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## Low-temperature behavior of the magnetoelastic characteristics of praseodymium ferroborate

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The behavior of the elastic moduli and sound absorption in a  $\text{PrFe}_3(\text{BO}_3)_4$  single crystal at low temperatures is studied. A transition of the magnetic subsystem into an antiferromagnetically ordered state is manifested in the temperature behavior of the sound velocities and absorption. The characteristic behavior of the elastic properties of  $\text{PrFe}_3(\text{BO}_3)_4$  in an external magnetic field is observed. A phenomenological theory that gives a qualitative description of the observed features is constructed. It is proposed that a weak magnetic moment exists in the crystal. © 2010 American Institute of Physics. [doi:10.1063/1.3420962]

### I. INTRODUCTION

Crystals belonging to the family  $\text{RFe}_3(\text{BO}_3)_4$  ( $\text{R} = \text{Y, La-Nd, Sm-Er}$ ) have extremely interesting optical, magnetic, and magnetoelectric properties and they show promise for developing new multifunctional materials. As a result such crystals have been drawing the attention of researchers in recent years.<sup>1,2</sup>

The specific nature of the magnetic properties of rare-earth ferroborates is due to the fact that these materials contain two types of magnetic ions ( $3d$  and  $4f$  elements).<sup>3,4</sup> It is determined by the special behavior of iron magnetic subsystem and the crystal-field formed electron structure of the rare-earth ion on the one hand and by the  $f-d$  interaction on the other. The magnetic structures realized in crystals of this group are very diverse: depending on the type of rare-earth ion these compounds can be easy-axis ( $\text{Tb, Dy}$  ferroborates) or easy-plane ( $\text{Nd}$  ferroborate) antiferromagnets or transition spontaneously from an easy axis to an easy plane state ( $\text{Gd}$  ferroborate).

For certain ferroborates ( $\text{R} = \text{Gd, Nd}$ ), a correlation has been established between their magnetic properties and the elastic and electric properties,<sup>5,6</sup> which enabled the authors of the works mentioned to classify these compounds as multiferroics, i.e. materials where at least two of three order parameters coexist: elastic, electric, or magnetic. Multiferroelectric effects are most clearly manifested with spontaneous or magnetic field induced phase transitions. This is why the study of the elastic properties of these compounds near structural and magnetic phase transitions is of indisputable interest.

The present work is devoted to investigating the elastic properties as well as magnetoelastic effects in praseodymium ferroborate  $\text{PrFe}_3(\text{BO}_3)_4$ .

### II. STRUCTURE AND MAGNETIC PROPERTIES OF $\text{PrFe}_3(\text{BO}_3)_4$

At room temperature  $\text{PrFe}_3(\text{BO}_3)_4$ , just as all rare-earth ferroborates, possesses a trigonal (orthorhombic) crystal lattice symmetry (space group  $R32 (D_3^7)$ ). A characteristic feature of the structure are helicoidal chains of  $\text{FeO}_6$  octahedra, bound by rare-earth ions  $\text{Pr}^{3+}$  and boron ions, along the trigonal  $c$  axis.<sup>7</sup>

Antiferromagnetic ordering of the iron subsystem occurs in all compounds of this family at sufficiently low temperatures ( $20 \text{ K} < T_N < 40 \text{ K}$ ). Investigations of the magnetic susceptibility of single crystal  $\text{PrFe}_3(\text{BO}_3)_4$  showed that an antiferromagnetic magnetic structure with easy-axis anisotropy is realized in this material at temperature  $T_N = 32 \text{ K}$ . The same conclusions were drawn in Ref. 9, whose authors performed a spectroscopic study of this compound by the rare-earth erbium probe method. In this case, while remaining in a paramagnetic state the rare-earth subsystem of the  $\text{Pr}^{3+}$  ions turns out to be an ordered iron system magnetized by a magnetic field.

In a sufficiently strong external magnetic field  $\mathbf{H}$  directed along the trigonal axis  $c$  ( $H_{\text{cr}} = 4.3 \text{ T}$  at  $T = 4.2 \text{ K}$ ,  $H_{\text{cr}} = 6.5 \text{ T}$  at  $T = 31 \text{ K}$ ), a spin-flop spin-reorientation transition is observed in the iron magnetic system. As a result of this transition the spins of the  $\text{Fe}^{3+}$  ions turn into the basal plane.<sup>8</sup>

TABLE I. Absolute values of the sound velocities measured at  $T \approx 77$  K in a  $\text{PrFe}_3(\text{BO}_3)_4$  single crystal

Wave vector $\mathbf{q}$	$S(m, n)$	$S \cdot 10^5$ , cm/s
[100]	$S(x, x)$	8.7
	$S(x, y)$	4.16
	$S(x, z)$	3.17
[010]	$S(y, y)$	9.26
	$S(y, x)$	3.32
	$S(y, z)$	4.26
[001]	$S(z, z)$	7.8
	$S(z, x)$	3.5
	$S(z, y)$	3.5

### III. EXPERIMENTAL RESULTS

Isometric  $\text{PrFe}_3(\text{BO}_3)_4$  single crystals were grown from a fluxed solution based on bismuth trimolybdate by the procedure described in Ref. 10. Sizes up to 10–12 mm were obtained. We worked with a green crystal which was a transparent, hexagonal, well-faceted, prism of the order of 4 mm high in a direction close to the three-fold symmetry axis. The experimental samples with characteristic dimensions  $\sim 1.5 \times 1 \times 1$  mm were prepared from this crystal.

The back-reflection Laue method was used to set the orientation of the samples.

