Eq. (19) the following inhomogeneous Volterra integral equation of the second kind for the time-differential attenuation coefficients

$$\langle G_{kk}(t) \rangle = G_{kk}^{(0)}(t) \cdot e^{-\lambda t} + \lambda \int_0^t \langle G_{kk}(t') \rangle G_{kk}^{(0)}(t-t') \cdot e^{-\lambda(t-t')} dt'. \quad (20)$$

The time-integral attenuation coefficients, however, are most easily calculated directly from Eq. (19). Due to the close relation between the definition of the Laplace transform and the time-integral attenuation coefficients, one has simply

$$\langle G_{kk}(\infty) \rangle \equiv \frac{1}{\tau} \int_0^{\infty} e^{-t/\tau} \langle G_{kk}(t) \rangle dt = p \cdot \langle \tilde{G}_{kk}(p) \rangle |_{p=1/\tau}. \quad (21)$$

#### 4. Results for the Time-Integral Attenuation Factors

##### 4.1. Gaussian Distribution of the Perturbing Field

In order to obtain the time-integral attenuation coefficients from (21), the Laplace transform of the static interaction term (12) has to be inserted into Eq. (19). The Laplace transform of Eq. (12) is

$$\tilde{G}_{kk}^{(0)}(p) = \frac{1}{2k+1} \left\{ \frac{1}{p} + \sum_{N=1}^k \sqrt{\frac{2\pi}{N^2 \langle \omega^2 \rangle}} \left[ 1 - \Phi \left( \frac{p}{\sqrt{2N^2 \langle \omega^2 \rangle}} \right) \right] \exp \left[ \frac{p^2}{2N^2 \langle \omega^2 \rangle} \right] \right\} \quad (22)$$

and the attenuation coefficients are found from (19) to be

$$\langle G_{kk}(\infty) \rangle_{\text{Gauss}} = \frac{\sqrt{2\pi}}{(2k+1) \langle \omega^2 \rangle^{\frac{1}{2}} \tau} \frac{\frac{z}{\sqrt{2\pi}} + \sum_{N=1}^k \frac{1}{N} \left[ 1 - \Phi \left( \frac{1}{\sqrt{2} N z} \right) \right] \cdot \exp \left[ \frac{1}{2N^2 z^2} \right]}{1 - \frac{\sqrt{2\pi} \cdot \tau}{(2k+1) \cdot \langle \omega^2 \rangle^{\frac{1}{2}} \tau} \left\{ \frac{z}{\sqrt{2\pi}} + \sum_{N=1}^k \frac{1}{N} \left[ 1 - \Phi \left( \frac{1}{\sqrt{2} N z} \right) \right] \cdot \exp \left[ \frac{1}{2N^2 z^2} \right] \right\}}, \quad (23)$$

with

$$z = \frac{\langle \omega^2 \rangle^{\frac{1}{2}} \tau}{1 + \frac{\tau}{\tau_c}}, \quad \langle \omega^2 \rangle = g_I^2 \frac{\mu_N^2}{\hbar^2} \langle H^2 \rangle, \quad (24)$$

where  $\tau_c = 1/\lambda$  is the correlation time,  $\tau$  the lifetime and  $g_I$  the  $g$ -factor of the intermediate nuclear state  $I$  and  $\langle H^2 \rangle$  the second moment of the Gaussian distribution (11) of the size of the perturbing magnetic field. In Eq. (23)  $\Phi(x)$  is the normalized error function

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-u^2} du$$

and the sum extends to all positive integers  $N=1, 2, \dots, k$ . Since the attenuation coefficients are independent of  $N_1$  and  $N_2$  and have the property  $k_1 = k_2 = k$ , the angular correlation function of Eq. (5) reduces, of course, to

$$W(\vartheta, \infty) = \sum_k \langle G_{kk}(\infty) \rangle \cdot A_{kk} \cdot P_k(\cos \vartheta). \quad (25)$$

