Nonreciprocal rotation of the polarization plane of light in the antiferromagnet Cr_2O_3 which is linear and quadratic in the electric field

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Effects of a nonreciprocal rotation of the polarization plane of light with a linear dependence and also with a quadratic dependence on the electric field have been observed. These effects are comparable in magnitude. The quadratic effect is most obvious during a switching of antiferromagnetic domains and during magnetic ordering near T_N.

A nonreciprocal rotation of the polarization plane of light is caused by an antisymmetric dielectric tensor ϵ_{ij}^{α}, and can be observed in all crystals in an external magnetic field or in magnetically ordered crystals when there is a magnetic moment. Upon the transition of the crystal to a magnetically ordered state, however, the time-reversal operation I' is lost, and it becomes possible to observe some new optical effects, in particular, a nonreciprocal rotation of the polarization plane of light in crystals in an external electric field.[1–3] Let us examine the effects which are determined by an expansion of the form

$$\epsilon_{ij}^{\alpha} = \gamma_{ijk}E_k + \nu_{ijkl}E_kE_l + \eta_{ijklm}H_kE_lE_m, \qquad (1)$$

where γ_{ijk} and ν_{ijkl} are polar c-tensors,[2] and η_{ijklm} is an axial i-tensor. The tensor γ_{ijk} is forbidden by the inversion $\overline{I}$ but allowed by the anti-inversion $\overline{I}'$, and a nonreciprocal rotation of the polarization plane of light, which is linear in the electric field, should be observed in crystals that exhibit a linear magnetoelectric effect.[4] In contrast, the tensor ν_{ijkl} is forbidden by the element $\overline{I}'$ but allowed in the presence of an element $\overline{I}$, i.e., in piezomagnetic materials. This effect has been observed previously in the ferrimagnet yttrium iron garnet.[5] The tensor η_{ijklm} is nonzero in all media.

To study the nonreciprocal rotation of the polarization plane of light in an electric field, we selected the antiferromagnet Cr_2O_3. Below $T_N \cong 307$ K, the spins of the sublattices are ordered antiferromagnetically along the trigonal axis, and the magnetic group of the crystal is[4] $\bar{3}'m'$. A report[6] of a linear effect in this crystal cannot be accepted as adequate at present, since that study was carried out with a thin crystal (4 μm thick), and surface effects may have been pronounced. The sensitivity and accuracy of the measurements were not high, since an intensity method was used. The effect was found to disappear not at T_N, as one would expect, but at a much lower temperature. We know of no other reports of the observation of a nonreciprocal rotation of the polarization plane of light of second order in the electric field in Cr_2O_3 or other antiferromagnets.

The present experiments are carried out on an apparatus similar to that described

in Ref. 5. The sensitivity to changes in the rotation of the polarization plane is no worse than 0.05″. The error is no worse than 15%. A signal is detected simultaneously at the first harmonic ($\omega = 700$ Hz) and at the second harmonic (2ω) of the alternating electric field $E_z = E_0 \cos \omega t$. The samples are polished parallel plates 0.6–1.1 mm thick, cut perpendicular to the optic (z) axis with an orientation error no worse than 1°. The electric field is applied to semitransparent platinum electrodes deposited on the plate. A magnetic field up to 3.3 kOe is applied along the z axis. The focused beam from a helium-neon laser, with a wavelength $\lambda = 1.15$ μm, passes along this direction through the crystal. As a result, the geometry $\mathbf{k}\|\mathbf{z}\|\mathbf{E}\|\mathbf{H}$ is achieved in this experiment; in this geometry, the nonreciprocal rotation of the polarization plane of the light is determined by the component ϵ^{α}_{xy}.

