

Theory of magnetic structure and electric polarization of the Cr_2BeO_4 system

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A theory is derived for the double exchange cycloidal structures with a long period in orthorhombic antiferromagnets (space group $D_{2h}^{16} - P_{bnm}$). Particular attention is given to the analysis of the compound Cr_2BeO_4 . The electric polarization of this system is attributed to the nonuniform magnetoelectric effect.

Neutron-diffraction experiments¹ have shown that at 28 K the compound Cr_2BeO_4 (space group D_{2h}^{16} ; see Fig. 1) undergoes magnetic ordering to a modulated phase with a superstructure vector directed along the c axis of the orthorhombic crystal (the modulation period is approximately 30 interatomic distances). The magnetic moments of Cr ions lie in the ac plane, i.e., a cycloidal magnetic structure is in force. More recently, Newnham *et al.*² have observed electric polarization in this compound in the temperature region 28–24 K, i.e., in the region in which the superstructure exists. In this letter we explain the magnetic structure and electrical polarization of Cr_2BeO_4 .

The crystallochemical unit cell of Cr_2BeO_4 (Fig. 1) contains two types of crystallographically equivalent chromium ions (Cr_1 in b positions and Cr_2 in c positions). Introducing for each type of ion the principle antiferromagnetism vector $\mathbf{G}_1 = \mathbf{M}_1 - \mathbf{M}_2 + \mathbf{M}_3 - \mathbf{M}_4$ and $\mathbf{G}_2 = \mathbf{M}_5 - \mathbf{M}_6 + \mathbf{M}_7 - \mathbf{M}_8$, we can show that in the exchange approximation the thermodynamic-potential density is

$$f = \delta_1 \mathbf{G}_1^2 + \delta_2 \mathbf{G}_2^2 + \Delta \left(\mathbf{G}_1 \frac{d\mathbf{G}_2}{dz} - \mathbf{G}_2 \frac{d\mathbf{G}_1}{dz} \right) + \alpha_1 \left(\frac{d\mathbf{G}_1}{dz} \right)^2 + \alpha_2 \left(\frac{d\mathbf{G}_2}{dz} \right)^2, \quad (1)$$

where δ_i, Δ , and α_i are the exchange-interaction constants, and the Cartesian z axis coincides with the c axis of the crystal.

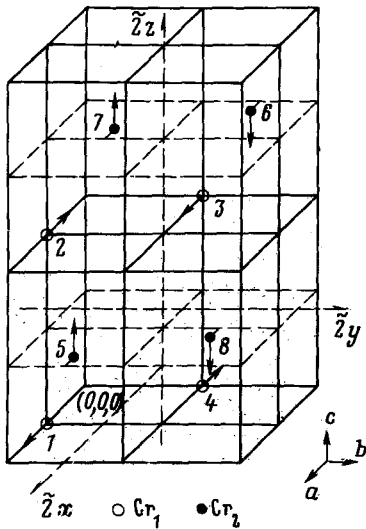


FIG. 1. Crystallochemical unit cell of Cr_2BeO_4 ($a = 4.555 \text{ \AA}$, $b = 9.792 \text{ \AA}$, and $c = 5.663 \text{ \AA}$). The arrows at the Cr ions show one example of a simple antiferromagnetic G -type structure.

We call attention to an important point. The term which contains the first derivatives and which is responsible for the superstructure is closely linked with the crystallographic nonequivalence of the two groups of magnetic ions. Since the minimum spacing between chromium ions in various positions is approximately 30% greater than their spacing in the b positions (Fig. 1), we can assume that the third term in (1) is much smaller than, for example, the fourth term. This circumstance accounts for the fact that the superstructure has a long period (see the discussion below).

We seek a solution of the problem in the form of a harmonic double cycloidal structure

$$\begin{aligned} G_{ix} &= G_1 \sin qz, & G_{2x} &= G_2 \sin(qz - \varphi), \\ G_{1z} &= G_1 \cos qz, & G_{2z} &= G_2 \cos(qz - \varphi). \end{aligned} \quad (2)$$

The absence in (1) of an invariant of the form $\mathbf{G}_1 \mathbf{G}_2$ accounts for the fact that the relative position of the superstructures in the first and second groups of magnetic atoms is determined by the invariant which is linear in the derivatives. At $\Delta > 0$ the minimum of the thermodynamic potential corresponds to $\varphi = 3\pi/2$, consistent with the result of Ref. 1.

