INTERACTION BETWEEN SPIN WAVES AND CARRIERS IN ANTIFERROMAGNETIC DIELECTRICS AND SEMICONDUCTORS

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The spin-wave relaxation times in antiferromagnetic dielectrics of the "easy plane" and "easy axis" are calculated. It is shown that the width of the antiferromagnetic resonance (AFMR) line due to the interaction of the spin waves with the freely moving polarons can exceed in order of magnitude the width of the AFMR line due to the spin-spin interaction. In the case of antiferromagnetic semiconductors, the relaxation is determined by the same formulas as the relaxation obtained for antiferromagnetic dielectrics. A difference is observed only in the carrier effective masses.

1. INTRODUCTION

RELAXATION processes and the line width of antiferromagnetic resonance (AFMR) in pure antiferromagnets are due to different interactions of the spin waves with one another and of the spin waves with the phonons. We shall consider relaxation processes in antiferromagnetic dielectric and semiconductors having a sufficiently large number of carriers (on the order of 10^{18}—10^{19} cm^{-3}). The width of the AFMR line will be governed not only by the aforementioned interactions, but also by the interaction between the spin waves and the carriers in these substances.

We consider for concreteness an antiferromagnetic dielectric in which the carriers are polarons[1]. The region of temperatures T will be chosen such as to be able to neglect the processes connected with jumps of polarons from one lattice site to another[2]: $T \ll \hbar\omega_0$ ($\omega_0 \sim 10^{14}$—10^{15} sec^{-1}—limiting frequency of optical phonons). This allows to regard the polaron as a freely moving particle obeying Boltzmann statistics. The polaron mass M* is assumed to be of the order of 10—100 electron masses[1,3].

2. HAMILTONIAN OF INTERACTION BETWEEN SPIN WAVES AND POLARONS

We separate two groups of electrons in the antiferromagnetic dielectric; a group of A-electrons, responsible for the production of the antiferromagnetism, and a group of P-electrons, responsible for the production of polarons in the material. The Hamiltonian of interaction between these groups of electrons can be written in the form of a sum of the following Hamiltonians: the Hamiltonian $\mathscr{H}_1$ describing exchange interaction between A- and P-electrons, the Hamiltonian $\mathscr{H}_2$ describing their magnetic dipole interaction, and the Hamiltonian $\mathscr{H}_3$ describing the interaction of the magnetic moment $\mathbf{M}_j(\mathbf{r}, t)$ ($j = 1, 2$) of the A-electrons with the polaron current **j**. However, $\mathscr{H}_3$ can be neglected compared with $\mathscr{H}_2$, since the polaron current is small, owing to the large polaron mass. Therefore the total Hamiltonian $\mathscr{H}$ of the antiferromagnetic dielectric will be written in the form of the sum of the Hamiltonian $\mathscr{H}_s$ connected with the magnetic energy of the antiferromagnetic dielectric, the Hamiltonian $\mathscr{H}_p$ characterizing the energy of the free polarons, and the interaction Hamiltonians $\mathscr{H}_1$ and $\mathscr{H}_2$:

$$\mathscr{H} = \mathscr{H}_s + \mathscr{H}_p + \mathscr{H}_1 + \mathscr{H}_2;$$

$$\mathscr{H}_s = \int d\mathbf{r} \Big\{ \delta \mathbf{M}_1\mathbf{M}_2 + \frac{1}{2}\alpha\Big[\Big(\frac{\partial \mathbf{M}_1}{\partial x_i}\Big)^2 + \Big(\frac{\partial \mathbf{M}_2}{\partial x_i}\Big)^2\Big] + \alpha_{12}\frac{\partial \mathbf{M}_1}{\partial x_i}\frac{\partial \mathbf{M}_2}{\partial x_i} + \frac{1}{2}\beta(M_{1z}^2 + M_{2z}^2) + \beta_{12}M_{1z}M_{2z} - (\mathbf{M}_1 + \mathbf{M}_2)\mathbf{H}_0\Big\},$$

