Nonreciprocal Optical Rotation in Antiferromagnets

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We consider nonreciprocal optical effects in classical Néel antiferromagnets, where the purely macroscopic electrodynamics does not provide any rotation. The nonzero result appears as a correction linear in the scattering theory parameter a/λ, atomic distance over wavelength. The recent experiment on Cr_2O_3, where optical nonreciprocity already exists with the macroscopic approach, is also discussed.

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In the last five years a number of attempts were made to detect time-parity violation in high-T_c superconductors, predicted within some theoretical models by measuring nonreciprocal optical rotation [1–3]. All of these attempts failed, but meanwhile nonreciprocal optical rotation was observed for the first time in a conventional antiferromagnet [4]. The general restrictions on nonreciprocal optical reflection and transmission imposed by time-reversal symmetry (T) were studied by Halperin [5], Shelankov and Pikus [6], and Canright and Rojo [7]. Here, we address the underlying physics of the phenomenon.

There is a cardinal difference between nonreciprocal effects in ferromagnets (or paramagnets in magnetic fields) and in antiferromagnets. In the former the nonreciprocal effects—the famous Faraday and Kerr rotations—are *macroscopic* and can be calculated if the antisymmetric part $\hat{\epsilon}^a$ of the dielectric function of the medium is known (see, e.g., [8]). In contrast, in a typical Néel antiferromagnet the time symmetry broken *microscopically* reemerges in the large-scale macroscopic picture. The potential nonreciprocal effects could be generated by the same antisymmetric part of the dielectric tensor ϵ_{ij}. It is subject to the well-known Onsager constraint:

$$\epsilon_{ij}(\omega, k, l) = \epsilon_{ji}(\omega, -k, -l), \tag{1}$$

which expresses the time reversal symmetry of interactions. Here k is the wave vector of the wave and l is the staggered magnetization. In the majority of antiferromagnets (some exceptions from this rule will be discussed later) the two Néel sublattices are chemically indistinguishable, and ϵ_{ij} is an even function of l, signifying that T is indeed macroscopically restored. With ϵ_{ij} even in l, the only way to construct an antisymmetric tensor is to make it odd in k, thus destroying not only time parity but space parity (P) as well. The corresponding rotation, so-called gyrotropic rotation, is of no concern to us here. To reveal the hidden *microscopically* violated time-parity we have to go beyond the conventional space dispersion (the k dependence of the dielectric function of an infinite homogenous sample) and take into account the underlying atomic structure. Let us consider a typical experiment: a slab of thickness L of a regular two-sublattice Néel antiferromagnet and a linearly polarized wave of frequency ω which is incident normally. The details of the geometry are shown in Fig. 1. The case of oblique incidence is considered elsewhere. To avoid unnecessary complications in formulas (it is easy to generalize the results), we assume that the medium response is given by a coordinate dependent dielectric tensor $\hat{\epsilon}(\mathbf{r})$ and, moreover, that $\hat{\epsilon}(\mathbf{r})$ is simply a sum of atomic polarizations $\hat{p}$:

$$\hat{\epsilon}(\mathbf{r}) = \sum_n \hat{p}_n \delta(\mathbf{r} - \mathbf{r}_n), \tag{2}$$

where the summation runs over all atomic sites n. The effects of atomic structure are small in the optical region, being suppressed by the factor of the scattering theory $\omega a/c$ (a is of the order of atomic distances). To calculate the zero and first order terms in $\omega a/c$ we follow the standard path of light scattering theory (see, e.g., [8]). In the zero order in $\omega a/c$ the coordinate dependent dielectric tensor (2) is substituted by its macroscopic value $\overline{\epsilon}$. As we have seen due to the Onsager relations (1), $\overline{\epsilon}$ is a symmetric tensor. To better separate nonreciprocal effects from birefringence, the propagation direction is taken to coincide with the high-symmetry axis of our uniaxial sample. Then only $\epsilon_{yy} = \epsilon_{zz} \equiv \epsilon$ appear in the problem, and the incident E^i, reflected E_0^r, and transmitted E_0^t zero-order electric fields are all polarized in the same direction, say, z. They are related by the well-known formulas (see, e.g., [8]).

