Perturbation of Angular Correlations by Randomly Oriented, Fluctuating Magnetic Fields*

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The perturbation factors in the angular-correlation function are obtained for multidomain ferromagnetic metals, taking into account the fluctuation of the local magnetic fields. The perturbation can be described by means of an equation for products of transition amplitudes, similar to the master equation, whose solution includes, as particular cases, the results for paramagnetic relaxation and for nonfluctuating fields.

I. INTRODUCTION

THE perturbation of angular correlations of γ-γ cascades from decaying nuclei by the internal fields in crystals furnishes information about the local magnetic and electric fields at the sites of the decaying nuclei.[1] This information is particularly valuable in the case of magnetic metals, where different models have been proposed to explain the hyperfine fields at impurities.[2,3] Measurements can be made without external magnetic fields, and extend throughout the whole sample.[4] In a magnetic crystal, the domains will be randomly oriented, and, within them, the magnetic fields will fluctuate in time about their average value. The angular-correlation probability at time t for each domain will be

$$W(\mathbf{k}_1,\mathbf{k}_2,t)=\sum_{k_1N_1}\sum_{k_2N_2}A_{k_1}(1)A_{k_2}(2)G_{k_1k_2}{}^{N_1N_2}(t)\times[(2k_1+1)(2k_2+1)]^{-1/2}Y_{k_1}{}^{N_1}(\theta_1\varphi_1)^*Y_{k_2}{}^{N_2}(\theta_2\varphi_2),\quad(1)$$

where $\mathbf{k}_i$ indicates the (θ_i,φ_i) direction of the ith radiation, the coefficients $A_{k_i}(i)$ depend on the interaction between the nuclei and the radiation, and the interaction between the crystal and the decaying nuclei is included in the perturbation factors

$$G_{k_1k_2}{}^{N_1N_2}(t)=\sum_{m_am_b}(-1)^{2I+m_a+m_b}[(2k_1+1)(2k_2+1)]^{1/2}\times\begin{pmatrix}I & I & k_1\\ n_a & -m_a & N_1\end{pmatrix}\begin{pmatrix}I & I & k_2\\ n_b & -m_b & N_2\end{pmatrix}\times\langle m_b|\Lambda(t)|m_a\rangle\langle n_b|\Lambda(t)|n_a\rangle^*,\quad(2)$$

and $\Lambda(t)$ is the time-evolution operator for the decaying nuclei in their intermediate state, perturbed by the hyperfine magnetic fields.

A special case of these equations has been used, choosing a reference frame in one domain and averaging over directions, for a situation with no fluctuations.[5] Above the Curie point, the average field in each domain disappears, and the influence of the electronic paramagnetic relaxation is described by choosing the Oz axis of the reference frame along the wave vector of the first radiation.[6] Studies have also been made on magnetic hyperfine interactions and their relaxation in europium-iron garnets.[7] Preliminary experiments have recently been performed to investigate the effects of both the average and the fluctuating field in metals, above and below the Curie temperature.[8] It is the purpose of this investigation to obtain the perturbation factors in the general case and discuss the fulfillment of the emerging restrictions.

II. THE PERTURBATION FACTORS

We shall describe the interaction between an intermediate-state nucleus and the hyperfine magnetic field by means of the time-dependent Hamiltonian

$$\mathcal{H}(t)=\mathcal{H}_0+\mathcal{V}(t),\quad(3)$$

$$\mathcal{H}_0=-g\mu_N\mathbf{I}\cdot\mathbf{H},\quad \mathcal{V}(t)=-g\mu_N\mathbf{I}\cdot\delta\mathbf{H}(t),\quad(4)$$

where μ_N is the nuclear magneton, $\mathbf{I}$ the nuclear spin, $\mathbf{H}$ the average magnetic field at the nucleus, with a fixed direction in each domain, and $\delta\mathbf{H}(t)$ the fluctuating part, with an isotropic distribution in time. Both $\mathbf{H}$ and $\delta\mathbf{H}(t)$ are temperature-dependent. The time-evolution operator can be written in the interaction picture as

$$\Lambda(t)=\Lambda_0(t)\Lambda_1(t),\quad(5)$$

$$\Lambda_0(t)=\exp(-i\mathcal{H}_0t/\hbar),\quad(6)$$

$$\Lambda_1(t)=1-i\hbar^{-1}\int_0^t\Lambda_0{}^\dagger(t)\mathcal{V}(t)\Lambda_0(t)\Lambda_1(t).\quad(7)$$