##### 4.2. Uniform Distribution

Attenuation coefficients for other distributions can be immediately calculated from Eq. (10). For a uniform (rectangular) distribution of the frequency of interaction

$$W(\omega) = \begin{cases} \frac{1}{2\omega_{\max}} = \frac{1}{\sqrt{12} \langle \omega^2 \rangle^{\frac{1}{2}}}, & -\omega_{\max} < \omega < \omega_{\max} \\ 0 & \text{elsewhere} \end{cases}$$

one obtains for the class of nuclei with a static interaction

$$G_{kk}^{(0)}(t) = \frac{1}{2k+1} \left\{ 1 + \sum_{N=1}^k 2 \cdot \frac{\sin(N \omega_{\max} t)}{N \cdot \omega_{\max} t} \right\}.$$

It is very convenient to use the second moment  $\langle \omega^2 \rangle$  instead of the parameter  $\omega_{\max}$  because this quantity facilitates a comparison with other stochastic models. The time-integral attenuation coefficients for the uniform distribution are found to be

$$\langle G_{kk}(\infty) \rangle_{\text{uniform}} = \frac{1}{(2k+1) \sqrt{3} \langle \omega^2 \rangle^{\frac{1}{2}} \tau} \frac{\sqrt{3} z + 2 \sum_{N=1}^k \frac{1}{N} \arctg(\sqrt{3} N z)}{1 - \frac{\tau/\tau_c}{(2k+1) \sqrt{3} \langle \omega^2 \rangle^{\frac{1}{2}} \tau} \left\{ \sqrt{3} z + 2 \sum_{N=1}^k \frac{1}{N} \arctg(\sqrt{3} N z) \right\}} \quad (26)$$

with the parameter  $z$  as defined in Eq. (24).

4.3. Delta Distribution

The limiting case of the  $\delta$  distribution

$$W(\omega) = \delta(\omega - \omega_0)$$

leads to a model, in which the direction of the interacting field is allowed to fluctuate at random, but the size  $H_0$  of the field remains fixed in time. The Laplace transform of the corresponding expression (10) is simply

$$\tilde{G}_{kk}^{(0)}(p) = \frac{1}{2k+1} \sum_{N=-k}^{+k} \frac{1}{p + iN\omega_0},$$

and the time-integral attenuation coefficients are

$$\langle G_{kk}(\infty) \rangle_{\delta} = \frac{\frac{1}{2k+1} \sum_{N=-k}^{+k} \frac{1}{1 + \frac{\tau}{\tau_c} + iN \langle \omega^2 \rangle^{\frac{1}{2}} \tau}}{1 - \frac{\tau/\tau_c}{2k+1} \sum_{N=-k}^{+k} \frac{1}{1 + \frac{\tau}{\tau_c} + iN \langle \omega^2 \rangle^{\frac{1}{2}} \tau}}, \tag{27}$$

where the sums extend to all integers  $-k, \dots, 0, 1, 2, \dots, k$ . For the  $\delta$  distribution we have  $\langle \omega^2 \rangle^{\frac{1}{2}} = \omega_0$ , but the second moment symbol will be retained for purpose of general notation. The attenuation coefficient  $\langle G_{22}(\infty) \rangle_{\delta}$  is shown in Fig. 1 (solid lines). As a function of the relative strength of fluctuation, given by the ratio of nuclear lifetime  $\tau$  and correlation time  $\tau_c$ , the amount of irreversible attenuation of the angular correlation generally decreases from static interaction (on the left-hand side of the figure) to extremely fast fluctuation (on the right-hand side). Typically, there are, however, intermediate regions of particularly strong perturbations.

