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SHORT NOTES

Low-temperature phase transitions in the rare-earth ferroborate $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$

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The low-temperature behavior of the elastic characteristics of a $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ single crystal has been studied. Features are found in the temperature and magnetic-field dependences of the velocity and absorption of transverse sound. These features are interpreted as being a manifestation of magnetic phase transitions in the compound studied. The H-T phase diagram is constructed. © *2010 American Institute of Physics*. [doi:10.1063/1.3331632]

The rare-earth ferroborates $RFe_3(BO_3)_4$ (R=Y; La–Nd; Sm–Er) with orthoborate structure (trigonal space group R32) are of interest is due to the coupling of the magnetic, electric and elastic subsystems that has been discovered in them, specifically, multiferroelectric effects in some of them. The specific nature of their optical, magnetic, and magnetoelectric properties is due to the exchange interaction between the magnetic subsystems of the iron and rare-earth ions.[1,2] It is believed that below the Néel point $T_N=20-40$ K antiferromagnetic ordering occurs in the magnetic subsystem of iron, while the rare-earth subsystem remains paramagnetic, being magnetized by magnetic field of the iron subsystem (see, for example, Ref. 2). The magnetic structure that must occur in crystals of this group are very diverse: depending on the type of rare-earth ion these compounds can be easy-axis (Tb, Dy ferroborates) or easy-plane (Nd ferroborate) antiferromagnets or spontaneously transition from the easy-axis to the easy-plane state (Gd ferroborate). This diversity of magnetic structures is due to the difference of the ionic radii and the ground states of the rare-earth ions in these compounds. In this connection it is of interest to study the competing contribution of different rare-earth ions to the magnetic anisotropy of the ferroborates of binary compounds of the type $R1_{1-x}R2_xFe_3(BO_3)_4$ (where R1=Dy, Tb and R2=Nd, Er), in which spontaneous orientational transitions from an easy-plane to an easy-axis state can be expected.

The first results from the study of ferroborates of substitute compounds, specifically $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$, appeared in 2008–2009.[3,4] Anomalies were found in the behavior of the magnetic susceptibility[3] at $T_N=32$ K and $T_R=25$ K. The special feature at $T_N=32$ K is attributed to a transition of the crystal into the antiferromagnetic, easy-plane state, and in the opinion of the authors of Ref. 3 the anomaly at $T_R=25$ K corresponds to a spin-reorientation transition into the easy-axis state (analogous to the Morin point in hematite[5]). In Ref. 3 anomalies were also found in the behavior of the magnetization, spontaneous electric polarization, and magnetostriction at a spin-flop transition induced by a magnetic field along the trigonal axis ($\mathbf{H}\parallel C3$) and the H-T diagram of possible magnetic phases arising in the crystal was constructed. The behavior of the magnetization and specific heat of this compound was studied in Ref. 4. The authors of Ref. 4 indicted the presence of three features at $T_1\approx 24$ K, $T_2\approx 22$ K, and $T_3\approx 16$ K, which, aside from T_N, were found in the temperature dependence of the magnetization. Maxima in the behavior of the specific heat were observed at the same temperatures. In addition, for each of the temperatures—2, 4, and 8 K—at least two features were recorded at H_1 and H_2 in the magnetic-field dependences of the magnetization near a spin-flop transition ($\mathbf{H}\parallel C3$), while only one anomaly is observed in thin similar dependences in Ref. 3. We note that in Ref. 6 the temperature of the spin-reorientation transition into the easy-axis state of $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ is determined as $T_R=16$ K.

However, analyzing the experimental data obtained by different methods it can be concluded that the behavior of the magnetic subsystem of the compound $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ is more complicated than proposed in Ref. 3. In the present work we studied the elastic properties of this compound by the ultrasonic method, since the behavior of the other elastic characteristics of magnets at a magnetic phase transition often gives more accurate information about the state of a magnetic subsystem than do other methods.

