calculated and the observed frequencies as a function of the external magnetic field. The value of the hyperfine field at different temperatures was obtained almost independently of the other constants. It was found that there is a slight deviation between the ratios of the hyperfine fields and the magnetization saturations which can be interpreted as a small temperature dependence of the canting angle.

The large field dependence of the linewidth at low magnetic field was found to arise from a large spread in the anisotropy field in the (111) plane.

Comparison between the integrated intensities in low and high fields has shown that the enhancement at low fields is larger than that calculated for the bulk. This increase of the enhancement may arise from the domain wall and further investigation is necessary to obtain conclusive results.

Saturation of the nuclear system in this canted spin-system has a different effect on the NMR signal from that in normal systems. This arises from the strong dependence of the position of the line on the nuclear temperature. As a result, for high enough rf power, the shape and position of the line may depend on the sense in which the frequency is swept. An additional line was observed at a frequency higher than that of the pulled line. Its origin is not clear and a few possibilities are suggested.

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Nuclear Spin-Lattice Relaxation in Ferromagnetic Insulators at Low Temperatures*

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A theoretical study of the nuclear spin-lattice relaxation in cubic ferromagnetic insulators at ultralow temperatures is presented. Calculations are performed for nuclei which belong to the magnetic atoms, considering only the direct processes. Three mechanisms are considered: the relaxation to mixed magnon-phonon modes, indirect nuclear-spin interaction modulated by lattice vibrations, and nuclear quadrupole energy modulated by lattice vibrations. The first two mechanisms lead to a relaxation time T_1 which depends on both the temperature T and the external field H_0, with $T_1 \sim H_0^2/T$ for $H_0 \gg$ magnetization and the anisotropy field. For a field comparable to the magnetization or the anisotropy field, the relaxation time is proportional to H_0^n, n being larger or smaller than 2 depending on the shape of the single-crystal sample. The last mechanism does not lead to a field-dependent T_1. Comparison with experiments performed on powdered EuS is also presented.

I. INTRODUCTION

THE temperature and external field dependence of the spin-lattice relaxation for nuclei which belong to magnetic atoms in a ferromagnetic insulator at very low temperatures and in fairly large external fields is of interest because of a recent experimental investigation on powdered EuS.[1] A convenient expression for the experimental temperature and field dependence is $T_1 \sim H_0^3/T^2$. Measurements were made in the ranges $0.05 \sim 0.15$°K for the temperature and $2 \sim 17$ kOe for the field.

We calculate here general expressions for T_1 for nuclei which belong to magnetic atoms with orbital S states and surroundings of cubic symmetry. The nuclear Hamiltonian H^N is written as

$$H^N = H_Z{}^N + H_{\mathrm{hf}} + H_{\mathrm{ind}} + H_Q\,, \tag{1.1}$$

where the first term is the Zeeman energy, the second the hyperfine interaction with atomic spins (assumed isotropic), the third the indirect nuclear spin interaction via the hyperfine interaction,[2] and the last the energy due to the presence of the nuclear electric quadrupole moment. The unperturbed nuclear-spin Hamiltonian is given by the sum of $H_Z{}^N$ and the static part of H_{hf}. The mechanisms of the nuclear spin-lattice relaxation come from the modulation of the remaining parts of H^N by the exchange motions and the lattice vibrations. Since we assume the temperature to be ultralow, direct processes should be dominant and so energy conserva-

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[1] R. I. Schermer and L. Passell, Bull. Am. Phys. Soc. **10**, 75 (1965).

[2] H. Suhl, Phys. Rev. **109**, 606 (1958); T. Nakamura, Progr. Theoret. Phys. **20**, 542 (1958).

tion between the unperturbed nuclear and the heat-bath systems should be required.

The mechanism due to modulation of H_{hf} by the exchange motions can be omitted because the energy conservation between the nuclear and magnon systems cannot be satisfied for a fairly large external field. The modulation of H_{hf} by the lattice vibrations gives two effects: one is the modulation of the hyperfine coupling constant and the other is the formation of mixed magnon-phonon modes[3] via the magnon-phonon interaction. The former mechanism leads to a Raman process and can be omitted for ultralow temperatures. The latter gives the relaxation mechanism of the nuclear-spin system to the mixed magnon-phonon modes and energy conservation is satisfied between the low-lying mixed mode and the nuclear system. Regarding the magnon-phonon interaction as a perturbation, the low-lying state of the mixed mode has a magnon component, the mixing amplitude being proportional to the inverse of the magnon energy (which is proportional to the external field for large field strength). In this case it is anticipated that the relaxation time will be proportional to the square of the external field. For the case of the external field comparable to the magnetization or the anisotropy field of the electronic system, the field dependence of the relaxation time will be complicated.

The indirect coupling between nuclear spins H_{ind} is obtained from the second order perturbation of H_{hf} with respect to the magnon system. Taking into account the dipolar interaction between electronic spins, H_{ind} includes terms like $I_j^{\pm}I_{j'}^{\pm}$, the coefficients of which depend on the hyperfine coupling constant and the magnon energy. The modulation of the hyperfine coupling constant by lattice vibrations gives a mechanism of relaxation and energy conservation is satisfied between the nuclear spin and the phonon systems. The relaxation time will depend on the external field in a way similar to the previous case.

The final mechanism comes from the modulation of H_Q by lattice vibrations. Expanding H_Q in powers of the displacements of nuclei and retaining only the linear terms, we get the relaxation time for direct process. Since the strength of the external field is very small compared to the hyperfine field acting on nuclei, the relaxation time obtained by this mechanism will not depend on field strength.

In Sec. 2, expressions for the mixed magnon-phonon modes for cubic crystals are obtained and, using them, we calculate the relaxation time given by Eq. (2.32) for the case that the anisotropy field is very small compared to the external field and the magnetization. For the case of the small magnetization compared to the external and the anisotropy fields, the expression of the relaxation time is given by Eq. (2.44). For both cases, the temperature dependence is given by the function $F_1(T)$ which is nearly proportional to temperature. In Sec. 3, the indirect nuclear spin interaction is derived by taking into account the dipolar interaction between electronic spins and the expression of the relaxation time, Eq. (3.6), is obtained. The order of magnitude of the relaxation time obtained here is longer than the one obtained in Sec. 2 for a rare-earth ion. In Sec. 4, we get the quadrupolar nuclear spin-lattice relaxation time given by Eq. (4.21). $F_4(T)$ is also proportional to temperature. The order of magnitude of the relaxation time can be comparable to the one obtained in Sec. 2. Finally in Sec. 5, the expressions of the relaxation times obtained in the previous sections are extended to the case that there are two isotopes and the comparison with the experiment on powdered EuS is presented. Taking the values of the magnon-phonon coupling constant G_1 and the ratio of the effective charge of the ion to the ionic charge γ as 10 cm^{-1} and 10^3, respectively, the order of magnitude of the relaxation times by the mechanisms stated in Secs. 2 and 4 is comparable to the experimental one, i.e., $T_1\sim$several hours at 0.05°K and 17 kOe. Before going to these problems, we discuss the electronic spin and the phonon systems.

The Hamiltonian of the electron-spin system for a ferromagnetic insulator is written as

$$H^e=H_{Z}{}^e+H_{\mathrm{ex}}+H_{\mathrm{d,p}}+H_{\mathrm{an}}, \tag{1.2}$$

where the first term is the Zeeman energy, the second the exchange interaction, the third the dipolar interaction between atomic spins and the last the anisotropy energy. They are given by

$$H_Z{}^e=g\mu_B H_0 \textstyle\sum_j S_j{}^{\zeta'}, \tag{1.2a}$$

$$H_{\mathrm{ex}}=2J\textstyle\sum_{\langle jj'\rangle}\mathbf{S}_j\cdot\mathbf{S}_{j'}, \tag{1.2b}$$

$$H_{\mathrm{dip}}=\tfrac{1}{2}\textstyle\sum_j\sum_{j'}D_{jj'}[\mathbf{S}_j\cdot\mathbf{S}_{j'}-3(\mathbf{S}_j\cdot\hat{r}_{jj'})(\mathbf{S}\cdot\hat{r}_{jj'})], \tag{1.2c}$$

$$H_{\mathrm{an}}=(\kappa/S^2)\textstyle\sum_j[S_j{}^{x^4}+S_j{}^{y^4}+S_j{}^{z^4}], \tag{1.2d}$$

$$D_{jj'}=(g\mu_B)^2/r_{jj'}{}^3,\quad \hat{r}_{jj'}=\mathbf{r}_{jj'}/r_{jj'}, \tag{1.2e}$$

where $\sum_{\langle jj'\rangle}$ denotes a summation over nearest-neighbor pairs. The external field H_0 is applied along the ζ' axis perpendicular to which the ξ' and η' axes are defined. The crystallographic axes are designated by x, y, z. We assume the sample to be an ellipsoid with symmetry axes x', y', z'. Finally we introduce the coordinate systems ξ, η, ζ with the ζ axis parallel to the equilibrium direction of the magnetization $\mathbf{M}_0$. The rotation matrices which relate these coordinate systems are defined by

$$\begin{bmatrix}\xi\\ \eta\\ \zeta\end{bmatrix}=\begin{bmatrix}\alpha_1'' & \alpha_2'' & \alpha_3''\\ \alpha_1' & \alpha_2' & \alpha_3'\\ \alpha_1 & \alpha_2 & \alpha_3\end{bmatrix}\begin{bmatrix}x\\ y\\ z\end{bmatrix}=\begin{bmatrix}\beta_1'' & \beta_2'' & \beta_3''\\ \beta_1' & \beta_2' & \beta_3'\\ \beta_1 & \beta_2 & \beta_3\end{bmatrix}\begin{bmatrix}x'\\ y'\\ z'\end{bmatrix}$$

$$=\begin{bmatrix}\gamma_1'' & \gamma_2'' & \gamma_3''\\ \gamma_1' & \gamma_2' & \gamma_3'\\ \gamma_1 & \gamma_2 & \gamma_3\end{bmatrix}\begin{bmatrix}\xi'\\ \eta'\\ \zeta'\end{bmatrix} \tag{1.3}$$

The direction cosines of the equilibrium magnetization with respect to the crystal axes are α_1, α_2, α_3. We intro-

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[3] P. Pincus and J. Winter, Phys. Rev. Letters, 7, 269 (1961).

duce the magnon variables by

$$\begin{aligned}
S_j^{+}&=S_j^{\xi}+iS_j^{\eta}=(2S)^{1/2}a_j^{*},\\
S_j^{-}&=S_j^{\xi}-iS_j^{\eta}=(2S)^{1/2}a_j,\\
S_j^{\zeta}&=-S+a_j^{*}a_j,\\
a_j^{*}&=(1/N)^{1/2}\textstyle\sum_k e^{-i\mathbf{k}\cdot\mathbf{j}}a_k^{*},\\
a_j&=(1/N)^{1/2}\textstyle\sum_k e^{i\mathbf{k}\cdot\mathbf{j}}a_k,\\
[a_k,a_{k'}^{*}]&=\delta_{k,k'}.
\end{aligned}\tag{1.4}$$

