ΔE effect in rare-earth phosphates RPO_4

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Experimental and theoretical results are reported on the influence of the magnetic field on Young's modulus (the ΔE effect) of rare-earth phosphates RPO_4 (R = Tb–Yb). A great diversity was observed in the magnitude and character of the field dependences of the ΔE effect in RE phosphates having different Youngs-modulus dependences. These include a giant ($\sim 3\cdot 10^{-1}$) positive ΔE effect of the real Jahn–Teller (JT) elastic $TbPO_4$, nonmonotonic field and temperature dependences of the ΔE effect following sign reversal for the virtual JT elastic $TmPO_4$ and nonmonotonic field and temperature dependences of the ΔE effect of a group of phosphates without JT correlations ($DyPO_4$). The field dependence of the strain susceptibilities, which determine the magnetoelastic renormalization of elastic moduli of varying symmetry, are calculated from the known crystal-field parameters at different magnetic-field orientations, and the elastic constants and directions for which the ΔE effect is a maximum in a crystal are identified.

1. INTRODUCTION

We have investigated experimentally and theoretically the effect of a magnetic field on the elastic moduli of a number of rare-earth (RE) phosphates RPO_4 (R = Tb–Yb) having the zirconium tetragonal structure. The RE zircons are among the compounds in which quadrupole [Jahn–Teller (JT)] interaction can play an important role beside the single-ion magnetoelastic interactions.[1]

Important information of one-ion and paired quadrupole interactions is provided by the study of the temperature dependence of the structural and elastic characteristics of RE compounds. The anomalies of these characteristics, which can be arbitrarily named spontaneous quadrupole effects because they are due to temperature dependences of the quadrupole moments or to strain susceptibilities of the RE ions, have been quite thoroughly investigated for RE intermetallic compounds.[2] The contribution of various RE ions to the thermal expansion and elastic moduli of a number of RE phosphates were investigated in Ref. 3 (where work on other oxide compounds is also cited).

Additional information on these interactions can be gained by studying magnetic-field-induced quadrupole effects such as magnetorestriction phenomena and field-induced changes of the elastic moduli; the latter are in fact the subject of the present paper. The peculiarities of the zirconium crystal structure (tetragonal local symmetry of the RE ion surrounding, the absence of nonequivalent sites in the structure), and the weakness of the magnetic interactions makes it possible to investigate these effects in RXO_4 compounds in the purest form.

Our aim was to study the influence of a magnetic field on the magnetoelastic renormalization of the elastic moduli of RE phosphates in the presence as well as in the absence of JT correlations. It is noteworthy that in JT systems, in contrast to the usual RE compounds, in the presence of degeneracy or quasidegeneracy in the RE-ion spectrum an external magnetic field acts more effectively on the spectrum and wave functions of RE ions, causing thereby more appreciable quadrupole effects. In addition, JT correlations are expected to lead to an additional enhancement of quadrupole effects in magnetic fields of certain direction, when strongly correlating local strains are induced. The contribution of JT correlations to the magnetostriction of RE compounds was investigated experimentally and theoretically in Refs. 4 and 5. To our knowledge, no such investigations of the contribution of JT correlations to the ΔE effect of RE compounds have been reported.

2. THEORY

To calculate the ΔE effect, i.e., the influence of a magnetic field on the elastic moduli of a system, we use the Hamiltonian

$$H = H_{CF} + H_Z + H_Q + H_{me}. \tag{1}$$

For a crystal with tetragonal symmetry, the Hamiltonian of the crystal field (CF) has the known form

$$H_{CF} = \alpha_J B_2^0 O_2^0 + \beta_J (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma_J (B_6^0 O_6^0 + B_6^4 O_6^4), \tag{2}$$

where $\alpha_J, \beta_J, \gamma_J$ are the Stevens parameters, B_n^m are the CF parameters, and O_n^m are equivalent operators. The Zeeman interaction between the magnetic moment of an RE ion and the magnetic field H is described by the usual expression

$$H_Z = g_J \mu_B J H, \tag{3}$$

where g_J is the Lande factor, J is the RE-ion angular-momentum operator, and μ_B is the Bohr magneton. The two-ion quadrupole Hamiltonian H_Q describing the JT correlations is expressed in the molecular-field approximation

$$H_Q = \alpha_J^2 \{ -K^\alpha \langle O_2^0 \rangle O_2^0 - K^\gamma \langle O_2^2 \rangle O_2^2 - K^\delta \langle Q_{xy} \rangle Q_{xy} - K^\varepsilon (\langle Q_{zx} \rangle Q_{zx} + \langle Q_{zy} \rangle Q_{zy}) \}. \tag{4}$$