The minima of the attenuation factors are an interesting feature of the exactly soluble "fluctuating orientation" models (for exceptions, however, see the note at the end of Section 4.4). Stochastic perturbation of the "fixed orientation" type does not show this behaviour. In Fig. 1 the attenuation coefficient of the fixed orientation Blume model [2] is shown for comparison. Since a jump of the magnetic field from  $+H_0$  to  $-H_0$  in the Blume model is equivalent to a jump of the initial direction of the unique field  $H_0$  to the antiparallel one in our model and since, furthermore, Poisson statistics (1) are also inherent to the Blume calculations, the two models only

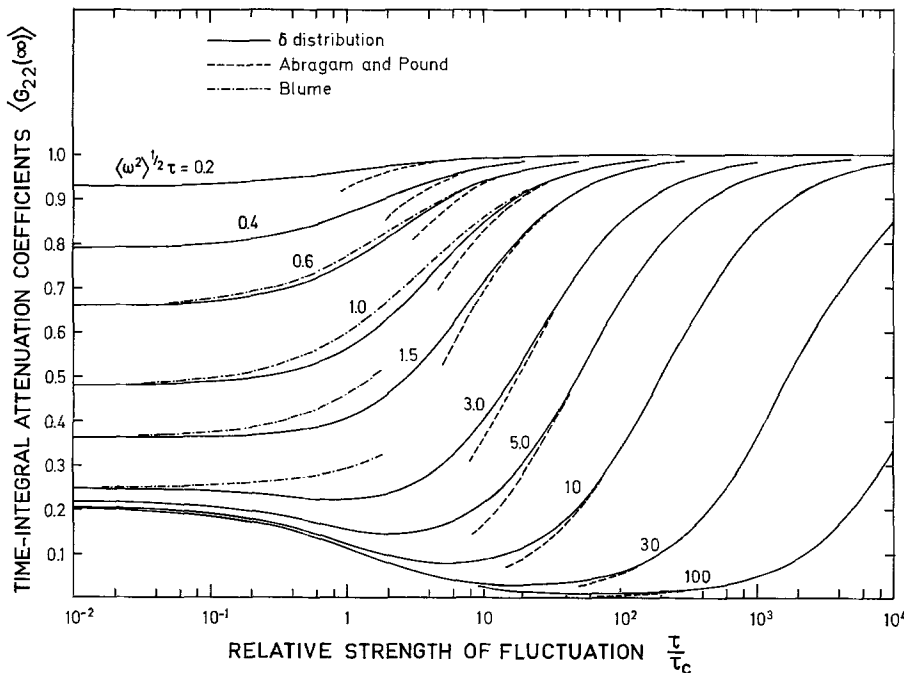


Fig. 1. Time-integral attenuation coefficients  $\langle G_{22}(\infty) \rangle_{\delta}$  from Eq. (27) for continual random reorientation of the axis of the perturbing magnetic field. The size of the field is assumed to be fixed in time ( $\delta$  distribution,  $\langle \omega^2 \rangle^{\frac{1}{2}} = \omega_0$ ). The attenuation coefficients from the "fixed orientation" Blume model [2] and the perturbation theoretic Abragam and Pound model [6] are shown for comparison

