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LINE WIDTH IN ANTIFERROMAGNETIC RESONANCE

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Relaxation of the uniform precession of the magnetization of an antiferromagnetic substance due to the interaction between the uniform precession and spin waves is considered. The model of localized spins with exchange interaction was employed in the calculations. Terms of the fourth order in the creation and annihilation operators were taken into account in the Hamiltonian. The relaxation time for uniform precession at low temperatures has been derived.

1. INTRODUCTION

WHILE the theory of line width in ferromagnetic resonance has progressed considerably,[1,2] the line width in antiferromagnetic resonance has not to date received any explanation that is at all satisfactory.

The object of the present paper consists of evaluating the linewidth (or the relaxation time associated with it) resulting from the interaction of the uniform precession of the magnetization with spin waves. A similar interaction in the case of ferromagnetic substances has been discussed in the papers by Akhiezer and co-workers.[2] However, the line width in the case of ferromagnetic substances turns out to be very small. This is associated with the fact that the interaction of spin waves with uniform precession (in the ferromagnetic case) is due to comparatively small relativistic effects, while the exchange interaction does not affect uniform precession. In the case of an antiferromagnetic substance the exchange interaction turns out to be the dominant one in the calculation of the relaxation brought about by the interaction between the uniform precession and spin waves, and, as will be seen from our estimates, leads to an appreciable line width.

2. THE INTERACTION HAMILTONIAN

We shall write the Hamiltonian for a system of spins in an antiferromagnetic substance in the following form* (cf., for example, [3])

$$\mathcal{H} = 2J \sum_{\langle lm\rangle} \mathbf{S}_l \mathbf{S}_m + g\beta H_A \Big(\sum_l S_l^z - \sum_m S_m^z \Big). \qquad (1)$$

Here $\mathbf{S}_l$ and $\mathbf{S}_m$ are spin operators of the first and the second sublattices respectively, $\langle lm \rangle$ denotes summation over the nearest neighbors, the exchange integral is $J > 0$, H_A is the effective anisotropic field (for simplicity we shall assume that there is no external magnetic field).

Further, we express $\mathbf{S}_l$ and $\mathbf{S}_m$ in the usual manner in terms of the spin deviation operators, restricting ourselves to terms of order not higher than the third:

$$S_l^+ = (2S)^{1/2} (1 - a_l^* a_l / 4S)\, a_l, \quad S_l^- = (2S)^{1/2} a_l^* (1 - a_l^* a_l / 4S),$$
$$S_l^z = S - a_l^* a_l, \quad S_m^+ = (2S)^{1/2} b_m^* (1 - b_m^* b_m / 4S),$$
$$S_m^- = (2S)^{1/2} (1 - b_m^* b_m / 4S)\, b_m, \quad S_m^z = - S + b_m^* b_m,$$
$$S_{lm}^{\pm} = S_{lm}^x \pm i S_{lm}^y. \qquad (2)$$

After these operators have been substituted into (1) the Hamiltonian can be separated into a sum of second- and fourth-order terms in the operators a and b. The second-order terms are subsequently diagonalized. They represent the unperturbed Hamiltonian for the spin waves. The fourth-order terms represent the interaction energy of the spin waves:

$$\mathcal{H}' = - 2J \sum_{\langle lm\rangle} \Big\{ \tfrac{1}{4} (a_l^* a_l a_l b_m + a_l b_m^* b_m b_m + b_m^* a_l^* a_l^* a_l + b_m^* b_m^* b_m a_l^*) + a_l^* a_l b_m^* b_m \Big\}.$$

We next transform to the Fourier components of the operators a and b:

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*We note that if the energy associated with the anisotropy is taken into account in a more general form (cf., for example,[2]) than in (1), then this leads to the appearance in (4) of a number of additional terms. However, it can be easily seen that some of these terms describe processes for which the law of conservation of energy does not hold, while the others have no effect on the uniform precession. From this it is clear that a more complete method of taking anisotropy into account, as has been done, in particular, by Akhiezer et al,[2] turns out to be superfluous in our calculation.

$$a_l = (2/N)^{1/2}\sum_k a_k e^{-i\mathbf{kl}}, \quad \overset{*}{a}_l = (2/N)^{1/2}\sum_k \overset{*}{a}_k e^{+i\mathbf{kl}},$$

$$b_m = (2/N)^{1/2}\sum_k b_k e^{i\mathbf{km}}, \quad \overset{*}{b}_m = (2/N)^{1/2}\sum_k \overset{*}{b}_k e^{-i\mathbf{km}}.$$

