Quadrupole magnetic field of magnetoelectric Cr_2O_3

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Experimental confirmation has been found for Dzyaloshinskiĭ's theoretical prediction [Solid State Commun. **82**, 579 (1992)] that a magnetoelectric crystal would have an intrinsic quadrupole magnetic field.

It has always been tacitly assumed that an antiferromagnetic crystal lacking a dipole magnetic moment could not have an intrinsic external magnetic field. However, Dzyaloshinskiĭ[1] has pointed out that such a crystal *can* have an intrinsic magnetic field, of a multipole order higher than dipole, and that this field should vary with distance in accordance with a power law. The order of the magnetic multipole which arises is intimately related to the magnetic symmetry of the crystal. In this paper, Dzyaloshinskiĭ showed, in particular, that the requirement which the crystal symmetry must meet for a quadrupole magnetic moment to exist was exactly the same as that for the existence of a magnetoelectric effect. In each case, both the magnetic symmetry of the magnetoelectric crystal[2] and the quadrupole magnetic moment are invariant under the simultaneous operations of spatial inversion and time reversal.

We have now carried out an experiment to detect a quadrupole field of magnetoelectric Cr_2O_3 (Ref. 4), for which Dzyaloshinskiĭ[1] wrote formulas describing the components of the external field along the trigonal axis of the crystal, H_z, and in its basal plane, $H_\perp$. Those formulas make possible a detailed comparison of experimental results with theoretical predictions.

Measuring the field components H_z and $H_\perp$ directly is a very complicated matter, but there is no difficulty in calculating and comparing with experiment the radial component of the quadrupole field of interest, H_R, or its tangential component H_t, which is directed along the tangent to the surface of a spherical crystal.

Using Dzyaloshinskiĭ's formulas, and measuring the angle θ from the crystal axis, we find some quantities which can conveniently be measured experimentally:

$$H_R = H_\perp \sin\theta + H_z \cos\theta = 3\times 10^{-5}\frac{V}{R^4}(3.3+\cos 2\theta),$$

$$H_t = H_\perp \cos\theta - H_z \sin\theta = 1.35\times 10^{-4}\frac{V}{R^4}\sin 2\theta, \qquad (1)$$

where R is the distance from the center of the crystal to the observation point (in centimeters), and V is the volume of the crystal (in cubic centimeters).

The experimental layout is shown in Fig. 1. The single-crystal Cr_2O_3 sample (1), which we have used previously,[3] is a sphere 6.4 mm in diameter. The deviations from

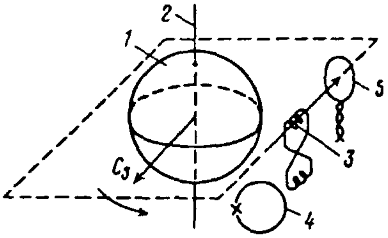

FIG. 1. Experimental layout, as explained in the text proper.

a spherical shape are 0.2 mm. The sample rotates 360° around an axis (2) which lies in the basal plane of the sample and which passes through the center of the sample. The error in the centering of the sample in the jig used to rotate it gives rise to beats which range in size up to 0.5 mm. At a short distance from the crystal is a fixed pickup coil (3), consisting of ten turns with a diameter of 3.5 mm. The length of the coil is 2 mm. This coil is part of a superconducting transformer of magnetic flux, which is measured (in ohms) by an rf SQUID (4). The pickup coil is oriented to measure either the tangential component H_t of the external field of the crystal, as shown in Fig. 1, or its radial component H_R. The axis of this coil lies in the plane perpendicular to the rotation axis and passes through the center of the crystal. The error in the orientation of the crystal and the coil is ~5°. A calibration coil (5), which generates a known magnetic field, is positioned near the pickup coil. This apparatus is enclosed in a superconducting shield. Before being installed in the apparatus, the crystal is put in a single-domain state by slow cooling through the point of the antiferromagnetic transition in a static electric field and a static magnetic field. The measurements are carried out at a temperature of 4.2 K.

Figure 2 shows the results of the measurements as a function of the angular position of the crystal. Curve *1* here is the angular dependence of the tangential component of the magnetic field calculated from expressions (1). Experimental curve *2* was recorded at the shortest possible distance between the crystal and the coil, 0.64

