

*RELAXATION OF THE MAGNETIC MOMENT IN AN ANTIFERROMAGNET WITH
ANISOTROPY OF THE "EASY PLANE" TYPE*

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The relaxation time for the magnetic moment of an antiferromagnet is calculated for the case when the nonequilibrium magnetic moment is perpendicular or parallel to the magnetic field in an "easy plane." In contrast to a uniaxial antiferromagnet, the main contribution to the relaxation at not very low temperatures is from processes involving three spin waves. The antiferromagnetic resonance (AFMR) line width due to these processes may depend strongly on the field if the antiferromagnet is weakly ferromagnetic. The results obtained are also applicable to the case of AFMR in antiferromagnets with "spin-flopped" sublattices.

AS Urushadze^[1] has shown, in an antiferromagnet with anisotropy of the "easy axis" type (MnF_2 , etc.), relaxation of the magnetic moment caused by spin-spin interactions proceeds principally via processes involving four spin waves; three-magnon processes are not possible because the conservation laws of energy and momentum are not fulfilled for them. This is due to the spin wave spectrum characteristic of these antiferromagnets.

In antiferromagnets with anisotropy of the "easy plane" type ($MnCO_3$, MnO , etc.) the situation is different. The spin wave spectrum of these substances consists of two branches:

$$\epsilon_{1k} = [\epsilon_{10}^2 + \Theta_c^2(ak)^2]^{1/2}, \quad \epsilon_{2k} = [\epsilon_{20}^2 + \Theta_c^2(ak)^2]^{1/2}, \quad (1)$$

where ϵ_{sk} is the energy of the spin wave with wave vector k ($s = 1, 2$); Θ_c is of the order of the Néel temperature T_N ; a is the lattice constant; ϵ_{10}/\hbar and ϵ_{20}/\hbar are the lower and upper frequencies of antiferromagnetic resonance (AFMR), and in moderate external fields ϵ_{10} is usually much less than ϵ_{20} . For this kind of spectrum three-magnon processes are possible: the spin wave ($k = 0, \epsilon_{10}$) that determines the transverse, non-equilibrium moment (see below) can disappear by uniting with the wave ($k_\alpha; \epsilon_{1k_\alpha}$) and transforming into the wave ($k_\alpha; \epsilon_{2k_\alpha}$). The magnitude of k_α is determined by the condition $\epsilon_{10} + \epsilon_{1k_\alpha} = \epsilon_{2k_\alpha}$ which gives

$$\begin{aligned} \Theta_c a k_\alpha &= \epsilon_{20} (\epsilon_{20}^2 - 4\epsilon_{10}^2)^{1/2} / 2\epsilon_{10}, \\ \epsilon_{2k_\alpha} &= \epsilon_{20}^2 / 2\epsilon_{10}, \quad \epsilon_{1k_\alpha} = (\epsilon_{20}^2 - 2\epsilon_{10}^2) / 2\epsilon_{10}. \end{aligned} \quad (2)$$

Estimating the corresponding wavelength $\lambda_\alpha = 2\pi/k_\alpha$, for example for $CoCO_3$ in medium fields

($H_0 \sim 5$ kOe),^[2] we obtain $\lambda_\alpha \approx 30a$. The calculation of the relaxation time τ , which, because of the relation $ak_\alpha \ll 1$, can be done by the spin wave theory,^[3] is of great interest, since in the indicated antiferromagnets $\Delta\omega \sim \tau^{-1}$ is more easily checked experimentally than in the usual uniaxial ones ($\Delta\omega$ is the width of the AFMR line).

Following the method used by Urushadze, we shall write the magnetic part of the Hamiltonian of the antiferromagnet with "easy plane" anisotropy for $T \ll T_N$ in the form

$$\begin{aligned} \mathcal{H} = \int dV &\left[\gamma M_1 M_2 + b_1 (M_{1z}^2 + M_{2z}^2) + 2b_2 M_{1z} M_{2z} \right. \\ &+ 2\beta (M_{1x} M_{1y} - M_{1y} M_{2x}) - (M_1 + M_2) H_0 \\ &\left. + \frac{\alpha}{2} \left(\frac{\partial M_{1i}}{\partial x_k} \right)^2 + \frac{\alpha}{2} \left(\frac{\partial M_{2i}}{\partial x_k} \right)^2 + \alpha_{12} \frac{\partial M_{1i}}{\partial x_k} \frac{\partial M_{2i}}{\partial x_k} \right]. \end{aligned} \quad (3)$$

