Magnetostriction of a $DyVO_4$ single crystal

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(Submitted 29 January 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **43**, No. 6, 287–290 (25 March 1986)

The magnetostriction properties of a $DyVO_4$ crystal are studied in the region of the structural phase transition resulting from a cooperative Jahn-Teller effect. The magnetostriction anomalies detected in these studies are explained in terms of the theory of the cooperative Jahn-Teller effect.

The magnetic fields in Jahn-Teller crystals have an appreciable effect on the properties of the crystal lattice.[1] The magnetostriction is a direct manifestation of this effect. It was shown in the theoretical studies of Vekhter and Kaplan[2,3] that the magnetostriction of crystals with cooperative Jahn-Teller effect if anomalously large and has characteristic magnetic-field and temperature dependences. However, the magnetostriction of Jahn-Teller crystals with structural phase transitions has so far not been measured directly. In the present letter we report the results of the first study of the field and temperature dependences of static magnetostriction in a $DyVO_4$ crystal—a representative of a large class of rare-earth compounds with a zircon structure RXO_4 ($R =$ Tm,Dy,Tb; $X =$ V,P,As) which undergo tetragonal phase-orthorhombic phase low-temperature phase transitions at $T = T_c$ as a result of the cooperative Jahn-Teller effect.

According to the data of Ref. 4, $DyVO_4$ has $T_c = 14$ K; below $T_N = 3$ K, this crystal exhibits antiferromagnetic ordering of Dy^{3+} ions. At $T > T_c$ the Kramers doublets Γ_7 and Γ_6, which are divided by a gap $2\Delta = 9\ \mathrm{cm}^{-1}$, are the lowest-lying electronic states of Dy^{3+}. On the basis of these states the Hamiltonian of the crystal in the molecular-field approximation is[5]

$$H = -A\bar{\sigma}_z\sigma_z + \Delta\gamma\sigma_x - h_xS_x - h_yS_y , \tag{1}$$

where A is the parameter of the molecular field which arises from the correlation of the Jahn-Teller distortions; $\sigma_{z,x}$ are the operators of the interaction with the molecular and crystal fields and $S_{x,y}$ are the operators of the interaction with the magnetic fields H_i ($h_i = g_\perp\mu_B H_i$), respectively; and γ is the factor of the vibron reduction. In (1) we ignored the slight difference in the g factors for the states $\Gamma_7(g_\perp = 9.9)$ and $\Gamma_6(g_\perp = 10.1)$ and the value of $g_\parallel$, which is small in comparison with $g_\perp$ (Refs. 4 and 6). Using Hamiltonian (1) for the equilibrium order parameter $\bar{\sigma}_z$, which determines the uniform deformation of the crystal U, we find

$$\bar{\sigma}_z = \frac{2}{Z}\Bigg[\left(A\bar{\sigma}_z - \frac{h}{2}\cos\varphi\right) \times E_-^{-1}\exp\left(-\frac{h\cos\varphi}{2kT}\right)\mathrm{sh}\frac{E_-}{kT} + \left(A\bar{\sigma}_z + \frac{h}{2}\cos\varphi\right)E_+^{-1}\exp\left(\frac{h\cos\varphi}{2kT}\right)\mathrm{sh}\frac{E_+}{kT}\Bigg]. \tag{2}$$

Here Z is a single-ion statistical sum, φ is the angle between the magnetic field h and the a axis in the ac plane of the crystal [the (010) plane], and

$$E_\pm = [(A\bar{\sigma}_z \pm {}^1/_2\, h\cos\varphi)^2 + \Delta^2\gamma^2]^{1/2} . \tag{3}$$

It follows from Eq. (2) that at $h = 0$ and $T < T_c = \Delta\gamma\,\mathrm{arctanh}^{-1}(\Delta\gamma/A)$ we have a nonvanishing order parameter $\bar{\sigma}_z$ which causes a spontaneous deformation. According to (2), the magnetic field $h_{x,y}$ increases the deformation. As a result, at $T \gtrsim T_c$ the magnetostriction of $DyVO_4$ can reach values comparable to the spontaneous deformation of the crystal lattice, i.e., on the order of 10^{-3}.

The magnetostriction ($\Delta l/l \equiv U$) was measured in $DyVO_4$ single crystals $2\times2\times1$ mm in size which were grown in different batches. The measurements were carried out in the temperature interval 1.8–25 K by the capacitive method.[7] The design of the apparatus was such that it allowed us to measure the longitudinal magnetostriction ($U_\parallel$) up to 50 kOe in a superconducting solenoid and the transverse magnetostriction ($U_\perp$) in a superconducting magnet having the geometry of Helmholtz coils (with a 3-kOe field).

Figure 1 is a plot of $U_\perp$ versus the orientation of the field $H = 10$ kOe in the (010) plane at temperatures above and below T_c. We see that at $T > T_c$ $U_\perp$ is maximum when $H \parallel [100]$ and is approximately equal to zero when $H \parallel [001]$ (the c axis). The experimental data are in qualitative agreement with the theoretical curve $U_\perp(\varphi)$ calculated for $T > T_c$ from Eqs. (2) and (3) with the parameters $A = 11.2$ cm^{-1}, $\Delta = 4.5$ cm^{-1}, and $\gamma = 1$.