Introducing a new reference frame with axis Oz' along $\mathbf{H}$ and Euler angles $(\alpha,\beta,0)$, and indicating with primes

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[1] For a survey, see H. Frauenfelder and R. M. Steffen, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), Vol. 2, p. 997.
[2] R. E. Watson and A. J. Freeman, Phys. Rev. **123**, 2027 (1961).
[3] D. A. Shirley and G. A. Westenbarger, Phys. Rev. **138**, A170 (1965).
[4] This seems an advantage over NMR experiments, where measurements correspond only to the nuclei in the domain walls.
[5] E. Matthias, S. S. Rosenblum, and D. A. Shirley, Phys. Rev. Letters **14**, 46 (1965).
[6] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).
[7] M. E. Caspari, S. Frankel, and G. T. Wood, Phys. Rev. **127**, 1519 (1962).
[8] T. Lindqvist (private communication).

the magnetic quantum numbers referred to it, we have

$$\mathcal{H}_0|m'\rangle=\omega_{PL}m'|m'\rangle, \quad (8)$$

where $\hbar\omega_L=-g\mu_N|\mathbf{H}|$. It is possible to express the perturbation factors in a basis defined in the new reference frame by using rotational matrices.[9] The average with respect to all possible Euler angles can then be calculated by taking into account the equation for reducing the products of rotational matrices, and their orthogonality properties. This average is

$$\bar{G}_{k_1k_2}{}^{N_1N_2}(t)=\delta_{k_1k}\delta_{k_2k}\delta_{N_1N_2}(-1)^{2I}\sum_{m'n'N'}(-1)^{m'+n'}e^{iN'\omega_Lt}$$
$$\times\begin{pmatrix} I & I & k \\ p' & -m' & N'\end{pmatrix}\begin{pmatrix} I & I & k \\ q' & -n' & N'\end{pmatrix}$$
$$\times\langle n'|\Lambda_1(t)|m'\rangle\langle q'|\Lambda_1(t)|p'\rangle^*. \quad (9)$$

In the absence of fluctuations, $\Lambda_1(t)=1$, and Eq. (9) reduces to the perturbation factors for randomly oriented (time-independent) fields.[5] To obtain the time behavior of the perturbation factors in Eq. (9), it is necessary to find the nondiagonal products $\langle n'|\Lambda_1(t)|m'\rangle \times\langle n'-N'|\Lambda_1(t)|m'-N'\rangle^*$ averaged with respect to all possible values of $\delta\mathbf{H}(t)$ at time t. Perturbation theory leads to a time-proportional dependence, which is not adequate to describe the correlation after long times. We must find the time dependence from an equation similar to the master equation for transition probabilities.[10] Dropping the primes, and denoting by $\Lambda_{nm}(t,t')$ the matrix elements of the $\Lambda_1(t,t')$ operator, and by $V_{nm}(t)$ those of the time-dependent perturbation, we can write

$$\Lambda_{nm}(t+\Delta t)\Lambda_{n-N\ m-N}{}^*(t+\Delta t)=\sum_{jk}\Lambda_{nj}(t+\Delta t,\,t)$$
$$\times\Lambda_{n-N\ k}{}^*(t+\Delta t,\,t)\Lambda_{jm}(t)\Lambda_{k\ m-N}{}^*(t). \quad (10)$$

Let us consider the expansion of $\Lambda_{nj}\Lambda_{n-N\ k}{}^*$ including terms of second order in $\mathcal{V}$. The integrals will multiply exponentials of the type $\exp[i\omega_L(n-j)t]$. Using the random-phase approximation, we shall replace these exponentials by δ_{nj}. The result is

$$\Lambda_{nj}(t+\Delta t,\,t)\Lambda_{n-N\ k}{}^*(t+\Delta t,\,t)$$
$$=\delta_{nj}\delta_{n-N\ k}[1+(i/\hbar)(J_{nn}+J_{n-N\ n-N}{}^*)+(i/\hbar)^2$$
$$\times(\sum_l K_{nl}+\sum_p K_{p\ n-N})]+\delta_{j-N\ k}\hbar^{-2}L_{njN}, \quad (11)$$

where

$$J_{nn}=\int_t^{t+\Delta t}V_{nn}(\tau)d\tau, \quad (12a)$$

$$K_{nl}=\int_t^{t+\Delta t}d\tau\int_t^{\tau}d\tau'\,e^{i\omega_L(n-l)(\tau-\tau')}V_{nl}(\tau)\times V_{nl}{}^*(\tau'), \quad (12b)$$

$$L_{njN}=\int_t^{t+\Delta t}d\tau\int_t^{t+\Delta t}d\tau'\,e^{i\omega_L(n-j)(\tau-\tau')}V_{nj}(\tau)\times V_{n-N\ j-N}{}^*(\tau'). \quad (12c)$$

