Effect of correlations of Jahn-Teller distortions on the magnetostriction of the virtual elastic $TmPO_4$

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Magnetostriction effects due to correlation of local Jahn-Teller distortions have been observed and investigated in the paramagnetic single crystal $TmPO_4$. It is shown that the correlations lead not only to a substantial increase of the magnetostriction $U = \alpha H^2$, but also to the appearance of a characteristic extremum on the $\alpha(T)$ plot at a temperature $T_0 \approx 14$ K. The experimental data are compared with results of theoretical calculations in the molecular-field approximation.

It follows from theoretical calculations [1–3] that correlation of local distortions in paramagnetic crystals containing Jahn-Teller (JT) ions should lead to an anomalously large magnetostriction and to a characteristic temperature of the derivative $\partial U/\partial H$ of the magnetostriction with respect to the field. Experiments[4,5] on real JT elastics (crystals with cooperative Jahn-Teller effect), however, have shown that these singularities are masked by reorientation of the crystallographic Jahn-Teller domains produced below the structural phase-transition temperature. We have chosen therefore to investigate in the present study the magnetostriction characteristics of the single crystal $TmPO_4$, in which there are no crystallographic domains.

The tetragonal crystal $TmPO_4$ (zirconium structure) is a virtual elastic, i.e., a compound in which no structural phase transition takes place, but strong correlations of local JT distortions exist around the Tm^{3+} ions and are due to phonon exchange and to electron-strain coupling. These correlations cause, obviously, the characteristic minimum of the elastic modulus C_{66}, observed in $TmPO_4$ in the region of $T = 20$ K (Ref. 6), and the anomalies of the magnetic properties near liquid-helium temperature.[3,7,8]

$TmPO_4$ single crystals measuring approximately $2\times1\times1$ mm were grown by crystallization form the molten solution, using lead pyrophosphate as the flux. The magnetostriction $U \equiv \Delta l/l$ was measured by a capacitive method [9] in the temperature interval 4.5–40 K and in longitudinal- and transverse-geometry magnetic fields of strenth up to 50 kOe.

Figure 1 shows plots of $U_{\parallel}(H)$ and $U_{\parallel}(H^2)$ of a $TmPO_4$ crystal for a longitudinal field $\mathbf{H}\parallel[110]$ (Fig. 1a shows by way of example only two isotherms). It can be seen that the magnetostriction reaches gigantic values ($> 10^{-3}$) near liquid-helium temperatures and is quadratic in the field for $H \leqslant 15$ kOe. The value of H at which the deviation from the relation $U = \alpha H^2$ sets in depends on temperature, and at $T \geqslant 28$ K the magnetostriction varies like H^2 in the entire magnetic-field interval investigated by us. The coefficient α has a nonmonotonic temperature dependence with a maximum near $T_0 = 13$ K. The field interval in which $U = \alpha H^2$ holds is significantly narrowed in the region of T_0.

The measurements have shown that the transverse magnetostriction of $TmPO_4$ [field in (110) crystal plane and $U\parallel[110]\perp\mathbf{H}$] has the usual angular dependence

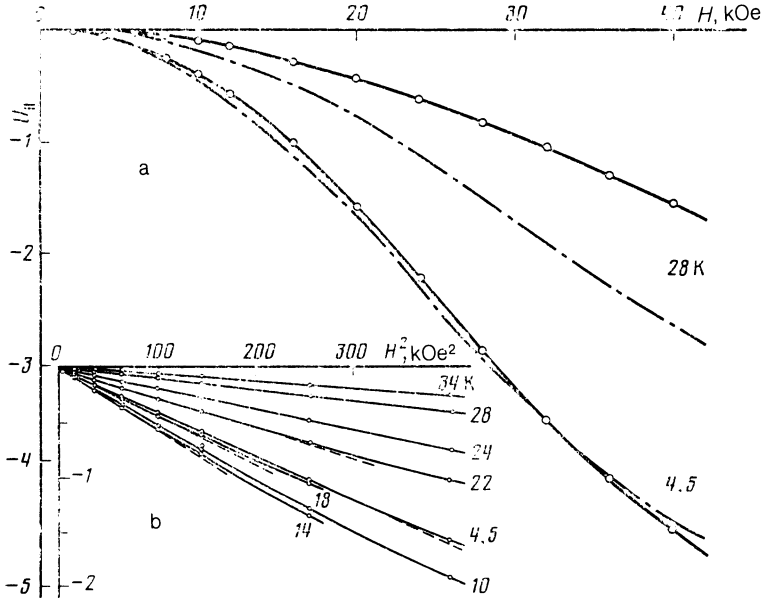

FIG. 1. Isotherms of the longitudinal magnetostriction of single-crystal $TmPO_4$ in a magnetic field $\mathbf{H}\parallel[110]$: a—$U_{\parallel}(H)$, b—$U_{\parallel}(H^2)$; points—experiment, dash-dot—calculation, dashed—linear extrapolation of the $U_{\parallel}(\mathbf{H}^2)$ dependence in weak field. The ordinate scale is 10^{-3}.