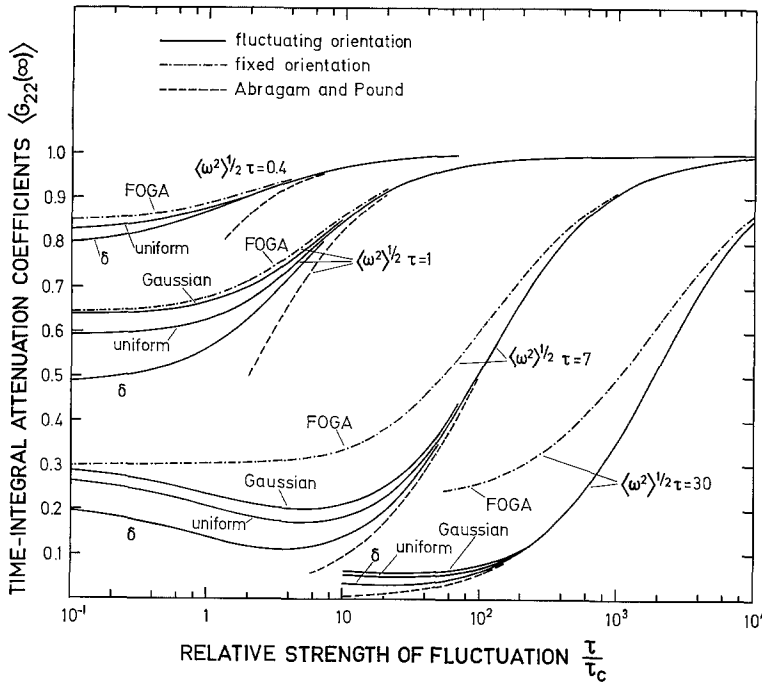


Fig. 2. Comparison of the attenuation coefficients for the three probability distributions discussed in Sections 4.1 to 4.3. The FOGA model [1] is shown as an example of a “fixed orientation” perturbation. In the proper limit the dependence of the attenuation coefficients on the specific distribution  $W(\omega)$  cancels

differ with respect to the assumptions of fixed or fluctuating orientation. As can be seen from the figure, the models exhibit a similar behaviour in the static and the fast fluctuation region. The main deviations occur in the intermediate region by filling up the minima. Similar remarks apply to the FOGA model [1] which should be compared to the fluctuating orientation model with Gaussian distribution, Eq. (23) (see Fig. 2).

#### 4.4. Relation to the Abragam and Pound Model

As can be easily calculated from Eqs. (23), (26) and (27), the time-integral attenuation factors reduce to

$$\langle G_{kk}(\infty) \rangle \rightarrow \frac{1}{1 + \frac{1}{3} k(k+1) \langle \omega^2 \rangle^{\frac{1}{2}} \tau \tau_c} \quad \text{if } \begin{cases} \tau_c \ll \tau \\ \langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1, \end{cases} \quad (28)$$

independent of the three probability distributions  $W(\omega)$ . Clearly, Eq. (28) is the well-known attenuation coefficient for a fluctuating magnetic interaction of the Abragam and Pound model [6]. It is interesting to note that in the perturbation theoretic treatment of Abragam and Pound the actual distribution of the frequency of interaction cancels, i.e. the parameter  $\langle \omega^2 \rangle$  is the second moment of a completely unspecified distribution. The mathematical background of this important fact will be discussed in Section 5.

The property specified by Eq. (28) is most clearly illustrated in Fig. 2 in which examples of the attenuation coefficients for the three distributions discussed in Sections 4.1 to 4.3 are compared with the Abragam and Pound model. All the attenuation factors of the fluctuating orientation model smoothly touch the Abragam and Pound curve with increasing relative strength of fluctuation. This behaviour is much less pronounced for the fixed orientation models, a fact which is also illustrated in the plot of  $\langle G_{22}(\infty) \rangle$  versus  $\langle G_{44}(\infty) \rangle$  in Fig. 3. Whereas all fluctuating orientation curves for  $\tau/\tau_c \gtrsim 10^2$  merge into the Abragam and Pound magnetic dipole limit, the fixed orientation model of Blume [2] yields a curve which is nearly independent of  $\tau/\tau_c$  (the dependence on  $\tau/\tau_c$  cannot be displayed in the figure). The FOGA model produces curves with a marked dependence on  $\tau/\tau_c$ , but they do not merge in the Abragam and Pound limit either, except for very weak perturbations.

It is also interesting to note the dependence of the hard-core values of the attenuation factors (marked by dots in Fig. 3) on the relative strength of fluctuation. Whereas the fixed orientation models invariably produce the well-known hard-cores  $(2k+1)^{-1}$  for isotropic magnetic dipole interaction, the hard-cores of the fluctuating orientation model decrease to zero with increasing  $\tau/\tau_c$ . The hard-core attenuation factors are independent of the distribution  $W(\omega)$  of the interaction frequency.



