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MAGNETOELECTRIC EFFECT OF Cr_2O_3 IN STRONG STATIC MAGNETIC FIELDS

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The magnetoelectric effect of Cr_2O_3 single crystals has been studied in magnetic fields up to 20 T in the range of liquid helium to room temperature. In the antiferromagnetic phase for low magnetic fields the well known results for the magnetoelectric effect below the Néel temperature were reproduced. In the spin-flop phase, for magnetic fields above a critical field ($H > H_{crit} = 10$ T), the magnetic field induced electric polarization has been measured along all three crystallographic directions. A magnetoelectric effect, linear in the magnetic field was found for the three orientations leading to the conclusion that the magnetic ground state symmetry of the spin-flop phase is either $\bar{1}'$ or 2/m', where 2'/m can be excluded.

Keywords: Cr_2O_3, magnetoelectric effect, high magnetic fields, spin-flop phase.

I. INTRODUCTION

The magnetoelectric (ME) effect in Cr_2O_3 is a well studied phenomenon. It was not only the first compound for which the ME effect was theoretically predicted in its antiferromagnetic phase,[1] it was also the first compound where the effect was experimentally shown to exist. First the electrically induced ME effect ($(ME)_E$) was discovered[2] quickly followed by the magnetically induced effect ($(ME)_H$).[3] The experimental studies concentrated increasingly on the $(ME)_H$ effect, not only because they are easier to realize, but also does Cr_2O_3 undergo a spin-flop (SF) transition in strong external magnetic fields. So far only few experimental studies are known to have been performed in high enough magnetic fields,[4–6] using mostly pulsed magnetic fields. We have studied the $(ME)_H$ effect in static magnetic fields up to 20 T, exceeding the critical field largely, in order to study the magnetic symmetry of the SF phase.

II. THEORY

Cr_2O_3 is a Heisenberg antiferromagnet with a Néel temperature of $T_N = 308$ K. In zero magnetic field the Cr^{3+} spins align themselves parallel to the threefold rhombohedral *c*-axis, forming an "*easy-axis*" antiferromagnet. The corresponding magnetic symmetry is $\bar{3}'m'$ which is compatible with a linear ME effect with two independent nonzero components of the ME tensor:

$$\alpha_{xx} = \alpha_{yy} = \alpha_{\perp} \quad \text{and} \quad \alpha_{zz} = \alpha_{\parallel}, \tag{1}$$

where $\alpha_{ij} = \delta P_i/\delta H_j$; $i, j = x, y, z$ and P being the electric polarization in an applied magnetic field H. A small external magnetic field parallel to the *c*-axis lowers the symmetry to 3m′ without affecting the form of the ME tensor.

When the external magnetic field is further increased and reaches a critical field, a SF transition occurs. Above this critical field ($H > H_{\text{crit}} = 10$ T) the spins are found to be oriented perpendicular to the *c*-axis in the basal plane. This SF transition has been studied by means of antiferromagnetic resonance[7] and ultrasonic attenuation.[8] These experiments were not able to determine the direction of the spins in the basal plane. The existence of two inequivalent spin directions in the basal plane would be reflected by different magnetic symmetry classes. Since the ME effect is sensitive to magnetic symmetries such measurements can help to clarify the magnetic structure.

For an antiferromagnetic ordering of the spins in the basal plane three spin configurations are possible. Either the spins order in the basal plane parallel to a mirror plane (*a*-direction) leading to a magnetic class of 2/m′ or parallel to a twofold axis (*b*-direction) leading to a magnetic class of 2′/m. A further possible magnetic structure has the spins in the SF phase ordered in an arbitrary direction in the basal plane. The corresponding magnetic class being $\bar{1}'$. The three possible magnetic ground states in the SF phasc would lead to different forms of the linear ME tensor:

$$2'/\text{m};\ \begin{pmatrix} 0 & \alpha_{xy} & 0 \\ \alpha_{yx} & 0 & \alpha_{yz} \\ 0 & \alpha_{zy} & 0 \end{pmatrix}, \quad 2/\text{m}':\ \begin{pmatrix} \alpha_{xx} & 0 & \alpha_{xz} \\ 0 & \alpha_{yy} & 0 \\ \alpha_{zz} & 0 & \alpha_{zz} \end{pmatrix}, \quad \bar{1}':\ \begin{pmatrix} \alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\ \alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\ \alpha_{zx} & \alpha_{zy} & \alpha_{zz} \end{pmatrix}. \tag{2}$$

Since the SF phase of Cr_2O_3 can only be reached by applying a magnetic field parallel to the *z*-axes, the only tensor elements which can be experimentally tested are α_{xz}, α_{yz}, and α_{zz}. According to (2) the behaviour of the ME effect of these three orientation should contain enough information to conclude on the magnetic symmetry of the SF phase.