Nonius's method was used to measure the absolute sound velocities in order to obtain adequate accuracy ( $\sim 1\%$ ).<sup>11</sup>

The absolute values of the velocities of longitudinal and transverse sound waves propagating along the  $x$ ,  $y$ , and  $z$  axes of a standard for trigonal crystals Cartesian coordinate system ( $y \parallel C_2$  and  $z \parallel C_3$ ), measured at liquid nitrogen temperature, are presented in Table I. Here and below, as a simplification, the following notation is introduced:  $S(m, n)$  is the velocity of a sound wave in which the wave vector  $\mathbf{q}$  is parallel to the direction  $\mathbf{m}$  and the vector  $\mathbf{u}$  of particle displacements in the sound wave is oriented in the direction  $\mathbf{n}$  ( $\mathbf{m}, \mathbf{n} = \mathbf{x}, \mathbf{y}, \mathbf{z}$ ).

The measurements of the relative changes of the velocity and damping of sound also were performed on an automatic apparatus which is described in Ref. 11. The accuracy of the relative measurements with sample thickness  $\sim 0.5$  mm was  $\sim 10^4$  for the velocity and  $\sim 0.05$  dB for the damping. The temperature range was 1.7–300 K and magnetic field range was up to 55 kOe.

#### A. Temperature dependences of the sound velocity and absorption

While a structural phase transition to the space group  $P3_121$  ( $D_3^4$ ) occurs in most  $\text{RFe}_3(\text{BO}_3)_4$  crystals (mainly in compounds with heavy rare earths), the compound  $\text{PrFe}_3(\text{BO}_3)_4$ , as is well known, retains  $R32$  symmetry to at least 2 K.<sup>7</sup> Our experiments showed that from 300 K to 32 K all longitudinal and transverse acoustic modes show monotonic behavior without any anomalies.

At  $T_N = 32$  K we detected for some modes features which can be interpreted as kinks (or weak jumps superposed on the temperature variation); see, for example, Figs. 1 and 2, which display the temperature variation of the velocity and

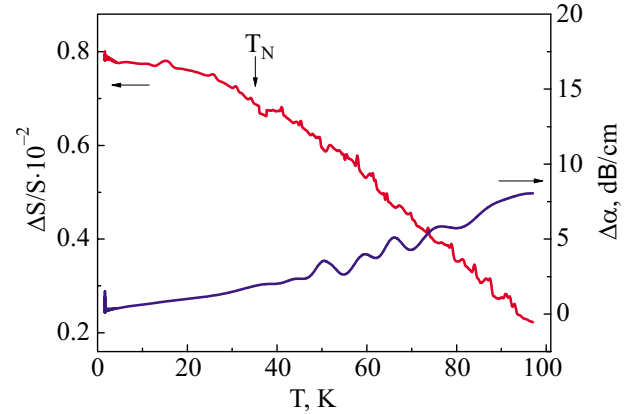


FIG. 1. Temperature dependence of the behavior of the velocity and absorption of the longitudinal acoustic mode  $C_{11}$ ,  $\mathbf{q} \parallel \mathbf{x}$ ,  $\mathbf{u} \parallel \mathbf{x}$ .

damping of the longitudinal and transverse acoustic modes ( $\mathbf{q} \parallel \mathbf{x}$ ,  $\mathbf{u} \parallel \mathbf{x}$  and  $\mathbf{q} \parallel \mathbf{y}$ ,  $\mathbf{u} \parallel \mathbf{x}$ ), where  $\mathbf{x}$ ,  $\mathbf{y}$ , and  $\mathbf{z}$  are the unit vectors in the direction of the corresponding axes. The features in the velocity at  $T_N$  are usually accompanied by weak anomalies in the sound absorption. Information about the fact that a transition into antiferromagnetic ordered state occurs in  $\text{PrFe}_3(\text{BO}_3)_4$  at this temperature (according to the magnetic and optical measurements<sup>8,9</sup>) permits associating the observed anomalies precisely to the Néel point. We note that in the case of terbium ferroborate the transition into the magnetically ordered state was less distinct: the features in the temperature dependence of the elastic characteristics in  $T_N$  were observed only for one transverse mode ( $\mathbf{q} \parallel \mathbf{x}$ ,  $\mathbf{u} \parallel \mathbf{z}$ ).<sup>12</sup>

#### B. Magnetic field dependences of the sound velocity and absorption

The magnetic field dependences of the sound velocity and absorption at fixed temperatures were measured from 0 to 55 kOe for two mutually perpendicular directions of the external magnetic field, coinciding with the direction of the three-fold symmetry axis  $C_3$  ( $z$ ) and lying in the basal plane  $xy$ . For  $\mathbf{H} \parallel \mathbf{z}$  all transverse velocities studied undergo a jump at a definite value of the magnetic field. The scale of the anomalies for different modes is 1–3%. A jump in the sound velocity is accompanied by absorption anomalies. The position of the velocity and absorption anomalies remains practically unchanged with an increase or decrease of the mag-

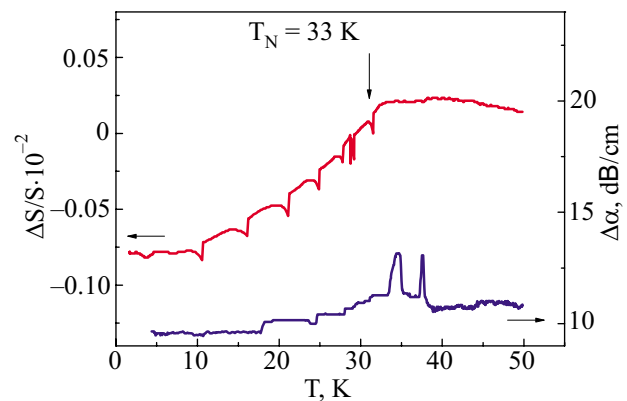


FIG. 2. Temperature dependence of the behavior of the velocity and absorption of the transverse acoustic mode  $C_{66}$ , when  $\mathbf{q} \parallel \mathbf{y}$ ,  $\mathbf{u} \parallel \mathbf{x}$ .