Applying Eqs. (1.3) and (1.4) to (1.2), we have

$$H^e=H_0^e+H_1^e+H_2^e,\tag{1.5}$$

where H_0^e, H_1^e, and H_2^e include the magnon operators a and a^* in zeroth, first and second order, respectively. H_0^e and H_1^e are given by

$$\begin{aligned}
H_0^e=-NS\Big[g\mu_BH_0\gamma_3+\tilde{z}JS\\
-g\mu_B2\pi M_0\sum_{i=1}^{3}\beta_i^2(N_i-\tfrac{1}{3})-\kappa S\sum_{i=1}^{3}\alpha_i^4\Big],
\end{aligned}\tag{1.6}$$

$$\begin{aligned}
H_1^e=(S/2)^{1/2}\Big[g\mu_BH_0(\gamma_3''+i\gamma_3')\\
-g\mu_B4\pi M_0\sum_{i=1}^{3}\beta_i(\beta_i''+i\beta_i')(N_i-\tfrac{1}{3})\\
-4\kappa S\sum_{i=1}^{3}\alpha_i^3(\alpha_i''+i\alpha_i')\Big]\sum_j a_j+\text{c.c.},
\end{aligned}\tag{1.7}$$

where $\tilde{z}$ is the number of the nearest neighbors and N_i are the principal values of the demagnetization tensor which are given by

$$4\pi N_i=(V/N)\textstyle\sum_{j'}r_{jj'}^{-3}(1-3\lambda_{ijj'}^2)+\tfrac{4}{3}\pi,\tag{1.8}$$

$\lambda_{ijj'}$ being the direction cosines of $\hat{r}_{jj'}$ in the x', y', z' system.

The equilibrium values of α_i are determined by minimizing H_0^e with respect to α_i or putting the coefficient of a_j in H_1^e equal to zero.[4] The results are

$$\begin{aligned}
g\mu_BH_0\gamma_3'-g\mu_B4\pi M_0\textstyle\sum_i\beta_i\beta_i'N_i\\
-4\kappa S\textstyle\sum_i\alpha_i^3\alpha_i'=0,\\
g\mu_BH_0\gamma_3''-g\mu_B4\pi M_0\textstyle\sum_i\beta_i\beta_i''N_i\\
-4\kappa S\textstyle\sum_i\alpha_i^3\alpha_i''=0.
\end{aligned}\tag{1.9}$$

H_2^e is given by

$$H_2^e=\textstyle\sum_k(A_ka_k^*a_k+\tfrac{1}{2}B_ka_ka_{-k}+\tfrac{1}{2}B_k^*a_k^*a_{-k}^*),\tag{1.10}$$

with

$$\begin{aligned}
A_k=g\mu_BH_0\gamma_3+2\tilde{z}JS(1-\Gamma_k)+g\mu_B4\pi M_0\sum_{i=1}^{3}\beta_i^2(\tfrac{1}{3}-N_i)\\
+g\mu_B2\pi M_0[A_{\xi\xi}(\mathbf{k})+A_{\eta\eta}(\mathbf{k})]\\
+2\kappa S(3-5\sum_{i=1}^{3}\alpha_i^4),
\end{aligned}\tag{1.11}$$

$$\begin{aligned}
B_k=g\mu_B2\pi M_0[A_{\xi\xi}(\mathbf{k})-A_{\eta\eta}(\mathbf{k})+2iA_{\xi\eta}(\mathbf{k})]\\
+6\kappa S\sum_{i=1}^{3}\alpha_i^2(\alpha_i''+i\alpha_i')^2\\
\equiv|B_k|e^{2i\phi_k},
\end{aligned}$$

where

$$\Gamma_k=\textstyle\sum_\rho\exp(i\boldsymbol{\rho}\cdot\mathbf{k}),\tag{1.11a}$$

$\boldsymbol{\rho}$ being the vectors to nearest neighbors and $A_{\xi\xi}(\mathbf{k})$, etc., are given by

$$\begin{aligned}
A_{\xi\xi}(\mathbf{k})&=\frac{V}{4\pi N}\sum_{j'}\frac{1}{r_{jj'}^3}(1-\alpha_{jj'}^2)\exp[i\mathbf{k}\cdot(\mathbf{j}-\mathbf{j}')],\\
A_{\eta\eta}(\mathbf{k})&=\frac{V}{4\pi N}\sum_{j'}\frac{1}{r_{jj'}^3}(1-\beta_{jj'}^2)\exp[i\mathbf{k}\cdot(\mathbf{j}-\mathbf{j}')],\qquad(1.11\text{b})\\
A_{\xi\eta}(\mathbf{k})&=-\frac{3V}{4\pi N}\sum_{j'}\frac{1}{r_{jj'}^3}\alpha_{jj'}\beta_{jj'}\exp[i\mathbf{k}\cdot(\mathbf{j}-\mathbf{j}')],
\end{aligned}$$

$\alpha_{jj'}$, $\beta_{jj'}$, $\gamma_{jj'}$ being the direction cosines of $\hat{r}_{jj'}$ in the ξ, η, ζ coordinate system. Using the canonical transformation

$$\begin{aligned}
a_k&=2^{-1/2}[(a_k^{(1)}+a_k^{(2)})\cosh\tfrac{1}{2}\varphi_k\\
&\qquad-(a_k^{(1)*}-a_k^{(2)*})\sinh\tfrac{1}{2}\varphi_k]e^{-i\phi_k},\\
a_{-k}&=2^{-1/2}[(a_k^{(1)}-a_k^{(2)})\cosh\tfrac{1}{2}\varphi_k\\
&\qquad-(a_k^{(1)*}+a_k^{(2)*})\sinh\tfrac{1}{2}\varphi_k]e^{-i\phi_k},\\
\tanh\varphi_k&=|B_k|/A_k,
\end{aligned}\tag{1.12}$$

where ϕ_k is defined by Eq. (1.10), H_2^e reduces to

$$\begin{aligned}
H_2^e&={\sum_k}'\sum_{\mu=1,2}\hbar\omega_k^ea_k^{(\mu)*}a_k^{(\mu)}+\text{const},\\
\hbar\omega_k^e&=(A_k^2-|B_k|^2)^{1/2},
\end{aligned}\tag{1.13}$$

where $\sum_k'$ is the summation over the half-space of $\mathbf{k}$. For the case of wavelengths small compared to crystal dimensions, $A_{\xi\xi}(\mathbf{k})$ etc. are expressed as[5]

$$\begin{aligned}
A_{\xi\xi}(\mathbf{k})&=k_\xi^2/k^2-\tfrac{1}{3},\\
A_{\eta\eta}(\mathbf{k})&=k_\eta^2/k^2-\tfrac{1}{3},\\
A_{\xi\eta}(\mathbf{k})&=k_\xi k_\eta/k^2.
\end{aligned}\tag{1.14}$$

Finally the Hamiltonian of the phonon system is written as

$$\begin{aligned}
H^P&=\sum_k\sum_{s=1}^{3}\hbar\omega_k^PN_{ks}+\text{const},\\
N_{ks}&=b_{ks}^*b_{ks},\quad[b_{ks},b_{k's'}^*]=\delta_{kk'}\delta_{ss'},
\end{aligned}\tag{1.15}$$

where b_{ks}^* and b_{ks} are the creation and annihilation operators of phonons of mode s and wave vector $\mathbf{k}$. We assume throughout this paper that the phonon fre-

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[4] J. Kanamori and K. Yosida, Progr. Theoret. Phys. **14**, 423 (1955).

[5] M. H. Cohen and F. Keffer, Phys. Rev. **99**, 1128, 1135 (1955).

quency is independent of s and given by

$$\omega_k{}^P = vk, \tag{1.16}$$

v being the velocity of propagation of the long-wavelength sound waves.

II. MIXED MAGNON-PHONON MODES AND NUCLEAR SPIN RELAXATION

In this section we consider the case that the energy of the nuclear spin system is transferred to the mixed magnon-phonon modes, which are assumed in excellent thermal contact with the heat bath.

The magnon-phonon coupling energy for a cubic crystal is written as

$$\begin{aligned} H^{eP} = {} & G_1 \sum_j \left(\epsilon_j{}^{xx}(S_j{}^x)^2 + \epsilon_j{}^{yy}(S_j{}^y)^2 + \epsilon_j{}^{zz}(S_j{}^z)^2\right) + G_2 \\ & \times \sum_j \left[\epsilon_j{}^{xy}(S_j{}^x S_j{}^y + S_j{}^y S_j{}^x) + \epsilon_j{}^{yz}(S_j{}^y S_j{}^z + S_j{}^z S_j{}^y) \right. \\ & \left. + \epsilon_j{}^{zx}(S_j{}^z S_j{}^x + S_j{}^x S_j{}^z)\right], \end{aligned} \tag{2.1}$$

where $\epsilon_j{}^{ab}$ are the strain-tensor components, given by

$$\begin{aligned} \epsilon_j{}^{ab} = {} & i(\hbar/2MN)^{1/2} \sum_k \sum_s \left(k_s{}^{ab}/(\omega_k{}^P)^{1/2}\right) \\ & \times (b_{ks}e^{ikj} - b_{ks}{}^* e^{-ikj}), \end{aligned} \tag{2.2}$$

$$k_s{}^{ab} = \tfrac{1}{2}(e_{ks}{}^a k^b + e_{ks}{}^b k^a), \quad a, b = x, y, z.$$

The $e_{ks}{}^a$ are the components of the unit vectors in the directions of polarization of the mode s and MN is the total mass of the crystal. Considering only the bilinear coupling between the magnons and phonons, and using the transformations (1.4), (1.12) for magnons and

$$\begin{aligned} b_{ks} &= 2^{-1/2}(b_{ks}{}^{(1)} + b_{ks}{}^{(2)}), \\ b_{-ks} &= 2^{-1/2}(b_{ks}{}^{(1)} - b_{ks}{}^{(2)}), \end{aligned} \tag{2.3}$$

for phonons, we have

$$\begin{aligned} H^{eP} = {} & -\sum_k{}' \sum_{\mu=1,2} \sum_s u_{ks}\left[a_k{}^{(\mu)*} b_{ks}{}^{(\mu)} \right. \\ & \left. + (-)^\mu a_k{}^{(\mu)*} b_{ks}{}^{(\mu)*}\right] + \text{c.c.}, \end{aligned} \tag{2.4}$$

with

$$\begin{aligned} u_{ks} &= t_{ks}e^{i\phi_k} \cosh\tfrac{1}{2}\varphi_k + t_{ks}{}^* e^{-i\phi_k} \sinh\tfrac{1}{2}\varphi_k \\ &\equiv |u_{ks}| e^{iv_{ks}}, \end{aligned} \tag{2.4a}$$

where t_{ks} is given by

$$\begin{aligned} t_{ks} = {} & (\hbar S^3/M\omega_k{}^P)^{1/2}\{G_1[\alpha_1(\alpha_1' + i\alpha_1'')k_s{}^{xx} \\ & + \alpha_2(\alpha_2' + i\alpha_2'')k_s{}^{yy} + \alpha_3(\alpha_3' + i\alpha_3'')k_s{}^{zz}] \\ & + G_2[(\alpha_1(\alpha_2' + i\alpha_2'') + \alpha_2(\alpha_1' + i\alpha_1''))k_s{}^{xy} \\ & + (\alpha_2(\alpha_3' + i\alpha_3'') + \alpha_3(\alpha_2' + i\alpha_2''))k_s{}^{yz} \\ & + (\alpha_3(\alpha_1' + i\alpha_1'') + \alpha_1(\alpha_3' + i\alpha_3''))k_s{}^{zx}]\} \\ \equiv {} & |t_{ks}| e^{ir_{ks}}. \end{aligned} \tag{2.5}$$