III. EXPERIMENT

The Cr_2O_3 crystals used for the measurements were two single crystals cut from the same specimen. One crystal was disc shaped with a diameter of 2.90 mm and 0.26 mm in thickness. The rhombohedral *c*-axis of the crystal was within ~3.5° along the disc axis as verified by a Laue X-ray technique. The second crystal was cube shaped of dimensions 2.25 × 0.96 × 1.28 mm, where the edges coincided within ~3° with the crystallographic axes. Electrodes were deposited by silver paint on the appropriated faces and the contacted samples were fixed to the sample holder by varnish. The polarization of the sample was measured by a dc-technique, where the current between the electrodes is measured and integrated leaving the sample at zero electric field. The integrated value is scaled and displayed as charge. In this way charges of 0.01 pC were detectable. The measurements were performed in a resistive Bitter magnet providing static magnetic fields up to 20 T. The temperature was varied in a helium flow cryostat and measured by a calibrated platinum-resistance thermometer.

We investigated all nine elements of the ME tensor, but only the components related to the symmetry of the SF phase will be presented. A ME annealing procedure, which could change the domain structure by applying simultaneously a magnetic and electric field while cooling the sample through T_N, did not have any influence on the amplitude or the sign of the ME signals in our experiment.

IV. RESULTS

The results for the antiferromagnetic phase of Cr_2O_3 agree very well with the results reported in the literature. Figure 1 shows a typical plot for the electric polarization versus the magnetic field for $P_z(H_z)$. For low magnetic field ($H < 6$ T) the polarization is a linear function of the magnetic field. The perpendicular ME effects

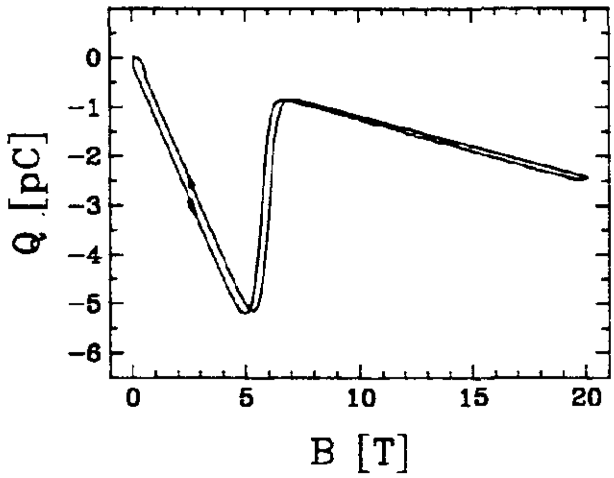

FIGURE 1 The electric polarization $P_z(H_z)$ in the z-direction versus the magnetic field applied parallel to the z-direction at 4.2 K.

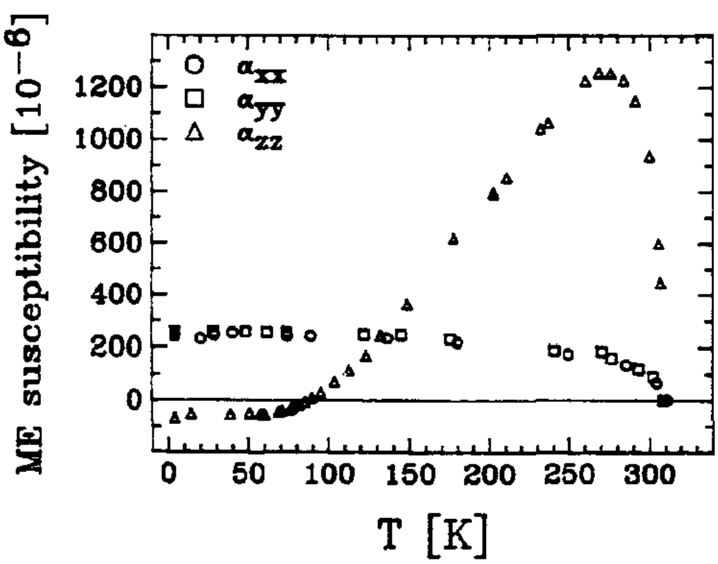

FIGURE 2 Temperature dependence of the magnetoelectric susceptibilities $|\alpha_{xx}|$, $|\alpha_{yy}| = |\alpha_{\perp}|$ and $\alpha_{zz} = \alpha_{\parallel}$ in fields below the spin flop transition.