$$\mathscr{H}_p = \int d\mathbf{r}\psi_p^+(\mathbf{r}, t)\Big\{\frac{p^2}{2M^*}\Big\}\psi_p(\mathbf{r}, t),$$

$$\mathscr{H}_1 = \mu\sum_{j=1}^{2}\sum_{\sigma}\int \psi_p^+(\mathbf{r}, t)\sigma\psi_p(\mathbf{r}, t)\mathbf{H}_j^e(\mathbf{r}, t)\,d\mathbf{r},$$

$$\mathbf{H}_j^e(\mathbf{r}, t) = \int J(\mathbf{r} - \mathbf{r}')\mathbf{M}_j(\mathbf{r}', t)\,d\mathbf{r}',$$

$$\mathscr{H}_2 = \mu\sum_j\sum_\sigma\int\int\psi_p^+(\mathbf{r}, t)$$

$$\times \frac{(\sigma \mathbf{M}_j(\mathbf{r}',t))(\mathbf{r}-\mathbf{r}')^2 - 3(\sigma\cdot(\mathbf{r}-\mathbf{r}'))(\mathbf{M}_j(\mathbf{r}',t)\cdot(\mathbf{r}-\mathbf{r}'))}{|\mathbf{r}-\mathbf{r}'|^5}$$

$$\times \psi_p(\mathbf{r},t)\,d\mathbf{r}\,d\mathbf{r}'. \quad (1)$$

Here M_1 and M_2 are the magnetic moments of the sublattices; J is the exchange integral between the electrons of type A and P; H_0 is the external magnetic field; α, α_{12}, and δ are the exchange constants, β and β_{12} the magnetic-anisotropy constants, μ the Bohr magneton, σ the spin operator of the P-electrons, and $\psi_p^+(\mathbf{r}, t)$ and $\psi_p(\mathbf{r}, t)$ are the operators for creation and absorption of a P-electron at the point $\mathbf{r}$ at the instant of time t. The operators ψ_p^+ and ψ_p can be expanded in plane waves characterizing the free polaron:

$$\psi_p(\mathbf{r},t) = \frac{1}{\sqrt{V}}\sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}}[\alpha(\sigma)d_{\mathbf{k}}(t) + \beta(\sigma)b_{\mathbf{k}}(t)],$$

$$\psi_p^+(\mathbf{r},t) = \frac{1}{\sqrt{V}}\sum_{\mathbf{k}} e^{-i\mathbf{k}\mathbf{r}}[\alpha^*(\sigma)d_{\mathbf{k}}^+(t) + \beta^*(\sigma)b_{\mathbf{k}}^+(t)], \quad (2)$$

where $\alpha(\sigma)$ and $\beta(\sigma)$ are spin function defined by $\sigma_\zeta\alpha = \alpha$, and $\sigma_\zeta\beta = -\beta$; d_k^+, b_k^+, d_k, and b_k are operators for the creation and absorption of a P-electron with momentum k and spin projections +1/2 and −1/2; V is the volume of the body.

It is convenient to represent the quantities $M_j(\mathbf{r}, t)$ in the form

$$\mathbf{M}_j = \mathbf{e}_{\zeta j}M_{\zeta j} + \mathbf{e}_{\eta j}M_{\eta j} + \mathbf{e}_{\xi j}M_{\xi j}, \quad (3)$$

with the unit vectors $\mathbf{e}_{\zeta j}$, $\mathbf{e}_{\eta j}$, and $\mathbf{e}_{\xi j}$ for the case of anisotropy of the "easy plane" type ($\beta - \beta_{12} > 0$) chosen as follows: the unit vector $\mathbf{e}_{\zeta j}$ is oriented in the direction of the magnetic moment of the j-th sublattice M_{j0} in the ground state, $\mathbf{e}_{\zeta j} = M_{j0}/M_{j0}$; the unit vector $\mathbf{e}_{\xi j}$ is oriented along the anisotropy axis, and $\mathbf{e}_{\eta j} = \mathbf{e}_{\zeta j} \times \mathbf{e}_{\xi j}$.