The mixed magnon-phonon modes are obtained by diagonalizing H^m;

$$H^m = H^e + H^P + H^{eP}. \tag{2.6}$$

We introduce the new boson operators $\xi_{ks}{}^{(\mu)}$, $\eta_{ks}{}^{(\mu)}$, defined by

$$\begin{aligned} a_k{}^{(\mu)}e^{-iv_{ks}} = {} & \tfrac{1}{2}\{[f_{ks}{}^+\xi_{ks}{}^{(\mu)} + (-)^\mu f_{ks}{}^-\xi_{ks}{}^{(\mu)*}] \cos\Phi_{ks} \\ & - [g_{ks}{}^+\eta_{ks}{}^{(\mu)} + (-)^\mu g_{ks}{}^-\eta_{ks}{}^{(\mu)*}] \\ & \times \sin\Phi_{ks}\}, \end{aligned} \tag{2.7}$$

$$\begin{aligned} b_{ks}{}^{(\mu)} = {} & \tfrac{1}{2}\{[l_{ks}{}^+\xi_{ks}{}^{(\mu)} + (-)^\mu l_{ks}{}^-\xi_{ks}{}^{(\mu)*}] \sin\Phi_{ks} \\ & + [m_{ks}{}^+\eta_{ks}{}^{(\mu)} + (-)^\mu m_{ks}{}^-\eta_{ks}{}^{(\mu)*}] \\ & \times \cos\Phi_{ks}\}, \end{aligned}$$

with

$$\begin{aligned} f_{ks}{}^\pm &= \left(\frac{\omega_k{}^e}{\omega_{ks}{}^-}\right)^{1/2} \pm \left(\frac{\omega_{ks}{}^-}{\omega_k{}^e}\right)^{1/2}, \\ g_{ks}{}^\pm &= \left(\frac{\omega_k{}^e}{\omega_{ks}{}^+}\right)^{1/2} \pm \left(\frac{\omega_{ks}{}^+}{\omega_k{}^e}\right)^{1/2}, \\ l_{ks}{}^\pm &= \left(\frac{\omega_k{}^P}{\omega_{ks}{}^-}\right)^{1/2} \pm \left(\frac{\omega_{ks}{}^-}{\omega_k{}^P}\right)^{1/2}, \\ m_{ks}{}^\pm &= \left(\frac{\omega_k{}^P}{\omega_{ks}{}^+}\right)^{1/2} \pm \left(\frac{\omega_{ks}{}^+}{\omega_k{}^P}\right)^{1/2}, \end{aligned} \tag{2.8}$$

and

$$\tan 2\Phi_{ks} = -\frac{4|u_{ks}|(\omega_k{}^e\omega_k{}^P)^{1/2}\hbar^{-1}}{(\omega_k{}^e)^2 - (\omega_k{}^P)^2}. \tag{2.9}$$

In these expressions, $\omega_{ks}{}^\pm$ are given by

$$\begin{aligned} (\omega_{ks}{}^\pm)^2 = {} & \tfrac{1}{2}[(\omega_k{}^e)^2 + (\omega_k{}^P)^2] \\ & \pm \tfrac{1}{2}\{[(\omega_k{}^e)^2 - (\omega_k{}^P)^2]^2 \\ & + (4|u_{ks}|/\hbar)^2\omega_k{}^e\omega_k{}^P\}^{1/2}, \end{aligned} \tag{2.10}$$

which are independent of μ, i.e., the same values for both **k** and −**k**. In terms of these new operators, H^m is diagonalized and written as

$$\begin{aligned} H^m = {} & \sum_k{}' \sum_s \sum_\mu [\hbar\omega_{ks}{}^- n_{ks}{}^{(\mu)} \\ & + \hbar\omega_{ks}{}^+ n_{ks}{}^{(\mu)\prime}] + \text{const}, \end{aligned} \tag{2.11}$$

$$n_{ks}{}^{(\mu)} = \xi_{ks}{}^{(\mu)*}\xi_{ks}{}^{(\mu)}, \quad n_{ks}{}^{(\mu)\prime} = \eta_{ks}{}^{(\mu)*}\eta_{ks}{}^{(\mu)};$$

n_{ks}, $n_{ks}{}'$ are the occupation number of the mixed magnon-phonon modes. As seen from Eq. (2.10), the branch with $\omega_{ks}{}^-$ for small k behaves like a phonon spectrum, so it is called the quasiphonon state.[3]

Next we calculate the relaxation time of the nuclear spin. The unperturbed nuclear Hamiltonian is

$$H_0{}^N = -\hbar\omega_0 \sum_j I_j{}^\zeta, \tag{2.12}$$

with

$$\hbar\omega_0 = A_0 S, \tag{2.13}$$

where A_0 is the static part of the hyperfine coupling coefficient and we assume

$$g_n\mu_n H_0 \ll A_0 S \tag{2.14}$$

throughout. ω_0 can be taken as positive, because it is seen that the relaxation time is independent of the sign of ω_0. The Hamiltonian which is responsible for the relaxation H' is

$$H'=\tfrac{1}{2}A_0\textstyle\sum_j(S_j^+I_j^-+S_j^-I_j^+). \tag{2.15}$$

Using the transformations (1.4), (1.12), (2.7) and considering only the quasiphonons, H' is written as

$$\begin{aligned}H'=\tfrac{1}{4}A_0(S/N)^{1/2}&\textstyle\sum_j\sum_k{}'\sum_s\sum_\mu e^{iv_{ks}}\\&\times[\exp(i\mathbf{k}\cdot\mathbf{j})-(-)^\mu\exp(-i\mathbf{k}\cdot\mathbf{j})]\cos\Phi_{ks}\\&\times(I_j^+e^{-i\phi_k}\cosh\tfrac{1}{2}\varphi_k-I_j^-e^{i\phi_k}\sinh\tfrac{1}{2}\varphi_k)\\&\times[f_{ks}^+\xi_{ks}^{(\mu)}+(-)^\mu f_{ks}^-\xi_{ks}^{(\mu)*}]+\text{c.c.}\end{aligned} \tag{2.16}$$

By standard perturbation theory, the probability per unit time W_j^+ of a transition from state $|m_j, n_{ks}^{(\mu)})$ to state $|m_j+1, n_{ks}^{(\mu)}+1)$ is

$$\begin{aligned}W_j^+(m_j, n_{ks}^{(\mu)}&\rightarrow m_j+1, n_{ks}^{(\mu)}+1)\\&=(2\pi/\hbar^2)|(m_j+1, n_{ks}^{(\mu)}+1|H'|m_j, n_{ks}^{(\mu)})|^2\\&\qquad\times\delta(\omega_{ks}^--\omega_0),\end{aligned} \tag{2.17}$$

where m_j is the quantum number of I_j^ζ. Considering that the nuclear spins are independent of each other in the unperturbed system and taking the thermal averages over the quasiphonons, we obtain the probability per unit time W^+ that the quantum number of a nuclear spin m increases by one:

$$\begin{aligned}W^+&=\textstyle\sum_k{}'\sum_\mu\sum_s N^{-1}\\&\qquad\times\textstyle\sum_j\langle W_j^+\rangle\text{ average over quasiphonons}\\&=\frac{\pi A_0^2S}{4\hbar^2}(m|I^-I^+|m)\frac{1}{N}\sum_k\sum_s|e^{iv_{ks}}f_{ks}^-\cosh\tfrac{1}{2}\varphi_k\\&\qquad+e^{-iv_{ks}}f_{ks}^+\sinh\tfrac{1}{2}\varphi_k|^2\\&\qquad\times\cos^2\Phi_{ks}(\bar{n}_{ks}+1)\delta(\omega_{ks}^--\omega_0),\end{aligned} \tag{2.18}$$

where $\bar{n}_{ks}$ is given by

$$\bar{n}_{ks}=(e^{\beta\hbar\omega_{ks}^-}-1)^{-1}, \tag{2.19}$$

β being $1/k_BT$. In a similar way, the probability per unit time that m decreases by one is obtained, the result being

$$\begin{aligned}W^-=(\pi A_0^2S/4\hbar^2)&(m|I^+I^-|m)N^{-1}\\&\times\textstyle\sum_k\sum_s|e^{iv_{ks}}f_{ks}^-\cosh\tfrac{1}{2}\varphi_k+e^{-iv_{ks}}f_{ks}^+\sinh\tfrac{1}{2}\varphi_k|^2\\&\qquad\times\cos^2\Phi_{ks}\bar{n}_{ks}\delta(\omega_{ks}^--\omega_0).\end{aligned} \tag{2.20}$$

Using these probabilities, the nuclear spin-lattice relaxation time T_1 is given by[6]

$$T_1^{-1}=\langle m(W^--W^+)\rangle/[\langle m^2\rangle-\langle m\rangle^2], \tag{2.21}$$

where

$$\begin{aligned}\langle f(m)\rangle&=\sum_{m=-I}^{I}f(m)e^{\beta\hbar\omega_0m}\Big/\sum_{m=-I}^{I}e^{\beta\hbar\omega_0m}\\&\equiv\langle\langle f(I^\zeta)\rangle\rangle.\end{aligned} \tag{2.22}$$

Now we assume

$$\omega_k^P/\omega_k^e,\quad |u_{ks}|/\hbar\omega_k^e,\quad\text{and}\quad |u_{ks}|^2/\hbar^2\omega_k^2\omega_k^P\ll 1, \tag{2.23}$$

for k which is given by the δ function appearing in Eqs. (2.18) and (2.20). In this case ω_{ks}^- and $\cos\Phi_{ks}$ are given by

$$\begin{aligned}\omega_{ks}^-&\simeq\omega_k^P=vk,\\ \cos^2\Phi_{ks}&\simeq\left(\frac{2|u_{ks}|}{\hbar\omega_k^e}\right)^2\frac{\omega_k^P}{\omega_k^e},\end{aligned} \tag{2.24}$$

and $f_{ks}^\pm$ are

$$f_{ks}^+\simeq f_{ks}^-\simeq(\omega_k^e/\omega_k^P)^{1/2}, \tag{2.25}$$

from Eqs. (2.8), (2.9), and (2.10). Then the relaxation time reduces to

$$\begin{aligned}\frac{1}{T_1}=\frac{\pi A_0^2S}{\hbar^2}F_1(T)\frac{1}{N}\sum_k\sum_s&\left(\frac{|t_{ks}|}{\hbar\omega_k^e}\right)^2\\&\times[\cosh2\varphi_k+\sinh2\varphi_k\cos2(\phi_k+r_{ks})]\\&\qquad\times\delta(vk-\omega_0),\end{aligned} \tag{2.26}$$

where

$$F_1(T)=\left[\frac{\langle\langle I^\zeta I^+I^-\rangle\rangle}{e^{\beta\hbar\omega_0}-1}-\frac{\langle\langle I^\zeta I^-I^+\rangle\rangle}{1-e^{-\beta\hbar\omega_0}}\right]\Big/[\langle\langle(I^\zeta)^2\rangle\rangle-\langle\langle I^\zeta\rangle\rangle^2]. \tag{2.27}$$

In Eq. (2.26) we use $|t_{ks}|$ and r_{ks} instead of $|u_{ks}|$ and v_{ks}, the relation between these quantities being given by Eqs. (2.4) and (2.5).

