Birefringence of light in antiferromagnets near the Neel temperature

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The manifestation of short-range order effects in the refractive index is considered for two-sublattice antiferromagnets on the basis of the mechanism of direct spin-electron interaction.

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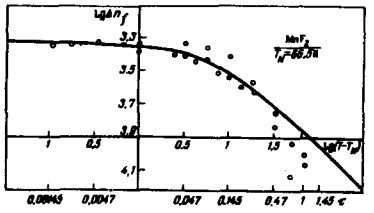
It was observed recently that a strong change takes place in the birefringence of light in a number of transparent ferrimagnets and antiferromagnets[1,3] near the magnetic-order temperature T_N, and that below T_N the effect acquires an increment proportional to $\langle S\rangle^2$—the square of the sublattice magnetization. An interesting circumstance is the fact that above T_N, owing to the presence of short-range magnetic order, the magnetic birefringence can constitute an appreciable fraction of the natural birefringence,[1,4] decreasing slowly with increasing temperature. Jahn, Dachs, and Jauch[3,4] attribute this behavior of the birefringence to an exchange-striction mechanism, which is known to introduce into the refractive index of light n_0 an increment proportional to the average magnetic energy $\epsilon_M(T)$ of the atom.[1,3] However, the function $\epsilon_M(T)$ does not account sufficiently well[4] for the temperature dependence of the effect. In this communication we discuss another mechanism of direct spin-electron interaction and show that this mechanism is capable in principle of explaining the experimentally observed anomalies.

We consider a system of Bloch electrons in a crystal. These electrons interact with a system of spins S_{n_i} localized on lattice sites $\mathbf{r}_{n_i}$, and execute interband transitions under the influence of the electric field of the light wave. To calculate the magnetic increment Δn_M^{xy} to the refractive index, we use the procedure described in[5], i.e., we find that, when account is taken of the electron-spin interaction, the density-matrix increment $\rho^{(1)}$ linear in the electric field $\mathbf{E} = \mathbf{E}_0 e^{i\omega t}$ is

$$\rho_{p\alpha s,\, p'\alpha' s'}(t) = \langle a^+_{p\alpha s}(t)\, a_{p'\alpha' s'}(t)\rangle \qquad (1)$$

where $a^+_{p\alpha s}(t)$ and $a_{p\alpha s}(t)$ are Heisenberg creation and annihilation operators of an electron with quasimomentum $\mathbf{p}$ and spin $s = \pm 1/2$ in the energy band α. Bearing in mind the application of the results primarily to fluorides of transition metals[2,3] (MnF_2, CoF_2, FeF_2, NiF_2), which are two-sublattice antiferromagnetics with two magnetic ions per unit crystal cell, we write the total Hamiltonian of the system in the form

$$H = \sum_{p\alpha s} \epsilon_\alpha(\mathbf{p})\, a^+_{p\alpha s}\, a_{p\alpha s} - \frac{e}{c}\, \mathbf{A} \sum_{p\alpha_1\alpha_2} \mathbf{v}_{\alpha_1\alpha_2}(\mathbf{p})\, a^+_{p\alpha_1 s}\, a_{p\alpha_2 s} + H_1, \qquad (2)$$



$$H_1 = -\frac{1}{N}\sum_{\substack{p\alpha\\ p'\alpha'\\ n_i}} I_i(p\alpha,\ p'\alpha')\exp\{i(p-p')r_{n_i}\}\{a^+_{p\alpha}a_{p'\alpha'}($$

$$-a^+_{p\alpha-}a_{p'\alpha'-})S^z_{n_i} + a^+_{p\alpha+}a_{p'\alpha'-}S^+_{n_i} + a^+_{p\alpha-}a_{p'\alpha'+}S^-_{n_i}\} \quad (3)$$

here $\mathbf{A}=(ic/\omega)\mathbf{E}$, $\epsilon_\alpha(\mathbf{P})$ is the dispersion law in band α, $i=1,2$ is the number of the magnetic sublattice, and the sum over n_i is taken over all N lattice sites in which there are magnetic ions. Since the local environment of non-equivalent magnetic ions is different in the sense of symmetry, and the symmetry of the Bloch wave function is determined by the total spatial symmetry of the crystal, the matrix elements of the exchange interval with the first and second sublattices are different for the given pair of Bloch states $\mathbf{p}\alpha$ and $\mathbf{p}'\alpha'$: $I_1(\mathbf{p}\alpha,\mathbf{p}'\alpha') \neq I_2(\mathbf{p}\alpha,\mathbf{p}'\alpha')$.