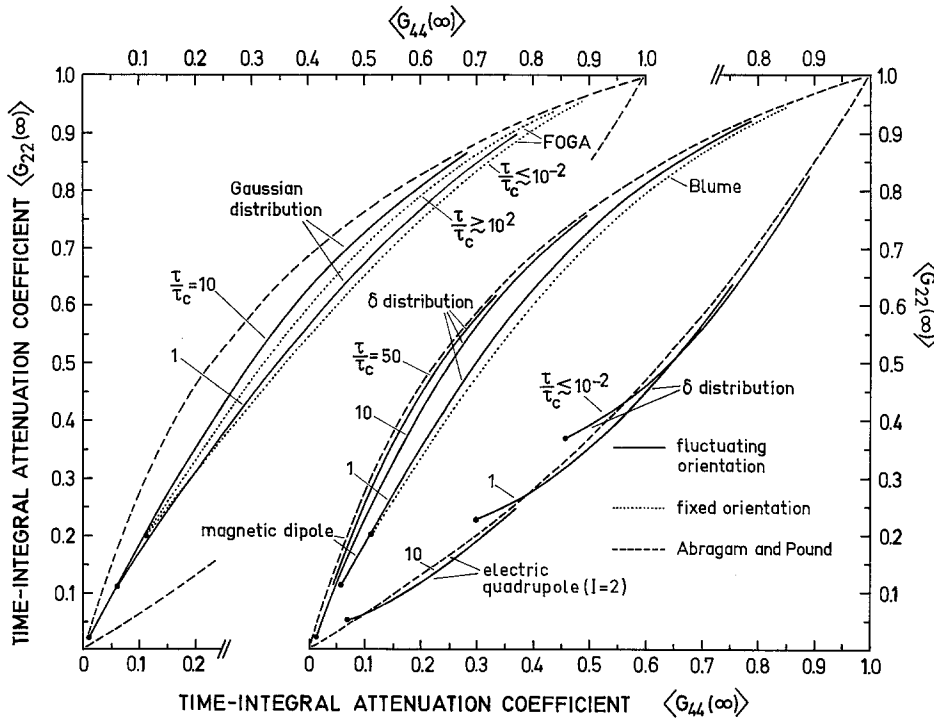


Fig. 3. Plot of  $\langle G_{22}(\infty) \rangle$  versus  $\langle G_{44}(\infty) \rangle$  for "fluctuating orientation" and "fixed orientation" models. The hard-core values for a very strong interaction are marked by dots. The Blume model [2] shows a rather weak dependence on the parameter  $\tau/\tau_c$  which cannot be displayed in the figure. Attenuation coefficients for an electric quadrupole interaction with  $I=2$  are also shown

The fluctuating orientation model is easily extended to the case of an electric quadrupole interaction. For a fluctuating field gradient having axial symmetry and with its magnitude fixed ( $\delta$  distribution), results for  $I=2$  are included in Fig. 3. Details are given in Ref. 10.

We finally note that not all probability distributions  $W(\omega)$  yield attenuation coefficients which merge in the Abragam and Pound coefficients in the proper limit. A distribution  $W(\omega)$  with a Lorentzian shape (Cauchy distribution), for example, with half width  $\omega_{HW}$ , will produce attenuation factors with the property

$$\langle G_{kk}(\infty) \rangle_{\text{Cauchy}} \rightarrow 0 \quad \text{for } \omega_{HW} \tau \gg 1,$$

even for  $\tau_c \rightarrow 0$ , in sharp contrast to the models previously discussed. This is due to the fact that the moments of the Cauchy distribution diverge. More details are given in Ref. 10.

### 5. Results for Time-Differential Attenuation Factors

Time-differential attenuation coefficients are obtained by numerically solving the integral equation (20), using an iteration procedure. In Fig. 4 typical results for three probability distributions of the interacting fields (Gaussian, uniform and  $\delta$  distribution) are compared. The attenuation coefficients of the fixed orientation Blume [2] and FOGA [1] models are also shown. As already pointed out in Section 4, the Blume curves should be compared with the curves for the  $\delta$  distribution whereas the FOGA curves correspond to the coefficients for the Gaussian distribution. It is interesting to note that all attenuation coefficients of the fluctuating orientation model are closely approximated by the Abragam and Pound coefficients in the limit  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1$ , except in the region close to  $t=0$ . The initial slope of the Abragam and Pound coefficients is not reproduced. This is not surprising because, strictly speaking, the Abragam and Pound coefficients should not be used for  $t \rightarrow 0$  due to the condition  $t \gg \tau_c$  inherent to this model.