TABLE I
A comparison of reported results for the ME susceptibilities in the low field antiferromagnetic phase of single crystal Cr_2O_3

Ref	$\alpha_\perp$ (4.2 K)	$\alpha_\parallel$ (4.2 K)	$\alpha_\parallel^{max}$	$T_{\alpha=max}$ [K]	$T_{\alpha=0}$ [K]
9	-0.8×10^{-4}	-0.4×10^{-4}	5.8×10^{-4}	255	80
10	$\lvert\alpha_\perp\rvert = 0.16\times10^{-4}$	—	1.8×10^{-4}	260	97
11	-4.7×10^{-4}	-0.8×10^{-4}	15.3×10^{-4}	255	87
12	—	-1.2×10^{-4}	12.4×10^{-4}	263	85
6	—	-1.0×10^{-4}	12.0×10^{-4}	260	100
This work	$\lvert\alpha_\perp\rvert = 2.2\times10^{-4}$	-0.7×10^{-4}	12.5×10^{-4}	270	87

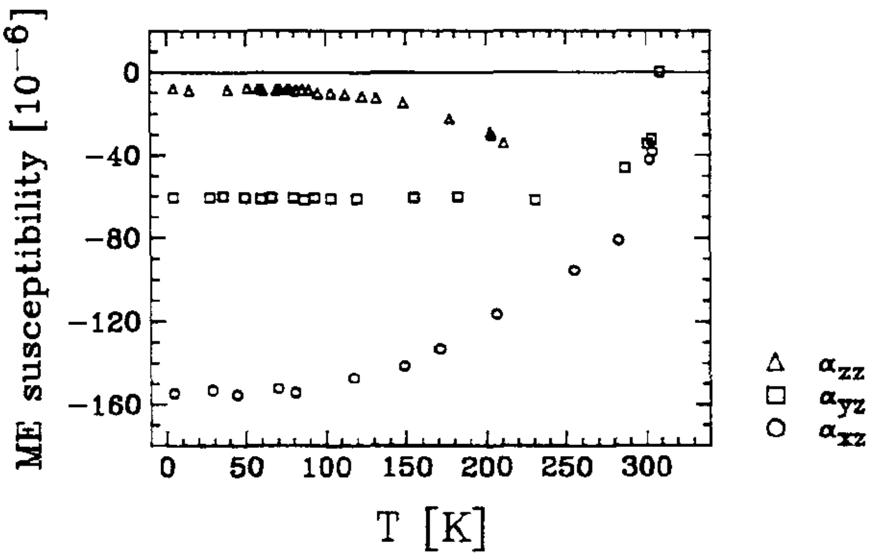

FIGURE 3 Temperature dependence of the magnetoelectric susceptibilities α_{xz}, α_{yz} and α_{zz} in fields above the spin flop transition.

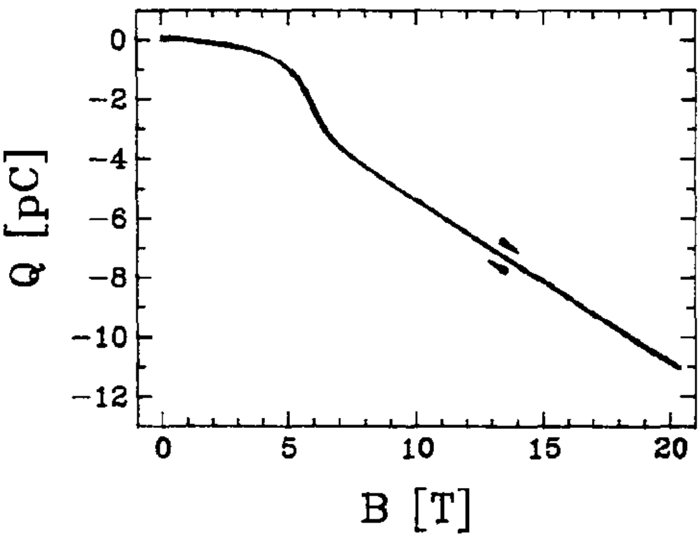

FIGURE 4 The electric polarization $P_y(H_z)$ in the y-direction versus the magnetic field applied parallel to the z-direction at 49.4 K.