Isometric $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ single crystals were grown from a fluxed solution based on bismuth trimolybdate by the procedure described in detail in Ref. 4; sizes up to 10–12 mm were obtained. We worked with a crystal consisting of a transparent hexahedral prism, green in color and of the order of 5 mm high, in a direction close to the three-fold axis of symmetry. Experimental samples with characteristic dimensions $\sim 1.5\times 1\times 1$ mm were prepared from it. The samples were oriented using the x-ray back-reflection method (Laue method).

The measurements of the changes of the velocity and damping of sound were performed on the automated apparatus described in Ref. 7. The accuracy of these measurements

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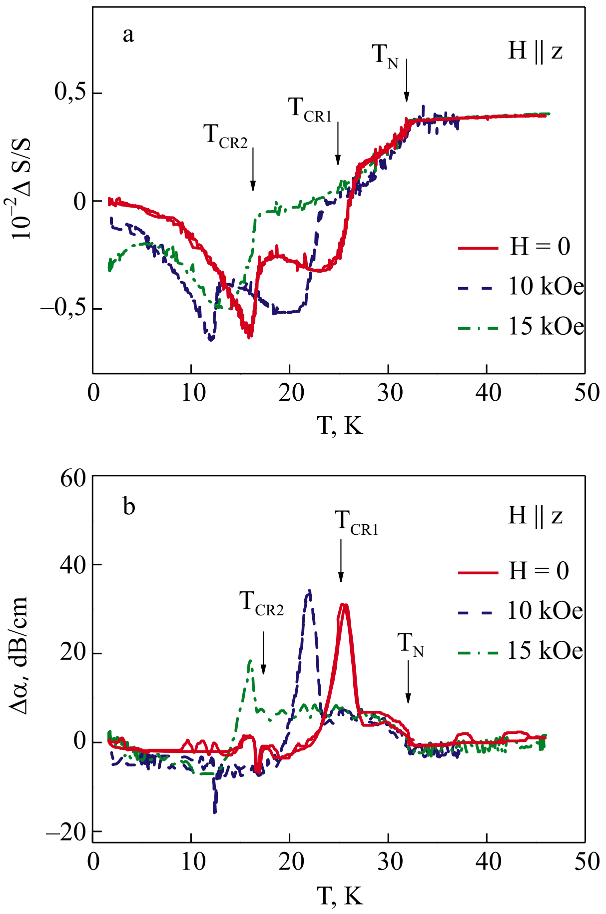

FIG. 1. Temperature dependences of the behavior of the velocity (a) and absorption (b) of the acoustic C_{44} mode ($\mathbf{q}\|\mathbf{z}$, $\mathbf{u}\|\mathbf{y}$) in an external magnetic field $\mathbf{H}$ applied along the trigonal axis: $H=0$ (solid curves), $H=10$ kOe (dashed), and $H=15$ kOe (dot-dash).

with sample thickness ~0.5 mm was $\sim 10^{-4}$ for velocity and ~0.05 dB for damping. We studied the behavior of the velocity and the absorption of the transverse C_{44} sound mode as a function of the temperature and magnetic field.$^{1)}$ The temperature range was 1.7–120 K and the magnetic field range was up to 55 kOe.

At temperatures from 120 to 32 K the propagation velocity and the absorption of the C_{44} mode exhibit typical solid-state behavior without any anomalies. At lower temperatures we observed three features in the behavior of the velocity of the C_{44} mode: at the temperatures $T_N=32$ K (kink), $T_{CR1}=25$ K, and $T_{CR2}=16$ K (jumps ~0.5%), accompanied by absorption anomalies. The application of a magnetic field $\mathbf{H}\|C3$ shifts the features at T_{CR1} and T_{CR2} to lower temperatures but the position of the feature at T_N remains virtually unchanged (Fig. 1).