The magnetostriction in the ordered phase (4.2 K) is considerably larger in magnitude, and the direction of H near the c axis has a sharp minimum whose position depends on the direction of rotation of the field. Such a behavior of magnetostriction is traceable, in our view, to the crystallographic "domains" which appears in $DyVO_4$

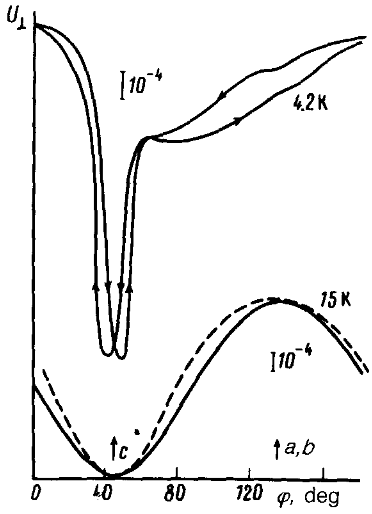

FIG. 1. Transverse magnetostriction of $DyVO_4$ single crystal versus the orientation of the magnetic field $H = 10$ kOe in the (010) plane above T_c (15 K) and below T_c (4.2 K). Solid curve—Experiment; dashed curve—theory.

crystals[8] below T_c. The angular dependence of $U_\perp$ in the (010) plane indicates that the $DyVO_4$ crystal becomes a single-domain crystal when there is a small component of H along the [100] axis and it decomposes into structural "domains" when $H \perp [100]$. The external magnetic field is able to change the orientation of the domains because of the large anisotropy of the g factor at $T < T_c$: $g_x = 19.4$ and $g_y \approx g_z \approx 0.4$ (Ref. 4).

The magnetostriction caused by the structural domains turned out to be markedly different in the four $DyVO_4$ crystals studied by us. The particular nature of the breakup of each crystal into domains below T_c is evidently determined by such difficultly controllable factors as crystal defects, mechanical stress caused by cementing of the sample to a holder and then cooling it down, small uniaxial stresses produced by the weight (~ 5 g) of the plate of the measuring capacitor, and other factors. An important point is that the crystals cooled below T_c in a magnetic field ~ 20 kOe do not remain single-domain crystals after the removal of the field at 4.2 K.

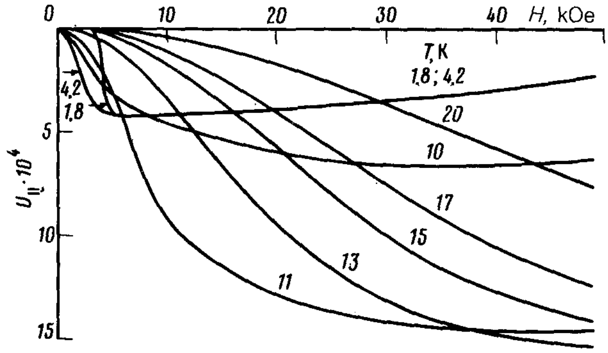

FIG. 2. Isotherms of the longitudinal magnetostriction for $H \parallel [010]$.

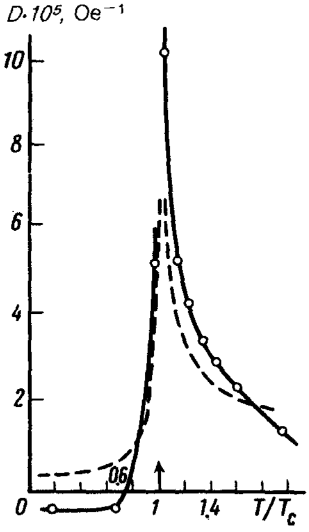

FIG. 3. Experimental temperature dependence (solid curve) and theoretical temperature dependence (dashed curve) of the magnetostriction coefficient.

One of the $DyVO_4$ crystals studied by us turned out to be a nearly single-domain crystal below T_c (at $H = 0$), as indicated by the isotherms $U_{\parallel}(H)$ of this sample for $H \parallel [010]$ in Fig. 2. In these isotherms the domain magnetostriction is seen as relatively small jumps in the field (on the order of 5 kOe) at $T = 1.8$ and 4.2 K. Of fundamental importance is the fact that $U_{\parallel}$ increases appreciably as the temperature is raised. Near 13 K in fields of ~40 kOe it reaches values on the order of spontaneous deformation of the lattice.

Another feature of these isotherms $U_{\parallel}(H)$ is the increase of the maximum magnetostriction coefficient $D \equiv \partial \bar{\sigma}_z/\partial H = 1/U_0(\partial U/\partial H)$ (U_0 is the saturation magnetostriction). Theoretical calculation shows that at small but finite values of H and $T \to T_c$ the coefficient D increases anomalously (at $T = T_c D \to \infty$). Figure 3, a plot of the theoretical and experimental curves of the coefficient D, shows that the theoretical curve is in qualitative agreement with the experimental curve.

The results of this study thus show that a Jahn-Teller $DyVO_4$ crystal has a large and unique magnetostriction. Since this magnetostriction is determined primarily by the specific electronic structure of magnetic ions, it would also be of interest to study the magnetostriction of other rare-earth paramagnetic elastic materials with a zircon structure.

We wish to thank B.V. Mill' for growing the $DyVO_4$ single crystals which were used in the experimental part of this study.

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Translated by S. J. Amoretty