For the case of anisotropy of the "easy axis" type ($\beta - \beta_{12} < 0$)

$$\mathbf{e}_{\zeta j} = \mathbf{M}_{j0}/M_{j0}, \quad \mathbf{e}_{\xi j} = \mathbf{e}_{x_1}, \quad \mathbf{e}_{\eta j} = \mathbf{e}_{\zeta j}\times\mathbf{e}_{\xi j}.$$

The operators $M_{\zeta j}$, $M_{\eta j}$, and $M_{\xi j}$ are connected with the Holstein-Primakoff operators a_j^+ and a_j by the formulas[4]

$$M_{\zeta j} = M_0 - \mu a_j^+ a_j, \quad M_{\eta j} = i(\mu M_0/2)^{1/2}(a_j - a_j^+),$$
$$M_{\xi j} = (\mu M_0/2)^{1/2}(a_j + a_j^+). \quad (4)$$

(We have confined ourselves in (4) to the first terms of the expansion of the operators of the moments M_j in terms of the operators a_j^+ and a_j, since we are not interested in the interaction of spin waves with one another.)

We now use relations (2), (3), and (4) and substitute them in (1). Then the Hamiltonian of the antiferromagnet $\mathscr{H}$ is expressed in terms of the operators a_{jk}^+, a_{jk}, d_k^+, d_k, b_k^+, and b_k (a_{jk}^+ and a_{jk} are the Fourier transforms of the operators $a_j^+(\mathbf{r})$ and $a_j(\mathbf{r})$). However, the operator $\mathscr{H}_s$ will not be diagonal in the operators a^+ and a. It is diagonalized with the aid of a canonical uv-transformation from the operators a_{jk} to the spin-wave creation and annihilation operators c_{jk}^+ and c_{jk}[5]:

$$a_{1\mathbf{k}} = u_{11}c_{1\mathbf{k}} + u_{12}c_{2\mathbf{k}} + v_{11}^*c_{1-\mathbf{k}}^+ + v_{12}^*c_{2-\mathbf{k}}^+,$$
$$a_{2\mathbf{k}} = u_{21}c_{1\mathbf{k}} + u_{22}c_{2\mathbf{k}} + v_{21}^*c_{1-\mathbf{k}}^+ + v_{22}^*c_{2-\mathbf{k}}^+. \quad (5)$$

The final form of the Hamiltonian $\mathscr{H}$ in terms of the spin-wave and polaron creation and annihilation operators is

$$\mathscr{H} = \mathscr{H}_s + \mathscr{H}_p + \mathscr{H}_{sp_1} + \mathscr{H}_{sp_2} + \mathscr{H}_{sp_3},$$
$$\mathscr{H}_s = W + \sum_{\mathbf{f}}[\mathscr{E}_{1\mathbf{f}}c_{1\mathbf{f}}^+c_{1\mathbf{f}} + \mathscr{E}_{2\mathbf{f}}c_{2\mathbf{f}}^+c_{2\mathbf{f}}],$$
$$\mathscr{H}_p = \sum_{\mathbf{k}}[E_{+1/2}(\mathbf{k})\,d_{\mathbf{k}}^+d_{\mathbf{k}} + E_{-1/2}(\mathbf{k})\,b_{\mathbf{k}}^+b_{\mathbf{k}}], \quad (6)$$

where $\mathscr{E}_{jf}$ is the energy of the spin wave of the j-th branch, $E_\sigma(\mathbf{k})$ is the energy of a polaron having a P-electron spin projection equal to σ:

$$E_\sigma(\mathbf{k}) = \Theta_p(ak)^2 + 2\sigma\mu M_0 J, \quad \sigma = \pm 1/2, \quad \Theta_p \equiv \hbar^2/2M^*a^2,$$

a is the interatomic distance;

$$\mathscr{H}_{sp_1} = 4\mu\sqrt{\frac{2\mu M_0}{V}}\,J\sum_{\mathbf{k}\mathbf{k}'\mathbf{f}} u_1(\mathbf{f})\,\{c_{1\mathbf{f}}^+\,d_{\mathbf{k}}b_{\mathbf{k}'}^+ + c_{1\mathbf{f}}d_{\mathbf{k}}^+b_{\mathbf{k}'}\}\,\Delta(\mathbf{k}-\mathbf{k}'-\mathbf{f}), \quad (7)$$