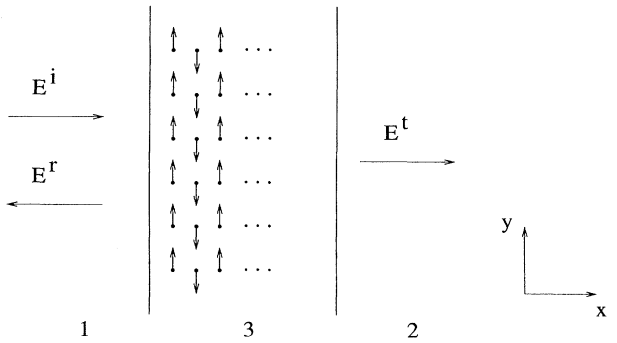

FIG. 1. Geometry of a typical optical experiment.

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The nonreciprocal rotation is generated (in first order in $\omega a/c$) by the antisymmetric part of the dielectric tensor $\hat{p}^a$. Instead of $\hat{p}^a_{ij}$ we use its dual vector u_k: $\hat{p}^a_{ij} = \epsilon_{ijk}u_k$. If crystalline electric fields are neglected then $u_k^{(n)}$ is oriented along the spin s_n on the site n: $u_k^{(n)} = \beta s_k(-)^n$ for our two-sublattice antiferromagnet, where s_k is the magnetization direction of the first layer on the left in Fig. 1.

Clearly, instead of (2) we may use the value of ϵ^a averaged over the yz plane;

$$\epsilon^a_{ij}(x) = \gamma a \epsilon_{ijk} s_k \sum_{n=0}^{N-1} (-)^n \delta(x - na), \tag{3}$$

where $\gamma = \beta/v$ (v is the volume of the unit cell) represents the rotating power of a ferromagnetically polarized layer.

The first order fields $\mathbf{E}$ satisfy the equations

$$\left(\epsilon \frac{\omega^2}{c^2} - \text{curl curl}\right)\mathbf{E} = -\frac{\omega^2}{c^2}\hat{\epsilon}^a(x)\mathbf{E}_3 \tag{4}$$

inside the slab with $\hat{\epsilon}^a(x)$ from (3), and $\mathbf{E}_3$ being the zero order field inside the slab. Outside the slab (regions 1 and 2 in Fig. 1) $\mathbf{E}$ satisfies the corresponding homogeneous equation with $\epsilon = 1$. The equations were solved using the Green function method. For y projections of reflected and transmitted fields which give the nonreciprocal rotation we write

$$E^r_y = \frac{\omega^2}{c^2} a\gamma s_x \sum_{n=0}^{N-1} \int_{x<0} (-)^n D_{yy}(x, na) E_3(na) dy\, dz, \tag{5}$$

and the analogous formula for E^t_y with $D_{yy}(x, na)$ for $x > L$. The Green functions in the geometry of Fig. 1 may be calculated as the line by line repetition of the corresponding calculations in Ref. [9]. In evaluating the sum in (5) we should keep only the leading order in $\omega a/c$.

The polarization of the reflected wave is given by E^r_y/E^r_z. Expressing E_3 in terms of $E^i \simeq E^r_z$ we find

$$\frac{E^r_y}{E^r_z} = \frac{\gamma a \omega s_x}{c(\epsilon - 1)} \frac{\epsilon - 1 - 2(-)^N\epsilon + \frac{1}{2}(1 + \sqrt{\epsilon})^2 e^{-2i\omega\sqrt{\epsilon}L/c} + \frac{1}{2}(\sqrt{\epsilon} - 1)^2 e^{2i\omega\sqrt{\epsilon}L/c}}{\sin(\omega\sqrt{\epsilon}L/c)[(1 + \sqrt{\epsilon})^2 e^{-i\omega\sqrt{\epsilon}L/c} - (\sqrt{\epsilon} - 1)^2 e^{i\omega\sqrt{\epsilon}L/c}]}. \tag{6}$$

In the case of a thin slab, $\omega L/c \ll 1$, the answer depends on the parity of the number of layers N.