A magnetoelastic Hamiltonian linear in the components of the strain tensor[6] ε^μ (harmonic approximation) is deter-

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mined in the quadrupole approximation by five independent magnetoelastic coefficients B^{μ}:

$$H_{\mathrm{me}}=\alpha_J\{-(B^{\alpha 1}\varepsilon^{\alpha 1}+B^{\alpha 2}\varepsilon^{\alpha 2})O_2^0-B^{\gamma}\varepsilon^{\gamma}O_2^2$$
$$-B^{\delta}\varepsilon^{\delta}Q_{xy}-B^{\varepsilon}(\varepsilon_1^{\varepsilon}Q_{zx}+\varepsilon_2^{\varepsilon}Q_{zy})\}. \qquad (5)$$

In expressions (2), (4), and (5) we use the notation $O_2^0=3Q_{zz}=3J_z^2-J(J+1)$, $O_2^2=J_x^2-J_y^2$, $Q_{ij}=\frac{1}{2}(J_iJ_j+J_jJ_i)$ $(ij=xy, zx, zy)$.

The influence of the JT correlations depends strongly on the magnetic-field direction. To take this dependence into account correctly, we calculated the spectrum and wave functions of the RE ion, solving numerically the secular equation for the Hamiltonian $H_0=H_{\mathrm{CF}}+H_ZH_Q$. The influence, leading to the ΔE effect, of the magnetoelastic interaction on the spectrum and wave functions of RE ions, was calculated in the very convenient strain-susceptibilities formalism already used by us.[3] The magnetoelastic contribution to the initial elastic constants C_0^{μ} is described by the expression

$$C^{\mu}=C_0^{\mu}-(B^{\mu})^2\chi_{\mu},\quad \mu=\alpha,\ \gamma,\ \delta,\ \varepsilon\ \ (\delta\equiv xy,\ \varepsilon\equiv xz), \qquad (6)$$

and the ΔE effect is determined by the change of the strain susceptibility in the field

$$\frac{C^{\mu}(H)-C^{\mu}(H=0)}{C_0^{\mu}}=-\frac{(B^{\mu})^2}{C_0^{\mu}}[\chi_{\mu}(H)-\chi_{\mu}(H=0)]$$
$$=-\frac{(B^{\mu})^2}{C_0^{\mu}}\Delta\chi_{\mu}. \qquad (7)$$

We note once more that the strain susceptibilities χ_{μ} defined by the equations

$$\chi_{\mu}=\alpha_J^2\Bigg[\frac{1}{Z}\sum_n \exp\{-E_n/k_BT\}$$
$$\Bigg\{\frac{|\langle n|Q|n\rangle|^2}{T}-2\sum_{m\neq n}\frac{|\langle n|Q|m\rangle|^2}{E_n-E_m}$$
$$+\delta_{\mu,\delta}\sum_{m\neq n}\frac{|\langle n|Q|m\rangle|^2}{T}\delta_{E_n,E_m}\Bigg\}-\frac{\langle Q\rangle^2}{T}\Bigg], \qquad (8)$$

$$Q=\begin{cases} O_2^0, & \mu=\alpha\\ O_2^2, & \mu=\gamma\\ Q_{xy}, & \mu=\delta\\ Q_{zx}, & \mu=\varepsilon \end{cases}$$

are calculated for an RE-ion electron structure determined by an Hamiltonian H_0 that includes a Zeeman term as well as the JT correlations. The form of the normalization of the elastic constants on account of the JT correlations (6) differs therefore from the one obtained by perturbation theory.[3]

The strain susceptibilities of RE phosphate were calculated numerically for the CF parameters systematized by us[7] for all RE ions. In the case of JT compounds, the coefficients K^{μ} that determine the JT correlations were varied between reasonable limits using the published data; for $\mathrm{TmPO_4}$ we used our estimates in Ref. 3.