Preliminary studies of the magnetoelectric annealing[7] of this crystal made it possible to quantitatively determine the conditions under which the crystal undergoes a transition to either of the two possible single-domain antiferromagnetic states, l^- or l^+. It was found that the crystal has a tendency toward the formation of a stable l^- domain, to which the crystal undergoes a transition without a bias electric field. A procedure has also been developed for causing a transition of the crystal from the unstable single-domain state l^+ to a multidomain (l^+,l^-) state by changing the temperature; this transition is followed by a transition to a stable l^- state. Figure 1 shows the results of a study of the nonreciprocal rotation of the polarization plane of light as a function of the frequency, $NR(\omega)$, at the first harmonic of the alternating electric field, when the crystal is in different states. When the crystal is in the l^- or l^+ single-domain state, the magnitude of the nonreciprocal rotation can be described quite accurately as a linear function of the electric field (see the inset in Fig. 1) The sign of

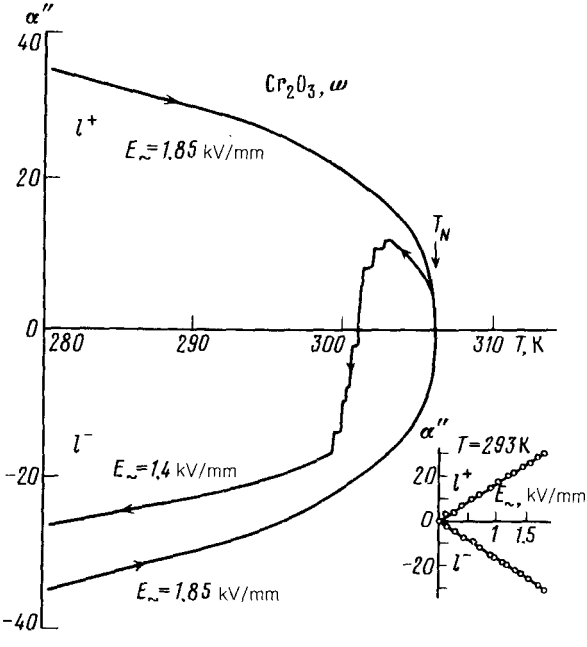

FIG. 1. Temperature dependence of $NR(\omega)$ for a Cr_2O_3 crystal in different antiferromagnetic states. The thickness of the crystal is 612 μm. The inset shows the field dependence (the dependence on the electric field) of $NR(\omega)$ in the two antiferromagnetic states, $l^{\pm}$.

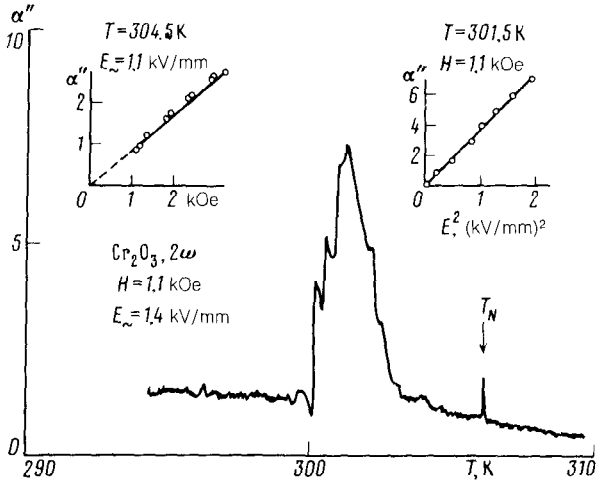

FIG. 2. Temperature dependence of $NR(2\omega)$ corresponding to the curve for $E = 1.4$ kV/mm in Fig. 1. The inset shows the $NR(2\omega)$ signal as a function of the magnetic and electric fields.

the effect is opposite in the l^- and l^+ states. There is no dependence on the magnetic field; i.e., this effect is determined by the tensor γ_{ijk} from (1). The magnitude of the nonreciprocal rotation at 293 K is 0.025″/V. The central curve in Fig. 1 shows the change in $NR(\omega)$ upon the transition from the unstable state to the stable l^- state as the temperature is changed. The transition temperature depends on the conditions of the magnetoelectric annealing.