Here M_1 and M_2 are the magnetic moments of the sublattices ($|M_1| = |M_2| = M_0$); H_0 is the external magnetic field; γ , α , α_{12} are the exchange interaction constants ($\gamma > 0$; $\alpha - \alpha_{12} > 0$); b_1 and b_2 are the anisotropy constants ($b_1 - b_2 > 0$, since in the ground state, the sublattice moments lie in the basal plane). For greater generality (application to antiferromagnets with weak ferromagnetism), the Dzyaloshinskii interaction (with the constant β) is included in the Hamiltonian in the form in which it exists in carbonates of the transition metals.^[4] The anisotropy in the basal plane and the field of the spin waves are not taken into account for the sake of simplicity. The field H_0 is applied in the basal plane xy (along the x axis); this is the most interesting case.

The configuration of the moments in the ground state is determined by the following formulas^[5]:

$$M_{1x} = M_{2x} \approx M_0 \psi,$$

$$M_{1y} = M_{2y} \approx M_0 (1 - \psi^2/2),$$

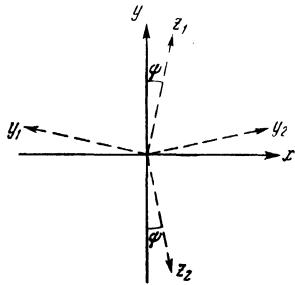
$$M_{1z} = M_{2z} = 0,$$

where $\psi = (H_0 + H_D)/2H_E$,

$$H_D \equiv 2\beta M_0,$$

$$H_E \equiv \gamma M_0 \quad (\psi \ll 1).$$

To transform to operators of second quantization we introduce supplementary orthogonal coordinate systems^[6] $x_S y_S z_S$ ($s = 1, 2$), which are oriented relative to the crystallographic system xyz in the fashion indicated in the figure so that



the axes z_1 and z_2 are the axes of quantization for the first and second sublattices. In these supplementary coordinate systems, the operators a_s^+ and a_s are defined in the usual way:

$$m_s^+ = M_{sx_s} + iM_{sy_s} = \sqrt{2\mu M_0} (a_s^+ - \mu a_s^+ a_s^+ a_s / 4M_0),$$

$$m_s^- = M_{sx_s} - iM_{sy_s} = \sqrt{2\mu M_0} (a_s - \mu a_s^+ a_s a_s^+ / 4M_0),$$

$$m_{sz_s} = M_{sz_s} - M_0 = -\mu a_s^+ a_s.$$

Then we express M_S via the components of the moments of the sublattices in the supplementary systems:

$$M_{1x} \approx \psi M_{1z_1} - (1 - \psi^2/2) M_{1y_1},$$

$$M_{2x} \approx \psi M_{2z_2} + (1 - \psi^2/2) M_{2y_2},$$

$$M_{1y} \approx (1 - \psi^2/2) M_{1z_1} + \psi M_{1y_1},$$

$$M_{2y} \approx -(1 - \psi^2/2) M_{2z_2} + \psi M_{2y_2},$$

$$M_{1z} = -M_{1x_1},$$

$$M_{2z} = -M_{2x_2};$$

we expand the operators $a_s^+(\mathbf{r})$ and $a_s(\mathbf{r})$ in Fourier series and transform the Hamiltonian (3) into the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{int}^{(3)} + \mathcal{H}_{int}^{(4)}, \quad (4)$$

$$\begin{aligned} \mathcal{H}_0 = & \sum_{\mathbf{k}} \left\{ \frac{1}{2} A_{\mathbf{k}} a_{1\mathbf{k}}^+ a_{1\mathbf{k}} + \frac{1}{2} A_{\mathbf{k}} a_{2\mathbf{k}}^+ a_{2\mathbf{k}} + B_{\mathbf{k}} a_{1\mathbf{k}} a_{2-\mathbf{k}} + C a_{1\mathbf{k}}^+ a_{2\mathbf{k}} + \right. \\ & \left. + \frac{1}{2} D a_{1\mathbf{k}}^+ a_{1-\mathbf{k}}^+ + \frac{1}{2} D a_{2\mathbf{k}}^+ a_{2-\mathbf{k}}^+ \right\} + \text{Herm. conj.}, \end{aligned} \quad (5)$$