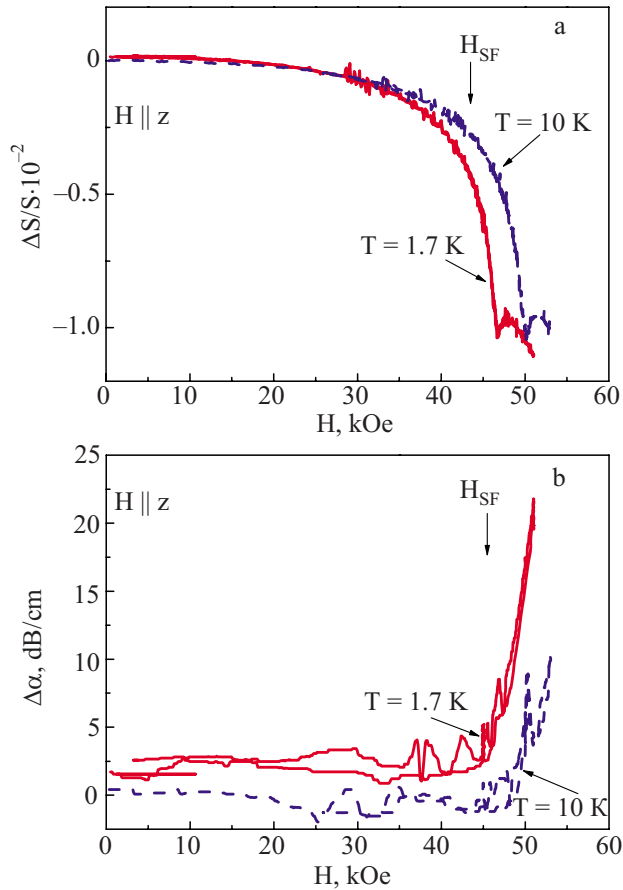


FIG. 3. Magnetic-field dependence of the behavior of the velocity (a) and absorption (b) of the acoustic wave ( $\mathbf{q}||\mathbf{y}$ ,  $\mathbf{u}||\mathbf{z}$ ) at  $T, K$ : 1.7 (—) and 10 (---).

netic field. Examples of the magnetic-field dependences of the transverse sound velocities and absorption are presented in Figs. 3 and 4. An increase of the temperature to 15 K, which almost does not change the magnitude of the velocity jump, results in a shift of the feature in the direction of stronger fields (Fig. 3).

The longitudinal modes likewise exhibit features in the magnetic-field dependences for  $\mathbf{H}||\mathbf{z}$  in the field  $H_{SF}$  and the

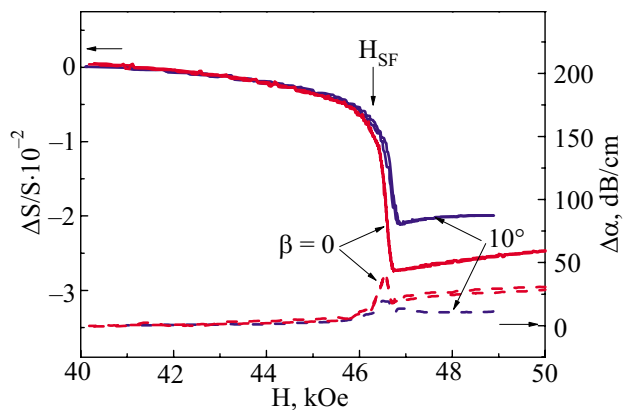


FIG. 4. Magnetic-field dependence of the velocity (—) and absorption (---) of the acoustic  $C_{66}$  mode when  $\mathbf{q}||\mathbf{y}$ ,  $\mathbf{u}||\mathbf{x}$  for angles of declination of the vector  $\mathbf{H}$  from the  $z$  axis  $\beta=0, 10^\circ$  and temperature 1.7 K.

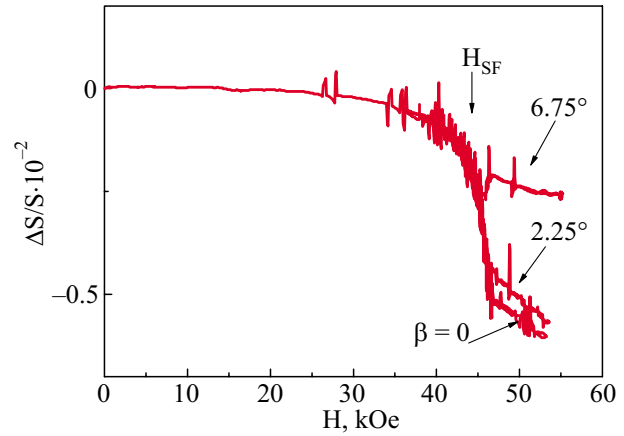


FIG. 5. Magnetic-field dependence of the velocity of the acoustic  $C_{11}$  mode ( $\mathbf{q}||\mathbf{x}$ ,  $\mathbf{u}||\mathbf{x}$ ) with angles of declination of the vector  $\mathbf{H}$  from the  $z$  axis  $\beta=0^\circ, 2.25^\circ, 6.75^\circ$  and temperature 1.7 K.

corresponding temperatures (Fig. 5). These anomalies are much weaker than for the transverse modes (jumps less than 0.5%).