The Hamiltonian $\mathcal{H}'$ can be put in the form

$$\mathcal{H}' = -\frac{Jz}{N}\sum_{1,2,3,4}\{\gamma_4[\overset{*}{b}_1 b_2 b_3 a_4 + \overset{*}{b}_3\overset{*}{b}_2 b_1\overset{*}{a}_4 + \overset{*}{a}_1 a_2 a_3 b_4$$

$$+ \overset{*}{a}_3\overset{*}{a}_2 a_1 b_4] + 4\gamma_{(\mathbf{k}_1-\mathbf{k}_2)}\overset{*}{a}_1 a_2\overset{*}{b}_3 b_4\}\,\Delta(\mathbf{k}_1 + \mathbf{k}_4 - \mathbf{k}_2 - \mathbf{k}_3),$$

$$\gamma_k = \frac{1}{z}\sum_{\langle lm\rangle} e^{i\mathbf{k}(\mathbf{l}-\mathbf{m})},$$

where 2z is the number of nearest spins.

We diagonalize the second order terms by introducing new variables:

$$a_k = \alpha_k \operatorname{ch}\theta_k - \overset{*}{\beta}_k \operatorname{sh}\theta_k, \quad b_k = -\overset{*}{\alpha}_k \operatorname{sh}\theta_k + \beta_k \operatorname{ch}\theta_k, \tag{3}$$

[*]

where $\tanh 2\theta_k = \gamma_k/D$; $D = 1 + g\beta H_A/zJS$. This operation, analogous to the Holstein-Primakoff[4] transformation brings the interaction Hamiltonian into the form

$$\mathcal{H}' = \frac{Jz}{N}\sum_{1,2,3,4}\{\Phi_{1(23)4}\alpha_1\overset{*}{\alpha}_2\overset{*}{\alpha}_3\alpha_4 + \Psi_{1(23)4}\beta_1\overset{*}{\beta}_2\overset{*}{\beta}_3\beta_4$$

$$+ \lambda_{1234}\overset{*}{\alpha}_1\alpha_2\overset{*}{\beta}_3\beta_4 - \Phi_{1234}\overset{*}{\alpha}_1\alpha_2\alpha_3\beta_4$$

$$- \Psi_{1234}\overset{*}{\beta}_1\beta_2\beta_3\alpha_4\}\,\Delta(\mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3 + \mathbf{k}_4). \tag{4}$$

Here we have omitted terms of the type $\alpha_1\alpha_2\beta_1\beta_2$ which cannot correspond to processes in which the energy of the spin waves is conserved. In formula (4) we have introduced the notation

$$\lambda_{1234} = \lambda'_{(1)2(3)4} + \lambda''_{42(31)} + \lambda'_{13(24)} + \lambda''_{(4)3(2)1}.$$

We shall not in future need the specific form of the coefficients Φ and Ψ, and, therefore, we do not give them here. We merely note that the omitted coefficients do not exceed λ in order of magnitude.

The quantities $\lambda_{(1)2(3)4}$ and $\lambda'_{12(34)}$ are of the form

$$\lambda'_{(1)2(3)4} = \gamma_4(\operatorname{sh}\theta_1\operatorname{sh}\theta_2\operatorname{ch}\theta_3\operatorname{sh}\theta_4 + \operatorname{ch}\theta_1\operatorname{ch}\theta_2\operatorname{sh}\theta_3\operatorname{ch}\theta_4)$$
$$- 2\gamma_{1-2}\operatorname{ch}\theta_1\operatorname{ch}\theta_2\operatorname{ch}\theta_3\operatorname{ch}\theta_4,$$

$$\lambda'_{12(3)(4)} = \gamma_4(\operatorname{sh}\theta_1\operatorname{ch}\theta_2\operatorname{sh}\theta_3\operatorname{sh}\theta_4 + \operatorname{ch}\theta_1\operatorname{sh}\theta_2\operatorname{ch}\theta_3\operatorname{ch}\theta_4)$$
$$- 2\gamma_{1-2}\operatorname{ch}\theta_1\operatorname{ch}\theta_2\operatorname{sh}\theta_3\operatorname{ch}\theta_4,$$

where $\gamma_{1-2} \equiv \gamma_{\mathbf{k}_1-\mathbf{k}_2}$; λ'' are obtained from the corresponding λ' by the substitutions $\cosh\theta_i \to \sinh\theta_i$ and $\sinh\theta_i \to \cosh\theta_i$.

