Anomalies of Magnetostriction of DyPO₄ Caused by Interaction of Levels

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Abstract—Anomalies of the linear magnetostriction caused by the interaction of energy levels of the rare-earth ion in DyPO₄ in a strong magnetic field along the [100] and [110] axes are investigated both experimentally and theoretically. Smeared jumps and maxima on the magnetostriction curve $\lambda(H)$ and its derivative $d\lambda(H)/dH$ along the three orthogonal axes in the critical fields $H_c = 140$ and 200 kOe, when the lower energy levels of the Dy$^{3+}$ ion converge, are discovered. Under the assumption of the adiabatic property of the magnetization process in pulsed fields, the quadrupolar moments $\langle O_2^- \rangle$, $\langle O_2^+ \rangle$, and $\langle P_{xy} \rangle$ are calculated, which give a rather accurate description of the anisotropy of the linear magnetostriction $\lambda(H)$ and its dependence on temperature and magnetic field. It is discovered that jump-like variation of the quadrupolar interaction of $\alpha$ and $\gamma(\delta)$ symmetry in DyPO₄ under crossover leads, according to the experiment, to the decrease of the critical field by about 20 kOe and a more sharp change of the linear magnetostriction $\lambda(H)$ and its derivative $d\lambda(H)/dH$ in the vicinity of the crossover. © 2001 MAIK “Nauka/Interperiodica”.

1. INTRODUCTION

Rare-earth paramagnetics with a tetragonal zircon structure $RXO_4$ ($X = P, V$) are very convenient for the study of quadrupolar ordering effects [1, 2] caused by quadrupolar interactions, for example, Yahn–Teller structural phase transitions (spontaneous or stimulated by a magnetic field). These paramagnetics are also convenient for the study of crystalline field effects, such as crossing of levels of rare-earth ions in a magnetic field. An analysis shows that the crossing or, in the more general case, convergence of the energy levels of rare-earth ions in a magnetic field (crossover) occurs for almost all rare-earth vanadates and phosphates with a zircon structure in strong and ultrastrong magnetic fields [3]. The crossover effect was discovered and studied in detail for rare-earth zircons HoVO₄ [4, 5], PrVO₄ [6] with Van Vleck ions, and for YbPO₄ [7] with a Kramer's ion when the magnetic field is oriented along the tetragonal axis. As a rule, such a geometry implies a true (without a gap) crossing of the lower levels, which is accompanied by sharp jumps on the magnetization curves at low temperatures. In the case when the magnetic field is oriented in the basal plane, the situation is qualitatively different. Indeed, the symmetry of the interacting levels is such that the magnetic field perpendicular to the tetragonal axis most often mixes their wave functions (mixes the states that differ by the projections $\Delta J_z = \pm 1$, i.e., results in a repulsion of the levels and the occurrence of a gap. This enables one to assume that there exist qualitatively different dependencies of the magnetic characteristics on the magnetic field and temperature when the levels cross.

The profound change of the electron structure of rare-earth ions (their spectra and wave functions) under crossover (in particular, the change of the ground state of the ion) is accompanied by a jump-like change not only of the magnetic moment, but also of various quadrupolar moments. This must lead to anomalies in magnetoelastic characteristics (e.g., magnetostriction) that depend on quadrupolar moments. Magnetostriction anomalies under crossover, which have never been studied, are of interest both from the experimental and theoretical points of view. In particular, crossover can lead to a variation of the contribution of quadrupolar interactions to magnetoelastic effects, which depend not only on quadrupolar constants but also on quadrupolar moments determined by the electron structure of the rare-earth ion. This paper is focused on the experimental and theoretical study of magnetostriction anomalies under crossover by the example of DyPO₄, which has been under intensive study in relation with its ideal Ising properties at low temperatures [8, 9].

2. SAMPLES AND MEASURING TECHNIQUE

We measured the magnetostriction of the crystal DyPO₄ under low temperatures. For the field oriented along the axis of hard magnetization [100], deformations of the crystal along the three mutually perpendicular axes [100], [010], and [001] were measured. These
deformations make it possible to give a complete description of the volume, tetragonal, and orthorhombic deformations of the crystal in this geometry. For $H \parallel [110]$, the crystal deformations were measured along the [110], [110], and [001] axes. In order to measure the magnetostriction in pulsed magnetic fields up to 300 kOe, the method of glued quartz piezoelectric sensor was used [10]. A thin monocrystal quartz (X cut) plate metallized on both sides was glued to the sample under study. The plate was 0.1–0.15 mm thick and had the dimensions of $1 \times 2 \text{ mm}^2$. Under these conditions, only the linear magnetostriction along the $Y$ axis can be measured, since the $X$ cut of quartz is not piezoelectric, and the piezosensitivity shear modulus gives no contribution when the field is oriented along the symmetric directions for which no shear deformations occur.