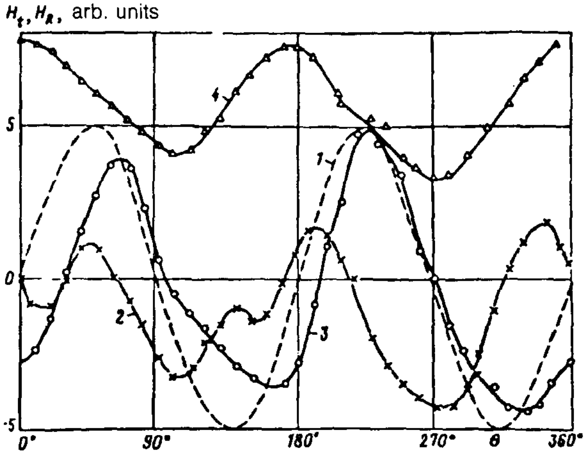

FIG. 2. Results of measurements of the tangential component of the magnetic field, H_t, at various distances from the crystal (× and ○), and of the radial component H_R (△). See the text proper for an explanation.

cm. The results in Fig. 1 are not drawn to scale along the vertical axis. Curve *3* is one of three typical curves of the angular dependence of the tangential component of the field, measured at large distances *R* from the center of the crystal, up to 0.96 cm. We did not carry out measurements at distances greater than 0.96 cm, since in this case the magnetic field distribution might have been distorted because of the proximity of the observation point to the superconducting shield.

We see that the angular variation of curve *3* has a period of 180°, as it should in the case of a quadrupole magnetic field, and that this variation differs only slightly from that expected on the basis of the theory. It can thus be assumed that the distortions (quite apparent on curve *2*), which arise as the observation point is moved closer to the sample, are not due to a magnetic contaminant or to the presence of twins in the crystal, which might give rise to an angular variation with a period of 360°. In addition, the signal from these two possible sources of a dipole magnetic field should fall off with the square of the distance, not with the fourth power, as in the case of a quadrupole field. It should thus have become predominant at large distances.

The deviation of the crystal from a spherical shape and the inadequately accurate centering of the crystal in the rotation jig are possible sources of a distortion of the signal at small distances. Distortions with this origin should give rise to a smooth curve with a period of 360°. Curve *2*, however, is considerably more complex. It suggests that at small distances we are seeing an admixture of a multipole state of order higher than quadrupole. The corresponding field was found by Dzyaloshinskiĭ for an observation point far from the sample. The field of such a multipole should fall off far more rapidly than $1/R^4$ with distance, as was observed experimentally. When the distance is increased by only 15%, the characteristic meandering seen on curve *2* disappears, at the reproducibility of our measurements.

Unfortunately, we were unable to make a detailed study of the field near the sample in this apparatus. In order to make such a study, it would be necessary to improve the spherical shape of the sample to a level that would reduce the beats several fold, and to use a much smaller measurement coil. In order to study the variation of the field component H_t with the distance, we would have to correctly choose the point inside the measurement coil to which this distance should be measured, since the field falls off rapidly with increasing *R*, and since it varies by a factor of more than 3 along the cross section of the coil at small distances from the test sample. We estimated the corresponding correction to *R*, which was measured within 0.3 mm, through a graphical integration.

The magnitude of the signal at the peaks of the angular variation, measured at the maximum difference, corresponds within 25% to a $1/R^4$ law. The signal magnitude itself varies by a factor of 4.8 as the distance is varied. However, a comparison with the field magnitude predicted by the theory revealed that the experimental values of H_t are smaller by a factor of 3.8 than expected.

Curve *4* in Fig. 2 shows the angular variation of the variable part of one of the typical curves of the radial component of the magnetic field of the crystal. We were unable to measure the constant component of the radial field, which should exist according to expressions (1); it is shown in arbitrary magnitude in Fig. 2. For this case the angular dependence corresponds very well to the theory, and the magnitude of the field agrees with the prediction within 15%. The variation of the field strength with increasing distance corresponds to the theoretical $1/R^4$ law within 15%.

It can thus be asserted that, at the accuracy of our measurements, the experimental results confirm the theoretical conclusion that a magnetoelectric Cr_2O_3 crystal has a quadrupole magnetic field. The experiments provide quantitative confirmation of conclusions regarding the angular variation of the radial and tangential components of the quadrupole field and regarding the dependence of the magnitude of both of these field components on the distance from the crystal to the observation point. However, while the agreement with the theory in terms of the amplitude of the radial component is within the experimental error, the experimental value of the tangential field component is smaller by a factor of 3.8 than the value predicted by the theory. This discrepancy probably indicates an error in the experiments, since the transcription of the theoretical formulas in a different coordinate system may not preserve the correct magnitude of one component and may greatly change the other. We have not yet found an explanation for this discrepancy.

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[1] I. Dzyaloshinskiĭ, Solid State Commun. **82**, 579 (1992).
[2] I. E. Dzialoshinskiĭ, JETP **10**, 626 (1960).
[3] D. N. Astrov, Zh. Eksp. Teor. Fiz. **40**, 1035 (1961) [Sov. Phys. JETP **13**, 729 (1961)].

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