To obtain the ensemble average over the random fluctuations $\delta\mathbf{H}(t)$, we introduce their magnitude $\delta H(t)$ and orientation angles $[\psi(t),\chi(t)]$. The perturbation can be written as

$$\mathcal{V}(t)=g\mu_N\delta H(t)\sum_q(-1)^qC_q{}^{(1)}(\psi,\chi)I_{-q}{}^{(1)}. \quad (13)$$

The average of the single integrals gives zero, while the averaged double integrals can be found, for a crystal in a stationary state, using the values

$$\langle C_q{}^{(1)}(\psi,\chi)C_{q'}{}^{(1)*}(\psi,\chi)\rangle=\tfrac{1}{3}\delta_{qq'}, \quad (14a)$$

$$\langle\delta H(t)\delta H(t')\rangle=\langle|\delta H(0)|^2\rangle\exp(-|t-t'|/\tau_c), \quad (14b)$$

where τ_c is the correlation time of the fluctuation correlation for $\delta H(t)$.[11] They lead to

$$\langle V_{nj}(t)V_{n-N\ j-N}{}^*(t')\rangle$$
$$=G_{njN}(0)\exp(-|t-t'|/\tau_c), \quad (15a)$$

$$G_{njN}(0)=\tfrac{1}{3}(\hbar\omega_c)^2(-1)^N$$
$$\times\begin{pmatrix} I & 1 & I \\ -n & -q & j\end{pmatrix}\begin{pmatrix} I & 1 & I \\ -n+N & -q & j-N\end{pmatrix}, \quad (15b)$$

with $\hbar\omega_c=-g\mu_N\langle|\delta H(0)|^2\rangle^{1/2}$. The double integrals are calculated by a standard procedure.[6] Changing to the variables $z=\tau-t$, $z'=\tau'-t$, and then to z, $z''=z-z'$, we obtain, for $\Delta t\gg\tau_c$,

$$\langle L_{njN}\rangle=\Delta t2\tau_c\frac{G_{njN}(0)}{1+[\omega_L(n-j)\tau_c]^2}, \quad (16a)$$

$$\langle K_{nl}\rangle=\Delta t\tau_c\frac{G_{nl0}(0)}{1-i\omega_L(n-l)\tau_c}. \quad (16b)$$

Equation (10) averaged over the fluctuations can now be obtained from (11), (15), and (16). We shall restrict the following discussion to the case $|\omega_L\tau_c|\ll 1$. Provided the quantity

$$\mathfrak{U}_{nmN}(t)=\langle\Lambda_{nm}(t)\Lambda_{n-N\ m-N}{}^*(t)\rangle \quad (17)$$

varies slowly over times $\Delta t\gg\tau_c$, we can let Δt go to

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[9] A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, New Jersey, 1957), Chap. 4.

[10] W. Pauli, in *Probleme der Modernen Physik*, edited by P. Debye, (Verlag S. Hirzel; Leipzig, 1928), p. 30.

[11] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley Publishing Company, Reading, Massachusetts, 1958), p. 374.

zero in the previous discussion, and get

$$\frac{1}{a}\frac{d\mathcal{U}_{nmN}}{dt}=-\mathcal{U}_{nmN}(t)+\sum_j(-1)^N(2I+1)\begin{pmatrix} I & 1 & I\\ -n & -q & j\end{pmatrix}\begin{pmatrix} I & 1 & I\\ -n+N & -q & j-N\end{pmatrix}\times\mathcal{U}_{jmN}(t)\,, \tag{18}$$

where $a=\frac{2}{3}\omega_c{}^2\tau_c I(I+1)$. For $N=0$, Eq. (18) reduces to the master equation for the transition probabilities, used to describe paramagnetic relaxation.[6] In the general case, the equation can be solved by using methods similar to those for $N=0$. Substituting in (18)

$$\mathcal{U}_{nmN}(t)=\mathcal{U}_{nmN}(0)\exp(-\lambda t)\,, \tag{19}$$

an eigenvalue equation is obtained, whose solutions are found by comparing it with a summation expression for 6-j symbols. The eigenvectors are given by