For N odd:

$$\frac{E^r_y}{E^r_z} \simeq \frac{\gamma a s_x}{\epsilon - 1}\left(\frac{1}{L} + \frac{1}{2} i(\epsilon - 1)\frac{\omega}{c}\right). \tag{7a}$$

For N even:

$$\frac{E^r_y}{E^r_z} \simeq -i\frac{\gamma a \omega s_x}{c(\epsilon - 1)}. \tag{7b}$$

In the odd case the first term in the large parentheses is dominant ($\omega L/c \ll 1$) and clearly represents the effect of the first ferromagnetic layer. It is proportional to the slab's total magnetization, and if absorption is absent then γ is imaginary and (7a) describes elliptically polarized light—the Kerr reflection from a ferromagnet [8]. For an even number of layers in the absence of dissipation (7b) gives a linearly polarized light. It might be stressed here that (7b) describes a "bulk" effect, with both components of the field proportional to the slab thickness L. In contrast, in the odd case the y component does not depend on L.

In the thick slab, $\omega L(\text{Im}\sqrt{\epsilon}/c) \gg 1$, the reflected wave polarization is

$$\frac{E^r_y}{E^r_z} \simeq -i\frac{\gamma a \omega s_x}{c(\epsilon - 1)}, \quad L \to \infty. \tag{8}$$

The polarization of the transmitted wave is given by

$$\frac{E^t_y}{E^t_z} = i\frac{1}{2}\gamma\frac{\omega a}{c} s_x[1 - (-)^N]\frac{(\sqrt{\epsilon} - 1)e^{i\omega L\sqrt{\epsilon}/c} + (\sqrt{\epsilon} + 1)e^{-i\omega L\sqrt{\epsilon}/c}}{(\sqrt{\epsilon} + 1)^2 e^{-iL\omega\sqrt{\epsilon}/c} - (\sqrt{\epsilon} - 1)^2 e^{i\omega L\sqrt{\epsilon}/c}}. \tag{9}$$

There is a Faraday type rotation which does not depend crucially on the slab thickness. For $\omega L/c \ll 1$:

$$\frac{E^t_y}{E^t_z} \simeq i\frac{1}{4}\gamma\frac{\omega a}{c} s_x[1 - (-)^N], \tag{10a}$$

for $\omega L(\text{Im}\sqrt{\epsilon}/c) \gg 1$:

$$\frac{E^t_y}{E^t_z} \simeq i\frac{1}{2}\frac{\gamma}{c}\gamma\frac{\omega a}{\sqrt{\epsilon} + 1} s_x[1 - (-)^N]. \tag{10b}$$

Inspection of the formulas (6)–(10) reveals their *microscopic* nature: The polarization in transmission and in reflection from this samples fluctuates wildly with the number N of ferromagnetic layers, and the sign of rotation is defined by the sign of the magnetization of the first layer on the left. Obviously, it implies that the experimental data will be extremely sensitive to the surface roughness and to antiferromagnetic domains inside the sample. Some of the physics contained in our model was also mentioned in Ref. [10]. Results qualitatively similar to ours, but using a different model, appeared in Ref. [11].

Next we considered another simple model: a spin-density wave (SDW), $s(x)$. The antisymmetric part of the dielectric function in (4) is now $\epsilon^a_{ij}(x) = \gamma\epsilon_{ijk}s_k(x)$, and identical calculations give for the reflection polarization of the thick slab

$$\frac{E^r_y}{E^r_z} = 2i\frac{\omega\gamma}{c(\epsilon - 1)}\int_0^\infty dx\, s_x(x)\exp\left(2i\frac{\omega}{c}\sqrt{\epsilon}x\right). \tag{11}$$

The exponential in (11) ensures convergence of the integral for a periodic $s(x)$. Equation (11) permits us to account for possible distortions of the SDW near the sample surface.