In fact, a rather general expression for the time-dependent attenuation factors for  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1$ , but *not* involving the additional condition  $t/\tau_c \gg 1$ , can be obtained as follows:

Eq. (10) for the attenuation factors for a static, isotropic interaction can be written in the form

$$G_{kk}^{(0)}(t) = \frac{1}{2k+1} \sum_{N=-k}^{+k} \int_{-\infty}^{+\infty} W(\omega) \cos(N \omega t) \cdot d\omega$$

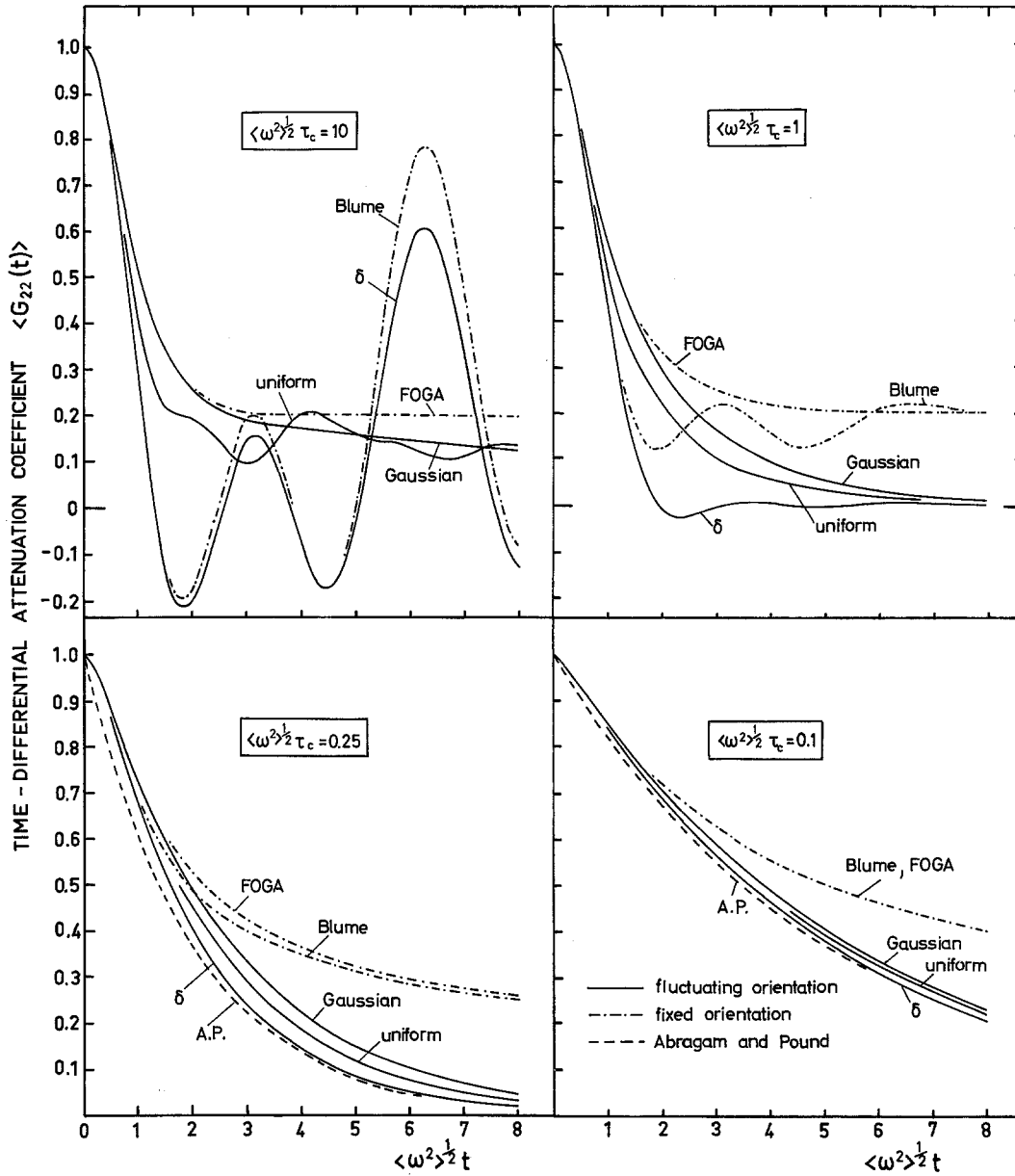


Fig. 4. Time-differential attenuation coefficients  $\langle G_{22}(t) \rangle$  for a Gaussian, a uniform and a  $\delta$  distribution of the interaction frequency. The data are obtained numerically from the integral equation (20). The corresponding "fixed orientation" Blume [2] and FOGA [1] attenuation coefficients as well as the limiting Abragam and Pound coefficients are shown for comparison

where  $W(\omega)$  is assumed to be an arbitrary distribution of the interaction frequency. By expanding the cosin-function in a power series we have

$$G_{kk}^{(0)}(t) = 1 - \frac{1}{2k+1} \left\{ \sum_N \frac{N^2 t^2}{2!} \langle \omega^2 \rangle - \sum_N \frac{N^4 t^4}{4!} \langle \omega^4 \rangle + \dots \right\},$$

where the moments

$$\langle \omega^n \rangle = \int_{-\infty}^{+\infty} \omega^n W(\omega) d\omega$$

have been introduced. According to Eq. (19) the Laplace transform of  $G_{kk}^{(0)}(t)$  is needed for the argument  $p + \lambda$ . We have

$$\tilde{G}_{kk}^{(0)}(p + \lambda) = \frac{1}{\lambda} \left\{ \frac{1}{1 + \frac{p}{\lambda}} - \sum_N \frac{N^2}{2k+1} \cdot \frac{\langle \omega^2 \rangle}{\lambda^2} \cdot \frac{1}{\left(1 + \frac{p}{\lambda}\right)^3} + \sum_N \frac{N^4}{2k+1} \cdot \frac{\langle \omega^4 \rangle}{\lambda^4} \cdot \frac{1}{\left(1 + \frac{p}{\lambda}\right)^5} - \dots \right\}.$$