3. SAMPLES AND MEASUREMENT PROCEDURE

We investigated the elastic properties of polycrystalline RE-phosphate specimen synthesized by a ceramic technology with double annealing in air. RE phosphate samples shown by x-rays to be single-phase were obtained by annealing at $T_1=1200$ °C a charge of stoichiometric oxides $\mathrm{R_2O_3}$ and $\mathrm{NH_4H_2PO_4}$. The annealing was repeated at $T_2=1400$ °C to improve the homogeneities and mechanical properties of the ceramics. To obtain single-phase $\mathrm{TbPO_4}$ with zircon structure, the first and second anneals were at $T_1=T_2=1200$ °C. The phase composition was monitored with a "Geigerflex" x-ray diffractometer accurate to $\sim 1\%$. The ceramic-sample density was (65–75)% of the x-ray value.

The Young's modulus E and the internal-friction coefficient q^{-1} were measured with a compound cavity at ~ 150 K in the temperature interval 2–100 K and in magnetic fields up to 40 kOe.

4. RESULTS AND DISCUSSION

A great variety of values and features of the field dependences of the ΔE-effect isotherms were observed for RE phosphates with different Young-modulus temperature dependences. The measured temperature dependences of Young's modulus of RE phosphates made it possible to divide them, depending on the magnitude of the JT interaction, into two groups, the ΔE effect in which is best considered separately.

The first and most numerous group includes those in which the JT correlations are not too substantial, and the renormalization of the elastic moduli can be described by using a single-ion magnetoelastic interaction.[3] The strongest magnetoelastic renormalization of the elastic moduli in this group is observed for $\mathrm{DyPO_4}$ (Ref. 3), which we shall use as the model in the treatment of the ΔE-effect features.

$\mathrm{DyPO_4}$. Figure 1 shows the relative change of Young's modulus $\Delta E(H)/E=[E(H)-E(H=0)]/E(H=0)$ of $\mathrm{DyPO_4}$ as a function of the squared magnetic field at various temperatures. The ΔE effect is evidently quadratic in the magnetic field and has a nonmonotonic temperature dependence, reaching a value $\Delta E(H)/E\simeq 1.2\cdot 10^{-3}$ at 25 K and 40 kOe.

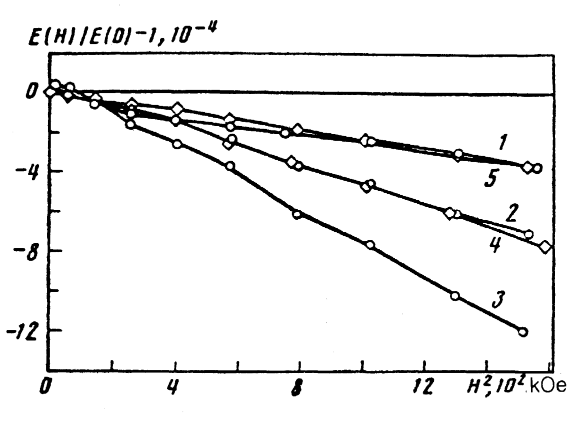

FIG. 1. Isotherms of ΔE effect of polycrystalline $\mathrm{DyPO_4}$ sample at low temperatures: *1*—$T=4.2$ K, *2*—$T=15$ K, *3*—$T=25$ K, *4*—$T=40$ K, *5*—$T=50$ K.