showed an electric polarization $P_x(H_x)$ and $P_y(H_y)$ linear in the magnetic field up to the highest fields of 20 T. The temperature dependences of the absolute values of the corresponding ME susceptibilities α_{xx}, α_{yy} and α_{zz} are shown in Figure 2. All ME effects disappear at T_N, which was in our crystal found to be at 307.9 K. For a direct comparison with reported results on Cr_2O_3 single crystals, some values characterizing the temperature dependence of the ME susceptibilities extracted from Figure 2 are shown in Table I, such as $\alpha_\perp$ and $\alpha_\|$ at 4.2 K, the maximum value of $\alpha\|$ at the temperature $T_{\alpha=\max}$ and the temperature for $\alpha_\| = 0$ ($T_{\alpha=0}$). All values are given in cgs-units to facilitate comparison with the literature. To pass from the value α_{ij} in cgs-units to the value of $\alpha_{ij}^{S.I.}$ one has simply to divide α_{ij} by the velocity of light c. The values become in S.I. units:

$$|\alpha_\perp^{S.I.}(4.2\ \mathrm{K})| = 0.73\ [\mathrm{ps/m}],$$

$$\alpha_\|^{S.I.}(4.2\ \mathrm{K}) = 0.23\ [\mathrm{ps/m}] \quad \text{and}$$

$$\alpha_\|^{\max,S.I.} = 4.14\ [\mathrm{ps/m}].$$

Applying the magnetic field along the z-direction, a SF transition takes place at a critical field ranging from 6.1 T at 4.2 K up to 12.5 T near the Néel temperature.[7,8]

Measuring $P_z(H_z)$, the polarization drops off to zero at the critical field (Figure 1). By increasing the magnetic field even further the polarization becomes again a linear function of the magnetic field with negative sign and stays linear up to the highest fields. At 4.2 K the ME susceptibility α_{zz} in the SF phase has a finite value and increases at 100 K considerably (Figure 3). For temperatures higher than 250 K we could no longer clearly identify the SF phase due to the increase of H_{sf} and the broadening of the transition. Lowering the magnetic field and reentering into the AF phase, no significant change in form and amplitude of $P_z(H_z)$ was observed. Foner *et al.*[4] and Ohtani *et al.*[5] reported also a linear contribution in α_{zz} in the SF phase, however in magnetic fields up to 10.5 T not exceeding far beyond the SF transition.

Measuring the polarization along the x- or y-direction ($P_x(H_z)$, $P_y(H_z)$) changes the behaviour of the ME effect, as shown in Figure 4. For $H < H_{sf}$ a linear ME effect is forbidden by symmetry, in fact only a small contribution is seen, which we relate to a residual contribution of α_{zz} due to misalignment. This interpretation is confirmed by the temperature dependence of α_{xz}, α_{yz} for $H < H_{sf}$. The SF transition can again be clearly observed by a sharp drop of the polarization at $H = H_{sf}$. In the SF phase the polarization $P_x(H_z)$ and $P_y(H_z)$ become again a linear function of the magnetic field up to highest fields. The temperature dependence of α_{xz}, α_{yz} is also shown in Figure 3 together with α_{zz}. At the lowest temperature (4.2 K) the value of α_{xz} is almost three times bigger than α_{yz} although they show roughly the same temperature dependence in contrast to α_{zz}. The ME susceptibility α_{xz} does hardly change up to ~250 K where it drops off quickly for higher temperatures and vanishes at T_N, whereas α_{yz} seems to approach T_N more smoothly. Reentering into the AF phase by lowering the magnetic field does not change the overall behaviour. A linear extrapolation of the electric polarization in the SF phase to $H = 0$ shows that the polarization for all three orientations $P_x(H_z)$, $P_y(H_z)$ and $P_z(H_z)$ would vanish.

The temperature dependence of the ME susceptibilities α_{xz}, α_{yz} in the SF phase seem to be proportional to the magnetization of the sublattices as it is the case for $\alpha_\perp$ in the low field antiferromagnetic phase. The temperature dependence of α_{zz} in the SF phase, in the accessible temperature range, shows a similar temperature dependence as $\alpha_\|$ in Figure 2 for the low field phase, leading to the conclusion that the ME effect for this orientation is proportional to the difference of the sublattice magnetization.

V. CONCLUSION

For all three crystallographic orientations of the electric polarization a linear ME effect has been observed in the SF phase for magnetic fields applied along the easy axis of the crystal. Due to the fact that we measured a linear ME effect in α_{zz} we can exclude the 2′/m magnetic symmetry as the ground state for the spin flop phase. Although the result for the ME tensor components is only compatible with the $\bar{1}'$ magnetic class in (2) for an antiferromagnetic alignment in the basal plane we cannot exclude the possibility that the magnetic symmetry is 2/m′. The existence of antiferromagnetic domains which are not correctly aligned along the mirror planes due to higher anisotropy terms can lead to a projection of α_{xz} into α_{yz}. The similar temperature dependence and the amplitudes of α_{xz} and α_{yz} do not clarify this question.

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