$$\begin{aligned} \mathcal{H}_{int}^{(3)} = & i\mu H_0 \sqrt{\mu/2M_0 V} \sum_{\mathbf{k}_1 \mathbf{k}_2 \mathbf{k}_3} (a_{1\mathbf{k}_1}^+ a_{1\mathbf{k}_2} a_{2\mathbf{k}_3}^+ - a_{2\mathbf{k}_1}^+ a_{2\mathbf{k}_2} a_{1\mathbf{k}_3}^+) \\ & \times \Delta(\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3) + \text{Herm. conj.}, \end{aligned} \quad (6)$$

where the following symbols have been introduced:

$$A_{\mathbf{k}} = \mu M_0 (\gamma + b_1 + 2\beta\psi + \alpha k^2);$$

$$B_{\mathbf{k}} = \mu M_0 (\gamma + b_2 + 2\beta\psi - \gamma\psi^2 + \alpha_{12}k^2),$$

$$C = \mu M_0 (b_2 + \gamma\psi^2 - 2\beta\psi),$$

$$D = \mu M_0 b_1.$$

The amplitude of the Hamiltonian $\mathcal{H}_{int}^{(3)}$ is determined only by the external magnetic field, without involving the quantities b_1 , b_2 , and β : the third-order terms in a_s^+ , a_s appear because of the presence of the tilt angle ψ of the sublattices as well as the Dzyaloshinskii interaction, and as a result $\mathcal{H}_{int}^{(3)}$ turns out to be proportional to the combination $(\gamma\psi - \beta) \sim H_0$. The Hamiltonian $\mathcal{H}_{int}^{(4)}$ incorporates terms containing products of four operators $a_{s\mathbf{k}}^+$, $a_{s\mathbf{k}}$, i.e., it describes processes involving four spin waves. Of these, those which are caused by exchange interaction, as usual, do not contribute to the relaxation of the magnetic moment, since the Hamiltonian of the exchange interaction commutes with the total magnetic moment of the body. Terms of the fourth order proportional to the constants b_1 and b_2 give a contribution to the relaxation similar to that calculated by Urushadze, and they must be considered at low H_0 and T . The part of the Hamiltonian (3) that pertains to the Dzyaloshinskii interaction does not commute with the total magnetic moment; however, as calculation shows, its contribution to relaxation by means of four-magnon processes turns out to be of the order of H_A/H_E times the contribution determined by the anisotropy energy.

To diagonalize the Hamiltonian (5) we transform from the operators $a_{s\mathbf{k}}^+$, $a_{s\mathbf{k}}$ to the operators $c_{s\mathbf{k}}^+$, $c_{s\mathbf{k}}$ (see Eq. (8) in [1]), and, using the equation of motion $i\hbar \dot{a}_{s\mathbf{k}} = [\mathcal{H}_0, a_{s\mathbf{k}}]$ we obtain the dispersion law:

$$\varepsilon_{1,2} = [A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2 + C^2 - D^2 \pm 2(A_{\mathbf{k}}C - B_{\mathbf{k}}D)]^{1/2}, \quad (7)$$

i.e., we obtain Eq. (1), with

$$\varepsilon_{10} = \mu \sqrt{H_0(H_0 + H_D)},$$

$$\varepsilon_{20} = \mu \sqrt{H_{AE}^2 + H_D(H_0 + H_D)},$$

$$H_{AE}^2 = 2H_A H_E, \quad H_A = 2(b_1 - b_2) M_0,$$

$$\Theta_c^2 = 2\gamma\mu^2 M_0^2 (\alpha - \alpha_{12})/a^2. \quad (1')$$

A calculation of the amplitudes U_{sr} , V_{sr} (see [1]) gives

$$\begin{aligned}
U_{11} &= U_{21}, \quad V_{21} = V_{11} = -P_k U_{21}, \\
U_{21} &= -U_{12}, \quad V_{12} = -V_{22} = Q_k U_{12}, \\
|U_{21}|^2 &= \frac{1}{2}(1 - P_k^2)^{-1}, \quad |U_{12}|^2 = \frac{1}{2}(1 - Q_k^2)^{-1}, \\
P_k &\equiv (B_k + D)/(A_k + \varepsilon_{1k} + C), \\
Q_k &\equiv (B_k - D)/(A_k + \varepsilon_{2k} - C).
\end{aligned} \tag{8}$$

