

Metamagnetoelasticity and metamagnetolectricity of Jahn-Teller crystals

M. D. Kaplan

Institute of Chemistry, Academy of Sciences of the Moldavian SSR

(Submitted 22 October 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **35**, No. 3, 89–91 (5 February 1982)

A microscopic theory for Jahn-Teller antiferroelastics predicts new phenomena: metamagnetoelasticity and metamagnetolectricity.

PACS numbers: 75.80. + q, 75.10. – b

1. Metamagnetism, a well-known effect, consists of a sharp nonlinear increase in the magnetic moment as the external magnetic field is increased. It is characteristic of Ising antiferromagnets. In the physics of elastic crystals exhibiting phase transitions for which the order parameter is the strain—by analogy with antiferromagnets in magnetism—one may study systems with an antiferrodistortion ordering. In the microscopic theory of structural phase transitions, which is based on the cooperative Jahn-Teller effect,¹ it has been shown that for a broad class of elastic materials a phase transition is a consequence of an Ising electron-electron interaction. For antiferroelastics of this type, by analogy with metamagnetism, there is naturally a characteristic metaelasticity: a highly nonlinear behavior of the strain as a function of the pressure. Several recent experimental results are evidence that such crystals have been observed.^{2,3} On the other hand, an antiferrodistortion phase of a system may also occur in ferroelastics subjected to external fields.⁴ There is accordingly particular interest in the specific properties of Jahn-Teller antiferroelastics. Analysis shows that such materials will exhibit a very unusual property: a metamagnetoelasticity accompanied by a metamagnetolectricity.

In Jahn-Teller systems there is an intimate relationship between the structural and magnetic properties, as was pointed out by Kugel' and Khomskii⁵ in a study of exchange-induced orbital and spin orderings in compounds of transition metals. In contrast with the situation in those compounds, the states of a Jahn-Teller ion in rare-earth compounds are characterized by the total moment because of a strong spin-orbit coupling. The magnetic moment operators and also the operators representing the pseudospins which become ordered upon a structural phase transition are expressed in terms of the components of this moment. The molecular fields, which arise below the critical temperature, serve as a magnetic anisotropy energy. On the other hand, there may be situations (in dysprosium compounds, for example) in which the magnetic fields acting in the basis plane of the crystal maintain a spontaneous deformation (the operators representing the magnetic moment and the order parameters commute). In such antiferroelastics, the magnetic field may invert the pseudospin sublattice, opposing the intersublattice molecular field and putting the crystal in a ferrodistortion phase. This effect would correspond to a sharp linear increase in the strain as a function of the magnetic field, an effect which may be labeled "metamagnetoelasticity." On the other hand, the distortion ordering in these

crystals may be accompanied by an ordering of the dipole moments,⁴ so that the metamagnetoelastic behavior would be accompanied by a metamagnetolectricity, i.e., a sharply nonlinear change in the polarization with increasing magnetic field. To illustrate these arguments, we consider the case of antiferroelastics having the zircon structure.

2. We consider a crystal of the DyVO₄ type. The two Jahn-Teller ions in the cell are coupled by the inversion operation. At $T > T_c$ the lowest-lying states of Dy³⁺ are two Kramers doublets Γ_7 and Γ_6 , which are separated by a gap and mixed by oscillations of symmetry b_2 , with the result that an electron-electron interaction arises and causes a phase transition. At $T < T_c$, molecular fields of different signs act in the two sublattices, giving rise to different anisotropies of the g factors, such that we have $g_x \gg g_{y,z}$ for the ground state and $g_y \ll g_{x,z}$ for an excited state in one sublattice and vice versa in the other. Correspondingly, a magnetic field $H \parallel x$ maintains the deformation along the x axis, while a field $H \parallel y$ maintains that along the y axis. In this case of tetragonal symmetry, however, the corresponding deformations differ in sign, so that the magnetic field may induce a metaelastic transition.

The Hamiltonian of the electron subsystem for the α th sublattice can be written as follows in the molecular field approximation:

$$H_\alpha = (A\bar{\sigma}_z^\alpha + B\bar{\sigma}_z^{\alpha'} \pm f\chi_0\bar{\epsilon})\sigma_z^\alpha + \Delta\gamma\sigma_x^\alpha - h_x S_x^\alpha - h_y S_y^\alpha, \quad (1)$$

where \mathcal{E} is the electric field, $h_i = (1/2)g\mu_B H_i$, γ is the vibron reduction or fact, and A and B are the constants of the intrasublattice and intersublattice interactions (in the system under consideration here, we have $B < 0$, in contrast with the real DyVO₄ crystal, in which we have $B > 0$, and at $T < T_c$ there is a ferrodistortion, rather than antiferrodistortion, phase). The spectrum of the system is determined by

$$E_{1,2,3,4}^\alpha = \pm \frac{h}{2} f_1(\theta) \pm \sqrt{\left[H_{mol}^\alpha - \frac{h}{2} f_2(\theta) \right]^2 + \Delta^2 \gamma^2}, \quad (2)$$

where $h = \sqrt{h_x^2 + h_y^2}$, $f_{1,2}(\theta) = \cos\theta \pm \sin\theta$, θ is the angle between the magnetic field and the x axis, and $H_{mol}^\alpha = A\bar{\sigma}_z^\alpha + B\bar{\sigma}_z^{\alpha'} \pm f\chi_0\epsilon$. Using (2), we can derive a system of transcendental equations for the sublattice order parameters:

$$\bar{\sigma}_z^\alpha = \frac{2}{z_\alpha} \left[\left(H_{mol}^\alpha - \frac{h}{2} f_2(\theta) \right) \left(E_\alpha^- \right)^{-1} \exp \left(-\frac{\beta h}{2} f_1(\theta) \right) \text{sh} \beta E_\alpha^- + \left(H_{mol}^\alpha + \frac{h}{2} f_2(\theta) \right) \left(E_\alpha^+ \right)^{-1} \exp \left(\frac{\beta h}{2} f_1(\theta) \right) \text{sh} \beta E_\alpha^+ \right], \quad (3)$$

where

$$E_\alpha^\pm = \sqrt{\left[H_{mol}^\alpha \pm \frac{h}{2} f_2(\theta) \right]^2 + \Delta^2 \gamma^2}, \quad (4)$$

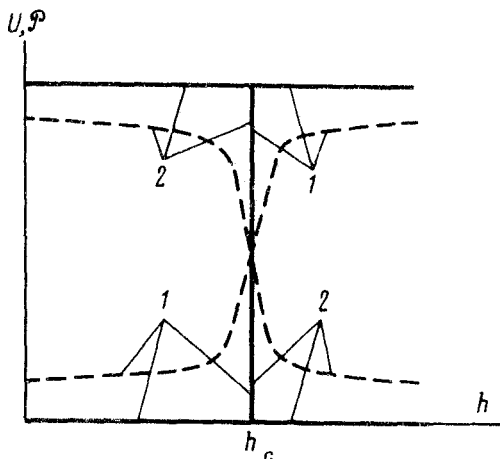


FIG. 1. Dependence of the spontaneous deformation and of the spontaneous polarization on the external magnetic field at $\Delta = 0$. 1— $U(h)$; 2— $P(h)$; solid lines— $T = 0$; dashed curves— $T \neq 0$.

$$Z_a = 2 \left[\exp \left(-\frac{\beta h}{2} f_1(\theta) \right) \operatorname{ch} \beta E_a^- + \exp \left(\frac{\beta h}{2} f_1(\theta) \right) \operatorname{ch} \beta E_a^+ \right]. \quad (5)$$

System (3) describes the temperature and field behavior of spontaneous deformations $U \sim (\bar{\sigma}_z^I + \bar{\sigma}_z^{II})$ and of the polarization $\mathcal{P} \sim (\bar{\sigma}_z^I - \bar{\sigma}_z^{II})$. It follows from an analysis of the free energy that at $H = \mathcal{E} = 0$ and at $T < T_c$ there is an antiferrodistortion phase. A magnetic field opposing the molecular field of one of the sublattices may invert that sublattice, putting the crystal in a ferrod distortion antiferroelectric phase (Fig. 1).

3. Can these effects be observed experimentally? We see two possibilities. The first is presented by real DyVO_4 and DyAsO_4 crystals. Although they are ferroelastics, an electric field can convert them into an antiferrodistortion ferroelectric phase.⁴ The metamagnetoelastic behavior of the crystals is exhibited in magnetic fields which oppose the intersublattice interaction and the electric field.

The second possibility involves the use of crystals of double tungstenates and molybdates. Jahn-Teller phase transitions have been observed in these crystals,⁶ and these transitions may be classified as antiferrodistortion transitions, judging from several of the results obtained in Refs. 2 and 3. In these compounds, however, the antiferrodistortion phase is apparently antiferroelectric, rather than ferroelectric. Although the situation here is slightly more complicated than that in the zircons, the results derived above are still applicable. Estimates put the critical magnetic fields (the fields which would elicit the metaelastic behavior of the crystal) at $H \sim 3\text{--}10\text{ T}$, which can be produced experimentally.

I wish to thank B. G. Vekhter for a discussion of these results.

1. G. A. Gehring and K. A. Gehring, Rep. Prog. Phys. **38**, 1 (1975).

2. A. H. Cooke, M. M. Davidson, N. J. England, M. J. M. Leask, J. B. Lowry, A. C. Tropper, and

- M. R. Wells, *J. Phys. C* **9**, L573 (1976).
3. S. D. El'chaninova, S. D. Anders, A. I. Zvyagin, M. I. Kobets, and Yu. G. Litvinenko, *Fiz. Nizk. Temp.* **7**, 187 (1981) [*Sov. J. Low Temp. Phys.* (to be published)].
 4. B. G. Vekhter and M. D. Kaplan, *Pis'ma Zh. Eksp. Teor. Fiz.* **29**, 173 (1979) [*JETP Lett.* **29**, 155 (1979)]; *Zh. Eksp. Teor. Fiz.* **78**, 1781 (1980) [*Sov. Phys. JETP* **51**, 893 (1980)].
 5. K. I. Kugel' and D. I. Khomskii, *Pis'ma Zh. Eksp. Teor. Fiz.* **23**, 264 (1976) [*JETP Lett.* **23**, 237 (1976)]; D. I. Khomskii and K. I. Kugel, *Phys. Status Solidi* **b79**, 441 (1977).
 6. E. Pytte, *Phys. Rev. B* **9**, 932 (1974).
 7. A. I. Zvyagin, T. S. Stestenko, V. G. Yurko, and R. A. Vaishnoras, *Pis'ma Zh. Eksp. Teor. Fiz.* **17**, 190 (1973) [*JETP Lett.* **17**, 135 (1973)].

Translated by Dave Parsons

Edited by S. J. Amoretty