The maximum value of 0.1 W for the acoustic power P_f was set by electric breakdown in the cavity. By increasing the breakdown field, one could make the harmonic of the same magnitude as the fundamental for $P_f = 0.5$ W. Harmonic generation can be useful in extending present techniques to the millimeter range, where high-power microwave sources are presently unavailable.

The author is indebted to Dr. J. deKlerk for fabricating the thin film CdS transducer, W. M. Lepsevich for constructing the cavities, and J. Silva for technical assistance.

*Also a doctoral candidate at Department of Physics, Brandeis University, Waltham, Massachusetts.

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OBSERVATION AND POSSIBLE MECHANISMS OF MAGNETOELECTRIC EFFECTS IN A FERROMAGNET

George T. Rado
U. S. Naval Research Laboratory, Washington, D. C.
(Received 13 July 1964)

In this Letter we report the first observation of magnetoelectric (ME) effects in a material which is ferromagnetic. Previous ME experiments in magnetically ordered substances were confined to chromic oxide[1] (Cr_2O_3) and titanium oxide[2] (Ti_2O_3), both of which are antiferromagnetic. In addition, we suggest an atomic model which qualitatively explains our experimental result that the direction of the induced magnetic (electric) polarization is perpendicular to the applied electric (magnetic) field. This is in contrast to earlier theories[3-6] whose applicability is restricted to situations in which the induced polarizations and the applied fields are coaxial.

Our experiments were performed on single crystals[7] of gallium iron oxide having the composition $Ga_{2-x}Fe_xO_3$, where x is around unity. This material is piezoelectric as well as ferromagnetic. Its magnetic structure and even its crystal structure are unknown, but it is known[8] that the crystals are orthorhombic and that the most probable crystallographic space group is C_{2v}^9. The axes of the unit cell are assigned according to $c < a < b$. If the crystallographic point group is indeed $2mm$, the possible magnetic point groups are $2mm$, $2m'm'$, $2'm'm$, and their subgroups. On the basis of the experimentally established magnetic properties[9] of this material, we eliminate the group $2mm$ because it does not allow ferromagnetism, and the group $2m'm'$ because it allows a spontaneous magnetic moment along the two fold axis (y axis) only. (We take the rectangular coordinates x, y, z to be along a, b, c, respectively.) Since in our experiments the crystal is exposed to a static biasing field H_0 which is applied along the $+z$ or $-z$ direction, and since there is no reason for lowering the symmetry, we suppose the correct magnetic-point group to be $2'm'm$. If we now expand some appropriate thermodynamic potential of the biased crystal in powers of the applied electric-field components E_i and the applied incremental magnetic field components H_j (where i and j denote x, y, z, and we assume $|H_j| \ll |H_0|$), then we easily see that the symmetry permits terms of the type E_yH_z and E_zH_y, both of which lead to linear ME effects. For the experimental configuration used in this work, only the E_yH_z term is applicable, so that the induced magnetization δM_z and the induced electric polarization P_y are given by

$$\delta M_z = \alpha E_y, \tag{1a}$$

$$P_y = \alpha H_z, \tag{1b}$$

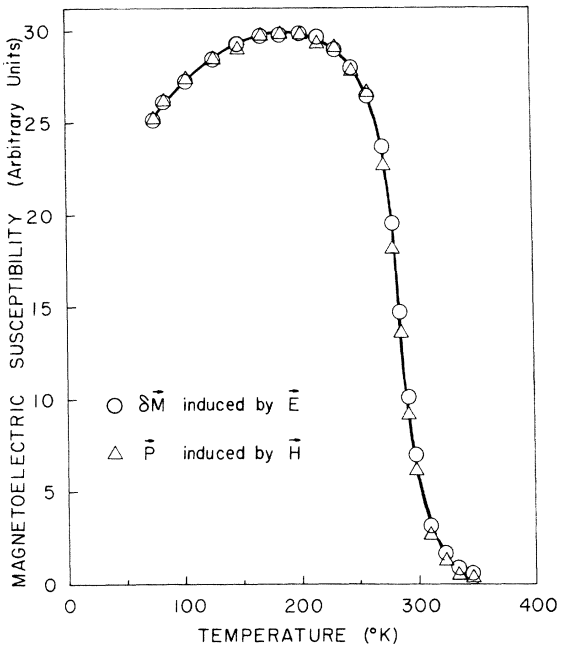

FIG. 1. Temperature dependence of the magnetoelectric susceptibility, α, as measured by means of the $(ME)_E$ effect (circles) and the $(ME)_H$ effect (triangles). The value of the biasing field, H_0, was 4450 oersteds.

where α is a (nondiagonal) magnetoelectric susceptibility. Both of these linear ME effects [which we denote by $(ME)_E$ and $(ME)_H$, respectively] were observed and measured in the present experiments by means of ac methods (using 1000 cps which will be described elsewhere.

The absolute magnitudes of the α's measured by either effect were found to be independent of the direction of $\vec{H}_0$. Furthermore, they were found to be independent of the value of H_0, provided H_0 is sufficiently large and T sufficiently small. (In the case of the crystal used for the data of Fig. 1, H_0 at 77°K must exceed 400 oersteds to achieve bias independence of the ME effects as well as approximate magnetic saturation.)

As expected on the basis of symmetry considerations, the signs of the alphas measured by either experiment were found to reverse upon reversal of the direction of $\vec{H}_0$. When the crystal was in a demagnetized state ($H_0 = 0$), the domain structure presumably leads to cancellations and no ME effect was detected experimentally.

