OBSERVATION OF THE MAGNETICALLY INDUCED MAGNETOELECTRIC EFFECT AND EVIDENCE FOR ANTIFERROMAGNETIC DOMAINS

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In this Letter we report the first observation of the magnetically induced magnetoelectric effect [henceforth denoted by $(\mathrm{ME})_H$] in an antiferromagnetic material. When correlated with the previously observed[1,2] electrically induced magnetoelectric effect [henceforth denoted by $(\mathrm{ME})_E$], this observation provides a necessary verification of the existence of EH-type terms[3,4] in the thermodynamic potential of a spin-ordered substance. We also report the discovery of magnetic annealing effects in an antiferromagnet. In conjunction with other structure-sensitive phenomena which we found by means of magnetoelectric (ME) measurements, these magnetic annealing effects will be shown to provide the first strong evidence for the existence of antiferromagnetic "domains." Previous work[5] on such "domains" was largely speculative.

The thermodynamically derived[4] constitutive relations appropriate for the $(\mathrm{ME})_H$ effect are

$$D_z = \epsilon_{\parallel} E_z + \alpha_{\parallel} H_z, \quad D_{x,y} = \epsilon_{\perp} E_{x,y} + \alpha_{\perp} H_{x,y}, \tag{1}$$

where $\alpha_{\parallel}/4\pi$ and $\alpha_{\perp}/4\pi$ are magnetoelectric susceptibilities and the z axis is chosen to be along the principal axis (c axis) of a uniaxial crystal. In our experiments on Cr_2O_3 the applied electric field was zero. Using x-ray-oriented single-crystal disks whose cylindrical axes z' were at an angle θ ($\theta = 0$, 90°, and $\theta_1 \approx 30°$) to the z axis, we switched on a magnetic field H along z' and measured the voltage V which was induced between the silvered plane surfaces of the disks. For fields up to the highest values used ($H \approx 2500$ oe), V was found to be proportional to H. Although V decays with a relaxation time τ determined by the conductivity and dielectric constant of Cr_2O_3, the value of τ was found to be sufficiently large to permit us to measure V with a vacuum tube electrometer (Keithley Instruments, Inc., Model 610R). In the vicinity of room temperature the low-voltage measurements could not be made because of drift in the apparatus. The actual voltage V may not be equal to the open-circuit voltage V^0 (because the resistance of the disks may not be small in comparison to the input impedance of the electrometer), but it is clear the V must be proportional to V^0 and hence [because of Eq. (1)] proportional to α.

Our experimental results show that the temperature dependence of $V_{\parallel}$ and $V_{\perp}$ is essentially the same as that of the $\alpha_{\parallel}$ and $\alpha_{\perp}$ which we deduced from our $(\mathrm{ME})_E$ effect[2] experiments and described satisfactorily in a recent theoretical[6] study. As shown in Fig. 1, the temperature dependence of $|V_{\theta_1}|$ and $|\alpha_{\theta_1}{}^{\mathrm{app}}|$ [which were measured by means of the $(\mathrm{ME})_H$ and $(\mathrm{ME})_E$ effects, respectively] are also in agreement. Since the values of antiferromagnetic permeabilities are close to unity, we find that $\alpha_\theta{}^{\mathrm{app}}$ (the measured value of α_θ) is given by the relation $\alpha_\theta{}^{\mathrm{app}} \approx [1 - (N_{z'}/4\pi)]\alpha_\theta$, where $N_{z'}$ is the axial demagnetizing factor of an oblate spheroid having the same axial ratio as the disks used in the ME measurements. Furthermore, we observed that the signs of V_{θ_1} and $\alpha_{\theta_1}{}^{\mathrm{app}}$ reverse at the same temperature. We also found that the three ratios $|V_{\parallel}|/|\alpha_{\parallel}{}^{\mathrm{app}}|$,