The temperature dependence of the relaxation time is given by the function $F_1(T)$ which is shown in Fig. 1 for

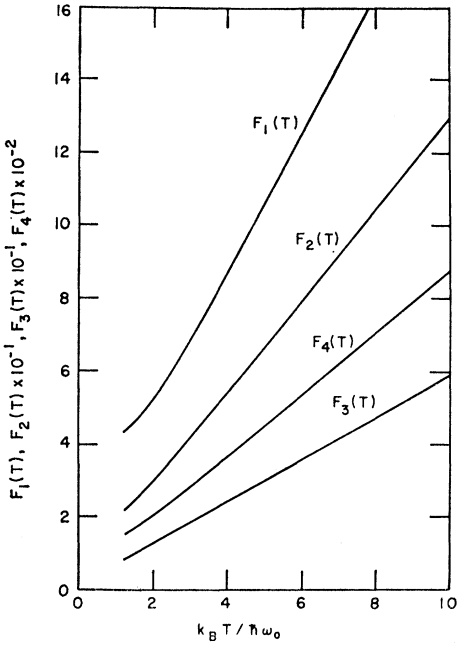

FIG. 1. Curves of $F_1(T)$, $F_2(T)$, $F_3(T)$, and $F_4(T)$ versus $k_BT/\hbar\omega_0$.

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[6] For example, C. P. Slichter, *Principles of Magnetic Resonance* (Harper & Row, New York, 1963), Chap. 5, without the assumption $\beta E_n\ll 1$.

$I=\frac{5}{2}$. $F_1(T)$ is proportional to temperature for $k_BT>6\hbar\omega_0$, represented by $2k_BT/\hbar\omega_0$, and deviates slightly from a straight line for lower temperatures. A typical value of $\hbar\omega_0$ is $\sim 10^{-2}$ cm^{-1}$=1.4\times 10^{-2}$ °K. The curve at ultralow temperatures near 0°K is not shown in Fig. 1, because the assumption that the quasiphonon system is in the thermal equilibrium will not be valid at such a low temperature.

The external field dependence of T_1 is given by the summation term in Eq. (2.26), which shows that T_1 is proportional to H_0^2 for high fields such that $H_0 \gg 4\pi M_0$, $\kappa S/g\mu_B$, but deviates from the H_0^2 curve for fields comparable with the magnetization or the anisotropy field. It is difficult to perform the summation rigorously, so we will consider the following two extreme cases:

$$\text{Case 1:}\quad H_0,\ 4\pi M_0 \gg \kappa S/g\mu_B\,, \tag{2.28}$$

$$\text{Case 2:}\quad H_0,\ \kappa S/g\mu_B \gg 4\pi M_0\,. \tag{2.29}$$

The value of the magnitude of vector k is determined by the δ function in Eq. (2.26), i.e., $k=A_0S/v\hbar$. For the values of $A_0S\sim 10^{-2}$ cm^{-1} and $v\sim 10^5$ cm/sec, $1/k$ is estimated as $\sim 10^{-4}$ cm which is very small compared to crystal dimensions and lies well outside the Walker-mode[7] region, showing that the spin-wave theory to represent the electron-spin system gives a good approximation for this k. The exchange energy for small k is given by $2\tilde{z}JS(ak)^2$ from Eq. (1.10) for cubic lattices, where a is the nearest-neighbor distance. A typical value of this energy is, at most, $\sim 10^{-4}J$, so we may omit this energy in the magnon spectrum.

Case 1. Introducing the angles θ_k and ψ_k of vector **k** in the ξ,η,ζ coordinate system and using Eqs. (1.10) and (1.14), we obtain

$$A_k=g\mu_BH_0\gamma_3-g\mu_B4\pi M_0\sum_{i=1}^{3}\beta_i^2N_i + g\mu_B2\pi M_0\sin^2\theta_k\,, \tag{2.30}$$

$$|B_k|=g\mu_B2\pi M_0\sin^2\theta_k\,,$$

$$\phi_k=\psi_k\,.$$

Replacing the summation over **k** by an integral and using the orthogonality relations

$$\begin{aligned}\textstyle\sum_s e_{ks}{}^a e_{ks}{}^b&=\delta_{ab}\,,\\ \textstyle\sum_a e_{ks}{}^a e_{ks'}{}^a&=\delta_{ss'}\,,\end{aligned} \tag{2.31}$$

we obtain, after some tedious calculations,

$$\frac{1}{T_1}=\frac{A_0^5S^7G_1^2}{8\pi\hbar^4v^5d}F_1(T)\frac{G_\alpha(H_0)}{(g\mu_BH_0)^2}\,, \tag{2.32}$$

where d is the density of the crystal. $G_\alpha(H_0)$ is a function of the magnon-phonon coupling constants G_i and the equilibrium direction of magnetization α_i as well as H_0 and M_0. It is expressed as

$$\begin{aligned}G_\alpha(H_0)=&\tfrac{1}{2}r_G^2(I_1+I_2-I_3+I_4)+(1-r_G)\\ &\times[(1+r_G)(I_1+I_2)-\tfrac{1}{2}(1+3r_G)(I_3-I_4)]K_\alpha\\ &+3(1-r_G)^2(I_1-3I_2-\tfrac{1}{2}I_3+\tfrac{1}{2}I_4)L_\alpha\,,\end{aligned} \tag{2.33}$$

with

$$\begin{aligned}r_G&=G_2/G_1\,,\\ K_\alpha&=\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2\,,\\ L_\alpha&=\alpha_1^2\alpha_2^2\alpha_3^2\,.\end{aligned} \tag{2.34}$$

The integrals I_i are defined as

$$\begin{aligned}I_1&=\int_0^\pi (g\mu_BH_0/\hbar\omega_k{}^e)^2\cosh2\varphi_k\sin\theta_k d\theta_k\,,\\ I_2&=\int_0^\pi (g\mu_BH_0/\hbar\omega_k{}^e)^2\cosh2\varphi_k\cos^2\theta_k\sin\theta_k d\theta_k\,,\\ I_3&=\int_0^\pi (g\mu_BH_0/\hbar\omega_k{}^e)^2\sinh2\varphi_k\sin\theta_k d\theta_k\,,\\ I_4&=\int_0^\pi (g\mu_BH_0/\hbar\omega_k{}^e)^2\sinh2\varphi_k\cos^2\theta_k\sin\theta_k d\theta_k\,.\end{aligned} \tag{2.35}$$

If the sample has a shape of an ellipsoid of revolution about z' axis,

$$N_1=N_2=N_\perp\,,\quad N_3=N_\parallel\,, \tag{2.36}$$

and I_i are given by

$$\begin{aligned}I_1=&(h_1-m_0N_\perp)^{-2}+(2(h_1-m_0N_\perp)[h_1+m_0(N_\parallel+N_\perp)])^{-1}\\ &+(2m_0^{1/2}[h_1+m_0(N_\parallel+N_\perp)]^{3/2})^{-1}\\ &\times\tanh^{-1}[m_0/(h_1+m_0(N_\parallel+N_\perp))]^{1/2}\,,\\ I_2=&(3(h_1-m_0N_\perp)^2)^{-1}+(2m_0(h_1-m_0N_\perp))^{-1}\\ &-(2m_0^{3/2}[h_1+m_0(N_\parallel+N_\perp)]^{1/2})^{-1}\\ &\times\tanh^{-1}[m_0/(h_1+m_0(N_\parallel+N_\perp))]^{1/2}\,,\\ I_3=&\frac{2}{(h_1-m_0N_\perp)^2}-I_1\,,\quad I_4=\frac{2}{3(h_1-m_0N_\perp)^2}-I_2\,,\end{aligned} \tag{2.37}$$

where

$$h_1=\gamma_3-m_0(N_\parallel-N_\perp)\beta_3^2\,, \tag{2.38}$$

and

$$m_0=4\pi M_0/H_0\,. \tag{2.39}$$

The equilibrium values of α_i are determined by Eq. (1.9) and given by

$$\begin{aligned}m_0(N_\parallel-N_\perp)\beta_3\beta_3''&=\gamma_3''\,,\\ m_0(N_\parallel-N_\perp)\beta_3\beta_3'&=\gamma_3'\,.\end{aligned} \tag{2.40}$$

From Eq. (2.40), the relation between γ_3 and β_3 is

$$\gamma_3=[1-m_0^2(N_\parallel-N_\perp)^2\beta_3^2(1-\beta_3^2)]^{1/2}\,, \tag{2.41}$$

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[7] L. R. Walker, Phys. Rev. **105**, 390 (1957).

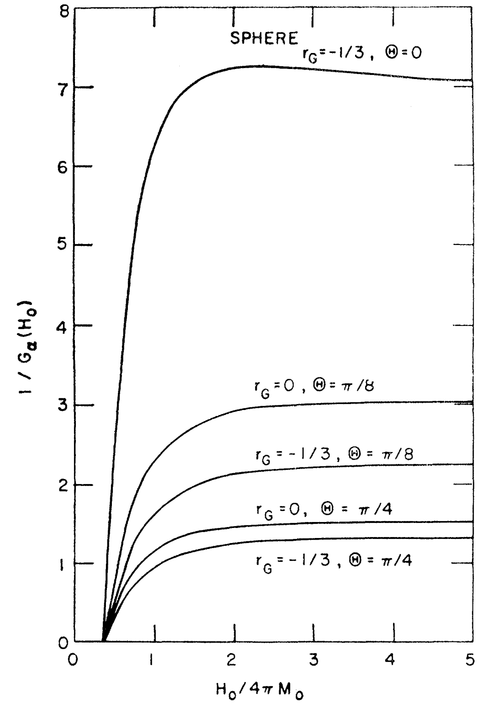

FIG. 2. External field dependence of $1/G_\alpha(H_0)$ for the sphere sample. The external field is applied in the plane constructed by the two crystal axes. Θ is measured from a crystal axis.

where only the positive value of γ_3 is necessary, because the magnon energy should have a real value.