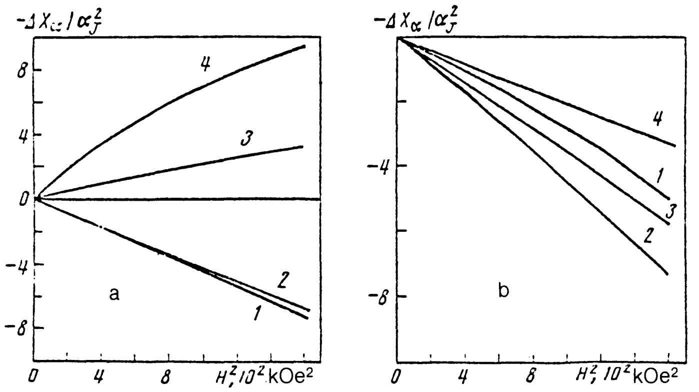

FIG. 2. Calculated field dependences of the strain susceptibility $-\Delta\chi_\alpha$ of $DyPO_4$ at low temperatures: (a) $\Delta\chi_\alpha(H^2)$ dependences at $T = 20$ K for different magnetic-field orientations (*1*—$H \| x$, *2*—$H \| xy$, *3*—$H \| xz$, *4*—$H \| z$); (b) $\Delta\chi_\alpha(H^2)$ dependences for $H \| x$ at different temperatures (*1*—$T = 4.2$ K, *2*—$T = 20$ K, *3*—$T = 30$ K, *4*—$T = 40$ K.

As shown earlier,[3] in the absence of JT correlations the largest contribution to the magnetoelastic renormalization of Young's modulus of a polycrystal is made by the strain susceptibility χ_α, so that the ΔE effect for the polycrystal is due in this case predominantly to the variation of χ_α in the field. Numerical calculations of $\Delta\chi_\alpha(H)$ for different field directions make it possible to analyze its orientational dependence and carry out the averaging for a polycrystal.

Figure 2a shows the calculated field dependences of the strain susceptibility $-\Delta\chi_\alpha(H^2)$ for $DyPO_4$ at 20 K. Evidently, $-\Delta\chi_\alpha$ is negative and practically isotropic in the basal plane (curves *1* and *2*). For a field along the tetragonal axis, the effect is positive (curve *4*), but decreases very rapidly on deviation from this axis. Analysis of the orientation dependence of $-\Delta\chi_\alpha$ and averaging over various magnetic-field orientations yield, in accord with experiment, a negative resultant ΔE effect. The minus sign of $-\Delta\chi_\alpha$ for magnetic-field orientation in the basal plane determines therefore the signs of the ΔE effect for a polycrystal.

Averaging, by a single procedure, $-\Delta\chi_\alpha(H)$ for $DyPO_4$ for various temperatures, it is possible to describe the experimentally observed nonmonotonic temperature dependence of the ΔE effect, with a maximum in the region of 25 K. Figure 2b shows the $-\Delta\chi_\alpha(H^2)$ dependence for a field orientation $H \| x$, which makes the largest contribution to the ΔE effect of a polycrystal at different temperatures. The nonmonotonic temperature dependence of $-\Delta\chi_\alpha(H^2)$ for a field oriented in the basal plane governs the experimentally observed variation of the ΔE effect of $DyPO_4$ effect with temperature.

A quantitative comparison of the experimental data with the theoretical results for magnetoelastic effects calls for knowledge of the magnetoelastic and elastic constants. It is possible, however, to compare the values of the ΔE effect and the temperature anomaly of Young's modulus,[3] which have like dependences on these potentials. As seen from Fig. 1, at $T = 4.2$ K and $H = 40$ kOe we have $\Delta E(H)/E \approx 3.7 \cdot 10^{-4}$. For the temperature anomaly we have $\Delta E(T)/E = [E(T = 4.2 \text{ K}) - E(T = 50 \text{ K})]/E(T = 4.2 \text{ K}) \approx 1.8 \cdot 10^{-2}$ (Ref. 3), and the ratio of these quantities amounts to ~ 49. The ratio of the corresponding averaged differences of the strain susceptibilities $-\Delta\chi_\alpha$ is ~ 33. In our opinion, the proximity of these numbers is evidence that the temperature and field dependences of the Young's modulus of $DyPO_4$ can be described quite adequately by a single set of magnetoelastic and elastic constants.