The magnetic field dependences of the velocity and absorption of the C_{44} mode were investigated at fixed temperatures in the range 1.7–15 K for $\mathbf{H}\|\mathbf{z}$. Two closely spaced features (jumps ~0.5%) exhibiting hysteresis were found in the magnetic field dependences of the velocity of the C_{44} mode in the fields H_{CR2} and H_{CD1}. The velocity jumps are accompanied by absorption anomalies. The critical fields H_{CR2} and H_{CR1} are determined as the average values between the positions of the velocity and absorption anomalies with increasing and decreasing magnetic field. An example of the magnetic field dependence of the velocity and absorption of the C_{44} mode for the lowest temperature of the experiment 1.7 K is presented in Fig. 2a. An increase of temperature shifts them in the direction of weaker fields (Fig. 2b) with virtually no change of scale of the anomalies. The critical fields of the features which we observed in the behavior of the C_{44} mode and the temperature corresponding to them are correlated with the values of the fields and temperatures at which magnetization and specific-heat anomalies were found according to the measurements performed in Ref. 4.

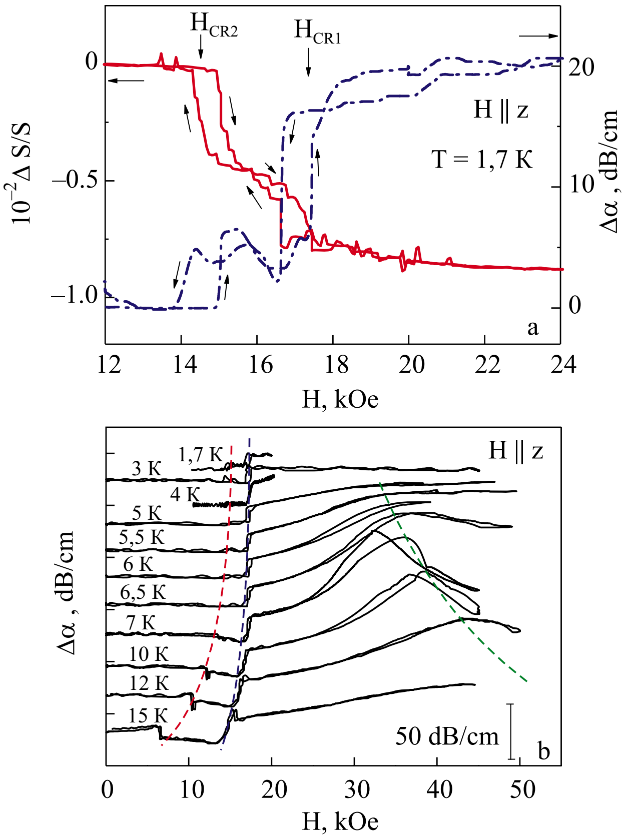

FIG. 2. Magnetic-field dependences of the characteristics of the acoustic C_{44} mode ($\mathbf{q}\|\mathbf{z}$, $\mathbf{u}\|\mathbf{y}$): velocity (solid curves) and absorption (dashed curve) at 1.7 K (a); absorption at different temperatures in the range 1.7–15 K. The dependences for different temperatures are shifted for clarity along the ordinate relative to one another (b).

The measurements performed were used to construct a low-temperature fragment of the *H-T* phase diagram of a $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ crystal for $\mathbf{H}\|C3$ (Fig. 3). The diagram was found to be more complex than the one presented in Ref. 3, and differs from the latter by the presence of the lines 3 and 4.