Case 2. In this case, A_k, $|B_k|$, and ϕ_k defined by (1.10) are independent of **k**, so we denote them divided by $g\mu_B H_0$ by a_α, b_α, and ϕ_α. They are written as

$$\begin{aligned} a_\alpha &= \gamma_3+\tilde{\kappa}(5K_\alpha-1), \\ b_\alpha &= 3\tilde{\kappa}(K_\alpha^2-3L_\alpha)^{1/2}, \\ \tan\phi_\alpha &= 2\sum_i \alpha_i^2\alpha_i'\alpha_i''/\sum_i \alpha_i^2(\alpha_i''^2-\alpha_i'^2), \end{aligned} \tag{2.42}$$

where

$$\tilde{\kappa}=4\kappa S/g\mu_B H_0. \tag{2.43}$$

Replacing the summation over **k** in Eq. (2.26) by the integral and using the relation (2.31), we obtain

$$\frac{1}{T_1}=\frac{A_0^5S^7G_1^2}{8\pi\hbar^4v^5d}F_1(T)\frac{G_\alpha'(H_0)}{(g\mu_B H_0)^2}, \tag{2.44}$$

where

$$G_\alpha'(H_0)=\frac{8}{3(a_\alpha^2-b_\alpha^2)^2}\{[\tfrac{1}{2}r_G^2+(1-r_G^2)K_\alpha](a_\alpha^2+b_\alpha^2) \\ -6\tilde{\kappa}a_\alpha(1-r_G^2)(K_c^2-3L_\alpha)\}. \tag{2.45}$$

The equilibrium values of α_i are given by

$$\begin{aligned} \tilde{\kappa}\textstyle\sum_i \alpha_i^3\alpha_i' &= \gamma_3', \\ \tilde{\kappa}\textstyle\sum_i \alpha_i^3\alpha_i'' &= \gamma_3'', \end{aligned} \tag{2.46}$$

from Eq. (1.9) and the relation between γ_3 and α_i is

$$\gamma_3=[1-\tilde{\kappa}^2(K_\alpha+3L_\alpha-4K_\alpha^2)]^{1/2}, \tag{2.47}$$

from Eq. (2.46).

G_α and G_α' depend on the external field vector H_0 and the ratio $r_G=G_2/G_1$ when M_0, κ, and G_1 are given. The dependences on these variables are shown in Figs. 2–6. In these figures, H_0 is applied in the planes constructed by the two crystal axes, and the symmetry axis of the ellipsoid of revolution z' coincides with the crystal [001] axis. The radios r_G are taken as 0, $-\frac{1}{3}$. Since the G_i are related with the usual spin-lattice coupling coefficients G_{11}, G_{44} for cubic crystals by

$$G_1=\tfrac{3}{2}G_{11},\quad G_2=2G_{44}, \tag{2.48}$$

r_G adopted here may be reasonable for the orbital S-state atoms, referring to the experimental values on Mn^{2+}.[8] As seen from these figures, G_α and G_α' depend strongly on the shape of the sample and the sign of the anisotropy constant and therefore T_1 is not proportional to H_0^2 for $H_0\sim 4\pi M_0$ or $\kappa S/g\mu_B$. The saturation values of H_0 above which G_α and G_α' are regarded as constants are $8\pi M_0$ for the Case 1 and $16\kappa S/g\mu_B$ for Case 2.

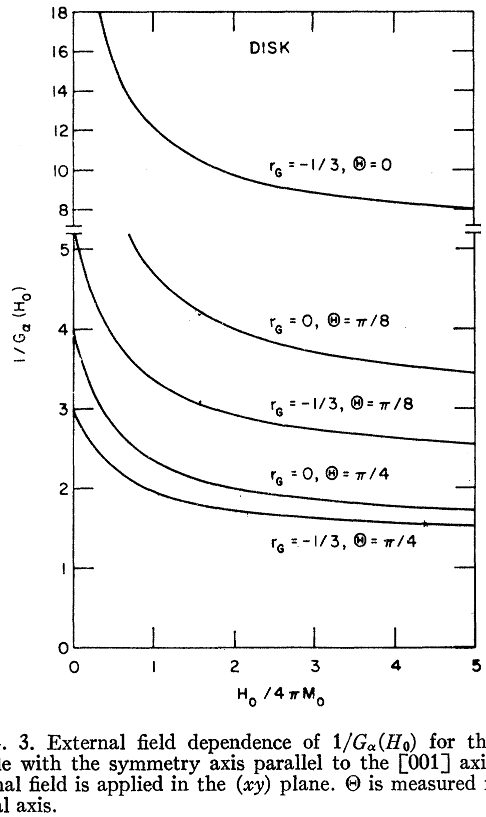

FIG. 3. External field dependence of $1/G_\alpha(H_0)$ for the disk sample with the symmetry axis parallel to the [001] axis. The external field is applied in the (xy) plane. Θ is measured from a crystal axis.

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[8] G. D. Watkins and E. Feher, Bull. Am. Phys. Soc. 7, 29 (1962); N. S. Shiren, Bull. Am. Phys. Soc. 7, 29 (1962).

III. INDIRECT NUCLEAR-SPIN INTERACTION AND RELAXATION

The indirect nuclear spin interactions in magnetic materials are given by Suhl and Nakamura.[2] In this section we will first derive the Hamiltonian responsible for the nuclear relaxation in ferromagnets and then calculate the relaxation time.

The indirect coupling between nuclear spins is obtained from the second-order perturbation of

$$H_{\mathrm{hf}}'=\tfrac{1}{2}\textstyle\sum_j A_j(S_j^+I_j^-+S_j^-I_j^+)\,, \tag{3.1}$$

with respect to the electronic spin system. A_j in Eq. (3.1) is the hyperfine coupling coefficient of site j and considered as a function of strain because the electrons of the atom have overlap and covalency with the electrons of the surrounding ions. A_j is, therefore, expanded in powers of the strain and is written as

$$A_j=A_0+\epsilon_j A_{j1}+\cdots\,, \tag{3.2}$$

where ϵ_j is, for simplicity, the average strain neglecting polarization effects. Applying transformations (1.4) and (1.12) to (3.1), we have

$$\begin{aligned}H_{\mathrm{hf}}'=\tfrac{1}{2}(S/N)^{1/2}&\textstyle\sum_j\sum_k{}'\sum_\mu\\ &\times A_j[e^{i\mathbf{k}\cdot}-(-)^\mu e^{-i\mathbf{k}\cdot\mathbf{j}}](e^{-i\phi_k}I_j^+\cosh\tfrac{1}{2}\varphi_k\\ &\qquad-e^{i\phi_k}I_j^-\sinh\tfrac{1}{2}\varphi_k)a_k^{(\mu)}+\text{c.c.}\end{aligned} \tag{3.3}$$

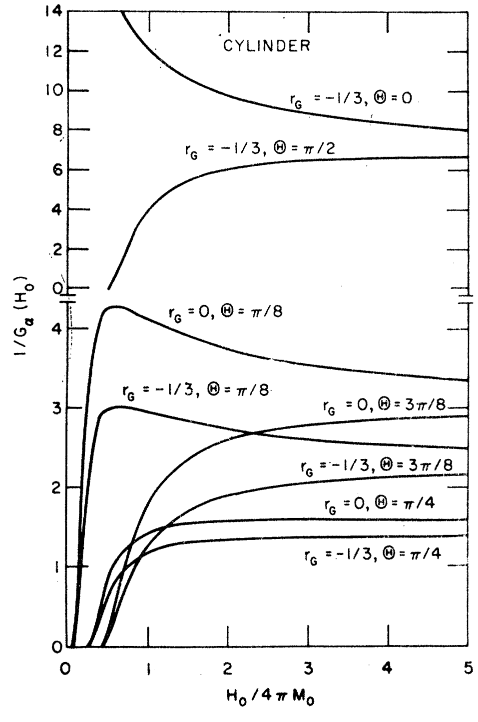

FIG. 4. External field dependence of $1/G_\alpha(H_0)$ for the cylinder sample with the symmetry axis parallel to the [001] axis. The external field is applied in the (zx) plane. Θ is measured from the [001] axis.

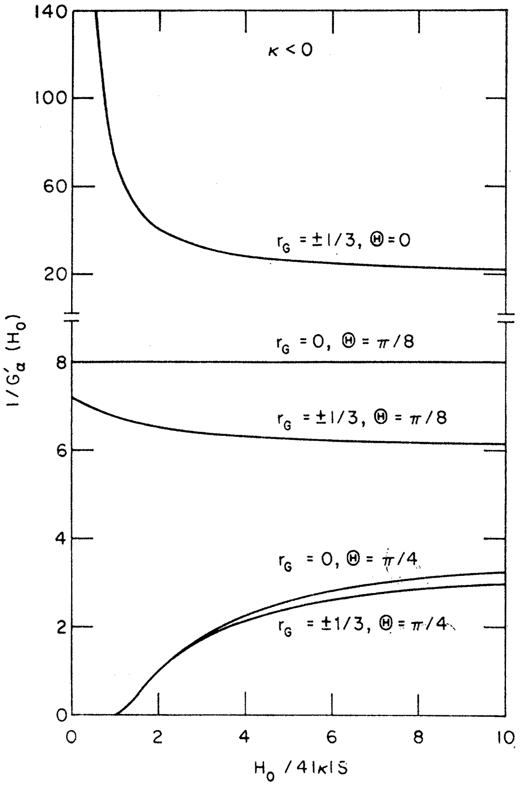

FIG. 5. External field dependence of $1/G_\alpha'(H_0)$ for the negative anisotropy constant, i.e., the easy axes are [100]. The external field is applied in the plane constructed by the two crystal axes and Θ is measured from a crystal axis.

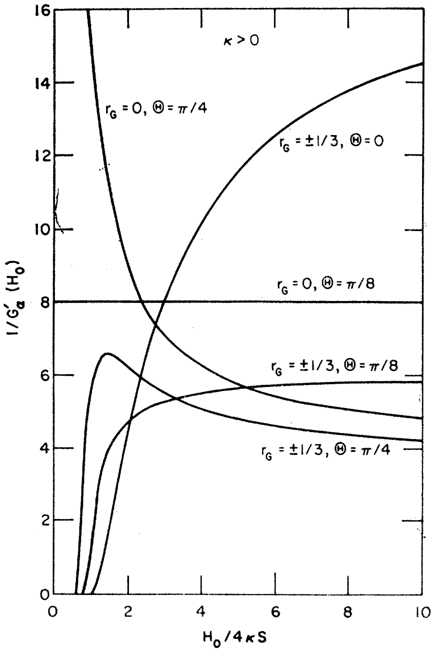

FIG. 6. External field dependence of $1/G_\alpha'(H_0)$ for the positive anisotropy constant, the easy axis being [111]. The external field is applied in the plane constructed by the two crystal axes and Θ is measured from a crystal axis.