$TbPO_4$. The ΔE effect for phosphates, which exhibit appreciable JT correlations, is substantially larger and is characterized as a rule by a strong anisotropy and nonmonotonic field and temperature dependences. The distinguishing features of the ΔE effect depend strongly on the magnitude and type of the JT correlations. In the real JT elastic $TbPO_4$, somewhat above the structural phase-transition temperature $T_c = 2.2$ K, the observed ΔE effect is gigantic and positive, $\sim 3 \cdot 10^{-1}$, and decreases with rise of temperature (Fig. 3a). This means that the softening of Young's modulus with decrease of temperature, observed at $H = 0$, decreases substantially to ~ 0.14 in a magnetic field $H = 30$ kOe (Fig. 3b).

The largest contribution to the magnetoelastic renor-

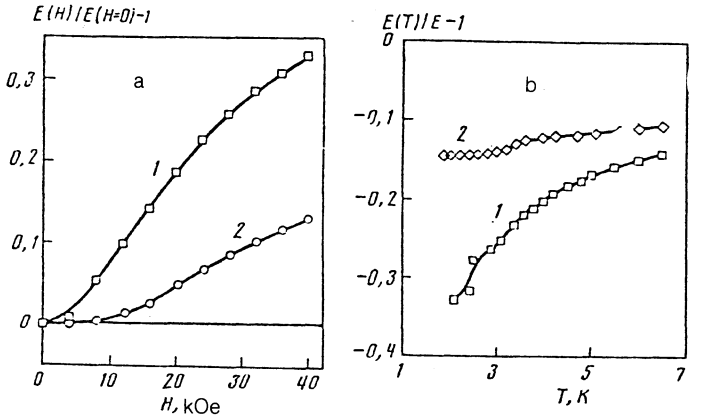

FIG. 3. ΔE effect of polycrystalline $TbPO_4$ sample at low temperatures: (a) isotherms of ΔE effect (*1*—$T = 2.6$ K, *2*—$T = 4.2$ K); (b) temperature dependences of the relative value of Young's modulus for $H = 0$ (*1*) and $H = 30$ kOe (*2*).

malization of the Young's modulus is made in the case of JT compounds by the correlationally enhanced component of the strain susceptibility. The preceding experiments and calculations[3] show that the most substantial for $TbPO_4$ is the strain susceptibility χ_{xz}. The field dependence of $-\Delta\chi_{xz}$ for different magnetic-field orientations at $T = 4.2$ K is shown in Fig. 4a. The magnitude of the ΔE effect depends naturally on the JT-correlations parameter K_ε. The $-\Delta\chi_\mu$ curves of Fig. 4 were calculated using the value $K_\varepsilon = 0.2\cdot 10^3$ K/ion, corresponding to the JT-correlations constant K_ε used in the pseudospin-operator formalism of Ref. 8. Allowance for JT interactions is essential only if the most strongly interacting local strains are induced by a nonzero quadrupole moment Q_{xz}. Analysis shows that for $TbPO_4$ it suffices to take into account the JT interactions for a field oriented in the xz plane.

It is seen from Fig. 4a that an effect that is maximal and positive is observed when the field is oriented in the direction of the spontaneous strain of the lattice in a structural phase transition, i.e., for $H \| xz$ (curve *3*). In fields $H \lesssim 10$ kOe the quantity $-\Delta\chi_{xz}$ is negative and small, but reverses sign and increases sharply in stronger fields. Analysis of the field dependences of the Tb spectrum shows that this is due to a singularity of the crossover type for the second and third levels at $H \simeq 10$ kOe for this field orientation. The effect for other field orientations is smaller and reverses sign at $H \| x$ sign and $H \| z$. It is interesting that the strongest and positive change of the strain susceptibility χ_{xz} takes place when the magnetic field is oriented in the xz plane. Therefore averaging over the magnetic-field orientation yields, in agreement with experiment, a positive ΔE effect for the polycrystal. This accounts also for the weak change of the Young's modulus in fields $H \lesssim 10$ kOe (curve *2* of Fig. 3a).

The variation of the ΔE effect with temperature is clearly seen in Fig. 4b, which shows the temperature dependences of $-\Delta\chi_{xz}$ for $H = 0$ (curve *1*) and $H = 40$ kOe (curves *2* and *3*) with the magnetic field directed along xz. Curves *2* and *3* were calculated in the presence and absence of JT correlations, respectively, so that the role of the JT interaction in the magnetoelastic renormalization of the elastic moduli can be quantitatively estimated. Since the constant K_ε of $TbPO_4$ is not very large, allowance for the JT correlations increases the ΔE effect by only ~25%.