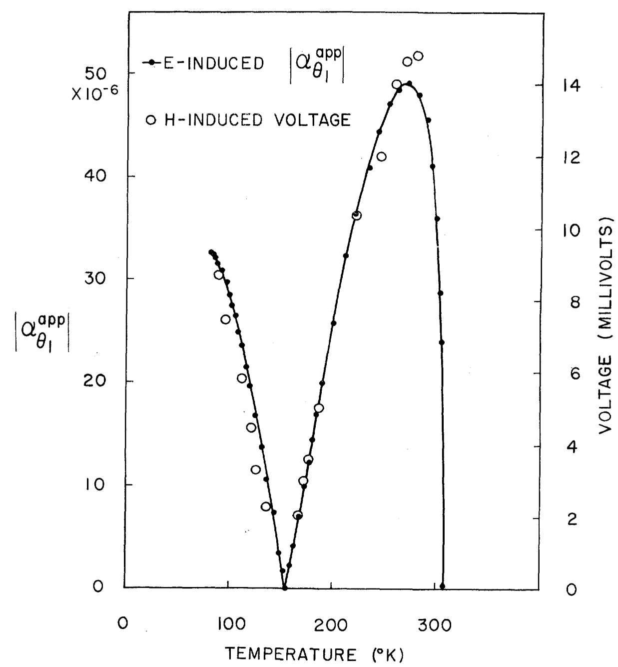

FIG. 1. Comparison of the temperature dependence of $|\alpha_{\theta_1}{}^{\mathrm{app}}|$ and the induced voltage ($|V_{\theta_1}|$). These quantities were measured by means of the $(\mathrm{ME})_E$ and $(\mathrm{ME})_H$ effects, respectively. Both $\alpha_{\theta_1}{}^{\mathrm{app}}$ and V_{θ_1} change sign near 154°K. The angle θ_1 is about 30°.

$|V_\perp|/|\alpha_\perp^{app}|$, and $|V_{\theta_1}|/|\alpha_{\theta_1}^{app}|$ are equal to each other provided the V and α^{app} contained in a given ratio are measured on the same sample.

Using both the $(ME)_H$ and the $(ME)_E$ effects, we observed that the signs and magnitudes of $\alpha_\parallel$ and $\alpha_\perp$, but not their temperature dependence, may vary (a) from sample to sample, and (b) upon cooling a sample through the Néel temperature either with or without the presence of a magnetic field. In view of (a), it is understandable that the relation $\alpha_\theta = \alpha_\parallel \cos^2\theta + \alpha_\perp \sin^2\theta$ (which follows from the form of the α tensor of Cr_2O_3) is not obeyed if the α's are measured on different samples. The magnetic annealing mentioned in (b) caused the α's to increase by factors as large as 3000. In addition, an almost complete "erasure" of $\alpha_\parallel$ was accomplished by cooling a $z \parallel z'$ sample through the Néel temperature in the presence of a 60-cycle/sec magnetic field.

All these observations may be interpreted by postulating the existence of antiferromagnetic "domains." As to the specific nature of the domain structure, we tentatively suggest that in the case of Cr_2O_3, there are two kinds of domains and that they are characterized, respectively, by each spin in a domain pointing toward, or away from, the nearest oxygen plane which is perpendicular to the c axis. The difference between these two kinds of domains results from the asymmetry of the Cr^{+++} sites with respect to the oxygen planes. This asymmetry produces linear terms in the crystalline electric potential, and in a possible mechanism[6] of the ME effects these terms play an essential role and determine the signs of the α's. Consequently, the suggested domain structure explains the fact that the observed α's can have either sign and that (due to partial cancellations resulting from the presence of two types of domains) they can have variations in their magnitudes. Since the two kinds of domains differ solely by a 180° reversal of all spins, it is clear that the intrinsic α's associated with each domain should have the same temperature dependence. Thus we have a natural explanation of the striking fact that the temperature dependence of each observed α_θ does not vary even though its sign and magnitude may vary. Finally, we note that the large increases in the magnitudes of the α's which can be produced by magnetic annealing from a temperature just above the Néel point are difficult to explain without postulating antiferromagnetic domains.

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