The existence of the line 3, aside from the previously known phase-transition lines 1 and 2, can cast doubt on the interpretation of the magnetic structural phase transition as a Morin transition between easy-plane and easy-axis phases. (We note that in some orthoferrites and orthochromates such orientational phase transitions have been observed as two second-order phase transitions with respect to temperature.$^{9)}$) Our investigations show that one more low-temperature magnetically ordered phase exists in the compound studied. The differences of the critical fields and temperatures at which this phase exists appear to be too high to consider this phase to be an intermediate state, associated, as is well

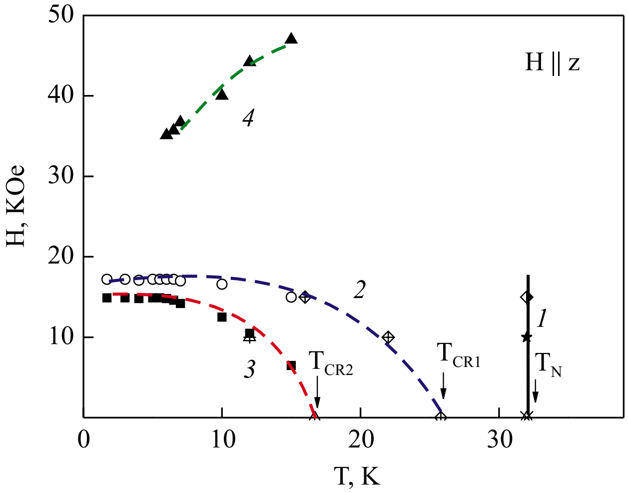

FIG. 3. Fragment of a phase diagram of a $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ single crystal (see text).

known, with the internal magnetization of the sample and the demagnetization factors (see, for example, Ref. 10). An important alternative explanation of the behavior of the ferroborate studied could as follows. Let us suppose that an antiferromagnetic interaction exists inside the magnetic sublattices, which results in the appearance of not two but four magnetic sublattices. Then the presence of two similar metamagnetic phase transitions (which were observed not only in the magnetic-field behavior of the elastic moduli but also in the characteristic low-temperature field dependence of the magnetization of ferroborate[4]) can be given a natural explanation. Such behavior is characteristic for many easy-axis multiple-sublattice antiferromagnets,[8] specifically, magnets containing iron-group magnetic ions as well as rare-earth ions. On the other hand if the supposition that a Morin transition occurs in the system, as in orhtoferrites,[9] is correct, then the behavior which we observed can be attributed to the presence of weak ferromagnetism in the experimental system. The magnetization due to the Dzyaloshinskiĭ interaction should be oriented in the basal plane of the crystal, since we observed the features precisely in the behavior of the modulus C_{44}. The phase-transition lines 2 and 3 can be interpreted as being due to an orientational phase transition between an antiferromagnetic phase of a four-sublattice magnet (the phase with the lowest temperature) and two weakly ferromagnetic phases, which differ from one another by the orientation of the antiferromagnetism and magnetization vectors.

The line 4 tracks the position of the velocity and absorption anomalies of the C_{44} mode, which we observed in fields considerably above H_{CR2} and H_{CR1} (Fig. 2b). It is similar to the analogous line which was observed in Dy orthoborate[3] with no Nd impurity.

In summary, investigating the behavior of the acoustic properties of the ferroborate $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ we discovered new phase transitions (as functions of the temperature and the magnetic field) and we constructed the low-temperature section of the phase diagram. In contrast to pure neodymium and dysprosium ferroborates, this compound characteristically exhibits several phase-transition lines and, correspondingly, several magnetic phases. Our investigations confirmed that the method of studying low-temperature magnetic-field dependences of the variation of the sound velocity and absorption makes it possible to determine the magnetic phase transitions in magnets with complex structure and different nature of the ordering to a high degree of accuracy, higher than that of conventional investigations of the behavior of the magnetic susceptibility and specific heat.

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[1] The C_{44} mode in a crystal with trigonal symmetry corresponds to sound waves propagating along a three-fold axis of symmetry $C3$ (sound wave vector $\mathbf{q}\|C3\|\mathbf{z}$). However, the polarization vector must be oriented in the direction of the axis $\mathbf{y}\|C2$ or the x axis ($\mathbf{u}\|\mathbf{y}$ or $\mathbf{u}\|\mathbf{x}$).

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