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$U_\perp \sim 1 + \cos\varphi$, with $U_\perp \approx 10^{-2}$ for $\mathbf{H}\|[1\bar{1}0]$ and $U_\perp < 10^{-6}$ for $\mathbf{H}\|[001]$. The longitudinal striction for $\mathbf{H}\|[100]$ is 10^{-5} for $T = 4.5$ K and $H = 40$ kOe. The anisotropy $U_\parallel$ in the basal plane is large: it changes by two orders when the direction of H is varied in this plane. So large an anisotropy is evidence of a weak electron-phonon coupling with B_{1g} oscillations and deformations. In our opinion, this constitutes the characteristic difference between the virtual elastic $TmPO_4$ and real JT elastics of $DyVO_4$ type, in which experiments reveal a substantially lower anisotropy.

The magnetoelastic properties of $TmPO_4$ can be adequately described with the aid of an electron Hamiltonian obtained by a shift transformation (without allowance for the electron-phonon coupling)[2]:

$$H = -\sum_{m,n} A_{mn}\sigma_z^m\sigma_z^n - \frac{\Delta\gamma}{2}\sum_m (1+\tau_z^m)\sigma_x^m - g\mu_B\sum_m (H_x s_x^m + H_y s_y^m). \quad (1)$$

In Eq. (1), $\Sigma_m A_{mn} \equiv A$ is the constant of the molecular field resulting from the correlation of the JT distortions; Δ is the energy gap between the doublet and the singlets; γ is the vibronic reduction constant; σ, τ, and s are electron operators specified on the basis of four states (singlet-doublet-singlet) of the Tm^{3+} ion.

In the molecular-field approximation it is easy to obtain from the Hamiltonian (1) equations for the energy spectrum and for the crystal's homogeneous deformation $U(T,H)$ which is proportional to the equilibrium value of the order parameter $\bar{\sigma}_z$. These equations, however have no exact analytic solution. We have therefore obtained $U(T,H)$ by a numerical calculation that yields the best agreement with experiment at the following parameter values: $A = 22$ cm^{-1}, $\gamma\Delta = 30$ cm^{-1}, and $g = 8$. The results of a comparison of the theory with experiment are illustrated in Fig. 1a. The possible cause of the quantitative difference between the theoretical and experimental data at $T > 10$ K is the influence of external (governed by the experimental conditions) and internal mechanical stresses in the investigated $TmPO_4$ crystal. Of course, this can be also the consequence of a number of other factors, including physical effects that are not taken into account in the employed theoretical model.

Using the Hamiltonian (1) in the approximation $g\mu_B H$, kT, $\Delta\gamma \gg A\sigma_z$, and considering only the terms with H^2, it is possible to obtain an analytic relation for the coefficient α in the relation $U = \alpha H^2$:

$$\alpha = \frac{g^2\mu_B^2}{\Delta^2\gamma^2}\left(\operatorname{ch}\frac{\Delta\gamma}{kT} - 1\right)\left(\operatorname{ch}\frac{\Delta\gamma}{kT} + 1 - \frac{A}{kT}\right)^{-1}. \quad (2)$$

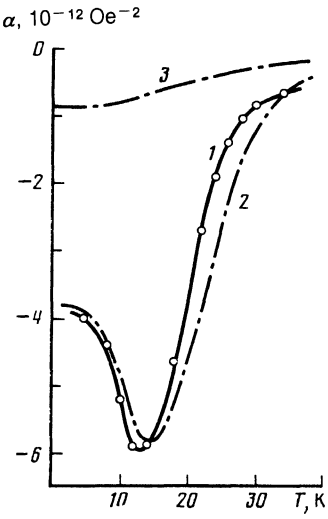

FIG. 2. Temperature dependence of the coefficient α: 1—experiment; 2—theory, $A = 22$ cm^{-1}; 3—theory, $A = 0$.

It follows from (2) that for the parameter values cited above the $\alpha(T)$ dependence has a maximum at $T_0 \simeq 14$ K. As seen from Fig. 2, the correlations of the local JT distortions ($A \neq 0$) not only enhance substantially the magnetostriction effects in $TmPO_4$ at low temperatures, but lead also to the appearance of an extremum of $\alpha(T)$ at $T_0 = 14$ K. Note that T_0 is considerably lower than $T = 20$ K, where a minimum of the elastic modulus C_{66} is observed, and that the characteristic singularities of the $\alpha(T)$ curve depend substantially on the value of the parameter A.

Virtual elastics are thus quite convenient objects for the experimental study of effects due to correlation of local distortions in Jahn-Teller crystals.

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