Linear magnetoelectric effect in Cr_2O_3 in strong magnetic fields

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The longitudinal and transverse magnetoelectric effects have been studied in Cr_2O_3 in a strong magnetic field. These are the first such experiments. The longitudinal effect disappears, and the transverse effect appears, in a spin-flip phase. The temperature dependence of the corresponding magnetoelectric susceptibilities, α_{zz} and α_{xz}, has been measured.

The magnetoelectric effect in Cr_2O_3 was first predicted theoretically in 1959, by Dzyaloshinskiĭ.[1] It was observed experimentally by Astro[2,3] in 1960. Since then, several experimental and theoretical studies[4,5] of this effect in Cr_2O_3 have been reported. This particular compound is regarded as a classic material exhibiting a magnetoelectric effect. Most of this research has been carried out on the magnetoelectric effect in a weak magnetic field, i.e., for the undeformed antiferromagnetic structure of Cr_2O_3. We know of only one study[6] in which the magnetoelectric susceptibility $dP(H)/dH$ of Cr_2O_3 has been measured in strong magnetic fields. Questions which we regard as particularly interesting are the relationship between the magnetoelectric effect and the magnetic structure, which can be altered by a strong magnetic field, and (particularly) the magnetoelectric effect in a spin–flip phase. In this letter we are reporting an experimental study of these problems.

We studied the longitudinal and transverse magnetoelectric effects, i.e., the electric polarization $\vec{P}$ induced by a pulsed magnetic field $\vec{H}$, in fields up to 150 kOe. The field was directed along various crystal axes. Measurements were carried out over the temperature range 4.2–310 K. The Cr_2O_3 test crystal was a small cube (with linear dimensions of $1.5\times1.5\times1.5$ mm). The edges of the cube coincided with the a, b, and c axes in the orthohexagonal coordinate system (a is one of the twofold axes in the basal plane, which is perpendicular to the rhombohedral c axis of the crystal). The crystal was oriented by an x-ray technique within ~2°; the rhombohedral c axis was distinguished within 1°. In this study, we did not carry out any special E–H annealing to stablize the domain structure.

To measure component i ($i = x,y,z$) of the polarization, P_i, in the plane perpendicular to the i axis, we used some epoxy resin with a conducting filler to deposit some electrodes. The voltage V_i from these electrodes was sent through an electrometer cascade to the Y plates of an oscilloscope. The voltage across the electrodes, $V_i \propto P_i$, was displayed as a function of the external magnetic field on the oscilloscope screen. In the experiments we actually used some existing apparatus for measuring magnetostriction by means of quartz piezoelectric transducers.[7] A similar measurement method was used in Ref. 8, in an independent study, carried out slightly before our own. By canceling the input capacitance of the electrometer cascade we were able to significantly improve the sensitivity of the apparatus. It then became possible to reliably detect a surface charge $\sim 10^{-7}$ C/m^2.

The inset in Fig. 1 shows a typical plot of the longitudinal magnetoelectric effect, $P_z(H_z)$, for the case in which H is oriented along the rhombohedral c axis, at $T \simeq 248$ K. Below the critical field, the polarization P_z is a linear function of the magnetic field.