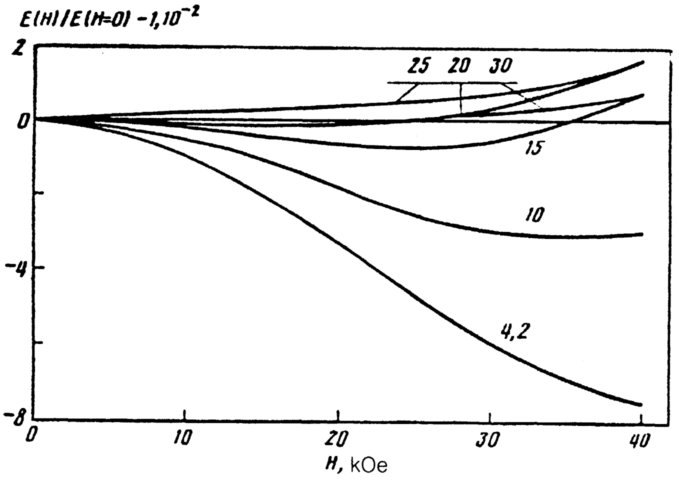

FIG. 5. Isotherms of ΔE effect of polycrystalline $TmPO_4$ sample at low temperatures.

$TmPO_4$. The effect of a magnetic field on Young's modulus of the virtual JT elastic $TmPO_4$ is weaker and differs qualitatively in character from $TbPO_4$ (Fig. 5). A negative ΔE effect amounting to ~0.07 is observed at low temperature ($T = 4.2$ K) in fields up to 40 kOe. The absolute value of the ΔE effect decreases at higher temperatures ($T = 10$, 15, 20 K) and it becomes positive in stronger fields. The ΔE effect of $TmPO_4$ at $T > 25$ K is positive in the entire range of investigated fields.

A magnetic field produces in $TmPO_4$ the largest change, as expected, in the strain susceptibility χ_{xy}, which determines the renormalization of the "soft" modulus C_{66}. Figure 6 shows the field dependence of the strain susceptibility $-\Delta\chi_{xy}$ for different magnetic-field orientations at $T = 4.2$ K. Note first of all the anisotropy of $-\Delta\chi_{xy}$ for H oriented in the basal plane. For $H \| x$ (curve *1*), $-\Delta\chi_{xy}$ is negative and is independent of the JT-correlation constant K_δ. This means that a field $H \| x$ softens C_{66} even more, in agreement with the theoretical[9] and experimental[10] results, according to which a field $H \| x$ induces in $TmPO_4$ a structural phase transition of type B_{2g}.