The signal coming from the quartz sensor caused by the sample deformation was amplified by a specially designed electrometric amplifier. To eliminate the detrimental effect of the connecting cable capacitance, a capacitance compensating circuit with a doubly screened cable (triax) was used. In order to make temperature characteristics more homogeneous, the sample with the quartz sensor attached to it was placed in a nonmagnetic ampoule with a dry mix (1 : 1) of kerosene and diffusion oil. The absolute value of the magnetostriction was calibrated by the known value of the magnetostriction of electrolytic nickel. During the field pulse, the signals coming from the piezosensor $\lambda = \Delta l/t$ and the field coil $H(t)$ were digitized and registered at 0.02 ms intervals (about 500 points). Then, the smoothed linear magnetostriction function $\lambda(H)$ and its derivative $d\lambda(H)/dH$ were calculated using computer.

Rare-earth zircon crystals are layered and brittle, which hinders measurements of their magnetostriction in pulsed fields. Preliminary study showed that the free DyPO$_4$ crystal with a sensor attached to it disintegrates along the cleavage planes after the first field pulse. To protect the sample against disintegration, it was placed in a blob of araldite. When the field pulses were not stronger than the critical crossover field (in this case, the deformation of the crystal does not exceed $5 \times 10^{-4}$, the signal coming from the piezosensor was stable and reproducible, which testified that the sample did not disintegrate. For stronger fields, the signal at the start and the end of the pulse was different, and the maximal value of the signal decreased during two or three pulses; then, the situation became stable. To our opinion, this means that the crystal was partially laminated, and the thickness of the sample coupled to the sensor reduced to about 0.3–0.5 mm. The stability of the signal made it possible to investigate the dependence of magnetostriction on field and temperature. However, since elastic constants of quartz are greater than those of the samples under study, measurements performed on a thin crystal result in a noticeable reduction of the magnetostriction deformation. To estimate the absolute value of the magnetostriction in fields stronger than the critical one, we used referencing to the first measurements made in weak fields before the crystal lamination.

3. EXPERIMENTAL RESULTS

Experimental dependencies of the magnetostriction deformation $\lambda_{[100]}$, $\lambda_{[110]}$, and $\lambda_{[100]}$ along the [100], [001], and [010] axes and their derivatives $d\lambda_{[100]}/dH$, $d\lambda_{[110]}/dH$, and $d\lambda_{[100]}/dH$ for the monocrystal DyPO$_4$ on the strength of magnetic field oriented along the [100] axis are shown in Figs. 1, 2, and 3, respectively. The
behavior of $\lambda_{100}^{100}$ and $\lambda_{100}^{001}$ along the [100] and [001] axes is qualitatively the same. The deformation is small in the fields less than 100 kOe. Then, it increases jump-like near the critical field $H_c = 140$ kOe and tends to saturation in the fields stronger than the critical one. The critical crossover field value $H_c$ can be determined more accurately by derivatives $d\lambda_{100}^{100}/dH$ and $d\lambda_{100}^{001}/dH$ (see inlets in the figures). The value of the critical field for the two geometries is the same; in this field, the magnetostriction reaches a huge value of about $10^{-3}$ and has different signs along two perpendicular axes. As the temperature increases, the dependencies $\lambda_{100}^{100}(H)$, $\lambda_{100}^{001}(H)$ become smeared and the critical field value slightly increases.

The dependencies $\lambda_{100}^{010}(H)$ are more complex: at low temperatures ($T < 15$ K), the magnetostriction is positive in weak fields and changes its sign in the vicinity of the critical field $H_c$. At higher temperature, the magnetostriction varies monotonically within the range of the field intensities examined. It will be shown below that such a behavior of $\lambda_{100}^{010}(H)$ is determined by the competition of different contributions to the magnetostriction.

A similar behavior of the magnetostriction is observed for the field orientation $H \parallel [110]$. By way of example, Fig. 4 presents the dependencies $\lambda_{100}^{001}(H)$ for this geometry. It is seen that the behavior of these curves is basically the same as of those in Fig. 2; the difference is that the critical field value is increased from 140 to 200 kOe.