We easily see that on the curve ABC of the magnetic phase diagram (expressed in terms of the variables γ_1, γ_2) (see Fig. 2), where

$$\gamma_1(T) = \delta_1(T) / \alpha_1, \quad \gamma_2(T) = \delta_2(T) / \alpha_2, \quad \text{and} \quad x = \Delta^2 / \alpha_1 \alpha_2, \quad (3)$$

which is described by the equation

$$(\gamma_1 - \gamma_2)^2 - 2(\gamma_1 + \gamma_2)x + x^2 = 0, \quad (4)$$

there is an uninterrupted phase transition from the paramagnetic state to superstruc-

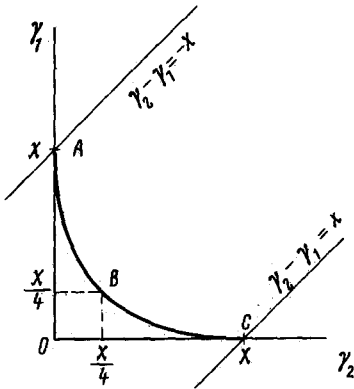


FIG. 2. Schematic representation (in γ_1, γ_2) of the magnetic phase diagram.

ture (2). On this curve we find

$$q^2 = \gamma_2 \frac{x - (\gamma_2 - \gamma_1)}{x + (\gamma_2 - \gamma_1)} = \gamma_1 \frac{x + (\gamma_2 - \gamma_1)}{x - (\gamma_2 - \gamma_1)} > 0. \quad (5)$$

In expression (1) we ignored the magnetic-anisotropy energy $\beta_1 G_{1z}^2 + \beta_2 G_{2z}^2$, which may render the existence of superstructure (2) impossible. The condition for the existence of a superstructure far from the phase transition can be written as follows:

$$|\beta_1| G_1^2 + |\beta_2| G_2^2 \leq \frac{2\Delta^2 G_1^2 G_2^2}{\alpha_1 G_1^2 + \alpha_2 G_2^2}. \quad (6)$$

We will now account for the appearance of electric polarization in Cr_2BeO_4 at 28 K. According to Bar'yakhtar *et al.*,³ electric polarization \mathbf{P} necessarily occurs in a magnetically ordered crystal of any symmetry if it has a macroscopic magnetic nonuniformity (a double cycloidal superstructure in our case). This is the so-called nonuniform magnetoelectric effect. By introducing into the thermodynamic-potential density the corresponding magnetoelectric invariants of an exchange-relativistic origin

$$P_x \left[d_1 \left(G_{1z} \frac{dG_{1x}}{dz} - G_{1x} \frac{dG_{1z}}{dz} \right) + d_2 \left(G_{2z} \frac{dG_{2x}}{dz} - G_{2x} \frac{dG_{2z}}{dz} \right) \right] \quad (7)$$

we can express \mathbf{P} in this case in the form

$$P_z = P_y = 0, \quad P_x = \chi \Delta \frac{(d_1 G_1^2 + d_2 G_2^2) G_1 G_2}{\alpha_1 G_1^2 + \alpha_2 G_2^2} \quad (8)$$

where χ is the dielectric susceptibility of the crystal. The relativistic invariants which contain P_i , on the other hand, account for the alternating component P_z , with $|P_z| \ll |P_x|$.

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¹D. E. Cox, B. C. Frazer, R. E. Newman, and R. P. Santoro, *J. Appl. Phys.* **40**, 1124 (1969).

²R. E. Newnham, I. I. Kramer, W. A. Schulze, and L. E. Cross, *J. Appl. Phys.* **49**, 6088 (1978).

³V. G. Bar'yakhtar, V. A. L'vov, and D. A. Yablonskiĭ, *Pis'ma Zh. Eksp. Teor. Fiz.* **37**, 565 (1983) [*JETP Lett.* **37**, 673 (1983)].

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