$$\mathcal{U}_{nmN}{}^{(r)}=(-1)^{r+n+m+N}(2r+1)^{1/2}\times\begin{pmatrix} I & I & r\\ n-N & -n & N\end{pmatrix}, \tag{20}$$

normalized so that $\sum_n \mathcal{U}_{nmN}{}^{(r)}\mathcal{U}_{nmN}{}^{(r')}=\delta_{rr'}$, and the eigenvalues are

$$\lambda_r=a\left[1+(-1)^{2I+r}(2I+1)\begin{Bmatrix} I & I & r\\ I & I & 1\end{Bmatrix}\right]=\tfrac{1}{3}\omega_c{}^2\tau_c r(r+1)\,. \tag{21}$$

Using them, we can write

$$\mathcal{U}_{nmN}(t)=\sum_r \alpha_{mN}{}^{(r)}\mathcal{U}_{nmN}{}^{(r)}e^{-\lambda_r t}\,, \tag{22}$$

and find $\alpha_{mN}{}^{(r)}$ from the value of $\mathcal{U}_{nmN}(0)$, which is given by (17) for $\tau_c\ll t\ll a^{-1}$. For these times, the contribution from nondiagonal transition amplitudes will be negligible, so that

$$\mathcal{U}_{nmN}(0)=\delta_{nm}(2k+1)^{-1}. \tag{23}$$

The proportionality constant cannot depend on m or N if the special cases mentioned before are to be included. The result is

$$\mathcal{U}_{nmN}(t)=(2k+1)^{-1}(-1)^{2I}\sum_r(-1)^{m+n}(2r+1)\times\begin{pmatrix} I & I & r\\ m-N & -m & N\end{pmatrix}\begin{pmatrix} I & I & r\\ n-N & -n & N\end{pmatrix}e^{-\lambda_r t} \tag{24}$$

which, substituted in (9), gives for the perturbation factors

$$\langle \bar{G}_{k_1k_2}{}^{N_1N_2}\rangle=\delta_{k_1k}\delta_{k_2k}\delta_{N_1N_2}G_k(t)\,,\qquad G_k(t)=(2k+1)^{-1}e^{-\lambda_k t}\sum_{N=-k}^{k}\cos N\omega_L t\,, \tag{25}$$

where the functions $G_k(t)$ are called attenuation factors. The final result includes a single exponential. It should be noticed that, unlike the case for quadrupole perturbations, both the relaxation constant λ_k and the fundamental frequency ω_L are independent of I. The integral attenuation coefficients are given by

$$G_k=\tau_N{}^{-1}\int_0^\infty G_k(t)e^{-t/\tau_N}dt=(2k+1)^{-1}\sum_N \gamma_k/[1+(N\omega_L\tau_N\gamma_k)^2]\,, \tag{26}$$

with $\gamma_k=(1+\lambda_k\tau_N)^{-1}$. Even if $\omega_L\tau_N\gg1$, the integral attenuation would show a "hard-core" value for $N=0$, depending on the temperature through λ_k.

III. DISCUSSION

An appreciable number of angular-correlation experiments in metals have been consistently analyzed assuming that the decaying impurities replace host atoms in an otherwise undistorted lattice, and that their positions are not affected by nuclear recoil following the first γ emission.[3,6] The recovery of the electronic cloud in metals following the nuclear change is expected to be very short compared with both τ_C and τ_N.[12] Consequently, the angular correlation will not be perturbed by quadrupole interactions in cubic-lattice host metals, nor by the rearrangement of the electronic cloud. The perturbation due to hyperfine magnetic fields on diamagnetic impurities is obtained from the Fermi contact term in the Hamiltonian. This perturbation, whether produced by core- or conduction-polarized electrons, is different from zero only for s orbitals, so that the hyperfine-interaction tensor reduces to a scalar.[13] The Hamiltonian (3) is valid for these cases. The mechanism responsible for the fluctuations is not known, but investigations on related systems suggest that the exchange interaction between magnetic electrons and conduction electrons gives an important contribution.[14]

The previous investigation applies, in particular, to the angular correlation of Cd^{111} in Ni at room temperatures. In this case, $\tau_N=(1.22\pm0.01)\times10^{-7}$ sec,[15] $g=-0.318\pm0.007$,[16] $|H|=65.3\pm1.6$ kG,[5] and we can take $|\delta H(0)|/|H|<1$. Supposing that the hyperfine magnetization is proportional to the magnetization per unit volume in Ni, the value of τ_c can be found from ferromagnetic resonance experiments to be $\tau_c\approx10^{-9}$ sec.[17] Using these values, it is seen that the restrictions

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[12] H. Frauenfelder, Phys. Rev. **82**, 549 (1951).