The second-order perturbation of Eq. (3.3) gives

$$\Delta E=(S/4N)\sum_j\sum_{j'}\sum_k A_jA_{j'}(\cos\mathbf{k}\cdot(\mathbf{j}-\mathbf{j}')/\hbar\omega_k{}^e)\times[(e^{2i\phi_k}I_j{}^-I_{j'}{}^-+e^{-2i\phi_k}I_j{}^+I_{j'}{}^+)\sinh\varphi_k-2I_j{}^\zeta\delta_{jj'}-(I_j{}^+I_{j'}{}^-+I_j{}^-I_{j'}{}^+)\cosh\varphi_k], \quad (3.4)$$

in which only the first term is responsible for the relaxation. Using (3.2) and (3.4), the relaxation Hamiltonian for the direct process is written as

$$H'=(A_0A_1S/4N)\sum_k\sum_j(\sinh\varphi_k/\hbar\omega_k{}^e)\times[2\epsilon_j(e^{-2i\phi_k}I_j{}^{+2}+e^{2i\phi_k}I_j{}^{-2})+\sum_{j'\neq j}(\epsilon_j+\epsilon_{j'})\cos\mathbf{k}\cdot(\mathbf{j}-\mathbf{j}')\times(e^{-2i\phi_k}I_j{}^+I_{j'}{}^++e^{2i\phi_k}I_j{}^-I_{j'}{}^-)]. \quad (3.5)$$

The relaxation time through the mechanism (3.5) is given by

$$\frac{1}{T_1}=\frac{6A_0{}^5A_1{}^2S^5}{\pi\hbar^4v^5d(g\mu_BH_0)^2}\times[L_1(H_0)F_2(T)+2L_2(H_0)F_3(T)], \quad (3.6)$$

where

$$L_1(H_0)=|N^{-1}\sum_k(g\mu_BH_0/\hbar\omega_k{}^e)\sinh\varphi_ke^{2i\phi_k}|^2, \quad (3.7)$$

$$L_2(H_0)=N^{-1}\sum_k((g\mu_BH_0/\hbar\omega_k{}^e)\sinh\varphi_k)^2, \quad (3.8)$$

$$F_2(T)=\left[\frac{\langle\langle I^\zeta I^{+2}I^{-2}\rangle\rangle}{e^{2\beta\hbar\omega_0}-1}-\frac{\langle\langle I^\zeta I^{-2}I^{+2}\rangle\rangle}{1-e^{-2\beta\hbar\omega_0}}\right]\Big/[\langle\langle(I^\zeta)^2\rangle\rangle-\langle\langle I^\zeta\rangle\rangle^2], \quad (3.9)$$

$$F_3(T)=\left[\frac{\langle\langle I^\zeta I^+I^-\rangle\rangle\langle\langle I^+I^-\rangle\rangle}{e^{2\beta\hbar\omega_0}-1}-\frac{\langle\langle I^\zeta I^-I^+\rangle\rangle\langle\langle I^-I^+\rangle\rangle}{1-e^{-2\beta\hbar\omega_0}}\right]\Big/[\langle\langle(I^\zeta)^2\rangle\rangle-\langle\langle I^\zeta\rangle\rangle^2]. \quad (3.10)$$

In deriving Eq. (3.6) we have used the fact that for $r=|\mathbf{j}-\mathbf{j}'|/a_0\gg1$, a_0 being lattice constant, the integration of the term $\cos[\mathbf{k}\cdot(\mathbf{j}-\mathbf{j}')]/\hbar\omega_k{}^e$ over the magnitude of $\mathbf{k}$ decreases exponentially with r,[2] so that we can take the value of $ra_0\omega_0/v$ as very small compared to unity.

$F_2(T)$ and $F_3(T)$ which are shown in Fig. 1, behave like $F_1(T)$ defined by (2.27), that is, for $I=\frac{5}{2}$ they are proportional to temperature for $k_BT>5\hbar\omega_0$ and deviate from the straight lines for lower temperature in a way similar to the $F_1(T)$ curve. At higher temperatures $F_2(T)$ and $F_3(T)$ are given by $2\{4I(I+1)-3\}k_BT/5\hbar\omega_0$ and $2I(I+1)k_BT/3\hbar\omega_0$, respectively. The field dependence of $L_i(H_0)$ may be similar to $G_\alpha(H_0)$ in Sec. 2.

We compare the order of magnitude of the relaxation time $T_{1\,\mathrm{Ind}}$ obtained here with that of T_{1M} in Sec. 2. For the Case 1 defined by Eq. (2.28), $L_1(H_0)$ in Eq. (3.6) can be neglected compared to $L_2(H_0)$ and T_{1M} is given by Eq. (2.32). The ratio is

$$\frac{T_{1M}}{T_{1\,\mathrm{Ind}}}=\frac{96}{S^2}\frac{F_3(T)}{F_1(T)}\frac{L_2(H_0)}{G_\alpha(H_0)}\left(\frac{A_1}{G_1}\right)^2=\frac{32}{S^2}I(I+1)\frac{L_2(H_0)}{G_\alpha(H_0)}\left(\frac{A_1}{G_1}\right)^2. \quad (3.11)$$

The last step in Eq. (3.11) is obtained by using the high-temperature approximation. Since the order of magnitude of $L_2(H_0)$ is the same as that of $G_\alpha(H_0)$ except for $r_G\sim0$ and some special values of α_i, A_1 should have a value of

$$A_1\sim G_1/10 \quad (3.12)$$

in order that $T_{1\,\mathrm{Ind}}$ be comparable to T_{1M}. For a rare-earth ion, G_1 has a value of 10 cm^{-1}, so A_1 from (3.12) becomes ~1 cm^{-1} which seems to be too large compared to $A_0\sim10^{-2}$ cm^{-1}. That $L_2(H_0)$ has the same order of magnitude as $G_\alpha(H_0)$ may be certainly true for $G_\alpha(H_0)$ averaged over all angles contained.

IV. QUADRUPOLAR NUCLEAR SPIN-LATTICE RELAXATION

The nuclear relaxation time arising from the quadrupolar energy has been calculated by Van Kranendonk[9] for the Raman process. In this section we calculate the expression of the relaxation time for the nuclei which belong to magnetic atoms in a ferromagnetic insulator, considering only the direct process. In this case the heat bath is the phonon system expressed by the Hamiltonian (1.15) and the unperturbed nuclear Hamiltonian is given by Eq. (2.12). The perturbation H' is the quadrupolar energy expanded in powers of the displacements of nuclei, retaining only the linear terms with respect to displacements:

$$H'=\sum_\mu Q_\mu\sum_j\mathbf{A}_{\mu j}\cdot\mathbf{u}_{j0}=(\hbar/2MN)^{1/2}\sum_\mu Q_\mu\sum_j\sum_k\sum_s(\mathbf{A}_{\mu j}\cdot\mathbf{e}_{ks}/(\omega_k{}^P)^{1/2})\times[b_{ks}(\exp(i\mathbf{k}\cdot\mathbf{j})-1)+b_{ks}{}^*(\exp(-i\mathbf{k}\cdot\mathbf{j})-1)], \quad (4.1)$$

where the summation over j extends over all the nuclei except the central nucleus $j=0$ and $\mathbf{u}_j$ is the relative displacement $\mathbf{u}_j-\mathbf{u}_0$ with respect to the central nucleus. Q_μ in Eq. (4.1) are given by

$$Q_0=B[3(I^\zeta)^2-I(I+1)],\quad Q_{\pm1}=B[I^\zeta I^\pm+I^\pm I^\zeta]/2,\quad Q_{\pm2}=BI^{\pm2}, \quad (4.2)$$

where

$$B=eQ/I(2I-1),\quad eQ=(II|Q_0|II). \quad (4.3)$$

In the point-charge model, the vectors $\mathbf{A}_{\mu j}$ in Eq. (4.1) are obtained as in Van Kranendonk's paper[8] and written

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[9] J. Van Kranendonk, Physica **20**, 781 (1954).

as

$$\textstyle\sum_\mu \mathbf{A}_{\mu j}(\mathbf{r})Y_\mu(\mathbf{r})=(3q/2R_j^4)[2(\hat{R}_j\cdot\mathbf{r})\mathbf{r}+(r^2-5(\hat{R}_j\cdot\mathbf{r})^2)\hat{R}_j], \quad (4.4)$$

where

$$Y_0(\mathbf{r})=3z^2-r^2,\quad Y_{\pm1}(\mathbf{r})=z(x\pm iy),\quad Y_{\pm2}(\mathbf{r})=(x\pm iy)^2, \quad (4.5)$$

and

$$\hat{R}_j=\mathbf{R}_j/R_j,$$

$\mathbf{R}_j$ being the relative position vector in equilibrium from the central nucleus. q in Eq. (4.4) is the effective charge of the ion and is assumed to be

$$q=\gamma q_0, \quad (4.6)$$

in which q_0 is the ionic charge and the multiplication factor γ includes both the Sternheimer antishielding factor[10] for the nuclear quadrupole moment, and the covalent and overlap effects on the central ion due to the surrounding ions. From Eq. (4.4) we obtain the relations

$$\mathbf{A}_{\mu-j}=-\mathbf{A}_{\mu j},\quad \mathbf{A}_{-\mu j}=\mathbf{A}_{\mu j}^*. \quad (4.7)$$

For the simple cubic lattice with lattice constant a and considering only the contributions from the nearest neighbors, $\mathbf{A}_{\mu j}$ are given by

$$\begin{aligned}&\mathbf{A}_{01}=C(\tfrac{1}{2},0,0), && \mathbf{A}_{11}=C(0,0,1), && \mathbf{A}_{21}=C(-\tfrac{3}{4},-\tfrac{1}{2}i,0),\\ &\mathbf{A}_{02}=C(0,\tfrac{1}{2},0), && \mathbf{A}_{12}=C(0,0,-i), && \mathbf{A}_{22}=C(-\tfrac{1}{2}i,\tfrac{3}{4},0),\\ &\mathbf{A}_{03}=C(0,0,-1), && \mathbf{A}_{13}=C(1,-i,0), && \mathbf{A}_{23}=0,\end{aligned} \quad (4.8)$$

with

$$C=3q/2a^4. \quad (4.9)$$

By standard perturbation theory, the probability per unit time $W_{m;m+\mu}$ of the transition that the quantum number of the central nuclear spin m changes by μ is

$$W_{m;m+\mu}=\textstyle\sum_k\sum_s\langle W(m,N_{ks}\to m+\mu,N_{ks}+1)+W(m,N_{ks}\to m+\mu,N_{ks}-1)\rangle_{\substack{\text{thermal average}\\ \text{over phonons}}}, \quad (4.10)$$

where

$$W(m,N_{ks}\to m+\mu,N_{ks}\pm1)=(2\pi/\hbar^2)|(m+\mu,N_{ks}\pm1|H'|m,N_{ks})|^2\times\delta(\omega_k{}^P\mp\mu\omega_0). \quad (4.11)$$