The critical fields of the features which we observed in the behavior of the acoustic modes and the corresponding temperature correlate well with the values of the fields and temperatures for which, according to the magnetic measurements,<sup>8</sup> a spin-flop type spin-reorientation transition occurs in  $\text{PrFe}_3(\text{BO}_3)_4$ .

Recent research on magnetoelastic effects in the isostructural single crystal terbium ferroborate, which possesses a similar magnetic structure, has shown that the behavior of certain transverse acoustic modes near a spin-flop transition is very sensitive to the direction of the external magnetic field.<sup>12</sup> For this reason we performed similar measurements for  $\text{PrFe}_3(\text{BO}_3)_4$  also.

It was found that declination of the vector  $\mathbf{H}$  from the  $z$  axis in the  $xz$  and  $yz$  planes by angle  $\beta$  more than 3 degrees at a fixed temperature shifts a feature in the direction of stronger fields. The magnitude of the jump decreases with increasing angle (Figs. 4–7). When the vector  $\mathbf{H}$  lies in the basal plane, the sound velocity and absorption do not exhibit any anomalies right up to the maximum field (55 kOe) in our experiment.

The most interesting result was observed while investigating the magnetic-field dependences of the behavior of the  $C_{44}$  mode in fields above the field  $H_{SF}$ . We recall that in crystals with trigonal symmetry the  $C_{44}$  mode corresponds to sound waves propagating along the three-fold symmetry axis (the sound wave vector  $\mathbf{q}||\mathbf{z}$ , polarization vector  $\mathbf{u}||\mathbf{y}$  or  $\mathbf{u}||\mathbf{x}$ ).<sup>13</sup> If the external magnetic field is directed strictly along the  $z$  axis, the behavior of the  $C_{44}$  mode in the entire experimental range of fields is practically identical to the behavior of other transverse modes. This is a jump in the velocity, a feature in the absorption at  $H=H_{SF}$ , and the absence of any anomalies in fields  $H>H_{SF}$ ; Figs. 8 and 9 ( $\beta=0$ ).

Let us examine the behavior of the  $C_{44}$  mode when the vector  $\mathbf{H}$  declines from the  $z$  axis.

### 1. Declination of $\mathbf{H}$ from the $z$ axis in the $xz$ plane

Additional features (jumps  $\sim 0.1^\circ$ ) appear in the magnetic field dependences of the velocity of the  $C_{44}$  mode after

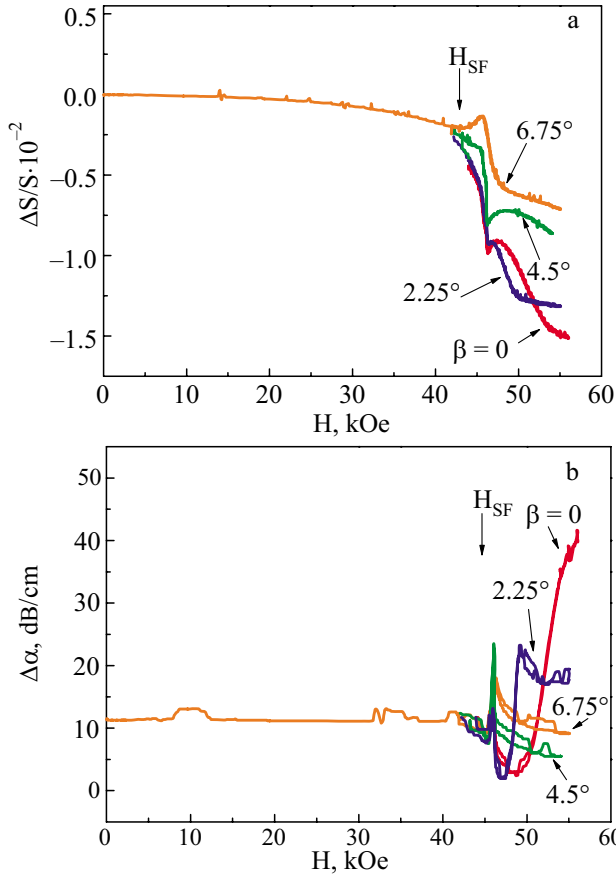


FIG. 6. Magnetic-field dependence of the velocity (a) and absorption (b) of an acoustic wave ( $\mathbf{q} \parallel \mathbf{x}$ ,  $\mathbf{u} \parallel \mathbf{z}$ ) for different angles of declination of the vector  $\mathbf{H}$  from the  $z$  axis and temperature 1.7 K.

the jump at  $H=H_{SF}$  in fields  $H_{cr} > H_{SF}$  starting at small angles ( $\sim 0.1^\circ$ ). The absorption likewise behaves anomalously. The features in the velocity and absorption are accompanied by hysteresis with respect to the magnetic field. An increase of the angle in the range  $0.1^\circ$ – $2^\circ$  shifts the additional features in the direction of stronger fields with practically no change in the position of the jump corresponding to the spin-flop transition (Fig. 8a). As the angle increased to  $15^\circ$  we were able to record only the anomalies corresponding to the spin-flop transition (Fig. 8b).

It should be noted that the behavior of the  $C_{44}$  mode with  $\mathbf{H}$  declining from the  $z$  axis in the positive and negative directions of the  $x$  axis is qualitatively the same (form and position of a feature).