4. THEORETICAL TREATMENT

The complete Hamiltonian for a single 4$f$ ion includes the Hamiltonian of the crystalline field $H_{CF}$, Zeeman’s term $H_Z$, the bilinear term $H_B$, which describe the interaction of the magnetic moment with the external field $H$ and the exchange field $H_B$, and the Hamiltonian of the quadrupolar interaction $H_{QT}$:

$$H = H_{CF} + H_Z + H_B + H_{QT}. \quad (1)$$

Using the equivalent operator method and the approximation of the molecular field for the pair bilinear and quadrupolar interactions, we can write these terms in the form (for details, see, e.g., [11])

$$H_{CF} = \alpha_J B^0 \langle O^0 \rangle + \beta_J (B^0 \langle O^0 \rangle + B^4 \langle O^4 \rangle) + \gamma_J (B^0 \langle O^0 \rangle + B^6 \langle O^6 \rangle), \quad (2)$$

$$H_Z = -g_J \mu_B H \cdot J, \quad (3)$$

$$H_B = -g_J \mu_B H \cdot J, \quad (4)$$

$$H_{QT} = -G^{\alpha} \langle O^{\alpha}_2 \rangle O_2^{\alpha} - G^{\beta} \langle O^{\beta}_2 \rangle O_2^{\beta} - G^{\gamma} \langle P_{xy} \rangle P_{xy},$$

where $n$ is the bilinear exchange parameter. Here $O^{\alpha}_m$, $\alpha_J$, $\beta_J$, and $\gamma_J$ are the Stevens operator and parameters; $B^\alpha_m$ are the parameters of the crystalline field; $g_J$ and $\mu_B$ are the Landé factor and the Bohr magneton, respectively; and

$$\langle O^{\alpha}_m \rangle = \alpha \sum_i \langle |O^\alpha_m| \rangle \exp(-E_i/k_B T)/Z,$$

$$O^{\alpha}_m = O^2_2, O^6_2, \langle P_{xy} \rangle$$
are the quadrupolar moments. The total quadrupolar constants
\[ G^\mu = G^\mu_{ME} + K^\mu = \left( B_\mu \right)^2/C^\mu + K^\mu \] (\( \mu = \alpha, \gamma, \delta \)) include terms both from the one-ion magnetoelectric interaction \( B^\mu \) and the pair quadrupolar interaction \( K^\mu \) (\( C^\mu_0 \) is the background elastic constant in the absence of interactions). In the Hamiltonian \( H_{QE} \) we dropped the \( \varepsilon \)-symmetry terms, which do not contribute to the orientation of the magnetic field in the basal plane and along the tetragonal axis.

Parameters of the pair interactions for DyPO₄ were determined from the measurements of the first- and third-order susceptibilities and of the parastriction at low temperatures in relatively weak fields for all symmetry modes [12]. For DyPO₄, bilinear interactions are not very small, and lead to the antiferromagnetic ordering of Dy³⁺ ions at \( T_N \sim 3.4 \) K [13]. Estimates show that for the zircon structure the contribution of superexchange and dipole–dipole interactions to the total constant of bilinear interactions are comparable in magnitude. The bilinear interaction constant \( n = \theta/C \) is reliably determined for the easy magnetization axis [001]; however, it is not clear whether or not bilinear interactions are completely isotropic. In the absence of the magnetic field, the quadrupolar interactions \( G^\mu \) in DyPO₄ do not lead to sizable quadrupolar effects, for example, to the spontaneous quadrupolar ordering; however, their role increases in the vicinity of the crossover. In subsequent calculations, we use the values \( \theta = -1.5 \) K \( (H \parallel [001]) \), \( G^\alpha = 1.5 \) mK, \( G^\gamma = 4.4 \) mK, and \( G^\delta = 16.6 \) mK obtained in [12].

