

Proceedings of the European Conference "Physics of Magnetism 96", Poznań 1996

QUADRUPOLEAR MAGNETIC FIELD OUTSIDE ANTIFERROMAGNETIC Cr_2O_3

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Predicted by Dzyaloshinskii quadrupolar magnetic field outside antiferromagnet was experimentally observed for Cr_2O_3 single crystal. The magnetic quadrupole sharply disappeared with heating the sample above $T_N = 317$ K and appeared again on cooling below T_N . The obtained value of the magnetic quadrupole is in reasonable agreement with theoretical estimation.

PACS numbers: 75.10.-b, 75.25.+z, 75.50.Ee

Recently Dzyaloshinskii [1] has pointed out that antiferromagnets which display the magnetoelectric effect (this magnetic structure does not possess the centre of symmetry) should produce quadrupolar magnetic field. Astrov and Ermakov [2] have confirmed this prediction experimentally for Cr_2O_3 single crystal. In the present paper we report the results obtained on the same sample in wide temperature range from 4.2 to 324 K using different from [2] method of measurements.

First of all we present formulas for the magnetic field and flux produced by quadrupole (coefficients given by Dzyaloshinskii in [1] are not correct). Generally, a quadrupole consists of two opposite dipoles m , $-m$ displaced by vector d . In the dipole approximation

$$H = \frac{3(mr)r}{r^5} - \frac{m}{r^3} - \frac{3[m(r+d)](r+d)}{|r+d|^5} + \frac{m}{|r+d|^3}, \quad (1)$$

where r is the vector from an observer to the dipole m . We find radial component of H by multiplying Eq. (1) by r/r and neglecting terms with d^2 :

$$H_r = \frac{3}{r^6} [3(mr)(dr) - (md)r^2]. \quad (2)$$

For the case $m \parallel d$

$$H_r = \frac{3(md)}{r^4} (3 \cos^2 \vartheta - 1), \quad (3)$$

where ϑ is the angle between r and d (the quadrupole axis).

Now we may calculate flux through a coil. If the quadrupole is placed at coil's axis at a distance z from its centre and φ is the angle between quadrupole's and coil's axes then the answer is

$$\Phi(z, \varphi) = 3\pi a^2 (md) \frac{z}{(z^2 + a^2)^{5/2}} (3 \cos \varphi - 1), \quad (4)$$

where a is the radius of a coil. From this formula it can be seen that at given z the flux for $\varphi = 90^\circ$ has an opposite sign and is exactly twice smaller than for $\varphi = 0^\circ$.

Let us estimate the quadrupole (md) for Cr_2O_3 from structural data. Cr_2O_3 has rhombohedral unit cell (Fig. 1a). Magnetic Cr^{3+} ions have positions at uh , $(1/2 - u)h$, $(1/2 + u)h$, $(1 - u)h$ along the trigonal axis C_3 with a length

$$h = 3a \sqrt{1 - \frac{4}{3} \sin^2 \frac{\alpha}{2}},$$

where $u = 0.145$, $a = 5.38 \text{ \AA}$, $\alpha = 54^\circ 50'$. Quadrupole of the cell is

$$\begin{aligned} (md)_{\text{cell}} &= g\mu_B S [-u + (1/2 - u) - (1/2 + u) + (1 - u)] h \\ &= g\mu_B S (1 - 4u) h, \end{aligned} \quad (5)$$

where g is the Landé factor, μ_B is the Bohr magneton. Quadrupole of the sample is

$$(md) = N_A \frac{m_{\text{sample}}}{2M} (md)_{\text{cell}}, \quad (6)$$

where N_A is the Avogadro number, m_{sample} is the mass of the sample, M is Cr_2O_3 molecular mass (unit cell contains two formula units). Taking $g = 2$, $S = 3/2$ and $m_{\text{sample}} = 0.7187 \text{ g}$ we get $(md)_{\text{cell}} = 1.0645 \times 10^{-27} \text{ G cm}^4$ and $(md) = 1.51 \times 10^{-6} \text{ G cm}^4$.