**2. Declination of the vector  $\mathbf{H}$  from the  $z$  axis in the  $yz$  plane**

The magnetic field behavior of the velocity and absorption of the  $C_{44}$  mode with  $\mathbf{H}$  declining in the positive and negative direction of the  $y$  axis differs considerably. For  $\mathbf{H}$  declining in the positive direction of the  $y$  axis the  $C_{44}$  mode shows qualitatively the same behavior as for  $\mathbf{H}$  declining in the  $xz$  plane. After the jump at  $H=H_{SF}$  in fields  $H_{cr} > H_{SF}$ , starting already at small angles ( $\sim 0.1^\circ$ ), additional anomalies of a hysteresis nature appear. However, in this case the velocity jumps at  $H_{cr}$  reach magnitude  $\sim 3.5\%$ , and the range of angles at which they are observed is much narrower— $0.1^\circ$ – $1^\circ$ . However, if the vector  $\mathbf{H}$  declines in the negative

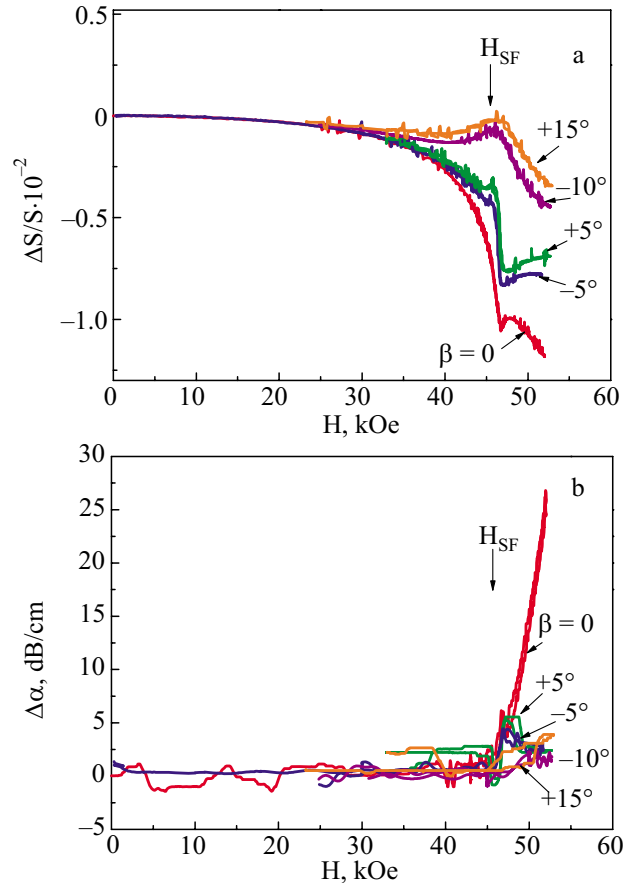


FIG. 7. Magnetic-field dependence of the velocity (a) and absorption (b) of an acoustic wave ( $\mathbf{q} \parallel \mathbf{y}$ ,  $\mathbf{u} \parallel \mathbf{z}$ ) for different angles of declination of the vector  $\mathbf{H}$  from the  $z$  axis and temperature 1.7 K.

direction of the  $y$  axis, then, aside from anomalies at  $H=H_{SF}$ , in fields  $H_{cr} > H_{SF}$  we did not observe any additional features of a hysteresis nature in the entire experimental range of angles ( $0.1^\circ$ – $15^\circ$ ); Fig. 9.

**IV. DISCUSSION**

We have shown in previous work<sup>12</sup> that the magnetic subsystem in rare-earth ferrobates interacts with the elastic subsystem as a result of, first and foremost, the change in the overlapping of the wave functions and, therefore, also the exchange integrals between the iron ions under the action of elastic deformation. However, the exchange interaction between the rare-earth ions, just as the interaction between the irons and the rare-earth ions, most likely makes a smaller contribution to the magnetoelastic characteristics under study. Just as in Ref 12 we assume that the renormalization of the elastic constants of the crystal as a result of relatively weak magnetic anisotropy is not so sharp in praseodymium ferrobates. Here, naturally, the magnetic anisotropy, which is of relativistic origin, is caused by the magnetic-dipole interaction and (or) the interaction of the rare-earth ions with the electric crystal field, since the iron ions are in the  $S$  state. However, we note that even though it is weak this magnetic anisotropy determines the anisotropy of the magnetoelastic response of the system, i.e. the dependence of the elastic characteristics of the response on the relative arrangement of