An unexpected result is the observation of a signal $NR(2\omega)$—at the frequency 2ω. The temperature and field dependence of this signal is shown in Fig. 2. The strongest $NR(2\omega)$ signal, with a strength comparable in magnitude to that of $NR(\omega)$, is observed near the transition from the l^+ state to the l^- state. The $NR(2\omega)$ signal

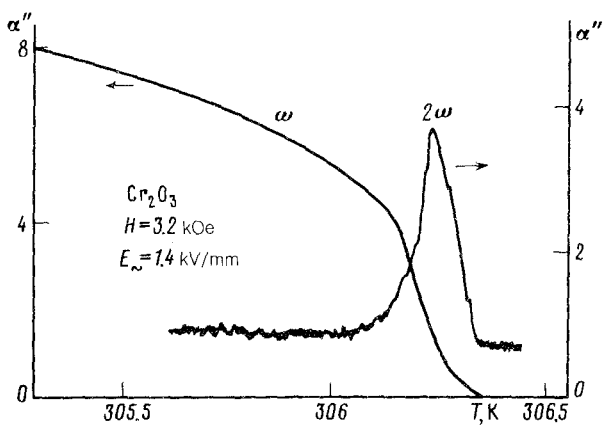

FIG. 3. Temperature dependence of $NR(\omega)$ and $NR(2\omega)$ near T_N.

depends linearly on the magnetic field and quadratically on the electric field (see the inset in Fig. 2); i.e., this effect is determined by the tensor η_{ijklm} in (1). Furthermore, the $NR(2\omega)$ signal is seen as a narrow peak in a temperature interval ~ 0.2 K near T_N, where there is a well-defined change in slope, and there is a sharp decrease in $NR(\omega)$ (Fig. 3). In this region, the $NR(2\omega)$ signal is again a linear function of the magnetic field and a quadratic function of the electric field; i.e., this effect is described phenomenologically by the same tensor that describes the $NR(2\omega)$ signal upon a temperature-induced switching of antiferromagnetic domains.

The appearance of an $NR(2\omega)$ signal, which is comparable in magnitude to the $NR(\omega)$ signal, can be explained in the following way. In a multidomain sample, the nonreciprocal rotation can be described by

$$\alpha = AE_0 \cos \omega t \, [S^+(E) - S^-(E)] , \tag{2}$$

where $S^{\pm}$ is the total area of the $l^{\pm}$ domains. As the temperature is changed, the areas of the $l^{\pm}$ domains change by virtue of the magneto-electric effect:

$$S^+(E) = S^+(0) + bE_0 \cos \omega t; \quad S^-(E) = S^-(0) - bE_0 \cos \omega t. \tag{3}$$

Substituting (3) into (2), we find the following expressions for the reciprocal rotation at the first and second harmonics:

$$\alpha(\omega) = AE_0 \cos \omega t \, [S^+(0) - S^-(0)]; \quad \alpha(2\omega) = AbE_0^2 \cos 2\omega t. \tag{4}$$

It follows from these results that the first harmonic, $NR(\omega)$, vanishes at that temperature at which the areas of the positive and negative antiferromagnetic domains are equal. At the same temperature, $NR(2\omega)$ reaches a maximum, when the changes in the areas of the $l^{\pm}$ domains are at their greatest. The narrow $NR(2\omega)$ peak near T_N is apparently due to an instability of the domain structure or an instability of the nucleating centers of the ordered phase in the paramagnetic phase. The jump in $NR(\omega)$ and the maximum of $NR(2\omega)$ near T_N may be evidence that the magnetic ordering in Cr_2O_3 is a first-order phase transition which is nearly of second order.

The mechanism proposed here for the nonlinearity should give rise to a nonlinear magnetoelectric effect (an invariant of the type H^2E^2) in Cr_2O_3, which is comparable in magnitude to the linear effect[8] in a static magnetic field and also in alternating fields in the sonic and rf frequency ranges.

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