The eigenvalues and eigenfunctions, which are necessary for the calculation of thermodynamic properties, were determined by the numerical diagonalization of the complete Hamiltonian in which the quadrupolar interactions of the \( \alpha \) and \( \gamma \) symmetry (which depend on the electron configuration) were taken into account self-consistently. The symmetrized magnetoelastic deformations \( \mathcal{E}^\mu \) \( (\mu = \alpha, \omega, \gamma, \delta) \) are linearly related to the quadrupolar moments \( \langle O^\mu_0 \rangle \) as
\[
\mathcal{E}^{\alpha,2} = A^{\alpha,2} \langle O^\mu_0 \rangle,
\]
\[
A^{\alpha,2} = \frac{B^{\alpha,2}}{C_0^{\alpha,2}} - \frac{B^{\omega,2}}{C_0^{\omega,2}} - \frac{C^{\alpha,2}}{C_0^{\alpha,2}} + \left( \frac{C^{\alpha,2}}{C_0^{\alpha,2}} \right)^2,
\]
\[
\mathcal{E}^\gamma = A^\gamma \langle O^\gamma_0 \rangle = \frac{B^\gamma}{C^\gamma} \langle O^\mu_0 \rangle,
\]
\[
\mathcal{E}^\delta = A^\delta \langle P_{y\gamma} \rangle = \frac{B^\delta}{C^\delta} \langle P_{x\gamma} \rangle.
\]
The coefficients \( A^{\alpha,2}, A^{\omega,2}, A^\gamma, \) and \( A^\delta \), which depend on the ratio of the magnetoelectric coefficients \( B^\mu \) and the elastic constants \( C^\mu_0 \), were determined by measuring parastriction in relatively weak fields. In subsequent calculations, we use the values \( A^{\alpha} = 17 \times 10^{-6}, A^{\omega} = -27 \times 10^{-6}, A^{\gamma} = -66 \times 10^{-6}, \) and \( A^{\delta} = 281 \times 10^{-6} \), which were determined experimentally in [12].

The linear magnetostriction \( \lambda^{\alpha\beta\gamma\delta}_{\beta\gamma\delta} \) oriented along the direction \( (\alpha, \omega, \gamma, \delta) \) induced by the magnetic field oriented along the direction \( (\beta, \gamma, \delta, \delta) \) is related to the symmetrized deformations as (see [14])
\[
\lambda^{\alpha1,01} = \frac{1}{\sqrt{3}} \varepsilon^{\alpha1} + \frac{2}{\sqrt{6}} \varepsilon^{\alpha2},
\]
\[
\lambda^{100,010} = \left( \frac{1}{\sqrt{3}} \varepsilon^{\alpha1} - \frac{1}{\sqrt{6}} \varepsilon^{\alpha2} \right) \pm \frac{1}{\sqrt{2}} \varepsilon^\gamma,
\]
\[
\lambda^{110,01} = \frac{1}{\sqrt{3}} \varepsilon^{\alpha1} + \frac{2}{\sqrt{6}} \varepsilon^{\alpha2},
\]
\[
\lambda^{110,10} = \left( \frac{1}{\sqrt{3}} \varepsilon^{\alpha1} - \frac{1}{\sqrt{6}} \varepsilon^{\alpha2} \right) \pm \frac{1}{\sqrt{2}} \varepsilon^\delta.
\]
The orientation \( (\beta, \gamma, \delta, \delta) \) of the magnetic field appears implicitly in the formulas above by determining the symmetrized deformations \( \varepsilon^{\alpha1}, \varepsilon^{\alpha2}, \varepsilon^\gamma (|H||100|), \) or \( \varepsilon^\delta (|H||100|) \).

5. DISCUSSION OF THE RESULTS
5.1. Crystalline Field and the Zeeman Effect
The crystalline field for DyPO₄ is believed to be reliably determined on the basis of numerous experimental data, including inelastic neutron scattering [15]. We used the parameters of the crystalline field from the paper [12] determined on the basic multiplet: \( B^\alpha = 202 \) K, \( B_\gamma = 22 \) K, \( B_\delta = 1024 \) K, \( B_\delta = -57 \) K, and \( B_\delta = 15 \) K. The Dy³⁺ ion spectra calculated with these parameters in the magnetic field oriented along the [100] and [110] axes (the Zeeman effect) are presented in Fig. 5. The general multiplet splitting at \( H = 0 \) is ~ 430 K. The figure shows only the six lower levels, which make the major contribution to the thermodynamic properties at low temperatures. In the crystalline phosphate field, the ground multiplet \( ^6H_{15/2} \) of the Dy³⁺ ion is split in such a way that the ground state is represented by the “isolated” Kramers doublet is with the almost maximal projection on the \( z \) axis (and the minimal projection on the \( x \) axis). The nearest excited doublet is separated by a gap of about 100 K, and its maximal component of the \( g \)-tensor is oriented along the \( x \) axis \( (g^x \gg g^z) \). In the \( J^-, J^+ \) representation, the wave function of the lower doublet is \( \pm 0.98|15/2 \rangle \); for the excited doublet, it is \( \{ \pm 0.57|\mp 9/2 \rangle \pm 0.62|\mp 7/2 \rangle \pm 0.51|\mp 11/2 \rangle \} \). This is exactly this specific feature of the spectrum and wave functions of the Dy³⁺ ion that determines its Ising prop-
properties and is favorable for the crossing of levels when the field is oriented in the basal plane.