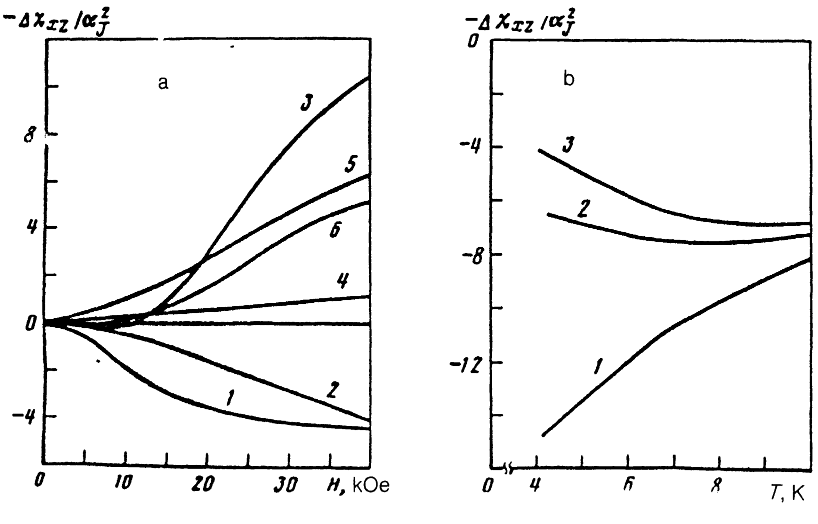

FIG. 4. Calculated field dependences of the strain susceptibility $-\Delta\chi_{xz}$ of $TbPO_4$ at low temperatures with allowance for the JT correlations $K_\varepsilon = 0.2\cdot 10^3$ K/ion; (a) $-\Delta\chi_{xz}(H)$ dependences for various magnetic field orientations (*1*—$H \| z$, *2*—$H \| x$, *3*—$H \| xz$, *4*—$H \| xy$, *5*—$H \| y$, *6*—$H \| xyz$); (b) $-\Delta\chi_{xz}$(T) dependences at $H = 0$ in the absence of JT correlations (*1*) and at $H = 40$ kOe in the absence (*2*) and presence (*3*) of JT correlations for $H \| xz$.

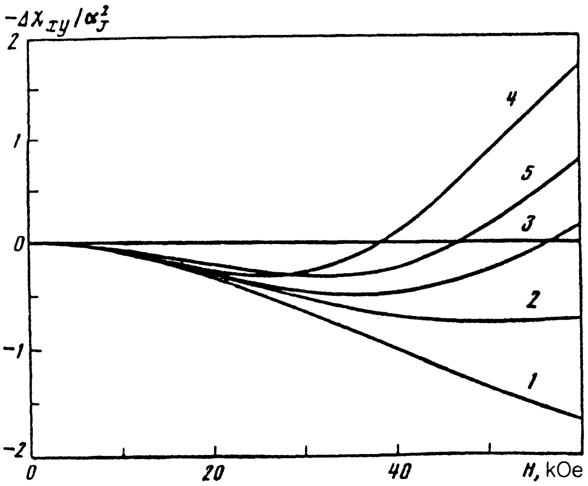

FIG. 6. Calculated field dependences of the strain susceptibility $-\Delta\chi_{xy}$ of TmPO$_4$ at $T = 4.2$ K for various magnetic-field orientations and various JT correlations K_δ: *1*—$H\|x$, *2*—$H\|xy$ and $K_\delta = 0$, *3*—$H\|xy$ and $K_\delta = 2K_\delta^0$, *4*—$H\|xy$ and $K_\delta = 5K_\delta^0$, *5*—$H\|xyz$ and $K_\delta = 5K_\delta^0$; $K_\delta^0 = 0.8\cdot 10^3$ K/ion.

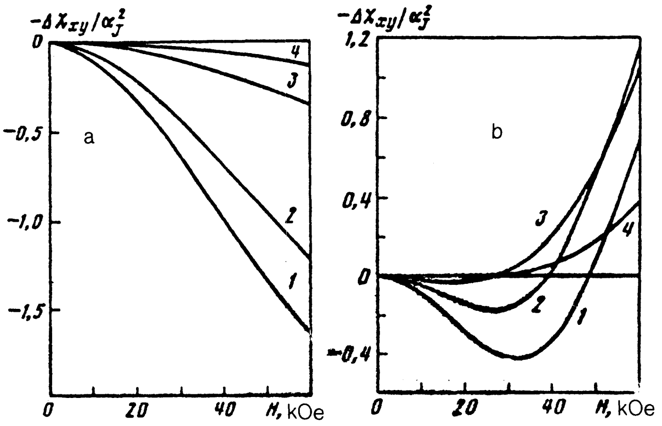

FIG. 7. Calculated field dependences of the strain susceptibility $-\Delta\chi_{xy}$ of TmPO$_4$ for magnetic fields $H\|x$ (a) and for $H\|xy$ and $K_\delta = 3K_\delta^0$ (b) at various temperatures: *1*—$T = 4.2$ K, *2*—$T = 10$ K, *3*—$T = 20$ K, *4*—$T = 30$ K; $K_\delta^0 = 0.8\cdot 10^3$ K/ion.