3. RELAXATION PROCESSES

The Hamiltonian (4) is responsible for the interaction of the spin waves. We note that this Hamiltonian satisfies Van Hove's condition of diagonal singularity.[5] This enables us in discussing the relaxation processes to utilize the ordinary method involving probabilities per unit time. We shall now consider the relaxation of the uniform precession of the magnetization which is, in particular, represented by the operators α_0 and α_0^*. In carrying this out we assume that the spin waves are in the state of thermodynamic equilibrium at the temperature T. We obtain the probabilities for the occurrence of processes involving a change in the number of spin waves and containing the operator α_k:

$$W(n_1, n_2, n_3, n_4 \to n_1 - 1, n_2 + 1, n_3 + 1, n_4 - 1)$$

$$= \frac{2\pi}{\hbar}\frac{\varepsilon_{\mathbf{ex}}^2}{4N^2}|\Phi_{1(23)4} + \Phi_{1(32)4} + \Phi_{4(23)1} + \Phi_{4(32)1} + \Phi_{3(14)2}$$

$$+ \Phi_{2(14)3} + \Phi_{2(41)3} + \Phi_{3(41)2}|^2\,\Delta(\mathbf{k}_1 + \mathbf{k}_4 - \mathbf{k}_2 - \mathbf{k}_3)\,\delta(\varepsilon_1$$

$$+ \varepsilon_4 - \varepsilon_2 - \varepsilon_3)(n_2 + 1)(n_3 + 1)n_1 n_4, \tag{5}$$

$$W(n_1, n_2, n_3, n'_4 \to n_1 + 1, n_2 - 1, n_3 - 1, n'_4 - 1)$$

$$= \frac{2\pi}{\hbar}\frac{\varepsilon_{\mathbf{ex}}^2}{4N^2}|\Phi_{1234}|^2\,\Delta(\mathbf{k}_1 + \mathbf{k}_4 - \mathbf{k}_2 - \mathbf{k}_3)\,\delta(\varepsilon_1 - \varepsilon_2$$

$$- \varepsilon_3 - \varepsilon'_4)(n_1 + 1)n_2 n_3 n'_4, \tag{6}$$

$$W(n_1, n_2, n'_3, n'_4 \to n_1 + 1, n_2 - 1, n'_3 + 1, n'_4 - 1)$$

$$= \frac{2\pi}{\hbar}\frac{\varepsilon_{\mathbf{ex}}^2}{4N^2}|\lambda_{1234} + \lambda_{2143}|^2\,\Delta(\mathbf{k}_1 + \mathbf{k}_4 - \mathbf{k}_2 - \mathbf{k}_3)\,\delta(\varepsilon_1$$

$$- \varepsilon_2 - \varepsilon'_3 - \varepsilon'_4)(n_1 + 1)n_2(n'_3 + 1)n'_4. \tag{7}$$

Here $\epsilon_{\mathrm{ex}} = 2Jz$, the primed quantities refer to spin waves described by the operators β; n_k and n'_k are the mean numbers of α- and β-spin waves in the state of momentum **k**; ϵ_k and ϵ'_k are the energies of magnons of momentum **k**.

In the rest of the paper we restrict ourselves to the low temperature case

$$kT < \varepsilon_0. \tag{8}$$

In this case we can neglect processes of type (5) since the corresponding matrix elements vanish. The contribution made by processes (6) will be small, since in order to satisfy the law of conservation of energy it is necessary that one of the ϵ_k should be greater than $3\epsilon_0$, and this corresponds to a relatively small value of n_k if condition (8) is satisfied. Thus, the principal contribution to the line width is made by processes of type (7). This leads to the following equation for the relaxation of the uniform precession of the magnetization:

$$\dot{n}_0 = \sum_{234}[W(n_0, n_2, n'_3, n'_4 \to n_0 + 1, n_2 - 1, n'_3 - 1, n'_4 - 1)$$

$$- W(n_0, n_2, n'_3, n'_4 \to n_0 - 1, n_2 + 1, n'_3 - 1, n'_4 + 1)]$$

$$+ \sum_{134}[W(n_1 - 1, n_0, n'_3 - 1, n'_4 + 1 \to n_1, n_0 + 1, n'_3, n'_4)$$

$$- W(n_1, n_0, n'_3, n'_4 \to n_1 + 1, n_0 - 1, n'_3 + 1, n'_4 - 1)]. \tag{9}$$

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[*] ch = cosh, sh = sinh.

Here we have everywhere taken $n_{k\neq 0} = \bar{n}_k = 1/[\exp(\epsilon_k/kT) - 1]$.

