Noncollinearity of the magnetic structure of Jahn-Teller antiferroelastic materials of the $KDy(MoO_4)_2$ type

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A theory based on the cooperative Jahn-Teller effect shows that a noncollinearity of a magnetic structure is a consequence of an antiferrodistortion phase transition in a crystal.

In crystals with a cooperative Jahn-Teller effect,[1] the mutual effects of the distortion correlations, the magnetic correlations, and the electric dipole correlations give rise to corresponding interrelationships among the acoustic, magnetic, and dielectric anomalies.[2–4] Working in this picture, we show below that in crystals of the $KDy(MoO_4)_2$ type a noncollinearity of the magnetic structure is a direct consequence of an antiferrodistortion order.

The $KDy(MoO_4)_2$ crystal has recently been the subject of active theoretical and experimental research [5–9] in connection with the antiferrodistortion ($T_c = 14$ K) and magnetic ($T_N = 1.1$ K) Jahn-Teller phase transitions that occur in it. According to the experiments of Refs. 7 and 9, the magnetically ordered phase is noncollinear: The magnetic moments of the two sublattices of the orthorhombic crystal lie in the ac plane, making angles of $\pm 50°$ with the a axis.

The physical reasons for this noncollinearity are that the anti-Teller molecular fields that arise at $T \leqslant T_c$ and act on the spin-orbit states of the rare-earth ion serve as a magnetic anisotropy which takes opposite signs for the antiferrodistortionally ordered sublattices. The corresponding easy axes of these sublattices are perpendicular to each other, and this situation is responsible for the noncollinearity. When the crystal fields are taken into account, we find that the magnetic moments rotate in the easy plane, and the magnetic interactions are responsible for an additional small change (corresponding to the small value of T_c/T_N) in the angles between the moments.

We write the Hamiltonian of a tetragonal crystal subjected to a pressure P that produces an orthorhombic deformation of symmetry B_{1g} as follows, in the basis of the lowest-lying states of the Dy^{3+} ion (two Kramers doublets, Γ_7 and Γ_6, separated by a gap Δ):

$$H = H_f + H_{str} + H_{el-str} - \sum_{m\kappa\beta} (V^{E_x}_{\kappa m\beta} E^{m\beta}_x + V^{E_u}_{\kappa m\beta} E^{m\beta}_y)(b_\kappa + b^+_{-\kappa})$$

$$- \Delta \sum_{m\beta} \sigma^{m\beta}_x - \sum_{\substack{mn \\ \beta\beta'}} [J^{\beta\beta'}_{mn\parallel} S^{m\beta}_z S^{n\beta'}_z + J^{\beta\beta'}_{mn\perp} (S^{m\beta}_x S^{n\beta'}_x + S^{m\beta}_y S^{n\beta'}_y)]. \quad (1)$$

The first three terms describe free phonons and the energy of the crystal associated

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with the orthorhombic and monoclinic deformations; the fourth term describes the interaction of electrons with E oscillations, which is responsible for the structural phase transition; the last term describes the magnetic interactions of the dipole-dipole type; the S_α are the components of the total moment; and β and β' are the indices of the four sublattices of the crystal. Subjecting the Hamiltonian to a displacement transformation[1,2] which eliminates the terms in (1) that are linear in the phonons, we find an intercenter interaction (in second order in the electron-phonon coupling constant) which gives rise to an antiferrodistortion phase transition. Noting that we have $\bar{E}_x^\beta \neq 0$, $\bar{E}_y^\beta = 0$ at $T \leqslant T_c$ by virtue of the orthorhombic symmetry, we write the electron part of the transformed Hamiltonian as follows in the molecular-field approximation:

$$\tilde{H}_{el} = -\sum_{m\beta\beta'} [(A^{\beta\beta}\bar{E}_x^\beta + A^{\beta\beta'}\bar{E}_x^{\beta'})E_x^\beta - g_0 P(c_0\Omega N)^{-1/2}\sigma_z^{m\beta} - \Delta\sigma_x^{m\beta}], \quad (2)$$

where $A^{\beta\beta}$ and $A^{\beta\beta'}$ are the parameters of the intra- and intersublattice Jahn-Teller interactions; $A^{\beta\beta'} < 0$; and g_0 is the coupling constant between electrons and the B_{1g} deformation. The electron operators E_α and σ_α are conveniently expressed in terms of[10] S_α; $E_x = S_xS_z + S_zS_x$, $\sigma_z = S_x^2 - S_y^2$, etc. Treating the moment operators at $T = 0$ as classical vectors whose spatial orientation is specified by the angles θ and φ, we can minimize the energy $\bar{H}_{el}(\theta, \varphi)$ and thereby find the equilibrium values of θ^β and φ^β. In the paramagnetic phase (with $J_{\|'\perp} = 0$ we find

$$\varphi^{\mathrm{I,II}} = 0, \qquad \theta^{\mathrm{I,II}} = \frac{1}{2}\arccos\left[\pm\frac{\Delta - g_0P(c_0\Omega N)^{-1/2}}{A^{\mathrm{I,I}} - A^{\mathrm{I,II}}}\right] \quad (3)$$

(the four sublattices under consideration are equivalent in pairs at $T > T_N$). In other words, the easy axes lie in the ac plane of the crystal and are rotated through 90° if $\Delta - g_0P(c_0\Omega N)^{-1/2} = 0$.