Our device (Fig. 1b) consisted of pick-up coil (two turns from Nb-Ti wire joined in opposite) connected to DC SQUID sensor. Distance between the turns was 4 cm and their radius $a = 1.2 \text{ cm}$. The coil was placed outside a dewar-insert made from glass. Inside it a tube glued from aluminium foil and paper with bifilar heater and Si-diode thermometer was mounted. Temperature was stabilised manually with an accuracy about 1 K. The main cryostat was made from fiberglass and shielded with three thin-wall permalloy tubes. The middle tube has longitudinal demagnetising coil, the inner one — longitudinal and toroidal demagnetising coils. No superconducting shield was used, even for SQUID sensor. Residual magnetic

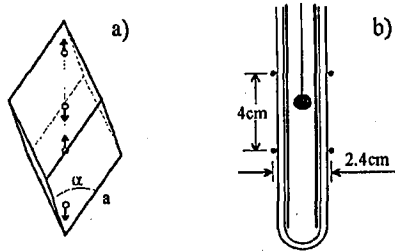


Fig. 1. (a) Crystallographic structure of Cr_2O_3 . (b) Schematic of the experimental setup.

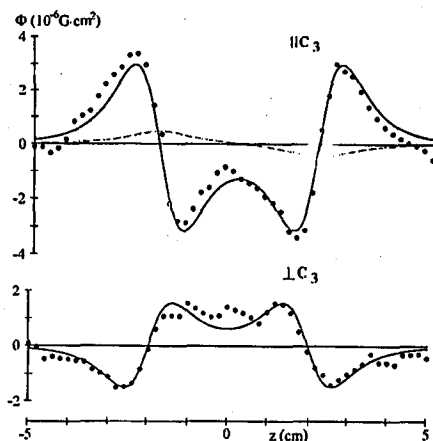


Fig. 2. Magnetic flux registered from Cr_2O_3 sample at $T = 4.2$ K when the sample is moved along the pickup coil's axis. Solid lines are the least squares fits to Eq. (7), dashed line at upper panel shows a dipole contribution.

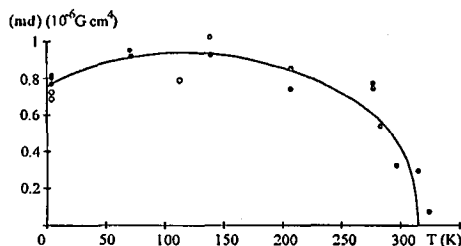


Fig. 3. Temperature dependence of the quadrupole of Cr_2O_3 sample. Experimental points were obtained for parallel (o) and perpendicular (●) orientations of the trigonal axis of the sample respectively to the coil's axis. Solid line is a guide for eyes.

field at the sample position did not exceed 3×10^{-4} Oe. The sample was ball-shaped single crystal Cr_2O_3 0.63 ± 0.01 cm in diameter. The sample was placed in a basket made from thin 0.1 cm wide paper tape and suspended with a cotton thread 65 cm long. No glue was used in preparation of this sample's holder. All these precautions permitted us to avoid parasitic magnetic signals. We moved the sample along the coil's axis and registered the change in flux as a function of distance. Calibration of the device was performed with known magnetic flux produced by dipole and quadrupole coils with dimensions similar to those of the sample.

In Fig. 2 the results obtained at $T = 4.2$ K for parallel ($\varphi = 0^\circ$) and perpendicular ($\varphi = 90^\circ$) orientations of the trigonal axis of the sample respectively to the coil's axis are shown. Solid lines are the least squares fit of the experimental points to the function

$$\Phi_s(z, \varphi) = \Phi(z + 2 \text{ cm}, \varphi) - \Phi(z - 2 \text{ cm}, \varphi) \quad (7)$$

where $\Phi(z, \varphi)$ is given by Eq. (4). As it is seen, the results nicely correspond to the expected quadrupole behaviour except that small dipole contribution is visible for parallel orientation. The same measurements were performed at higher temperatures. It was seen that quadrupole keeps up to the Néel point ($T_N = 317$ K), disappears above T_N and arises again with cooling below T_N . All measured curves were fitted to Φ_s (Eq. (7)) and the dependence of (md) on temperature was obtained, see Fig. 3. The observed maximal value of quadrupole was only 1.5 times smaller than estimated with Eq. (6).

In conclusion we must emphasise that such measurements of magnetic quadrupole are an unique way for determining the configuration of spins in the antiferromagnetic state.

References

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