$$\mathscr{H}_{sp_2} = 8\pi\mu\sqrt{\frac{2\mu M_0}{V}}\sum_{\mathbf{k}\mathbf{k}'\mathbf{f}} u_1(\mathbf{f})\,\frac{f_\zeta f_\xi}{f^2}\{c_{1\mathbf{f}}^+\,(d_{\mathbf{k}'}^+d_{\mathbf{k}} - b_{\mathbf{k}'}^+b_{\mathbf{k}}) + c_{1\mathbf{f}}\,(d_{\mathbf{k}}^+d_{\mathbf{k}'} - b_{\mathbf{k}}^+b_{\mathbf{k}'})\}\,\Delta(\mathbf{k}-\mathbf{k}'-\mathbf{f}), \quad (7')$$

$$\mathscr{H}_{sp_3} = 2\,\frac{\mu^2}{V}\,J\sum_{\mathbf{k}\mathbf{k}'\mathbf{f}\mathbf{f}'} u_j(\mathbf{f})\,u_j(\mathbf{f}')\,\{c_{j\mathbf{f}}^+c_{j\mathbf{f}'}\,(d_{\mathbf{k}'}^+d_{\mathbf{k}} - b_{\mathbf{k}'}^+b_{\mathbf{k}}) + c_{j\mathbf{f}}c_{j\mathbf{f}'}^+\,(d_{\mathbf{k}'}d_{\mathbf{k}}^+ - b_{\mathbf{k}'}b_{\mathbf{k}}^+)\}\,\Delta(\mathbf{k}+\mathbf{f}'-\mathbf{k}'-\mathbf{f}). \quad (8)$$

Here $u_j(\mathbf{f})$ are the coefficients of the canonical uv-transformation (5), and if we neglect the magnetic dipole interaction of the spins, then, according to[6-8],

$$u_1(\mathbf{f}) = u_{11} = u_{21} = -v_{11} = -v_{21} = (\delta\mu M_0/4\mathscr{E}_{1\mathbf{f}})^{1/2},$$
$$u_2(\mathbf{f}) = u_{12} = -u_{22} = v_{12} = -v_{22} = (\delta\mu M_0/4\mathscr{E}_{2\mathbf{f}})^{1/2}. \quad (9)$$

(We note that in our approximation the formulas (9) for the uv-transformation coefficients hold true both for "easy axis" and "easy plane" anisotropy.)

The Hamiltonians $\mathscr{H}_{sp_1}$ and $\mathscr{H}_{sp_2}$ describe the creation and absorption of a spin wave with and without change in the P-electron spin projection, and the Hamiltonian $\mathscr{H}_{sp_3}$ describes scattering of a spin wave without change of the P-electron spin projection.

3. INCREASE OF AFMR LINE WIDTH BY SPIN-POLARON INTERACTION

The dependence of the AFMR line width on the temperature and on the external magnetic field is determined, besides by the character of the interaction, by the spectrum of the spin waves $\mathscr{E}_{jf}$.

Let us consider anisotropy of the "easy plane" type. The spin-wave spectrum is of the form

$$\mathscr{E}_{jf} = [\Theta_N^2 (af)^2 + \mathscr{E}_{j0}^2]^{1/2}, \quad j = 1, 2, \tag{10}$$

where $\mathscr{E}_{10} = \mu M_0 [2\delta(\beta - \beta_{12}]^{1/2}$, $\mathscr{E}_{20} = \mu H_0$, H_0 is the external magnetic field, lying in the basal plane, and $\Theta_N = \mu M_0 [2\delta(\alpha - \alpha_{12}]^{1/2}/a$ is a temperature on the order of the Neel temperature.