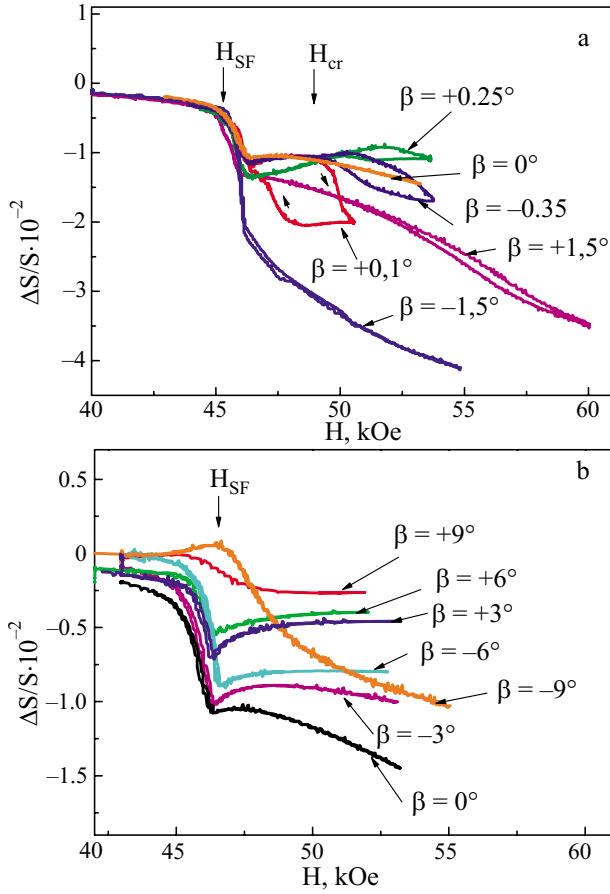


FIG. 8. Magnetic-field dependence of the velocity of the acoustic  $C_{44}$  mode when  $\mathbf{q} \parallel \mathbf{z}$ ,  $\mathbf{u} \parallel \mathbf{x}$  at temperature 1.7 K: for small angles of declination of the vector  $\mathbf{H}$  from the  $z$  axis; the position of  $H_{cr}$  is indicated for  $\beta = +0.1^\circ$  (a) and for large angles of declination of  $\mathbf{H}$  from the  $z$  axis (b).

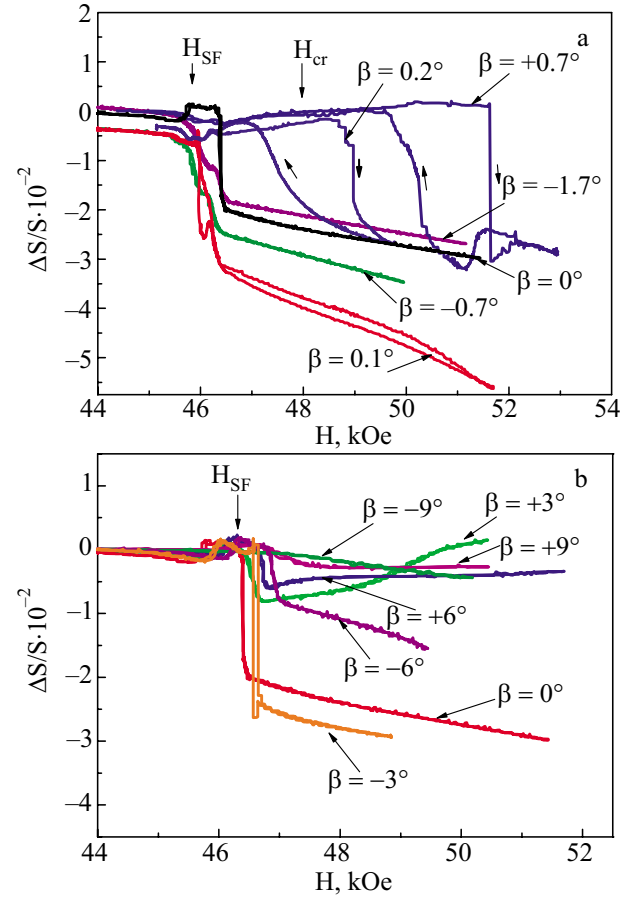


FIG. 9. Magnetic-field dependence of the velocity of the acoustic  $C_{44}$  mode ( $\mathbf{q} \parallel \mathbf{z}$ ,  $\mathbf{u} \parallel \mathbf{y}$ ) at temperature 1.7 K: for small angles of declination of the vector  $\mathbf{H}$  from the  $z$  axis; the position of  $H_{cr}$  is indicated for  $\beta = +0.2^\circ$  (a) and for large angles of declination of  $\mathbf{H}$  from the  $z$  axis (b).

the external magnetic field vector, the wave vector, and the polarization vector of the sound wave.

Let us examine a simple phenomenological model of a two-sublattice easy-axis antiferromagnet interacting with the elastic subsystem.

The expression for the energy of the systems has the form

$$F = A(\mathbf{M}_1 \mathbf{M}_2) - \frac{K}{2M_0^2} ((M_1^z)^2 + (M_2^z)^2) - \mathbf{H}(\mathbf{M}_1 + \mathbf{M}_2) + u_{yz} [B_1(M_1^y M_2^z - M_1^z M_2^y) + B_2(M_1^x M_1^z - M_2^z M_2^x)] + 2C_{44} u_{yz}^2.$$

Here  $A$  is the exchange constant of the antiferromagnetic inter-sublattice interaction,  $K$  is the easy-axis magnetic anisotropy constant ( $K, A > 0$ ),  $B_1$  and  $B_2$  are the magnetoelastic interaction constants, and  $u_{yz}$  is the deformation in the  $yz$  plane. Let the magnetic field  $\mathbf{H}$  make an angle  $\beta$  with the  $z$  axis, and let the magnetizations  $\mathbf{M}_1$  and  $\mathbf{M}_2$  of the sublattices make with this axis angles  $\theta_1$  and  $\theta_2$ , respectively; see Fig. 10.