As would be expected, the ground doublet with the maximal component $g_{\tau}\mu_{\mathrm{B}}$ for $H$ $|$ [100] and $H$ $|$ [110] splits weakly, whereas the first excited doublet splits much stronger and differently for the two symmetric field directions in the basal plane. As a result, in the field of about 140 and 200 kOe, respectively, the split sublevels of excited doublets with large projections of the magnetic moment ($M_\tau$) approach the ground level with a small projection ($M_\mu$); this phenomenon results in a sharp increase of the magnetic moment.

A characteristic feature of the Zeeman effect in this geometry is the presence of a substantial gap of about 30 K between the interacting levels, which is retained in the critical field $H_c$. This is due to the fact that the field mixes the wave functions of those levels. For the field oriented along the tetragonal axis, there is usually no gap between the lower levels under crossover in the framework of the Hamiltonian used. The gap appears only at a small deviation of the field from the symmetry axis; i.e., when the component of the field in the basal plane becomes nonzero. The presence of a finite gap under crossover explains the specific features of the magnetic properties in this case, in particular, their dependence on temperature and magnetic field. It is clear that a disorientation of the field both in the basal plane and its going out of the basal plane is less important than for the field oriented along the tetragonal axis.

The comparison of spectra calculated with regard (solid curves) and without regard (dashed curves) to quadrupolar interactions (Fig. 5) shows that their role becomes much more important in the fields stronger than the crossover one and that they cause a sizable change of the magnetic and magnetoelastic characteristics in the crossover region. In this case, even in the paramagnetic and quadrupolar disordered phases, the spectrum depends not only on the magnetic field, but also on temperature, since the field dependencies of the quadrupolar moments $\langle O^5_\tau \rangle (H)$, $\langle O^5_\mu \rangle (H)$, and $\langle P_{xy} \rangle (H)$ vary with temperature. In the absence of the field, the dependence of the spectrum on temperature is caused by the quadrupolar moment $\langle O^5 \rangle (T)$. We stress that the family of rare-earth zircons is a unique class of Jahn–Teller magnetics, in which full-symmetric quadrupolar interactions cause the effects observed.

**5.2. Magnetization Curves and the Magnetocaloric Effect**

In order to interpret magnetic and magnetoelastic properties in pulsed fields with sufficiently small pulse duration, one must calculate adiabatic magnetization processes. In our experiment, the rate of the field input was close to the estimate of the upper bound of the adiabaticity condition given in [16]; thus, we assume the magnetization process of the DyPO$_4$ monocrystal in our experiment to be adiabatic. The subsequent comparison of the calculation results with the experimental ones confirms this assumption.

When calculating the magnetic characteristics for every field value in the range from 0 to 400 kOe with the step $\Delta H = 2$ kOe, the complete Hamiltonian was numerically diagonalized in order to determine the spectrum and the wave functions of the Dy$^{3+}$ ion, and the “elementary” magnetocaloric effect $\Delta T$ was calculated when the field changed from $H$ to $H + \Delta H$:

$$\Delta T = -\frac{\partial M}{\partial T} \frac{T \Delta H}{C_H}.$$  (8)