Equation (9) can be written in the form

$$\dot{n}_0 = -\lambda(n_0 - \bar{n}_0).$$

λ^{-1} is equal to the average relaxation time of uniform precession. It can be easily obtained from (7) and (9) that

$$\lambda = \frac{\pi}{\hbar}\frac{\varepsilon_{\mathbf{ex}}^2}{N^2}\sum_{234}|\lambda_{0234} + \lambda_{2043}|^2\,\Delta(\mathbf{k}_4 - \mathbf{k}_3 - \mathbf{k}_2)\,\delta(\varepsilon_0 - \varepsilon_2 + \varepsilon_3' - \varepsilon_4')(n_3' - n_2n_4' + n_2n_3' + n_3'n_4'). \qquad (10)$$

By utilizing the expression for $\lambda_{ik l m}$, we obtain

$$|\lambda_{0234} + \lambda_{2043}|^2 = D^2\varepsilon_{\mathbf{ex}}^4|\mathbf{k}_1\mathbf{k}_2 l^2|^2/\varepsilon_0\varepsilon_2\varepsilon_3\varepsilon_4, \qquad (11)$$

where $\epsilon_k^2 = \epsilon_{ex}^2(h_A + k^2l^2/3)$; $h_A = g\beta H_A/zJS$, l is the lattice constant, and we have taken into account the fact that $\mathbf{k}\cdot\mathbf{l} \ll 1$.

We carry out the summation over $\mathbf{k}_4$, replace the summation over $\mathbf{k}_2$ and $\mathbf{k}_3$ by integration, substitute (11), integrate over the angles θ_2, φ_1, φ_2, and as a result obtain

$$\lambda = \frac{27}{\hbar(2\pi)^3\varepsilon_{\mathbf{ex}}^4}\int\frac{(\varepsilon_2^2 - \varepsilon_0^2)^{3/2}(\varepsilon_3^2 - \varepsilon_0^2)^{3/2}\cos^2\theta\,d\theta\sin\theta}{\varepsilon_0\varepsilon_{3+2}} \times e^{-\varepsilon_3'/kT}\,\delta(\varepsilon_0 - \varepsilon_2 + \varepsilon_3 - \varepsilon_{3+2})\,d\varepsilon_3\,d\varepsilon_2. \qquad (12)$$

Here $\epsilon_{3+2} \equiv \epsilon_{\mathbf{k}_3+\mathbf{k}_2}$ and $D^2 \approx 1$. In going over from formula (10) to formula (12) we have taken into account condition (8), and we have retained only the term $n_3' \approx \exp(-\epsilon_3/kT)$. Integration over ϵ_2 reduces, as follows from the presence of the δ-function, to the replacement

$$\varepsilon_2 = \varepsilon_0\frac{\varepsilon_3 + \varepsilon_0 + \cos^2\theta(\varepsilon_3 - \varepsilon_0)}{\varepsilon_3 + \varepsilon_0 - \cos^2\theta(\varepsilon_3 - \varepsilon_0)},$$

$$\varepsilon_{3+2} = \frac{\varepsilon_3(\varepsilon_0 + \varepsilon_3)\sin^2\theta + 2\varepsilon_0^2\cos^2\theta}{\varepsilon_3 + \varepsilon_0 - \cos^2\theta(\varepsilon_3 - \varepsilon_0)}.$$

From the condition $\epsilon_{2+3} < \epsilon_3$ (since $\epsilon_0 < \epsilon_2$) we obtain

$$\pi/2 < \theta < \pi.$$

After integrating over θ and ϵ_3 we finally obtain

$$\frac{\lambda}{\omega_0} = h_A^2\frac{e^{-\varkappa_0}}{\varkappa_0^5}(16\varkappa_0^4 + 30\varkappa_0^3 + 46\varkappa_0^2 + 54\varkappa_0 + 37),$$

where $\kappa_0 = (\epsilon_0/kT)$, $h_A = g\beta H_A/zJS = H_A/H_E$, H_E is the intensity of the "exchange force field," $\omega_0 = \epsilon_0/\hbar$ is the antiferromagnetic resonance frequency.

We give estimates of λ for the case of MnF_2. In this case[6] $H_A = 9 \times 10^3$ gauss; $H_E = 6 \times 10^5$ gauss, and at $T = 6°K$, $\lambda = 160$ gauss, at $T = 4°K$, $\lambda = 12$ gauss, i.e., even at such low temperatures considerable broadening is present. It is obvious that at higher temperatures the line width increases.

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