The interaction Hamiltonian $\mathcal{H}_{int}^{(3)}$ can now be expressed in terms of c_{Sk}^+ , c_{Sk} :

$$\mathcal{H}_{int}^{(3)} = \sum_{k_1 k_2 k_3} \Phi_{12;3} c_{1k_1}^+ c_{1k_2}^+ c_{2k_3} \Delta(k_1 + k_2 - k_3) + \text{Herm. conj.}$$

$$\Phi_{12;3} = i\mu H_0 \sqrt{2\mu/M_0 V} (1 - P_{k_1} P_{k_2} Q_{k_3}) U_{21k_1}^+ U_{21k_2}^+ U_{12k_3}. \tag{9}$$

Since low-frequency oscillations of the magnetic moment are easier to excite experimentally (AFMR at frequency ϵ_{10}/\hbar), we shall at first be interested in the mean value of the square of the component of the magnetic moment that is perpendicular to the magnetic field H_0 :¹⁾

$$\begin{aligned}
\langle \mathfrak{M}_y^2 + \mathfrak{M}_z^2 \rangle &= \left\langle \left[\int (M_{1y} + M_{2y}) dV \right]^2 \right\rangle \\
&+ \left\langle \left[\int (M_{1z} + M_{2z}) dV \right]^2 \right\rangle = 2(1 - \psi^2) \mu_{10}^{\text{eff}} M_0 V n_{10}; \\
\mu_{10}^{\text{eff}} &= \mu (1 - P_0)/(1 + P_0).
\end{aligned}$$

Thus the relaxation of the transverse magnetic moment is determined by the change in the number n_{10} of spin waves in the lower branch with wave vector $\mathbf{k} = 0$.

In calculating the time τ_{\perp} of this relaxation we shall take account only of processes involving three spin waves, since the four-magnon collisions give a contribution that is an order of magnitude smaller. Then, starting from Eq. (9) it is possible to obtain the following kinetic equation for n_{10} :

$$\begin{aligned}
\dot{n}_{10} &= \mathcal{L}_{10} \{n_1, n_2\}; \\
\mathcal{L}_{10} \{n_1, n_2\} &= \frac{8\pi}{\hbar} \sum_{k_2 k_3} |\Phi_{0k_2; k_3}|^2 [(n_{10} + 1)(n_{1k_2} + 1)n_{2k_3} \\
&- n_{10}n_{1k_2}(n_{2k_3} + 1)] \delta(\varepsilon_{10} + \varepsilon_{1k_2} - \varepsilon_{2k_3}) \Delta(k_2 - k_3).
\end{aligned}$$

Since the number n_{10}^{ex} of non-equilibrium spin waves is large ($n_{10}^{\text{ex}} \gg 1$), the collision operator \mathcal{L}_{10} can be approximately represented in the form $\mathcal{L}_{10} \{n_1, n_2\} = -n_{10}^{\text{ex}}/\tau_{10}$, which after the corresponding transformations gives

$$\begin{aligned}
\tau_{10}^{-1} &\sim \frac{V}{\pi^2 \hbar} |\Phi_{0k_2; k_3}|^2 4\pi k_{\alpha}^2 (n_{1k_{\alpha}} - n_{2k_{\alpha}}) \\
&\times \left[\frac{d}{dk} (\varepsilon_{10} + \varepsilon_{1k} - \varepsilon_{2k}) \right]_{k=k_{\alpha}}^{-1}; \\
n_{sk} &= (e^{\varepsilon_{sk}/T} - 1)^{-1}.
\end{aligned}$$