If $H\|xy$, the $-\Delta\chi_{xy}(H)$ dependence is nonmonotonic and depends substantially on the JT correlations (curves *2*, *3*, *4*). As seen from Fig. 6 , the larger the constant K_δ the weaker the field in which the decrease of $-\Delta\chi_{xy}$ give way to an increase and the steeper the growth of $-\Delta\chi_{xy}$ in strong fields. Our calculations show that this character of the $\Delta\chi_{xy}$ dependence is due to the nonmonotonic field dependence of the lower component of the first excited doublet of the Tm ion.

When the magnetic field leaves the basal plane, the character of $-\Delta\chi_{xy}$ remains unchanged, and the effect decreases the larger the field component along the tetragonal axis. Figure 6 shows by way of example a plot of $-\Delta\chi_{xy}$ at $H\|xyz$ (curve *5*).

Thus, the $-\Delta\chi_{xy}$ dependences for $H\|x$ and $H\|xy$ determine the main features of the behavior of the ΔE effect of a polycrystalline sample. Since the directions x and xy have the same periodicity, the averaging reduces in this case to simple summation of these two dependences. From a comparison of the averaged $\overline{\Delta\chi_{xy}}(H)$ dependence with the experimental $\Delta E(H)/E$ we can estimate the JT correlations K_δ. The best agreement of theory with experiment is for $K_\delta = (2\text{–}3)\cdot K_\delta^0$ ($K_\delta^0 = 0.8\cdot 10^3$ K/ion), in accord with the available published data for TmPO$_4$ (Refs. 4 and 11).

Figures 7a and 7b show plots of $-\Delta\chi_{xy}(H)$ at $K_\delta = 3K_\delta^0$ for $H\|x$ and $H\|xy$, respectively, at various temperatures. These plots describe the variation of the ΔE effect of TmPO$_4$ with temperature. The $-\Delta\chi_{xy}$ dependences for $H\|x$ remain monotonic and decrease in absolute value as the temperature drops. The variation of $-\Delta\chi_{xy}$ at $H\|xy$ with rise of temperature is such that the absolute value of the effect and the field in which the sign of the derivative $d(\Delta\chi_{xy})/dH$, is reversed decrease in weak fields (<10–20 kOe). It is important here that the rate of change of $\Delta\chi_{xy}$ in stronger fields (>30–40 kOe) remains changed with rise of temperature all the way to $T\sim 30$ K. This leads initially to the onset of nonmonotonicity and then to reversal of the sign of the ΔE effect of polycrystalline TmPO$_4$ with rise of temperature.

5. CONCLUSION

We have thus reported experimental and theoretical investigations of the ΔE effect in the RE phosphates RPO$_4$(R = Tb–Yb). Let us formulate briefly the main results.

The variation of the Young's modulus of RE phosphates in a field is adequately described by the changes, calculated using known crystal-field parameters, of strain susceptibilities $\Delta\chi_\mu(H)$ with various symmetries. In the absence of JT correlations (DyPO$_4$) the decisive role is assumed for the ΔE effect by the strain susceptibility $\chi_\alpha(H)$, while the magnitude of the ΔE effect is described, accurate to $\sim 30\%$, by magnetoelastic coefficients determined beforehand from the thermal expansion and the temperature anomalies of Young's modulus.

In the presence of JT correlations (TbPO$_4$, TmPO$_4$), the ΔE effect is determined by the corresponding correlation-enhanced strain susceptibility $\Delta\chi_\mu$, with a strongly pronounced anisotropy of the magnitude and character of the $-\Delta\chi_\mu(H)$ dependence for different magnetic-field orientations. Since the magnitude of the effect, as well as the characteristic fields and temperatures at which the singularities of the ΔE are observed, depend strongly on the JT-correlation parameter, it is possible to determine (or refine) the quadrupole-quadrupole interaction parameters by investigating the ΔE effect.

It must be specially emphasized that in the present paper, unlike in most theoretical studies of JT compounds, we used in the numerical calculations the real spectrum and the wave functions of the RE. This permits not only qualitative but also quantitative comparisons with experiments. Measurements on single crystals are certainly preferable, since they make it possible to invest not only the strongest JT correlations (which are manifested also in polycrystals), but also weaker JT interaction with strains and phonons having various symmetries.

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