The only experiment [4] on the nonreciprocal rotation in antiferromagnets was performed on Cr_2O_3. The rotation angle ϕ was found to be of the order 10^{-4}–10^{-5}, in rough agreement with (6) which predicts the rotation of the order $\omega a/c$, 10^{-3}–10^{-4} in the optical range. For detailed comparisons one has to take into account a specific property of Cr_2O_3. This oxide is the so-called magnetoelectric; it may be electrically polarized by magnetic fields and magnetized in electric fields. A great deal is known about magnetoelectrics (see, e.g., [8,9,12]. In all of them, both time and space parity are violated on the *macroscopic* scale, thus activating two new macroscopic mechanisms of nonreciprocal rotation. First, due to P,T violation, the linear space dispersion is now permitted by the Onsager relations (1). The corresponding rotation was calculated by Hornreich and Shtrikman [13]; however, the result is virtually indistinguishable from (8), because generally the strength of the bulk space dispersion is suppressed by the same factor $\omega a/c$. Obviously, you cannot easily test the *macroscopic* T,P violation in this way.

The second mechanism is truly macroscopic. In a genuine magnetoelectric, the inductions D,B and fields E,H are related by

$$\begin{aligned} D_i &= \epsilon_{ij}E_j + \alpha_{ij}H_j\,, \\ B_i &= \alpha_{ij}E_j + \mu_{ij}H_j\,, \end{aligned} \tag{12}$$

where α_{ij} is the so-called magnetoelectric tensor (see, e.g., [8]). Onsager's constraints for the tensor $\hat{\mu}$ can be written in exactly the same way as (1), while for the magnetoelectric tensor we may write

$$\alpha_{ij}(\omega,k,l) = -\alpha_{ji}(\omega,-k,-l)\,. \tag{13}$$

Applying P to the right hand side of (1) and (13) (l changes sign under P), we find that all three tensors, $\hat{\epsilon}$, $\hat{\mu}$, and $\hat{\alpha}$, are symmetrical. The lack of any antisymmetric part forbids the *bulk* optical rotation, and therefore there is no rotation in transmission (see, e.g., [9,12]). The constitutive relations (12) give rise to nonreciprocal effects in propagation and reflection (see, e.g., [14]), and the calculations in the geometry of Fig. 1 for the wave propagating along the threefold axis of Cr_2O_3 give the following value for the rotation in reflection from the thick sample [14]:

$$\frac{E_y^r}{E_z^r} \simeq -\frac{2\alpha}{\epsilon - 1}\,, \quad \alpha \ll 1\,, \tag{14}$$

where α is the only relevant component of the tensor α_{ij}. The experimental values of α in all known magnetoelectrics do not exceed 10^{-2} [11]. The difference between (8) and (13) is clearly visible. If T (and P) is broken macroscopically, the rotation (13) remains finite at low frequencies and is defined by the static value of α. The contribution (8) of microscopic nature goes to zero with $\omega \to 0$. The combination of both can be written as

$$\frac{E_y^r}{E_z^r} = -\frac{2\alpha(\omega) + i\overline{\gamma}(\omega)a\omega/c}{\epsilon(\omega) - 1}\,, \tag{15}$$

where $\overline{\gamma}(\omega)$ may be treated as the new phenomenological quantity, roughly representing the rotating power of a single ferromagnetically polarized layer. The truly theoretical distinction between the two terms in the numerator in (14) is in the microscopic factor $a\omega/c$. Unfortunately, nature plays a trick and gives the majority of the known magnetoelectric coefficients α of the same order of magnitude as $a\omega/c$ in the optical region.

That is a challenge for experimentalists. They prefer to work with magnetoelectric samples, because it is possible to get rid of antiferromagnetic domains using the well developed thermal treatment in electric and magnetic fields [11]. To separate the macroscopic and microscopic contributions to rotation they have to abandon the classic Cr_2O_3, where the experimental data [4] can be accounted for by $\approx$50% of the static value of α and to turn to its more exotic brethren with weaker static magnetoelectricity. (The known values of α are in the broad range 10^{-6}–10^{-2}.)

To summarize, we discuss how the hidden microscopic time parity violation in classic Néel antiferromagnets is revealed in optical experiments measuring nonreciprocal rotation. The rotation angles found are small, $\sim\omega a/c \approx 10^{-3}$–$10^{-4}$ in the optical region. The results are extremely sensitive to the sample surface roughness and to the presence of antiferromagnetic domains.

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