Substituting in Eqs. (1), (2), (8), (9), $\mu/a^3 \sim M_0$, and $\mu\gamma M_0 \sim \Theta_C$, we obtain finally

$$\tau_{10}^{-1} \sim \left(\frac{\mu H_0}{\Theta_c} \right)^2 \left(\frac{\varepsilon_{20}}{\varepsilon_{10}} \right)^5 \left(\varepsilon_{20}^2 - 4\varepsilon_{10}^2 \right)^{1/2} \frac{n_{1k_{\alpha}} - n_{2k_{\alpha}}}{8\pi\hbar}. \tag{10}$$

For temperatures $\epsilon_{2k_{\alpha}} \ll T \ll T_N$ (this same condition, together with (2) and (1'), determines also the lower limit of the field region for which Eq. (11) is valid) and when $\epsilon_{10} \ll \epsilon_{20}$, one can obtain the simpler expression

$$\tau_{10}^{-1} \sim \left(\frac{\mu H_0}{\varepsilon_{10}} \right)^2 \left(\frac{\varepsilon_{20}}{\Theta_c} \right)^2 \frac{T}{2\pi\hbar}. \tag{11}$$

For comparison with experiment we estimate from (1') and (11) the line width of the low-frequency AFMR in the indicated temperature interval ($\Delta\omega \sim \tau_{\perp}^{-1} \sim 1/2 \tau_{10}^{-1}$):

$$\begin{aligned}
\Delta H &\sim \frac{\hbar}{\mu} \frac{2\sqrt{H_0(H_0 + H_D)}}{2H_0 + H_D} \\
\times \Delta\omega &\sim \frac{T}{2\pi\mu} \left(\frac{\varepsilon_{20}}{\Theta_c} \right)^2 \frac{H_0 \sqrt{H_0}}{(2H_0 + H_D) \sqrt{H_0 + H_D}}.
\end{aligned} \tag{12}$$

From this it can be seen that in antiferromagnets with anisotropy of the "easy plane" type and with a significant Dzyaloshinskii interaction (e.g., in CoCO_3 , where $H_D \sim 25$ kOe) the width of the lower AFMR line can depend strongly on the frequency at which the measurements are made. And, although the temperature region in which Eq. (12) is valid is not too large, the dependence of ΔH on H_0 is maintained for these substances also at other temperatures, which follows from Eq. (10). This conclusion, as well as the order of magnitude of ΔH determined from Eq. (12) is evidently confirmed by experiment (private communication from E. M. Rudashevskii, and [7]).

In the high-frequency AFMR branch (frequency ϵ_{20}/\hbar) the components $M_{1x} + M_{2x}$ and $M_{1z} - M_{2z}$ take part; hence the width of the upper AFMR line is obviously due to relaxation of the longitudinal component of the non-equilibrium magnetic moment. The latter is determined principally (since $\psi \ll 1$) by the occupation number of the spin waves of the upper branch with wave vector $\mathbf{k} = 0$:

$$\begin{aligned}
\langle \mathfrak{M}_x^2 \rangle &= \left\langle \left[\int (M_{1x} + M_{2x}) dV \right]^2 \right\rangle \\
&= (2M_0\psi V)^2 + 2(1 - 3\psi^2) \mu_{20}^{\text{eff}} M_0 V n_{20} - \\
&- 4\psi^2 M_0 V \sum_{k \neq 0} (\mu_{1k}^{\text{eff}} n_{1k} + \mu_{2k}^{\text{eff}} n_{2k}); \\
\mu_{20}^{\text{eff}} &= \mu (1 - Q_0)/(1 + Q_0).
\end{aligned}$$

¹⁾The fact that in the low-frequency oscillations just these components of the magnetic moment take part is easy to see, for example, from Eq. (13) in the paper by Borovik-Romanov.^[5]