In formula (8), the total heat capacity of the crystal, $C_H$, includes the lattice heat capacity $C_{\text{latt}} = (12\pi^2 k_B^3 v/(T/T_D)^3$ (the Debye temperature for the phosphate lattice is $T_D = 275$ K [17] and $v = 6$) and the magnetic heat capacity $C_{\text{mag}}$ calculated for every value of the field and temperature on the basis of the rare-earth ion spectrum. These data make it possible to calculate the adiabatic magnetization of DyPO$_4$ and the sample temperature for given directions of the field. The isothermal and adiabatic magnetization curves along the three symmetric directions [001], [100], and [110] and the corresponding curves of the magnetocaloric effect $\Delta T$ for the initial temperatures $T = 5$ and 20 K are shown in Fig. 6. The smeared jumps of magnetization in the field oriented along the axes [100] and [110] correspond to approaching lower energy levels shown in Fig. 5.
For $H \parallel [100]$ in the critical field $H_c \sim 140$ kOe, the magnetization of DyPO$_4$ increases in a jump from about 1$\mu_B$ up to about 10$\mu_B$, which is close to the saturation value. For $H \parallel [110]$, the critical field $H_c \sim 200$ kOe is stronger, the jump is smaller, and the magnetic moment does not reach the saturation level after the crossover. This fact gives reason to assume that there exists another crossover in stronger fields. Indeed, calculations show that for $H_{c2} \sim 3000$ kOe another crossing of the levels occurs, at which the magnetic moment increases in a jump by about 2$\mu_B$ and reaches the saturation level. At low temperatures, the magnetization jumps in the isothermal mode vary insignificantly. Due to a finite magnitude of the spectrum gap, the jump on the curves $M(H)$ at $T = 5$ K for both field directions remains more smeared than, for example, for HoVO$_4$ under similar conditions [18]. In the case of the crossover without a gap, the magnetization jump and, respectively, the differential susceptibility maximum in the framework of the Hamiltonian used become infinitely sharp while approaching the absolute zero.

For $H \parallel [001]$ and $H \parallel [100]$, the adiabatic magnetization curves are flatter than the isothermal ones; this is due to the heating of the DyPO$_4$ crystal in the field. The maximal magnetocaloric effect $\Delta T \sim 25$ K is observed in the field oriented along the easy magnetization axis [001]. For both orientations of the field in the basal plane, the variation of the sample temperature is small in fields weaker than the crossover field ($\Delta T < 5$ K), since the magnetization is small. In fields stronger than $H_c$, the sample heats noticeably for $H \parallel [100]$ and remains almost at the initial temperature for $H \parallel [110]$. Thus, for $H \parallel [100]$, the sample temperature varies monotonically with the field starting from the initial temperature $T_a = 5$ K; however, the rate of the temperature variation is different. For higher starting temperatures, the magnetocaloric effect becomes nonmonotonic for both field orientations in the basal plane, and it is accompanied by cooling of the sample in fields weaker than $H_c$ (see dashed curves in Fig. 6). This is related to a complex behavior of $(\partial M/\partial T)_H$ in the crossover region.

### 5.3. Magnetostriction

The magnetostriction deformation $\lambda_{\parallel/[100]}^{[110]}$ for $H \parallel [100]$ is described by two quadrupolar moments $\langle O_2^2 \rangle$ and $\langle O_2^2 \rangle$, whose linear combination with the corresponding coefficients $A^x$, $A^y$, and $A^z$ determines the crystal deformation along the three mutually perpendicular axes [100], [010], and [001] (see formulas (6) and (7)). The variation of the quadrupolar moments $\langle O_2^2 \rangle$ and $\langle O_2^2 \rangle$ in the field $H \parallel [100]$ in the isothermal mode at $T = 5$ K is shown in Fig. 7 (solid curves). The quadrupolar moment $\langle O_2^2 \rangle$ for the tetragonal crystal is nonzero even in the absence of the field; we see from
Fig. 7 that it slightly varies in weak fields, but decreases jump-like in the critical field, changes its sign, and tends to saturation in the fields stronger than the critical one. On the contrary, the quadrupolar moment \(<O_2^g>\) is zero in the absence of the field, but increases sharply in the critical field and reaches the magnitude of \(\sim 50\) in fields stronger than \(H_c\); this value is comparable with the one that is observed under quadrupolar ordering in rare-earth zircons. Since these quadrupolar moments have different dependencies on the field, their linear combinations can lead to nonmonotonic field dependencies of the linear magnetostriction along certain directions in the crystal.

The deformation along the tetragonal axis \(\lambda_{100}^{001}\) is completely described by the quadrupolar moment \(<O_2^g>\), and its sign is determined by the relation of the coefficients \(A^{01}\) and \(A^{02}\), which have different signs. In addition, the deformations \(\lambda_{100}^{001}\) and \(\lambda_{010}^{100}\) along the [100] and [010] axes include a contribution of the quadrupolar moment \(<O_2^g>\), which is the same in magnitude but has different signs for two axes. Due to relations between various magnetoelastic and elastic coefficients, the linear magnetostriction in DyPO\(_4\) is highly anisotropic and is maximal along the [100] axis when all three terms in (7) have the same sign; it is minimal along the [010] axis.