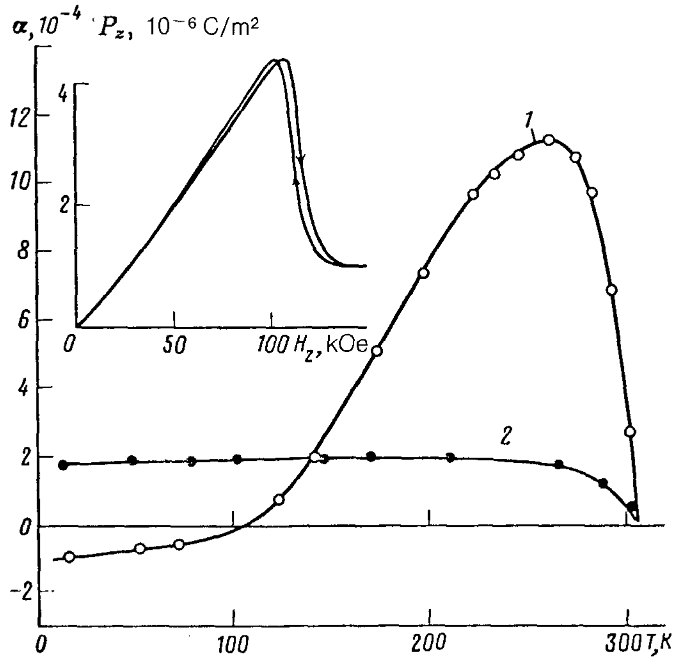

FIG. 1. 1—Temperature dependence of the longitudinal magnetoelectric susceptibility, α_{zz}, in fields below the spin–flip transition; 2—transverse magnetoelectric susceptibility α_{xz}, in fields above the spin–flip transition. The magnetic field was oriented along the rhombohedral axis of the Cr$_2$O$_3$ crystal. The inset shows the magnetic-field dependence of the longitudinal magnetoelectric effect, $P_z(H_z)$, along the rhombohedral axis at 248 K.

It decreases abruptly at $H = H_{cr} = 120$ kOe. When the magnetic field is reversed, the polarization also changes sign, i.e., the contribution to P_z stems from a magnetoelectric effect which is linear in H. The critical field H_{cr} coincides with the spin-flip field which has been observed previously in measurements of the magnetic properties, the antiferromagnetic resonance,[9] magnetostriction,[10] and the magnetoelectric susceptibility.[6]

We think that the small residual polarization on the $P_z(H_z)$ curves at $H > H_{cr}$ is due to a small twin in the crystal used in the measurements. Unfortunately, we were not able to isolate this twin by an x-ray method. The presence of a twin complicates a quantitative interpretation of the experimental results. Nevertheless, if we use the abrupt change in the polarization at the spin–flip transition, normalized to the field value H_{cr}, to calculate the magnetoelectric susceptibility $\alpha_{ij} = P_i/H_j$, we can essentially eliminate the twin contribution to the polarization, $\Delta P'_z$, which is linear in the magnetic field.

As the temperature is lowered, the abrupt drop on the $P_z(H_z)$ curve decreases in magnitude, reaching zero at $T \simeq 100$ K and then changing sign at even lower temperatures. The spin–flip transition exhibits a significant hysteresis, ~8 kOe. The slight deviation ~(5–6)° of the magnetic field from the rhombohedral axis has no great effect on the magnitude or shape of $P_z(H_z)$. The temperature dependence of the magnetoelectric susceptibility α_{zz} in fields below the spin-flip transition, as found from the abrupt drop on the $P_z(H_z)$ curve (Fig. 1), agrees well with results found in measurements of both electrically induced and magnetically induced (in weak fields) magnetoelectric effects.[4]

These results thus show that the longitudinal magnetoelectric effect $P_z(H_z)$, along the rhombohedral axis of the crystal, disappears in the spin-flip phase, and the corresponding magnetoelectric susceptibility α_{zz} vanishes. We know that the magnetoelectric susceptibility tensor of Cr_2O_3 in weak magnetic fields has only two independent longitudinal components, α_{zz} and $\alpha_{xx} = \alpha_{yy}$. The particular method which we used to measure the magnetoelectric effect is incapable of studying the behavior of α_{xx} and α_{yy} in the spin–flip phase. Nevertheless, it can be concluded from symmetry considerations that α_{xx} and α_{yy} vanish at $H > H_{cr}$.