[13] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961), p. 199.

[14] A. H. Mitchell, Phys. Rev. **105**, 1439 (1957); K. Yosida, *ibid.* **106**, 893 (1957).

[15] P. S. Simms and R. M. Steffen, Phys. Rev. **108**, 1459 (1957).

[16] E. Matthias, L. Boström, A. Maciel, M. Salomon, and T. Lindqvist, Nucl. Phys. **40**, 656 (1963).

[17] N. Bloembergen, Phys. Rev. **78**, 572 (1950).

$\tau_c \ll \tau_N$, $a\tau_c \ll 1$, and $|\omega_L \tau_c| \ll 1$, under which Eq. (25) is valid, are all verified. This example shows that the perturbation under discussion is present in some physical systems, even though the attenuation could be undetectable if, for example, $\lambda_k \tau_N$ were too small to produce a measurable effect.

It should also be noticed that exponentially decreasing attenuation coefficients could also be obtained if there were no fluctuations, but a certain distribution of magnetic fields $w(H)$ (e.g., Gauss or Lorentz distributions) at the impurities sites. They could be calculated from Eq. (25) by putting $\lambda_k = 0$ and averaging over $w(H)$. Nevertheless, the new result, unlike Eq. (25), should have a nonzero temperature-independent value for $t \to \infty$ arising from the term with $N=0$.

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Interaction Range, The Goldstone Theorem, and Long-Range Order in the Heisenberg Ferromagnet

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We derive a sufficient condition on the exchange-interaction range in the Heisenberg ferromagnet for the spin-wave excitations to have no energy gap. We show that there exists a class of exchange interactions which are consistent with long-range order and no energy gap in the spin-wave spectrum in one and two dimensions.

In a previous paper,[1] we showed that if the exchange interaction in the Heisenberg ferromagnet is of finite range, then the Goldstone theorem can be used to prove that the spin-wave excitation energy goes to zero in the zero-k or long-wavelength limit. The proof involved an analysis of the moments of the function $\lambda(\omega)$ defined by

$$\lambda(\omega) = \lim_{k\to 0} \int d(t-t') e^{i\omega(t-t')} \times \sum_j e^{-i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)} \langle [S_i^x(t), S_j^y(t')] \rangle, \quad (1)$$

where the angular brackets denote the thermodynamic average, and the subscripts i and j refer to the sites in the Heisenberg model.

In this paper, we will use the more convenient function[2] $\Lambda(\omega)$ defined by

$$\Lambda(\omega) = \lim_{k\to 0} \int d(t-t') e^{i\omega(t-t')} \times \sum_j e^{-i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)} \langle [S_i^+(t), S_j^-(t)] \rangle. \quad (2)$$

Since $\Lambda(\omega)$ is real, and

$$\Lambda(\omega)\ \mathrm{sgn}\omega \geqslant 0 \quad (3)$$

for all ω, it is sufficient, for our present purposes, to analyze the zeroth and first moments of $\Lambda(\omega)$, M_0, and M_1 where

$$M_0 = \int \frac{d\omega}{2\pi} \Lambda(\omega), \quad (4)$$

$$M_1 = \int \frac{d\omega}{2\pi} \omega \Lambda(\omega). \quad (5)$$

If M_0 is nonzero and M_1 is zero, it is a simple matter to show that there must be an excited state with zero energy in the $k \to 0$ limit which appears as an intermediate state in the commutator found in Eq. (2).

We establish the zero-k limit as was done in Ref. 1. That is,

$$M_0 = \lim_{V\to\infty} \sum_{i\in V} \langle [S_i^+(t), S_j^-(t)] \rangle \quad (6)$$

and

$$M_1 = \lim_{V\to\infty} \sum_{i\in V} \left\langle \left[\left(i\frac{\partial}{\partial t} \right) S_i^+(t), S_j^-(t) \right] \right\rangle. \quad (7)$$

Here, we sum i over all sites in a volume V centered at site j and then let this volume become infinitely large.

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[1] R. V. Lange, Phys. Rev. **146**, 301 (1966).

[2] A related analysis of the Goldstone theorem is found in H. Wagner, Z. Physik **195**, 273 (1966).