We conveniently introduce the parameter  $\alpha_n$  according to

$$\langle \omega^{2n} \rangle = \alpha_n \cdot \langle \omega^2 \rangle^n,$$

where  $\alpha_n$  depends, of course, on the distribution  $W(\omega)$ . For the  $\delta$  and the Gaussian distribution, for example, we have  $\alpha_n = 1$  and  $\alpha_n = (2n - 1)!!$ , respectively. Using this parameter, it is seen that in the Laplace space a series expansion in powers of  $\langle \omega^2 \rangle / \lambda^2 = \langle \omega^2 \rangle \tau_c^2$  is generated. If  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1$ , we may neglect all terms comprising the parameters  $\alpha_n$ ,  $n > 1$ , with the result that the attenuation coefficients prove to be independent of the distribution  $W(\omega)$ . If this approximation is used in the nominator and denominator of Eq. (19), it is a simple procedure to invert the Laplace transform by finding the poles of  $\langle \tilde{G}_{kk}(p) \rangle$ . The result for the time-differential coefficients is

$$\langle G_{kk}(t) \rangle = \exp \left[ -\frac{1}{2} \frac{t}{\tau_c} \right] \left\{ \cosh \left( \frac{1}{2} \frac{t}{\tau_c} \sqrt{1 - 8c_k \langle \omega^2 \rangle \tau_c^2} \right) + \frac{1}{\sqrt{1 - 8c_k \langle \omega^2 \rangle \tau_c^2}} \cdot \sinh \left( \frac{1}{2} \frac{t}{\tau_c} \sqrt{1 - 8c_k \langle \omega^2 \rangle \tau_c^2} \right) \right\}, \quad (29a)$$

$$c_k = \sum_{N=1}^k \frac{N^2}{2k+1} = \frac{1}{6} k(k+1); \quad \langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1.$$