Figure 1 shows the experimental data on the temperature dependence of α as determined by means of the $(ME)_E$ effect as well as the $(ME)_H$ effect. In accordance with the thermodynamic prediction, these two sets of data (which are normalized to each other at about 77°K) agree over the whole temperature range investigated, and both sets tend toward zero as the magnetic ordering disappears at the Curie temperature. Analogous results were obtained on three different crystals whose Curie temperatures differed. Since the experiments were carried out by a dynamic rather than a static method, we wish to point out that the equality of the α's measured by the $(ME)_E$ and $(ME)_H$ effects can be derived alternatively from a generalization[10] of the Onsager reciprocity relations.

Because of the poor geometry resulting from the smallness of the crystals (whose dimensions are roughly 1×1×4 mm), the actual magnitude of α could not be obtained reliably. A very rough estimate shows, however, that for the data of Fig. 1 the (dimensionless) value of α as determined by either effect is of the order of 3×10^{-4} at 77°K. This exceeds the largest value observed in Cr_2O_3 by an order of magnitude.

Additional experiments, which will be described elsewhere, employed a purely static method for measuring the induced voltage arising from the P_y produced by $\vec{H}_0$ as the latter is rotated in the xz or yz plane. It was found that at 77°K this voltage is a linear function of $|\cos\theta|$, where θ is the angle between $\vec{H}_0$ and the z direction, and that if $\vec{H}_0$ is applied along the x axis, the induced voltage varies as the square of H_0. All previous observations of ME effects in magnetically ordered materials were confined to linear phenomena.

To construct a theoretical model (see Fig. 2) which predicts a purely nondiagonal α, we assume that the material is a weak ferromagnet. We further assume, for the sake of simplicity, that the deviation of the sublattice magnetizations $\vec{M}_1$ and $\vec{M}_2$ from antiparallelism is small. The spontaneous magnetization M_z (which roughly equals the actual magnetization in the field H_0) is then given by $M_z = M_0(p/\lambda)$, where M_0 equals $|\vec{M}_1|$ and $|\vec{M}_2|$, λ is a molecular field coefficient, and p denotes the perturbation responsible for the weak ferromagnetism. Thus p may be of the order of (anisotropy constant/M_0^2), or else be equal to the magnitude of the Dzyaloshinskii vector. If the spin Hamiltonian describing the ME effects is of order no higher than second in the spin operators $\vec{S}$, which seems probable for a material whose symmetry is low, then we may replace the applied electric field E_y by a fictitious magnetic field h introduced[3,5] earlier for

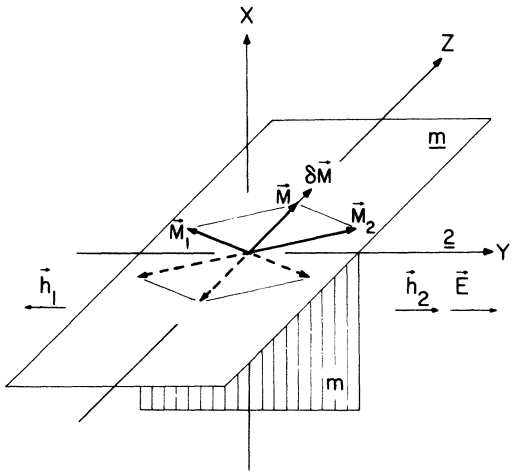

FIG. 2. Possible theoretical model (see text) of nondiagonal ME effects and weak ferromagnetism in a material whose magnetic point group is $2'm'm$. The dashed arrows refer to a domain whose magnetization is opposite to that containing $\vec{M}_1$ and $\vec{M}_2$.

Cr_2O_3. In contrast to the case of Cr_2O_3, however, we now propose two oppositely directed fictitious fields, $\vec{h}_1$ and $\vec{h}_2$, one of which acts on each of the sublattices. The effect of these fields on the two-sublattice system is easily shown to be equivalent to a change in λ given by $\delta\lambda = h/(2M_0)$, where h equals $|\vec{h}_1|$ and $|\vec{h}_2|$. Thus the fictitious fields induce the magnetization $\delta M_z = -(M_z/\lambda)\delta\lambda = -(p/2\lambda^2)h$. Recalling that h is of the form[3,5] $h = a_0\langle\langle S_y\rangle\rangle_{av}E_y$, and that $\langle\langle S_y\rangle\rangle_{av} \approx 2M_0/(Ng\mu_B)$ applies because the canting angle is assumed to be small, we now use Eq. (1a) to obtain the result

$$\alpha = -(a_0/\lambda Ng\mu_B)M_z, \qquad (2)$$

where a_0 is a temperature-independent constant of the material, g is the g factor, μ_B is the Bohr magneton, and N is the number of magnetic ions per cm^3. In a strongly piezoelectric material, like $Ga_{2-x}Fe_xO_3$, it is reasonable to expect that the odd-parity crystalline potentials are considerably larger than in Cr_2O_3 so that they may give rise to a relatively large a_0, thus yielding an α in agreement with the measurements. According to Eq. (2), the temperature dependence of α is given by that of M_z. Since M_z is known experimentally to have an approximate Brillouin function behavior,[9] there is a discrepancy between this prediction and the data of Fig. 1. However, the approximation involving a small canting angle is probably not valid in this material, and in most weak ferromagnets[11] the temperature dependence of even the magnetic susceptibility is not fully understood. The detailed predictions of the present model may have to be modified once the actual magnetic structure of $Ga_{2-x}Fe_xO_3$ is known.

The author wishes to thank V. J. Folen, R. D. Myers, and E. Prince for valuable discussions, R. V. Jones for making available the single crystals and information on some of their properties, and J. M. Ferrari for excellent technical assistance with the experiments.

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