

where  $h_{\pm} = h_x \pm ih_y$ ,  $\omega_e$ ,  $\omega_a$ , and  $\omega_H$  are the frequencies corresponding to the exchange field, the anisotropy field, and the constant magnetic field, respectively,  $\omega_r$  is the relaxation frequency, and  $\gamma$  is the gyromagnetic ratio. It is seen from (8) that  $\Delta P$  contains terms that do not depend on the time, as well as terms oscillating at double the frequency  $2\omega$ , which lead to magnetoelectric frequency doubling. Estimate of the effect off resonance yields  $\Delta P = \gamma^2 D |h|^2 / \omega_e^2$ .

We note also that two-magnon absorption at double the antiferromagnetic-resonance frequency, i.e., production of magnons with  $k = 0$ , is possible in ferroelectric-antiferromagnets. Magnons with nonzero quasimomenta  $k$  and  $-k$  appear in fluorides of iron-group transition elements in two-magnon absorption, since the symmetry group of the crystal contains an inversion and  $\sum_m d_{mm} = 0$ .

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#### INDUCED FERROELECTRIC MAGNETISM IN MAGNETICALLY ORDERED PIEZOELECTRICS

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The crystallographic symmetry group of a magnet is determined from its magnetic group if the transformation  $R$  [1], which consists of the reversal of the signs of all the currents and magnetic field, is formally regarded as identical. Such a symmetry group is as a rule lower than the symmetry of the paramagnetic phase, i.e., a realignment of the electric structure of the crystal takes place in the magnetic transition as a result of interactions of the spin-orbit or magnetostriction type. This means that the thermodynamic potential is in the region of the transition point a functional not only of the microscopic spin density, but also of a certain electric ordering parameter. We are interested in the case when this parameter can be chosen to be the spontaneous electric polarization  $\vec{P}$  of the crystal, i.e., when the crystallographic group resulting from the magnetic transition belongs to the pyroelectric class. To describe the magnetic phase transition we introduce, following Dyaloshinskii [2], the vectors  $\vec{S}_{n\alpha}$ . The connection between  $\vec{S}_{n\alpha}$  and the microscopic spin density  $s(r)$  inside the unit cell was considered in detail in [2], and there is no need to repeat the conclusion derived there. We note only that when the symmetry operations are applied, the  $S_{n\alpha}$  with different  $\alpha$  and specified  $n$  are transformed in terms of one another, realizing a crystallographic-representation which is a direct product of the representation of the realized pseudovector and the  $n$ -th reducible one. The thermodynamic potential can be expanded in the region of the transition point in a power series in the projections of  $\vec{S}_{n\alpha}$  and  $\vec{P}$  (and not a functional series, as when  $\vec{s}(r)$  is used). The terms of the series are determined as invariants relative to the symmetry group of the paramagnetic phase. Since the paramagnetic phase as an operation  $R$  by itself, and the  $\vec{S}_{n\alpha}$  are invariant with respect to

this transformation, all the terms of the expansion will be bilinear in  $s_{n\alpha}^i$ .

If account is taken of only exchange interaction, then the magnetic part of the thermodynamic potential, which is quadratic in the spin projection, will contain only the sums of the squares of the vectors  $\vec{S}_{n\alpha}$  with coefficients  $A_n$  that are positive in the paramagnetic phase

$$\phi_{II} = \sum_n A_n \sum_{\alpha} S_{n\alpha}^2.$$

The transition to the magnetically ordered state occurs when one of the  $A_{n_0}$ , with  $n = n_0$ , becomes negative. This corresponds to characterizing the magnet by a set of vectors  $\vec{S}_{n_0\alpha}$ . The vectors  $\vec{S}_{n_0\alpha}$  that differ from zero already in the exchange approximation, will be designated  $\vec{L}_{\alpha}$ .

If the paramagnetic phase has an inversion center, then, inasmuch as the even powers of the projections of  $\vec{L}_{\alpha}$  are invariant against coordinate inversion, the expansion must be carried out in even powers of the projections of  $\vec{P}$ .