Using the matrix elements of H' and the relation

$$\textstyle\sum_s(\mathbf{A}_{\mu j}\cdot\mathbf{e}_{ks})(\mathbf{A}_{\mu j'}{}^*\cdot\mathbf{e}_{ks})=\mathbf{A}_{\mu j}\cdot\mathbf{A}_{\mu j'}{}^*, \quad (4.12)$$

we obtain

$$W_{m;m+\mu}=(\pi/MN\hbar)|Q_{\mu m}|^2\textstyle\sum_j\sum_{j'}\sum_k(\mathbf{A}_{\mu j}\cdot\mathbf{A}_{\mu j'}{}^*)\times\sin\mathbf{k}\cdot\mathbf{R}_j\sin\mathbf{k}\cdot\mathbf{R}_{j'}[(\bar{N}_k+1)\delta(\omega_k{}^P-\mu\omega_0)+\bar{N}_k\delta(\omega_k{}^P+\mu\omega_0)]/\omega_k{}^P, \quad (4.13)$$

where

$$Q_{\mu m}=(m+\mu|Q_\mu|m),\quad \bar{N}_k=1/(e^{\beta\hbar\omega_k{}^P}-1). \quad (4.14)$$

Expanding $\sin\mathbf{k}\cdot\mathbf{R}_j$ in powers of $\mathbf{k}\cdot\mathbf{R}_j$ and replacing the summation over $\mathbf{k}$ by an integral, we have

$$W_{m;m+\mu}=\frac{|Q_{\mu m}|^2M_\mu(\mu\omega_0)^3}{6\pi\hbar v^5d(1-e^{-\mu\beta\hbar\omega_0})}, \quad (4.15)$$

where

$$M_\mu=\textstyle\sum_j\sum_{j'}(\mathbf{R}_j\cdot\mathbf{R}_{j'})(\mathbf{A}_{\mu j}\cdot\mathbf{A}_{\mu j'}{}^*), \quad (4.16)$$

and

$$d=MN/V.$$

The nuclear spin-lattice relaxation time T_1 is given by

$$\frac{1}{T_1}=\frac{\sum_\mu\mu\langle mW_{m;m-\mu}\rangle}{\langle m^2\rangle-\langle m\rangle^2}, \quad (4.17)$$

where $\langle\ \rangle$ means the same operation as Eq. (2.22). Using Eq. (4.15), we obtain

$$\frac{1}{T_1}=\frac{A_0{}^3S^3B^2}{6\pi\hbar^4v^5d}\sum_\mu{}'\mu^4M_\mu\left[\frac{\langle\langle I^\zeta\tilde{Q}_\mu\tilde{Q}_{-\mu}\rangle\rangle}{e^{\mu\beta\hbar\omega_0}-1}-\frac{\langle\langle I^\zeta\tilde{Q}_{-\mu}\tilde{Q}_\mu\rangle\rangle}{1-e^{-\mu\beta\hbar\omega_0}}\right]\Big/[\langle\langle(I^\zeta)^2\rangle\rangle-\langle\langle I^\zeta\rangle\rangle^2], \quad (4.18)$$

where $\sum_\mu{}'$ means the summation over the positive values of μ and $\tilde{Q}_\mu$ are defined by

$$Q_\mu=B\tilde{Q}_\mu, \quad (4.19)$$

B being the nuclear quadrupole moment expressed by Eq. (4.3).

For the simple cubic lattice, the M_μ are calculated by Eq. (4.8)

$$M_1=16C^2,a^2\quad M_2=\tfrac{13}{2}C^2a^2. \quad (4.20)$$

Using these values in Eq. (4.18), we obtain

$$\frac{1}{T_1}=\frac{8(aBC)^2(A_0S)^3}{3\pi\hbar^4v^5d}F_4(T), \quad (4.21)$$

in which

$$F_4(T)=\left[\frac{\langle\langle I^\zeta\tilde{Q}_1\tilde{Q}_{-1}\rangle\rangle}{e^{\beta\hbar\omega_0}-1}-\frac{\langle\langle I^\zeta\tilde{Q}_{-1}\tilde{Q}_1\rangle\rangle}{1-e^{-\beta\hbar\omega_0}}+\frac{13}{2}\left(\frac{\langle\langle I^\zeta\tilde{Q}_2\tilde{Q}_{-2}\rangle\rangle}{e^{2\beta\hbar\omega_0}-1}-\frac{\langle\langle I^\zeta\tilde{Q}_{-2}\tilde{Q}_2\rangle\rangle}{1-e^{-2\beta\hbar\omega_0}}\right)\right]\Big/[\langle\langle(I^\zeta)^2\rangle\rangle-\langle\langle I^\zeta\rangle\rangle^2]. \quad (4.22)$$

The relaxation time obtained here is independent of the external field, because of the assumption (2.4). The $F_4(T)$-versus-T curve which is shown in Fig. 1 is similar to $F_1(T)$ in Sec. 2, that is, for $I=\frac{5}{2}$ $F_4(T)$ is proportional to T for $k_BT>4\hbar\omega_0$ and deviates slightly from the line for lower temperatures. The high-temperature value of $F_4(T)$ is expressed by $27\,[4I(I+1)-3]k_BT/10\hbar\omega_0$.

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[10] R. M. Sternheimer, Phys. Rev. **84**, 244 (1951); **95**, 736 (1954); **130**, 1423 (1963); R. M. Sternheimer and H. M. Foley, Phys. Rev. **102**, 731 (1956).

V. ISOTOPE EFFECT AND COMPARISON WITH EXPERIMENT

In order to compare with the experiment on the powder EuS,[1] in which the nuclear spin-lattice relaxation time was determined by measuring the transmission of polarized neutrons through the sample, we extend our results obtained in the previous sections to the case that there are two isotopes. We assume these isotopes have the same lattice vibrations and are distributed at random on the lattice sites. Specifying them by the superscripts a and b and introducing the projection operator P_j^ν $(\nu=a,b)$:

$$\begin{aligned} P_j^\nu &= 1, \quad \text{if the site } j \text{ contains a } \nu \text{ nucleus}, \\ &= 0, \quad \text{otherwise}, \end{aligned} \tag{5.1}$$

the unperturbed nuclear Hamiltonian H_0^N is written as

$$H_0^N = -\sum_j \sum_\nu P_j^\nu \hbar\omega_\nu I_{j\nu}^\zeta, \tag{5.2}$$

where

$$\hbar\omega_\nu = A_0^\nu S. \tag{5.3}$$

Since the relaxation process through the indirect nuclear spin interaction is not effective for a rare-earth atom as stated in Sec. 3, we consider two other mechanisms. In deriving the expressions of the relaxation time, we will assume that the cross relaxation time[11] between these two isotope systems is short compared to the spin-lattice relaxation time of the individual system, i.e., these two systems have a common spin temperature.

A. Relaxation to the Quasiphonon System

The perturbation H' is given by

$$H' = \tfrac{1}{2} \sum_j \sum_\nu P_j^\nu A_0^\nu (S_j^+ I_{j\nu}^- + S_j^- I_{j\nu}^+). \tag{5.4}$$

Using the transformation (1.4), (1.12), (2.7) and considering only the quasiphonons with the assumption (2.23), H' becomes

$$\begin{aligned} H' = \tfrac{1}{4}(S/N)^{1/2} \sum_j {\sum_k}' \sum_s \sum_{\mu=1,2} \sum_{\nu=a,b} P_j^\nu A_0^\nu e^{i v_{ks}} \\ \times [e^{i\mathbf{k}\cdot\mathbf{j}} - (-)^\mu e^{-i\mathbf{k}\cdot\mathbf{j}}](\omega_k^e/\omega_k^P)^{1/2} \\ \times \cos\Phi_{ks} (I_{j\nu}^+ e^{-i\phi_k} \cosh\tfrac{1}{2}\varphi_k - I_{j\nu}^- e^{i\phi_k} \sinh\tfrac{1}{2}\varphi_k) \\ \times [\xi_{ks}^{(\mu)} + (-)^\mu \xi_{ks}^{(\mu)*}] + \text{c.c.}, \end{aligned} \tag{5.5}$$

which is the extension of Eq. (2.16) to the case considering here. As in Sec. 2, we obtain the relaxation time T_1 for the case that the anisotropy energy can be neglected, i.e., under the condition (2.28), as follows:

$$\frac{1}{T_{1M}} = \frac{S^7 G_1^2 G_\alpha(H_0)}{8\pi\hbar^4 v^5 d (g\mu_B H_0)^2} \sum_{\nu=a,b} (A_0^\nu)^5 F_{1\nu}(T), \tag{5.6}$$

where

$$F_{1\nu}(T) = c_\nu \omega_\nu^2 Z_\nu \left[\frac{\langle\langle I_\nu^\zeta I_\nu^+ I_\nu^- \rangle\rangle_\nu}{e^{\beta\hbar\omega_\nu} - 1} - \frac{\langle\langle I_\nu^\zeta I_\nu^- I_\nu^+ \rangle\rangle_\nu}{1 - e^{-\beta\hbar\omega_\nu}} \right] \Big/ \sum_\nu c_\nu \omega_\nu^2 Z_\nu [\langle\langle (I_\nu^\zeta)^2 \rangle\rangle_\nu - \langle\langle I_\nu^\zeta \rangle\rangle_\nu^2], \tag{5.7}$$

which corresponds to Eq. (2.32). In Eq. (5.7) c_ν is the natural abundance of the isotope ν, i.e., the ensemble average of P^ν, and $\langle\langle\ \rangle\rangle_\nu$ means

$$\begin{aligned} \langle\langle O_\nu \rangle\rangle_\nu &= \mathrm{Tr} O_\nu e^{\beta\hbar\omega_\nu I_\nu^\zeta} / Z_\nu, \\ Z_\nu &= \mathrm{Tr} e^{\beta\hbar\omega_\nu I_\nu^\zeta}. \end{aligned} \tag{5.8}$$