Let  $AM_0 = H_E$ ,  $K = M_0 H_A$ , and  $B_{1,2} = M_0 H_{B1,2}$ , where  $M_0$  is the magnetic moment of the sublattice. Then the expression for the energy can be written in the form

$$F = M_0 \left[ H_E \cos(\theta_1 - \theta_2) - \frac{H_A}{2} (\sin^2 \theta_1 + \sin^2 \theta_2) - H(\cos(\theta_1 - \beta) + \cos(\theta_2 - \beta)) + u_{yz} \times \left[ H_{B1} \sin(\theta_1 - \theta_2) + \frac{H_{B2}}{2} (\sin 2\theta_1 - \sin 2\theta_2) \right] + 2C_{44} u_{yz}^2 \right].$$

We shall consider two limiting cases:

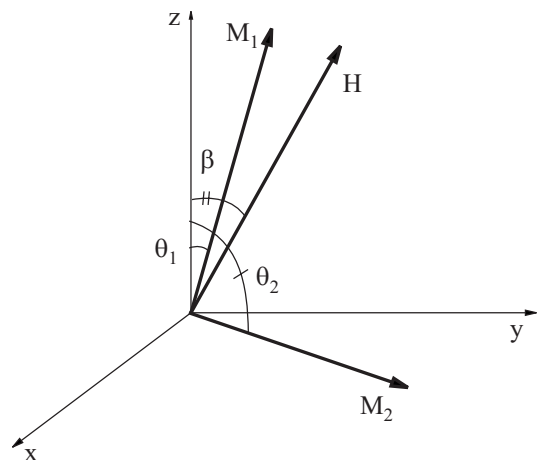


FIG. 10. Illustration of the two-lattice model of an antiferromagnet.

- magnetic field  $\mathbf{H}$  directed along the  $z$  axis ( $\beta=0$ );
- magnetic field  $\mathbf{H}$  directed perpendicular to the  $z$  axis ( $\beta = \pi/2$ ).

Let us find the minimum of the energy  $F$  with respect to  $\theta_1$ ,  $\theta_2$ , and  $u_{yz}$  assuming  $B_1, B_2 \ll A, K, H$ .

The ground state of the magnetic subsystem for the case  $\beta=0$  is realized in three possible configurations:

- 1) paraphase; here  $\theta_2 = \theta_1$  and  $F_{ME} + F_E = 2C_{44}u_{yz}^2$ ;
- 2) antiferromagnetic phase (AF); here  $\theta_2 = \pi - \theta_1$ ,

$$F_{ME} + F_E = 2C_{44}u_{yz}^2 + u_{yz}M_0 \sin(2\theta_1)(H_{B2} - H_{B1}),$$

$$\frac{\partial^2 F_{ME}}{\partial \theta_1 \partial u_{yz}} = 2M_0(H_{B2} - H_{B1})\cos 2\theta_1;$$

- 3) spin-flop phase (SF); here  $\theta_2 = -\theta_1$ ,

$$F_{ME} + F_E = 2C_{44}u_{yz}^2 + u_{yz}M_0 \sin(2\theta_1)(H_{B2} + H_{B1}),$$

$$\frac{\partial^2 F_{ME}}{\partial \theta_1 \partial u_{yz}} = 2M_0(H_{B2} + H_{B1})\cos 2\theta_1.$$

The renormalization of the elastic modulus  $C_{44}$  due to the interaction with the magnetic subsystem is

$$C_{44}^* = C_{44} - \frac{\left(\frac{\partial^2 F}{\partial \theta_1 \partial u_{yz}}\right)^2}{\frac{\partial^2 F}{\partial \theta_1^2}} \Bigg|_{\theta_1 = \theta_{1eq}},$$

where the equilibrium value in the corresponding phase is used for  $\theta_1$ :

in the paraphrase and in the AF phase  $-\theta_1=0$ , and in the SF phase

$$\cos \theta_1 = \frac{H}{2H_E - H_A}.$$

In the paraphrase we take  $C_{44}^* = C_{44}$ , i.e. the modulus  $C_{44}$  does not change in this phase. In the AF phase

$$C_{44}^* = C_{44} - \frac{2M_0(H_{B1} - H_{B2})^2}{(2H_E + H_A)}.$$

In the SF phase we obtain

$$C_{44}^* = C_{44} - \frac{2M_0(H_{B1} + H_{B2})^2}{(2H_E - H_A)} \frac{\left[1 - 2\frac{H^2}{(2H_E - H_A)^2}\right]^2}{1 - \frac{H^2}{(2H_E - H_A)^2}}.$$

Thus the elastic modulus  $C_{44}$  in the SF phase changes abruptly in the field at which a transition occurs from the antiferromagnetic phase into the spin-flop phase. We note that the critical field below which the AF phase is stable equals  $H_2 = \sqrt{(2H_E + H_A)H_A}$ , while the field above which the SF phase is stable equals  $H_1 = (2H_E - H_A)\sqrt{H_A}/\sqrt{2(H_E + H_A)}$  (we note that the expression for  $H_1$  in Ref. 12 contains an error), so that a first-order phase transition occurs at  $H_{SF} = \sqrt{H_1 H_2} = \sqrt{H_A(2H_E - H_A)}$ .

It is evident from the theory presented above that the change of the elastic modulus  $C_{44}$  in the antiferromagnetic

phase, as compared with the paraphase, is proportional to the squared difference of the effective magnetic fields  $(H_{B1} - H_{B2})^2$ , while in the spin flop phase such a change is considerably greater, since it is proportional to the squared sum of such fields.