The situation is entirely different if the crystallographic group has no inversion center prior to the phase transition. In this case the expansion of the thermodynamic potential can contain invariants of the form

$$\gamma_{ik}^{\alpha\beta} L_{\alpha}^i L_{\beta}^k P^i, \quad (1)$$

and spontaneous electric polarization must be observed in the crystal, depending on the direction of  $\vec{L}_{\alpha}$  (which is determined by the external magnetic field).

Such a situation always arises in the case of a magnetic transition in piezoelectrics.

Indeed, the representation realized by the set  $L_{\alpha}^i L_{\beta}^k P^j$  is the cube of the vector representation multiplied by the square of the representation  $n_0$ . The square of the representation  $n_0$  always contains invariants. In piezoelectrics, the cube of the vector representation contains invariants by itself, i.e., terms of the form (1) are always present in the expansion of the thermodynamic potential of magnetic piezoelectrics.

In pyroelectric crystals, the invariants (1) always include the exchange invariant  $\gamma_i^{\alpha\beta} L_{\alpha}^i L_{\beta}^i P^i$ .

The resultant "induced" electric dipole ordering should have a number of features compared with the usual ferroelectric one.

First, in magnetic fields  $\vec{H}$  strong enough to make the magnetic-polarization vector  $\vec{M}$  parallel to  $\vec{H}$ , but weaker than the exchange fields, the spontaneous electric polarization will depend strongly on the direction of  $\vec{H}$  and weakly on its magnitude. This is precisely the spontaneous-polarization dependence observed in [3].

Second, the electric polarization, as seen from (1), will vary linearly with the temperature near the point of the magnetic transition.

Let us consider the case of the symmetry Td [4] and exchange spin density with  $N_0 = A_2$  (this is apparently the situation in boracites [5]).

We write the expansion of the thermodynamic potential in powers of the direction cosines of the vector  $\vec{L}$  (we denote them  $l_x, l_y,$  and  $l_z$ ) and the projections of the vectors  $\vec{M}$  and  $\vec{P}$ :

$$\begin{aligned} \Phi = & \Phi + \frac{1}{2\chi} M^2 + \frac{B}{2} (IM)^2 + \frac{G}{2} (l_x^2 l_y^2 + l_x^2 l_z^2 + l_y^2 l_z^2) - MH + \\ & + \alpha (l_x l_y P^z + l_y l_x P^x + l_x l_z P^y) + \beta I M \cdot P + \frac{1}{2\kappa} P^2 - PE. \end{aligned}$$

We have confined ourselves to exchange terms of order not higher than the second in the powers of the ferromagnetism vector  $\vec{M}$ . The term with the introduced constant  $\beta$  indicates that in this substance there exists a linear magnetoelectric effect of exchange origin (since  $\beta \mathbf{l}_x \vec{M} \cdot \vec{P}$  does not depend on the orientation of the magnetic vectors relative to the crystallographic axes). Incidentally, a linear magnetoelectric effect should be observed for any antiferromagnetic ordering in the  $T_d$  group.

If  $G > 0$ , then the vector  $\vec{L}$  is parallel to one of the fourfold axes and  $P_0 = 0$ . If  $G < 0$ , then  $\vec{L}$  is parallel to one of the  $C_3$  [4] and

$$P_0^z = -\kappa a l_x l_y; \quad P_0^y = -\kappa a l_z l_x; \quad P_0^x = -\kappa a l_y l_z.$$

Another interesting effect occurring if invariants of the type (1) are present in the expansion of the thermodynamic potential is the rotation of the vector  $\vec{L}$  by the electric field. Thus, for example, in the case under consideration, directing the field parallel or antiparallel to the z axis reverses the sign of the product  $\mathbf{l}_x \mathbf{l}_y$ , i.e., it rotates  $\vec{L}$  through  $90^\circ$  (this was observed in [5]).

This and similar questions will be considered in detail separately.

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Last line reads "paramagnetic phase as an operation..." should read "paramagnetic phase has an operation..."