From (3) we see that it is possible to distinguish between the contributions of the tetragonal and orthorhombic fields to the total energy gap

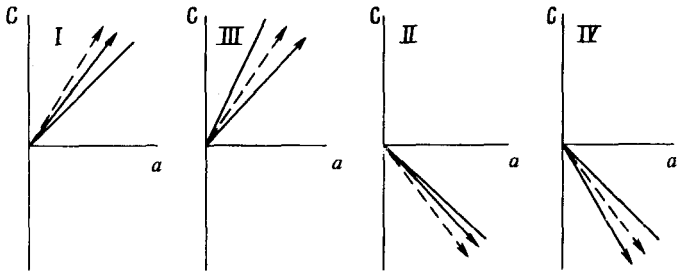

FIG. 1 Orientation of the magnetic moments in the four sublattices of the crystal. Dashed lines—without magnetic interactions; solid lines—with magnetic interactions.

$\Delta_\infty = [\Delta^2 + g_0^2 P^2 (c_0 \Omega N)^{-1}]^{1/2}$ that separates the Kramers doublets at $T > T_c$. Taking into account the definition of the critical temperature, $kT_c = \Delta_\infty \operatorname{arctanh}^{-1}[\Delta_\infty / (A^{I,I} - A^{I,II})]$, along with (3), and using the experimental values[9] $\Delta_\infty = 9$ cm^{-1} and $\theta^I = 40°$, we find $\Delta = 6.7$ cm^{-1} and $g_0 P (c_0 \Omega N)^{-1/2} = 6.0$ cm^{-1}.

To determine the magnetic configuration at $T < T_N$, we must also take the magnetic interactions into account. Noting that the magnetic interactions along the z axis are of an antiferromagnetic nature, while those along the x axis are of a ferromagnetic nature, under the condition[9] $T < T_N$, we find that the changes in the angles upon magnetic ordering are $\pm \delta\theta^{I,II} \sim (J_\perp \pm J_\parallel)/(A^{I,I} - A^{I,II})$ or, numerically, 1–3° [$J_{\parallel,\perp} \approx 1$–$2$ cm^{-1} (Ref. 9); $J \ll A$]. Consequently, at $T < T_N$ the crystal breaks up into four sublattices, not the two that were observed experimentally in Ref. 9, since each structural sublattice consists of two magnetic sublattices with a small angle $\delta\theta^\beta$ between the magnetic moments (Fig. 1).

The noncollinear magnetic structure of crystals of the $KDy(MoO_4)_2$ type can be acted upon effectively by means of uniaxial pressures, by rotating the sublattice magnetic moments in the *ac* plane or even causing them to depart from this plane (if the experimental situation allows the application of the corresponding pressures. Finally, we note that by applying a sufficiently strong external magnetic field ($\sim$20 kOe) in the *ac* plane, at an angle of 45° from the c axis, we can induce a metamagnetoelastic structural transition from an antiferrodistortion phase to a ferrodistortion phase.[4] At $T < T_N$, this transition is accompanied by a magnetic phase transition to a quasicollinear ($\delta\theta^\beta \ll 1$) phase.

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[1]G. A. Gehring and K. A. Gehring, Rep. Prog. Phys. **38**, 1 (1975).
[2]B. G. Vekhter and M. D. Kaplan, Zh. Eksp. Teor Fiz. **78**, 1781 (1980) [Sov. Phys. JETP **51**, 893 (1980)]; **87**, 1774 (1984) [Sov. Phys. JETP **60**, No. 5 (1984)].
[3]K. I. Kugel' and D. I. Khomskiĭ, Usp. Fiz. Nauk **136**, 621 (1982) [Sov. Phys. Usp. **25**, 231 (1982)].
[4]M. D. Kaplan, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 89 (1982) [JETP Lett. **35**, 105 (1982)].
[5]A. I. Zvyagin, T. S. Stetsenko, V. G. Yurko, and R. A. Vaĭshnoras, Pis'ma Zh. Eksp. Teor. Fiz. **17**, 190 (1973) [JETP Lett. **17**, 135 (1973)].
[6]I. V. Skorobogatova and A. I. Zvyagin, Fiz. Nizk. Temp. **4**, 801 (1978) [Sov. J. Low Temp. Phys. **4**, 381 (1978)].
[7]M. I. Leask, A. C. Tropper, and M. R. Wells, J. Phys. C **14**, 3481 (1981).
[8]L. N. Pelikh and A. I. Zvyagin, Fiz. Tverd. Tela (Leningrad) **24**, 1536 (1981) [Sov. Phys. Solid State **24**, 880 (1981)].
[9]A. H. Cooke, M. M. Davidson, M. J. England, J. B. Lowry, A. C. Tropper, and M. R. Wells, J. Phys. C **9**, L573 (1976).
[10]R. J. Ellyott, R. T. Harley, W. Hayes, and S. R. P. Smith. Proc. R. Soc. London **A328**, 217 (1972).

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