For \(H \parallel [110]\) \((H_c \sim 200\) kOe), the quadrupolar moments \(<O_2^g>\) and \(<P_{xy}>\) exhibit a similar behavior. Moreover, the variation of the moment \(<O_2^g>\) is almost the same, and that of the moment \(<P_{xy}>\) is by an order of magnitude less than the variation of \(<O_2^g>\) for \(H \parallel [100]\). We note that the quadrupolar moment \(<P_{xy}>\) does not tend to saturation when \(H > H_c\), in contrast to the moment \(<O_2^g>\). The field dependencies of the moments \(<O_2^g>, <O_2^g>, \) and \(<P_{xy}>\) become smeared as the temperature increases, and, in the process, the critical field \(H_c\) increases for both orientations of the field. Thus, the critical field is minimal along the [100] axis and increases when the field deviates from this axis both in the basal plane and in the direction of the tetragonal axis. For the deviation angles \(\Delta\varphi = 5^\circ\) and \(\Delta\theta = 5^\circ\), the increase of \(H_c\) is \(\Delta H_c = 7\) and 10 kOe, respectively.

Since the approach of the energy levels in DyPO\(_4\) is accompanied by substantial increase or variation of the quadrupolar moments, calculations must take into account quadrupolar interactions. The account for quadrupolar interactions of the \(\alpha\) and \(\gamma(\delta)\) symmetry results in a noticeable decrease of the critical field and makes the jumps sharper for both orientations of the field (cf. the solid and dashed curves in Fig. 7). We note that the dependencies calculated with and without taking into account quadrupolar interactions are almost identical in weak fields and begin to deviate in the vicinity of the critical field. This means that the contribution of quadrupolar interactions is much greater in fields stronger than the critical one.

The adiabatic curves of linear magnetostriction along the [100], [010], and [001] axes calculated at \(H \parallel [100]\) and along the [001] axis at \(H \parallel [110]\) are presented in Figs. 1–4 with the purpose of comparing them with the experimental data. The calculation results are in good agreement with the field dependencies of the linear magnetostriction along the three mutually perpendicular directions, including the nonmonotonic behavior in weak fields and the change of sign for \(\lambda(H)\) at \(H \parallel [100]\) (Fig. 3). Such nonmonotonic dependencies of \(\lambda(H)\) are observed only for a certain relationship of the contributions of the quadrupolar moments \(<O_2^g>\) and \(<O_2^g>\), i.e., for a certain relationship between the coefficients \(A^{01}, A^{02}, \) and \(A^\gamma\). In particular, to describe the experimental curves \(\lambda(H)\), one must increase the coefficient \(A^\gamma\) by about 10% compared to that determined by the measurements made in weak fields.

The absolute values of the calculated linear magnetostriction are regularly greater than the experimental ones by a factor of 1.5–2. As has already been discussed above, this can be explained by the lamination of the sample along the cleavage planes in pulsed fields. The small thickness of the sample compared to the sensor result in the experimental underestimate of the magnetostriction magnitude.

### 5.4. Magnetostriction Susceptibility

Experimental data and the theoretical analysis show that the behavior of the curves \(\lambda(H)\), as well as \(M(H)\), is very sensitive to quadrupolar interactions. This dependence can be conveniently analyzed on the differential curves \(dQ_2^2/dH\) and \(dQ_2^g/dH\), which are shown in Fig. 8.

It is seen that the field dependencies \(dQ_2^2/dH\) and \(dQ_2^g/dH\) are similar in the vicinity of the maximum, but are slightly different when the field is substantially less or greater than the critical one. We examine how the behavior of the dependencies changes with temperature by the example of the quadrupolar moment \(Q_2^2\). At temperatures less than 1 K, the field dependencies \(dQ_2^2/dH\) in the crossover region coincide; a small difference is observed only in weak fields. As the temperature increases, the maxima move to the region of stronger fields and become broader for both quadrupolar moments \(Q_2^0\) and \(Q_2^2\). The dependencies of the critical field \(H_c(T)\) and the breadth of the maximum \(\Delta W(T)\) (determined at the half the height of the peak \(dQ_2^2/dH\) on temperature are nonmonotonic (see the inlet), but exhibit extrema at \(T \approx 3\) and 5 K, respectively. These
The calculated adiabatic curve dependence of the initial magnetic susceptibility along the \( [100] \) axis at low temperatures [12]. Calculations were performed without any adjustable parameters of the quadrupole interactions determined and were based only on the magnitudes determined by measurements in relatively small fields.