If we take account of the Dzyaloshinskiĭ energy $dM_1 \times M_2$ in the Hamiltonian $\mathscr{H}_S$, then it is necessary to replace H_0^2 in the expression for $\mathscr{E}_{2f}$ by $H_0(H_0 + dM_0)$, and in the expression for $\mathscr{E}_{1f}$ it is necessary to add under the square root the term $\mu^2 dM_0(H_0 + dM_0)$.

The change per unit time in the number of spin waves n_{jf} of the j-th branch with wave vector **f** is described by the kinetic equation

$$\dot{n}_{jf} = \mathscr{L}_{j1f}\{n, N\} + \mathscr{L}_{j2f}\{n, N\} + \mathscr{L}_{j3f}\{n, N\}, \quad j = 1, 2. \tag{11}$$

The collision integrals $\mathscr{L}_{j1f}$, $\mathscr{L}_{j2f}$, and $\mathscr{L}_{j3f}$ are connected respectively with the Hamiltonians $\mathscr{H}_{sp_1}$, $\mathscr{H}_{sp_2}$, and $\mathscr{H}_{sp_3}$:

$$\mathscr{L}_{11f}\{n, N\} = 0, \quad \mathscr{L}_{12f}\{n, N\} = 0,$$

$$\begin{aligned}\mathscr{L}_{21f}\{n, N\} = \frac{16\pi}{\hbar}\frac{\delta(\mu M_0 J)^2}{\mathscr{E}_{1f}}\frac{\mu^2}{V}\sum_{kk'}\{(n_{1f}+1)N_{k+}(1-N_{k'-}) \\ - n_{1f}(1-N_{k+})N_{k'-}\}\delta(\mathscr{E}_{1f} - E_+(k) \\ + E_-(k))\Delta(k - k' - f),\end{aligned} \tag{12}$$

$$\begin{aligned}\mathscr{L}_{22f}\{n, N\} = \frac{64\pi^3}{\hbar}\frac{\delta(\mu M_0)^2}{\mathscr{E}_{1f}}\frac{\mu^2}{V}\sum_{\sigma kk'}\{(n_{1f}+1)N_{\sigma k}(1-N_{\sigma k'}) \\ - n_{1f}(1-N_{\sigma k})N_{\sigma k'}\}\delta(\mathscr{E}_{1f} - E_\sigma(k) \\ + E_\sigma(k'))\Delta(k - k' - f),\end{aligned} \tag{12'}$$

$$\begin{aligned}\mathscr{L}_{j3f}\{n, N\} = \frac{8\pi}{\hbar}u_j^2(f)J^2\frac{\mu^4}{V^2}\sum_{\sigma kk'f'}u_j^2(f')\{(n_{jf}+1) \\ \times n_{jf'}(1-N_{\sigma k})N_{\sigma k'} - n_{jf}(n_{jf'}+1)N_{\sigma k'}(1-N_{\sigma k})\} \\ \times \delta(E_\sigma(k) - E_\sigma(k') - \mathscr{E}_{jf} + \mathscr{E}_{jf'})\Delta(k + f' - k' - f).\end{aligned} \tag{13}$$

Here $N_{\sigma k}$ is the distribution function of polarons having P-electron spin projections equal to σ.

Formulas (11), (12), (12′), and (13) make it possible to determine the spin-wave damping τ_{jf}^{-1} as a function of the wave vector **f**.

To determine the relaxation of the magnetic moment (and also the AFMR line width) it is necessary to find the change in the number $n_{jf\to 0}$ of the spin waves of the j-th branch with wave vector $f = 0$. Recognizing that in the equilibrium state $n_{j0}^{(0)} = 0$ and $n_{j0} \gg 1$, we get from (11) for the AFMR line width

$$\tau_{j0}^{-1} = -\sum_{\alpha=1}^{3}\left(\frac{\delta\mathscr{L}_{j\alpha 0}}{\delta n_{j0}}\right)_0, \tag{14}$$

where the variational derivative is taken at the equilibrium values of $n_{j0}^{(0)}$ and $N_{\sigma k}^{(0)}$.