The equation for $\rho^{(1)}$ can be obtained and solved by perturbation theory, using the smallness of the parameter $\eta=|I/\beta_p|\ll 1$, where

$$\beta_p = \epsilon_{\alpha_1}(p) - \epsilon_{\alpha_2}(p) - \hbar\omega. \quad (4)$$

Inasmuch as in antiferromagnets we have $\langle S_{n_1}\rangle = -\langle S_{n_2}\rangle = \langle S\rangle$, the small magnetic increment to n_0 appears in second order, and, in addition to a part proportional to $\langle S\rangle^2$, it will contain also a fluctuating part Δn_f^{xy}.

$$\Delta n_M^{xy} = A^{xy}\langle S\rangle^2 + \Delta n_f^{xy}, \quad A^{xy}\sim \eta^2 e^2 v^2/\beta\omega^2. \quad (5)$$

If $|\tau|\gg\eta$, where $\tau=(T-T_N)/T_N$, then in the latter we can confine ourselves to allowance for only the pair correlators of the quantities $\delta S_n = S_n - \langle S\rangle$. In accordance with the phase-transition theory based on the hypothesis of similarity of the correlations, we assume that the properties of these correlators are such that

$$\sum_{n_1'}\langle \delta S_{n_1}\delta S_{n_1'}\rangle e^{ik(r_{n_1}-r_{n_1'})} = -\sum_{n_2}\langle \delta S_{n_1}\delta S_{n_2}\rangle e^{ik(r_{n_1}-r_{n_2})} \cong \frac{4\pi a^2}{k^2+r_c^{-2}}. \quad (6)$$

We have neglected in (6) the difference between the critical exponent of the spin correlator and unity. As $\tau\to 0$, the correlation radius $r_c\sim a|\tau|^{-\mu}$ increases (a is the characteristic interatomic distance). It can be assumed that the main contribution to Δn_f^{xy} is made by the pair of bands the energy gap between which is closest to $\hbar\omega$. Near the absorption threshold, which is usually connected with a high-symmetry point $\mathbf{p}_0$ in the Brillouin zone, we can write, neglecting the anisotropy,

$$\epsilon_{\alpha_j}(p) = \epsilon_{\alpha_j}(p_0) + \frac{(p-p_0)^2}{m_j}; \qquad \beta_p = \beta + \frac{(p-p_0)^2}{M}. \quad (7)$$

The calculation yields

$$\Delta n_f^{xy} = A_1^{xy}\left[B\frac{\kappa}{1+\kappa} + F(\kappa, C)\right],$$

$$A_1^{xy} = -\frac{e^2 v_{12}^x v_{21}^y (l_1 m_1 - l_2 m_2)^2}{2^5\omega^2 n_0 \beta}, \qquad B = \frac{2M(l_1^2 m_1 - l_2^2 m_2)}{(l_1 m_1 - l_2 m_2)^2}, \quad (8)$$

$$C = 4M^2/|m_1 m_2|, \quad \kappa = 2\sqrt{M\beta}\, r_c \sim |\tau|^{-\mu},$$

$$I_{\alpha_j} = I_1(p_0\alpha_j,\ p_0\alpha_j) - I_2(p_0\alpha_j,\ p_0\alpha_j).$$

For a threshold of the type $m_1 m_2 > 0$ we have

$$F(\kappa, C) = \frac{\kappa^2}{1+\kappa} - \frac{\kappa^2}{(1+C)^{1/2}+(\kappa^2+C)^{1/2}}. \quad (9)$$

For a threshold of the type $m_1 m_2 < 0$ the function $F(\kappa, C)$ has a more complicated form. However, $\kappa\ll 1$, in both cases, i.e., a strong temperature anomaly should be observed in $\Delta n_f^{xy}\sim \kappa/\beta\sim r_c\sim|\tau|^{-\mu}$ close to the absorption line. Little is known at the present concerning the band structure of fluorides of transition metals.[6] If we assume $|m_1|\ll|m_2|$, i.e., the upper band is much broader than the lower one, then $C\ll 1$ and

$$\Delta n_f^{xy} = A_1^{xy}\left(B + \frac{C}{2}\right)\frac{\kappa}{1+\kappa}. \quad (10)$$

In accordance with (5) and (10), Δn_M^{xy} should have a break near T_N, and such a break was observed in[2,3]. In the experiments of[2,3] they used light with $\lambda = 6328$ Å, which corresponds in MnF_2 to a distance $\beta\sim 0.2$ eV to the nearest threshold of the absorption line.[7] In the region $\eta\lesssim|\tau|\lesssim 0.5$, where the approximation $r_c\sim|\tau|^{-2/3}$ is reasonable, the experimental points from[2] agree fairly well with the relation (10) if it is assumed that the interband mass M is such that $(Ma^2)^{-1} = 4.5$ eV (see the figure).

In accordance with the result (8), the character of the temperature dependence of Δn_f^{xy} should change strongly with frequency. On the other hand, this change is not characteristic of the exchange-striction contribution, and therefore the variation of the frequency of the light in experiments of the type of[1-3] may turn out, in our opinion, to be useful in ascertaining the mechanism of magnetic birefringence in antiferromagnets.

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