For the isothermal curve \( d\lambda_{100}^{100}/dH \), the maximum is slightly shifted to stronger field values compared to the adiabatic curve. The main difference of the isothermal and adiabatic curves \( d\lambda_{100}^{100}/dH \) is in their shape. The isothermal curve is almost symmetric; i.e., the location of the peak center is the same at any height. In the adiabatic mode, the curve is asymmetric and is more extended for \( H > H_c \); this is due to the heavy heating of the sample in the fields stronger than the critical one. The comparison shows that the experimental curve is asymmetric as the adiabatic curve \( d\lambda_{100}^{100}/dH \). Thus, the experiment testifies that the magnetization process of the samples under consideration in pulsed fields of the indicated duration is close to adiabatic.

For the field orientation \( H \parallel [110] \), the curves \( d\lambda_{100}^{100}/dH \) exhibit a similar behavior. Quadrupole interactions also decrease the critical field by about 20 kOe, and the maximum of the curves \( d\lambda_{110}^{001}/dH \) becomes greater and narrower approximately by a factor of two, which is in much better agreement with the experiment. When the calculations are performed with the known parameters of the quadrupole interactions determined...
from independent experiments, the critical field \( H_c \) is almost the same or very close to the experimental value both for \( H \parallel [100] \) and \( H \parallel [110] \). We stress that the values of the constants \( G^\mu \) were determined on the basis of the magnetoelastic coefficients \( B^\mu \) found by measuring the parastriction and the elastic constants \( C^\mu \) averaged over the rare-earth phosphate series. In the process, only the contribution to \( G^\mu \) caused by the one-ion magnetoelastic interaction was taken into account, since no reliable data on the parameters of the pair quadrupolar interaction (which usually yields a considerably smaller contribution in zircons) are available. Taking into account this fact, we should admit that the agreement with the experimental data is very good.

6. CONCLUSIONS

In conclusion, we formulate the main results obtained in this study. The investigation of DyPO\(_4\) shows that the crossover in rare-earth compounds at low temperatures is accompanied not only by a jump of the magnetic moment, but also causes anomalies of the linear magnetostriction, which are determined by variations of quadrupolar moments of the rare-earth ion. To our opinion, the investigation of the magnetostriction anomalies under crossover is rather informative, since it makes it possible to determine or refine the values of magnetoelastic coefficients in addition to determining the critical field \( H_c \). In addition, the magnetostriction anomalies, which are directly related to the variation of the quadrupolar moments, enable one to estimate the role of quadrupolar interactions in the effects under study.

A specific feature of DyPO\(_4\) is that there exists a finite gap of \(-30\) K between the approaching levels under crossover when the field \( H \) is oriented in the basal plane. Due to this fact, the magnetization and magnetostriction curves remain rather smeared down to very low temperatures.

A considerable advantage of the systems under study is that they are relatively simple, and there is reliable information on the parameters of interactions available. This enables one to compare theoretical and experimental data not only qualitatively, but also quantitatively. In particular, this comparison shows that under pulsed fields the magnetization process is close to the adiabatic one and is accompanied by a considerable magnetoacaloric effect.

It is established that the contribution of the quadrupolar interactions of the \( \alpha \) and \( \gamma(\delta) \) symmetry for \( H \parallel [100] \) and \( H \parallel [110] \) greatly increases under crossover, which opens up a new method for investigating these interactions and determining or refining the quadrupolar interaction constants. In the absence of the field, the DyPO\(_4\) crystal does not belong to the class of Jahn–Teller compounds; however, for \( H \parallel [100] \) in the vicinity of the crossover, the contribution of the quadrupolar interactions of the \( \gamma \) symmetry becomes comparable in magnitude with that observed for the Jahn–Teller magnetic DyVO\(_4\), which exhibits spontaneous quadrupolar ordering. The magnetic field forms an electronic structure such that the quadrupolar effects for the Dy ion in the phosphate in the vicinity of the crossover field \( H_c \) and in the vanadate at \( H = 0 \) become comparable. In this case, not only the magnetic moment increases jump-like in the critical field \( H_c \), but also the quadrupolar moment \( \langle Q^2 \rangle \), which is characteristic of the stimulated Jahn–Teller transition. It follows from the calculations that the Zeeman effect for lower levels in the presence of quadrupolar interactions is of a more complex nature and considerably depends on the constants of the quadrupolar interactions. In this connection, of interest is not only the study thermodynamic characteristics, but also the direct investigation of the Zeeman effect in DyPO\(_4\) under crossover.

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