If we now put $f = 0$ in (12) and (12′) and calculate (14), then we get, owing to the impossibility of satisfying simultaneously the momentum and energy conservation laws,

$$\left(\frac{\delta\mathscr{L}_{j10}}{\delta n_{j0}}\right)_0 = \left(\frac{\delta\mathscr{L}_{j20}}{\delta n_{j0}}\right)_0 = 0. \tag{15}$$

Therefore when $f = 0$ the relaxation τ_{j0}^{-1} is determined only in terms of the variational derivative of the collision integral $\mathscr{L}_{j30}$:

$$\begin{aligned}\frac{1}{\tau_{j0}} = \frac{8\pi}{\hbar}u_j^2(0)J^2\frac{\mu^4}{V^2}\sum_{\sigma kk'f}(N_{\sigma k'}^{(0)}N_{\sigma k}^{(0)} - N_{\sigma k'}^{(0)}) \\ \times\delta(E_\sigma(k) - E_\sigma(k') - \mathscr{E}_{j0} + \mathscr{E}_{jf})\Delta(k - k' + f).\end{aligned} \tag{16}$$

It follows further from (15) that the number of P-electrons with given spin projection does not change. This allows us to effect the normalization

$$\sum_k N_{\sigma k}^{(0)} = N_p/2, \quad \sigma = \pm 1/2, \tag{17}$$

N_p is the total number of polarons in the body.

Going over in (16) from summation to integration and using (10) and (17), we obtain the following value for damping at temperatures $\mathscr{E}_{j0} \le T \ll \Theta_N$:

$$\frac{1}{\tau_{j0}} \approx \frac{C^2}{16\pi}\left(\frac{\mu^2}{a^3\Theta_N}\right)^2\left(\frac{\Theta_p}{\Theta_N}\right)^2\frac{\delta^2(\mu M_0 J)^2}{\hbar T}, \tag{18}$$

where $C \equiv N_p/N$ is the relative polaron concentration.

Let us obtain a numerical estimate for τ_{j0}^{-1} at the same temperatures as in Ozhogin's papers[8]. Putting $T \sim 1 - 10°K$, $C \sim 10^{-4}$ (polaron concentration $\sim 10^{19}\ cm^{-3}$), $\delta \sim 10^3$, $\mu M_0 J \sim 300°K$, we obtain $\tau_{j0}^{-1} \sim 10^7 - 10^8\ sec^{-1}$, which coincides in order of magnitude with the AFMR line width due to the spin-spin interaction.

Let us consider anisotropy of the "easy axis" type. The relaxation τ_{j0}^{-1} is determined also by Eq. (16). If the external magnetic field H_0 is applied along the anisotropy axis and lies in the interval $H_e \gg H_0 > H_a$, ($H_e \approx 2\delta M_0$, $H_a = (2\delta|\beta - \beta_{12}|)^{1/2}M_0$), then the spin-wave spectra $\mathscr{E}_{jf}$ have the form given by Eq. (10) (see[4]). Consequently, the damping is determined by formula (18) derived above. A similar situation is observed also in an external magnetic field $H_0 \ll H_e$ perpendicular to the anisotropy axis. According to[4], the spectrum $\mathscr{E}_{jf}$ coincides in form also with expression (10) and the damping is determined by formula (18).

In antiferromagnetic semiconductors, in which the carriers are electrons obeying Boltzmann statistics, the increase in the AFMR line width is obviously given by the same formulas (18). The only difference lies in the value of Θ_p, in which the polaron mass M^* must be replaced by the electron mass; this, according to (18), leads to an essential increase in the width of the AFMR line. This increase in the AFMR line width greatly exceeds the spin-spin width of the antiferromagnetic resonance.

In conclusion we point to antiferromagnetic semiconductors in which there is a sufficiently large number of carriers and in which we can expect, consequently, a noticeable increase in the AFMR line width. These are $CuFeS_2$ (carrier density $\sim 10^{19}$ cm^{-3}) and UTe_2 (density $\sim 10^{18}$—10^{19} cm^{-3}).[9]

The required polaron density can be produced in antiferromagnetic dielectrics by irradiating the sample with light from a laser or by injection of electrons to pass a large current through the dielectric.[1]

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