In calculating the time $\tau_{||}$ of longitudinal relaxation it is necessary to consider that when $\epsilon_{20} > 2\epsilon_{10}$, i.e., in practice even for very large fields, a splitting of the spin wave ($k = 0; \epsilon_{20}$) into two: $(k_\beta; \epsilon_{1k_\beta})$ and $(-k_\beta; \epsilon_{1-k_\beta})$ is possible; k_β is easily determined from

$$\Theta_c a k_\beta = \frac{1}{2} (\epsilon_{20}^2 - 4\epsilon_{10}^2)^{1/2}.$$

These processes were discussed by Bar'yakhtar and Kovalev^[8] in a consideration of the establishment of equilibrium in a system of spin waves of antiferromagnets with weak ferromagnetism and are also described by the Hamiltonian (9). Calculation by the above scheme leads to

$$2\tau_{||}^{-1} \sim \tau_{20}^{-1} \sim (\mu H_0 / \Theta_c)^2 (\epsilon_{20}^2 - 4\epsilon_{10}^2)^{1/2} \quad (13)$$

$$\times (2n_{1k_\beta} + 1) / 2\pi\hbar.$$

It is obvious that at very weak H_0 , the calculation of ϵ_{20} requires the inclusion of processes involving a large number of spin waves.

Three-magnon processes can have a significant value for relaxation in antiferromagnets with anisotropy of the "easy axis" type if the sublattices are "flopped" (i.e., for $H_{||} > H_{AE}$), since the spectrum of spin waves in this case has the form $\epsilon_{1k} = \Theta_c a k$, $\epsilon_{2k} = [\epsilon_{20}^2 + \Theta_c^2 (ak)^2]^{1/2}$, where $\epsilon_{20}/\hbar = \mu (H_0^2 - H_{AE}^2)^{1/2}/\hbar$ is the upper AFMR frequency (the lower frequency $\epsilon_{10}/\hbar = 0$ if anisotropy in the basal plane is ignored). In oscillations at frequency ϵ_{20}/\hbar , it can be easily shown that the participating components are $M_{1x} + M_{2x}$, $M_{1y} + M_{2y}$, and $M_{1z} - M_{2z}$, the mean values of which turn out to be proportional to the occupation numbers n_{20} . Hence the width of the AFMR line at this frequency is due to the relaxation time τ_{20} , which is determined by the rate equation $\dot{n}_{20}^{ex} = \mathcal{L}_{20} \{ n_s \}$.

Writing the Hamiltonian of a uniaxial antiferromagnet for $H_{||} > H_{AE}$ in the form ($\gamma > 0$, $b_1 - b_2 > 0$, $b_2 > 0$, $\alpha - \alpha_{12} > 0$)

$$\begin{aligned} \mathcal{H} = & \int dV \left\{ \gamma M_1 M_2 - b_1 (M_{1z}^2 + M_{2z}^2) - 2b_2 M_{1z} M_{2z} \right. \\ & - (M_1 + M_2) H_0 + \frac{\alpha}{2} \left(\frac{\partial M_{1i}}{\partial x_k} \right)^2 + \frac{\alpha}{2} \left(\frac{\partial M_{2i}}{\partial x_k} \right)^2 \\ & \left. + \alpha_{12} \frac{\partial M_{1i}}{\partial x_k} \frac{\partial M_{2i}}{\partial x_k} \right\}, \end{aligned}$$

we obtain for τ_{20} Eq. (13), in which $\epsilon_{10} = 0$ and $\Theta_c a k_\beta = \epsilon_{20}/2$. This expression may also prove useful in estimating the line width of AFMR of antiferromagnets with cubic anisotropy^[1] in those cases when $H_0 \parallel [111]$, since the anisotropy constant then enters only through ϵ_{20} . It is also interesting to note that the line width due to the proc-

ess considered here does not disappear as $T \rightarrow 0^\circ K$.

Thus, for antiferromagnets whose spin wave spectrum permits three-magnon processes (i.e., those with "easy plane" anisotropy or "spin-flopped" sublattices), one obtains Eqs. (10) and (13), which determine the relaxation time of the magnetic moment and in addition the line width of the corresponding resonances as a function of temperature and magnetic field. These expressions are valid in the framework of the spin-wave theory ($T \ll T_N$, $a k_\alpha \ll 1$, $a k_\beta \ll 1$), and their contribution to the relaxation at not too low fields and temperatures (see above) is found to be distinct in comparison with the contribution of processes of higher order. The conditions for experimental verification of the dependences (10) and (13) are favorable, since the frequencies of the corresponding resonances can lie in a convenient microwave region; however, in each case it is necessary to make sure that the "real" line width is being observed.

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