For  $\beta = \pi/2$  the ground state of the magnetic subsystem is realized in the configuration  $\theta_2 = \pi - \theta_1$  (antiferromagnetic phase). Here two values of  $\theta_1$  are possible:

$$\sin \theta_1 = \frac{H}{2H_E + H_A},$$

$$\cos \theta_1 = 0, \quad \theta_1 = \frac{\pi}{2}.$$

The renormalization of the elastic modulus  $C_{44}$  for the case  $\beta = \pi/2$  with the corresponding equilibrium values of  $\theta_1$  is

$$1) \cos \theta_1 = 0, \quad C_{44}^* = C_{44} - \frac{2M_0(H_{B1} - H_{B2})^2}{(H - 2H_E - H_A)};$$

$$2) \sin \theta_1 = \frac{H}{2H_E + H_A},$$

$$C_{44}^* = C_{44} - \frac{2M_0(H_{B1} - H_{B2})^2}{(2H_E + H_A)} \frac{\left[1 - 2\frac{H^2}{(2H_E + H_A)^2}\right]^2}{1 - \frac{H^2}{(2H_E + H_A)^2}}.$$

In the general case  $\beta \neq 0, \pi/2$  the behavior of the magneto-elastic, just as the magnetic, characteristic depends strongly on the angle of declination of the magnetic field vector  $\mathbf{H}$  from the  $z$  axis. If the angle  $\beta$  is less than the critical value  $\beta_c$  determined from the equation

$$(H^2 \sin 2\beta_c)^{2/3} + [2H_A H_E - H^2 \cos 2\beta_c (1 + [H_A/2H_E]^2)]^{2/3} = H_A(2/H_E)^{1/3},$$

then the behavior of the magnetic<sup>14</sup> and magnetoacoustic characteristics will be similar to the case  $\beta=0$ , while for angles  $\beta > \beta_c$  the behavior will be similar to  $\beta = \pi/2$ .

Evidently (Figs. 8 and 9) the change in the modulus  $C_{44}$  can be described qualitatively on the basis of the proposed simple phenomenological model.

The fact that two jumps (first-order phase transitions) in fields  $H_{cr} > H_{SF}$  (Figs. 8a and 9a) were observed in the field dependences of the change in the velocity and absorption of the acoustic  $C_{44}$  mode can be explained, for example, by the appearance an intermediate state in the crystal. It is known<sup>15</sup> that an intermediate state appears because the magnetization of the sample near the sample surface strives to be parallel to the surface. In consequence, the sample separates into domains. Of course, in an antiferromagnet a nonzero magnetization appears at  $\beta=0$  only in the spin-flop phase, while for  $\beta > \beta_c$  it appears for any nonzero field. It is shown in Ref. 16 that the intermediate state for an antiferromagnetic plate manifests as follows: together with a jump at the spin-flop field  $H_{SF}$  another jump appears in the field dependence of the magnetization at the critical field  $H_{cr} > H_{SF}$ , whose magnitude depends on the demagnetization factor of the plate. For

this reason it is logical to suppose that the two jumps which we observed in the magnetic field dependences of the velocities of certain transverse modes for fields  $H \geq H_{SF}$  are due to the appearance of an intermediate state in the sample. But hysteresis phenomena accompanying the anomalies in the velocity and absorption can be explained by the presence of domain structure in the intermediate state of the experimental antiferromagnetic system.<sup>16</sup>

Finally, the dependence of the behavior of the  $C_{44}$  mode with the vector  $\mathbf{H}$  declining in the  $yz$  plane on whether  $\mathbf{H}$  declines in the positive or negative direction of the  $y$  axis could be due to, for example, the presence of a weak ferromagnetic moment due to the Dzyaloshinskiĭ–Moriya interaction. A symmetry analysis for an orthorhombic system to which the experimental crystal belongs admits the existence of the Dzyaloshinskiĭ vector directed along the  $z$  axis ( $C_3$ ). Here, the weak ferromagnetic moment due to the Dzyaloshinskiĭ–Moriya interaction should lie in the basal plane  $xy$ . Then the difference in the behavior of the  $C_{44}$  mode with the  $\mathbf{H}$  vector declining in the positive or negative directions of the  $y$  axis can be explained by spontaneous symmetry breaking caused by the weak spontaneous magnetization due to the Dzyaloshinskiĭ–Moriya interaction. The presence of weak magnetic anisotropy in the basal plane could be an alternative explanation of such behavior of the  $C_{44}$  mode.

## V. CONCLUSIONS

In summary, the following conclusions can be drawn on the basis of our investigation of the temperature and magnetic-field dependences of the behavior of the velocity and absorption of sound waves in praseodymium ferroboration.

First, the magnetoelastic coupling in this substance, just like in terbium ferroboration, is manifested most strongly not because of the influence of the displacements of the nonmagnetic neighbors (ligands) of rare-earth ions but rather because of the renormalization of the exchange interaction between the iron ions, this exchange being indirect rather than direct. The magnetic-field behavior of the acoustic modes can be described qualitatively on the basis of a phenomenological model of two sublattices of iron ions interacting with the elastic vibrations of the lattice.

Second, the features observed in the magnetic-field behavior of the elastic  $C_{44}$  mode could be a consequence of the presence in the crystal of weak spontaneous magnetization due to a Dzyaloshinskiĭ–Moriya interaction.

Third, the jump-like features observed in the magnetic-field dependences of the velocity of the transverse  $C_{44}$  mode, which exhibit hysteresis, are probably due to the possible

appearance in the crystal of an intermediate state accompanied by the formation of domain structure in fields stronger than the spin-flop transition field.

Of course, investigating only magnetoelastic properties does not make it possible to choose between the proposed mechanisms (magnetic anisotropy or Dzyaloshinskiĭ–Moriya interaction), whose consequences are the features which we observed. For this reason additional studies of the low-temperature characteristics of the ferroboration studied, for example the magnetic-field behavior of the magnetization in the basal plane, that would make it possible to answer the question of whether or not a domain structure characteristic for the intermediate state is present in the crystal are needed.

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