B. Relaxation Through Nuclear Quadrupole Energy

The perturbation for the central nucleus is written as

$$\begin{aligned} H' &= \sum_{\nu=a,b} \sum_\mu P^\nu Q_\mu^\nu \sum_j \mathbf{A}_{\mu j} \cdot \mathbf{u}_{j0} \\ &= (\hbar/2MN)^{1/2} \sum_\nu \sum_\mu P^\nu Q_\mu^\nu \sum_j \sum_k \sum_s (\mathbf{A}_{\mu j} \cdot \mathbf{e}_{ks} / (\omega_k^P)^{1/2}) \\ &\quad \times [b_{ks}(e^{i\mathbf{k}\cdot\mathbf{j}} - 1) + b_{ks}^*(e^{-i\mathbf{k}\cdot\mathbf{j}} - 1)], \end{aligned} \tag{5.9}$$

where P^ν is the projection operator for the central nucleus and Q_μ^ν are given by

$$\begin{aligned} Q_0^\nu &= B_\nu [3(I_\nu^\zeta)^2 - I_\nu(I_\nu + 1)], \\ Q_{\pm 1}^\nu &= B_\nu [I_\nu^\zeta I_\nu^\pm + I_\nu^\pm I_\nu^\zeta]/2, \\ Q_{\pm 2}^\nu &= B_\nu (I_\nu^\pm)^2, \\ B_\nu &= eQ_\nu / I_\nu(2I_\nu - 1), \\ eQ_\nu &= (I_\nu I_\nu | Q_0^\nu | I_\nu I_\nu). \end{aligned} \tag{5.10}$$

These equations correspond to Eqs. (4.1)–(4.3). By the same method stated in Sec. 4, we obtain for the simple cubic lattice

$$\frac{1}{T_{1Q}} = \frac{8(aC)^2 S^3}{3\pi\hbar^4 v^5 d} \sum_{\nu=a,b} B_\nu^2 (A_0^\nu)^3 F_{4\nu}(T), \tag{5.11}$$

where

$$\begin{aligned} F_{4\nu}(T) = c_\nu \omega_\nu^2 Z_\nu \Bigg[& \frac{\langle\langle I_\nu^\zeta \tilde{Q}_1^\nu \tilde{Q}_{-1}^\nu \rangle\rangle_\nu}{e^{\beta\hbar\omega_\nu} - 1} - \frac{\langle\langle I_\nu^\zeta \tilde{Q}_{-1}^\nu \tilde{Q}_1^\nu \rangle\rangle_\nu}{1 - e^{-\beta\hbar\omega_\nu}} \\ & + \frac{13}{2} \left(\frac{\langle\langle I_\nu^\zeta \tilde{Q}_2^\nu \tilde{Q}_{-2}^\nu \rangle\rangle_\nu}{e^{2\beta\hbar\omega_\nu} - 1} - \frac{\langle\langle I_\nu^\zeta \tilde{Q}_{-2}^\nu \tilde{Q}_2^\nu \rangle\rangle_\nu}{1 - e^{-2\beta\hbar\omega_\nu}} \right) \Bigg] \Big/ \\ & \sum_\nu c_\nu \omega_\nu^2 Z_\nu [\langle\langle (I_\nu^\zeta)^2 \rangle\rangle_\nu - \langle\langle I_\nu^\zeta \rangle\rangle_\nu^2], \end{aligned} \tag{5.12}$$

with

$$\tilde{Q}_\mu^\nu = Q_\mu^\nu / B_\nu. \tag{5.13}$$

The Eq. (5.11) is the extension of Eq. (4.21).

To apply our results to the powder sample, it is necessary to take averages of Eq. (5.6) over all angles and the shapes of the single crystals composing the powder sample. We assume that the powder sample consists of a number of single crystals, each of which has, for simplicity, the sphere-shape with the crystal axes orientated at random with respect to the direction of the external field. In this case γ_3 determined by Eq.

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[11] A. Abragam and W. G. Proctor, Phys. Rev. **109**, 1441 (1958); N. Bloembergen, S. Shapiro, P. S. Pershan, and J. L. Artman, Phys. Rev. **114**, 445 (1959).

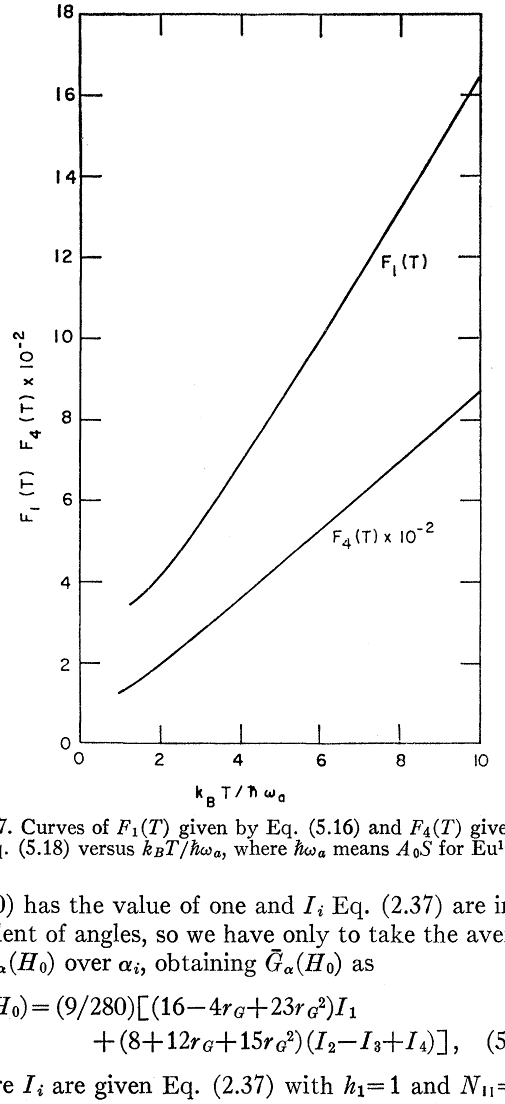

FIG. 7. Curves of $F_1(T)$ given by Eq. (5.16) and $F_4(T)$ given by Eq. (5.18) versus $k_BT/\hbar\omega_a$, where $\hbar\omega_a$ means A_0S for Eu^{151}.

(2.40) has the value of one and I_i Eq. (2.37) are independent of angles, so we have only to take the average of $G_\alpha(H_0)$ over α_i, obtaining $\bar{G}_\alpha(H_0)$ as

$$\bar{G}_\alpha(H_0) = (9/280)[(16-4r_G+23r_G^2)I_1 + (8+12r_G+15r_G^2)(I_2-I_3+I_4)], \quad (5.14)$$

where I_i are given Eq. (2.37) with $h_1=1$ and $N_{\parallel}=N_{\perp}=\frac{1}{3}$.

In EuS, the isotopes a and b correspond to Eu^{151} and Eu^{153} with $I_a=I_b=\frac{5}{2}$, and $S=\frac{7}{2}$. c_a and c_b are 0.478 and 0.522, respectively. Taking $A_0{}^a=0.298\times10^{-2}$ cm^{-1}, $A_0{}^b=0.144\times10^{-2}$ cm^{-1}, $d=5.7$ g/cm^3, $4\pi M_0=14$ kOe,[12] $Q_a=1.2\times10^{-24}$ cm^2, $Q_b=2.5\times10^{-24}$ cm^2,[13] lattice constant $a_0=2a=5.97A^0$,[14] $q_0=2e$ and $v=v_0\times10^{+5}$ cm/sec, we have from Eq. (5.6) with Eq. (5.14) and Eq. (5.11)

$$T_{1M}{}^{-1}=0.351\times10^{-6}(G_{11}{}^2/v_0{}^5) \times(4\pi M_0/H_0)^2\bar{G}_\alpha(H_0)F_1(T), \quad (5.15)$$

with

$$F_1(T)=F_{1a}(T)+2.66\times10^{-2}F_{1b}(T), \quad (5.16)$$

and

$$T_{1Q}{}^{-1}=1.05\times10^{-12}(\gamma^2/v_0{}^5)F_4(T), \quad (5.17)$$

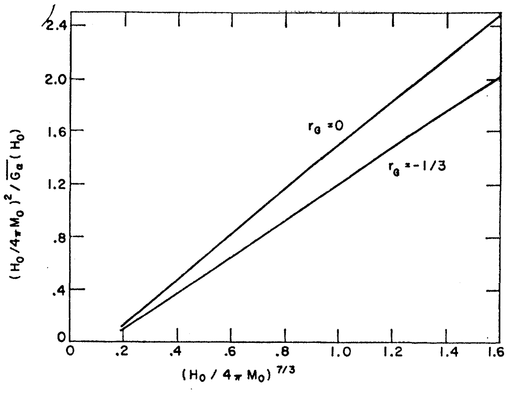

FIG. 8. External field dependence of T_{1M} given by Eq. (5.15).

with

$$F_4(T)=F_{4a}(T)+0.49F_{4b}(T). \quad (5.18)$$

In Eq. (5.15) we use G_{11} instead of G_1 related by Eq. (2.48) and G_{11} measures in cm^{-1}. According to the experiment,[1] T_1 is expressed by

$$T_1 \sim H_0{}^3/T^2, \quad (5.19)$$

for the ranges of $T=0.05$–0.15°K and $H_0=7$–17 kOe and has the value of several hours at 0.05°K and 17 kOe. $F_1(T)$ and $F_4(T)$ given by Eqs. (5.16) and (5.18) are shown in Fig. 7 as the functions of $k_BT/\hbar\omega_a$, where $\hbar\omega_a$ corresponds to 0.015°K. $F_1(T)$ is proportional to T for $T>0.1$°K and convex towards the T axis. $F_4(T)$ has the similar behavior as $F_1(T)$. The field-dependent part of T_{1M}, namely $(H_0/4\pi M_0)^2/\bar{G}_\alpha(H_0)$, is shown in Fig. 8 as the function of $(H_0/4\pi M_0)^{7/3}$, $4\pi M_0$ being 14 kOe. We see the T_{1M} is proportional $H_0{}^{7/3}$ in the range of $H_0=7$–17 kOe. The values of T_{1M} and T_{1Q} at 0.05°K and 17 kOe are

$$\frac{1}{T_{1M}}=\frac{G_{11}{}^2}{v_0{}^5}\times10^{-6}\begin{cases}0.855 & \text{for } r_G=0,\\ 1.06 & \text{for } r_G=-\frac{1}{3},\end{cases} \quad (5.20)$$

and

$$T_{1Q}{}^{-1}=3.17\times10^{-10}(\gamma^2/v_0{}^5). \quad (5.21)$$

Taking $v_0=2$, which may be a reasonable value from $\Theta_D=204$°K, T_{1M} and T_{1Q} are the same order as the experiment when $G_{11}=20$ and $\gamma=10^3$. Since the experimental relaxation time depends strongly on the external field, γ may have a value of 10^2 or less.[15] That the value of G_{11} is of the order 10 is reasonable for a rare earth ion. The agreement of the calculated T_1 with the experimental one seems to be fairly good.

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[12] S. H. Charap and E. L. Boyd, Phys. Rev. **133**, A811 (1964).

[13] For example, G. E. Pake, *Solid State Physics* (John Wiley & Sons, Inc., New York, 1956).

[14] G. Busch, P. Junod, M. Risi, and O. Vogt, in *Proceedings of the International Conference on the Physics of Semiconductors, Exeter, 1962* (The Institute of Physics and The Physical Society, London, 1962), p. 727. S. Van Houten, Phys. Letters **2**, 215 (1962).

[15] R. M. Sternheimer, Phys. Rev. **132**, 1637 (1963).