We would naturally ask about the form of the magnetoelectric susceptibility tensor α_{ij} in the spin–flip phase. It was pointed out in the first paper by Astrov[3] that when the magnetic structure is altered, and a projection of the magnetic moment of the Cr^{3+} ions onto the basal plane appears (this plane is perpendicular to the rhombohedral axis), terms of the type $\alpha_{xy}E_xH_y$ and $\alpha_{xz}E_xH_z$ arise in the interaction Hamiltonian, and a transverse magnetoelectric effect should arise.

Figure 2 shows isotherms of the transverse magnetoelectric effect, $P_x(H_z)$, for the case in which H is along the rhombohedral c axis. Above the critical field H_{cr}, a negative polarization arises abruptly. It then increases in absolute value as the field is raised further. We believe that the nonzero polarization $P_x(H_z)$ at $H < H_{cr}$ as well as the longitudinal magnetoelectric effect $P_z(H_z)$ result from the crystal twin. The actual $P_x(H_z)$ dependence, found by subtracting the linear component of the polarization, from the twin, $\Delta P'_x$ (curve 3), turns out to be proportional to the magnetic field in fields above H_{cr}: $P_x(H_z) = \alpha_{xz}H_z$ (the dashed line in Fig. 2).

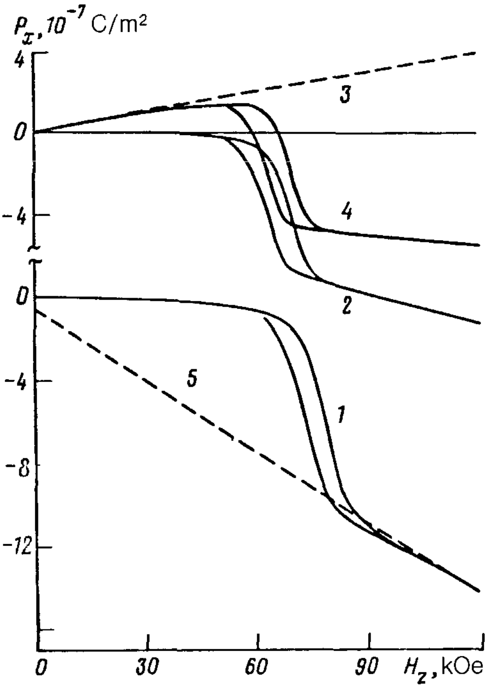

FIG. 2. Isotherms of the transverse magnetoelectric effect $P_x(H_z)$ along the *a* axis for the case in which the magnetic field is along the rhombohedral axis. 1—$T = 139$ K; 2, 3, 4—$T = 11$ K; 3—Contribution from the crystal twin; 4—experimental $P_x(H_z)$ curve with the contribution from the crystal twin; 5—linear extrapolation of $P_x(H_z)$ to $H > H_{\rm cr}$.

As the temperature is raised, the size of the abrupt drop on the $P_x(H_z)$ curve and also the field of the spin–flip transition increase in such a way that the magnetoelectric susceptibility α_{xz}, as determined from the abrupt drop in the polarization, and normalized to the field $H_{\rm cr}$, depends only weakly on the temperature up to 250 K. It gradually vanishes near $T_N \simeq 308$ K (Fig. 1). Consequently, the value of α_{xz} in the spin–flip phase varies with the temperature in the same way as α_{xx} in weak fields.[3] In other words, it is proportional to the magnetization of the sublattice. Interestingly, the absolute values of α_{xz} are also close to those of α_{xx} (in comparison with α_{zz}).

Correct measurements of the temperature dependence of α_{xz} near T_N, where the longitudinal magnetoelectric susceptibility α_{zz} is more than an order of magnitude larger than the transverse susceptibility α_{xz}, and where it also changes abruptly at the spin–flip transition, require that the direction of the polarization $\vec{P}$ in the basal plane be measured carefully (within 1°). Since the overall error in the orientation in our experiments was 2°–3°, the results found on the temperature dependence of α_{xz} in the temperature range 200–290 K should be regarded as qualitative.

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