The corresponding time-integral coefficients are simply

$$\langle G_{kk}(\infty) \rangle = \frac{1}{1 + \frac{\tau_c}{\tau}} \frac{2c_k \langle \omega^2 \rangle \tau \tau_c}{1 + \frac{\tau_c}{\tau}}; \quad \langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1. \quad (29b)$$

It should be noted that the condition  $t \gg \tau_c$  or  $\tau \gg \tau_c$  is not required in deducing Eqs. (29). Eq. (29b) reduces to Eq. (28) if this condition is introduced. Furthermore, all physically "reasonable" distributions  $W(\omega)$  yield the same attenuation factors due to the fact that the parameters  $\alpha_n$  (which represent the distribution) are not involved in this first order approximation. The only relevant parameter is  $\langle \omega^2 \rangle$ , the second moment of a completely unspecified distribution. Obviously, in a higher order approximation only distributions with the same parameter  $\alpha_2$  will produce identical attenuation coefficients. By extending step by step the scope of the theory with respect to the strength of interaction  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c$ , one inevitably produces attenuation coefficients which depend more and more on the finer details of the distributions. The slight variations for  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c = 0.1$  and  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c = 0.25$  for the three distributions shown in Fig. 4 may be said to be due to the differences in the fourth moments. It is obvious that distributions with diverging moments (such as the Lorentzian distribution mentioned in Section 4.4) have to be excluded from these considerations.

It can easily be seen that Eq. (29a) closely reproduces the Abragam and Pound coefficients except for the initial slope. This behaviour can be understood by noting that within a limited time interval,  $t \ll \tau_c$ , most of the excited nuclei witness a static field. The chaotic disorder, which highly simplifies the treatment, gradually develops only in course of time. Therefore, the initial decrease of anisotropy should be similar to the behaviour encountered in static, isotrop interactions, i.e. vanishing initial slope.

## 6. Conclusion

An exactly soluble stochastic model for the perturbation of angular correlations has been developed in which classical magnetic (or electric) fields are assumed to fluctuate randomly both in direction and size. The stochastic process for the perturbing field is assumed to be homogeneous in time and isotropic in space. As a consequence of the latter assumption the angular correlation function is given by (25). Explicit time-integral attenuation

coefficients are given in Eqs. (23), (26) and (27) for a Gaussian, a uniform and a  $\delta$  distribution for the magnitude of the perturbing magnetic fields. Time-differential attenuation coefficients have been calculated numerically from the integral equation (20). Results are shown in Fig. 4. The approximation (29) is obtained from a series expansion of the Laplace transform (19).

It should perhaps be emphasized that one has to be very careful in applying results of stochastic models to actual physical situations, for three main reasons. First of all, there is a considerable amount of speculation inherent in the specific choice of the distribution of the perturbing fields. A glance at Fig. 4 shows the drastic dependence of the time-differential attenuation coefficients on the distribution  $W(\omega)$ . It is of invaluable importance that in the limit  $\langle \omega^2 \rangle^{\frac{1}{2}} \tau_c \ll 1$  this dependence disappears (for physically "reasonable" distributions). This fact has been illustrated in Section 4.4 and discussed in connection with Eq. (29). Secondly, the classification of the interaction according to "fixed orientation" and completely random "fluctuating orientation" is, of course, a limiting concept. The results of the present paper may facilitate, however, the understanding of intermediate situations. And, finally, all the stochastic models published so far assume statistically stationary interactions. For free ions recoiling into gas or vacuum [11], for example, this assumption is expected to hold approximately if a sufficiently short time interval for the observation of the perturbation process can be selected experimentally. This last restriction is irrelevant for liquid or gaseous sources in thermal equilibrium. For such cases the fluctuating orientation model appears to be an extension of the Abragam and Pound model to interactions of arbitrary strength and arbitrary correlation time.

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