Low Temperature Physics

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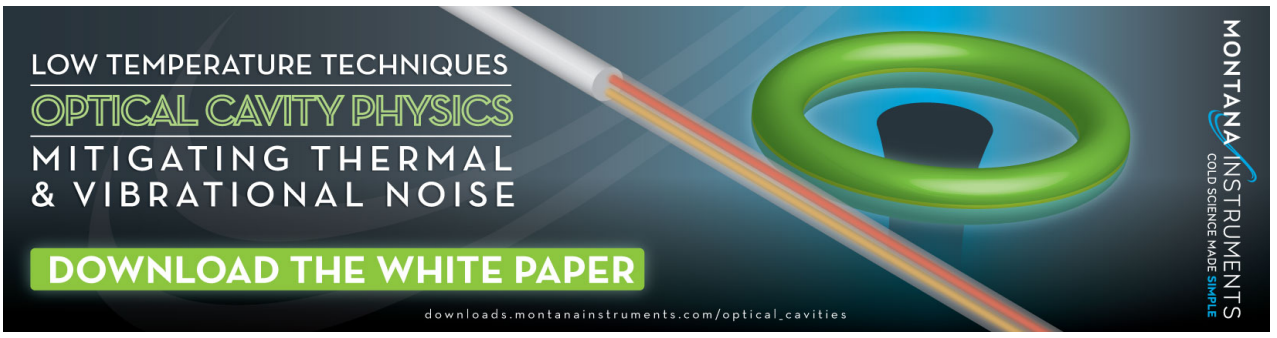




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## LOW-TEMPERATURE MAGNETISM

# Low-temperature magnetic phase transitions in the multiferroic $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$. Part 1. Transitions induced by magnetic fields directed along the trigonal symmetry axis. Spontaneous transitions with temperature changes

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The elastic and magnetic characteristics of single-crystal $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ are studied at low temperatures in zero magnetic field and in external fields $\mathbf{H}||C_3$. The temperature dependences of the acoustic mode velocities and the magnetic susceptibility manifest a transition of the magnetic subsystem into a magnetically ordered state and two successive, spontaneous spin-reorientation phase transitions. The possibility of a spontaneous transition into an incommensurate (spiral) magnetic phase in the crystal is discussed. It is shown that an external magnetic field directed along the trigonal axis of the crystal induces a sequence of spin-reorientation phase transitions. An *H-T* phase diagram ($\mathbf{H}||C_3$) is constructed for this compound. *Published by AIP Publishing.*
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The possibility of controlling the electrical and magnetic properties of materials has stimulated the synthesis and study of the multiferroics—compounds with simultaneous magnetic and electrical, and in some cases, elastic, ordering. It has been found relatively recently that some representatives of the family $RFe_3(BO_3)_4$ of rare earth ferroborates (R = Y:La-Nd; Sm-Er) with a rhombohedral (sp. gr. *R*32) structure and no inversion center manifest a significant magnetoelectric effect,[1,2] so they may have practical applications.

The features of the magnetic and electric properties of the ferroborates are caused by interactions of the *d*- and *f*-magnetic ion subsystems in these compounds.[1,2] Below the Neel temperature $T_N \approx 30$–40 K an antiferromagnetic (AFM) order develops in the iron ion subsystem. The rare earth subsystem plays a key role in the formation of the magnetic configuration of these compounds. Depending on the rare earth ion in a ferroborate, they can be easy-plane (EP), as are the Sm, Er, and Nd compounds, or easy-axis (EA) antiferromagnets, as are the Pr, Tb, and Dy crystals. As the temperature is lowered, in the Gd and Ho ferroborates the magnetic structure changes spontaneously from an easy-plane to an easy-axis state.

The magnitude of the electric polarization, which develops spontaneously in a number of ferroborates in the magnetically ordered phase or upon exposure to an external magnetic field, also depends significantly on the type of rare earth. The highest spontaneous polarizations are attained in the easy-plane ferroborates of Sm and Ho, and the highest magnetoelectric polarizations, in the Nd and Ho compounds.

Substitution of one rare earth ion by others in the confines of a single compound opens up the possibility of changing the properties of its magnetic and electric subsystems, as well as the coupling between them. In particular, it has been found[3,4] that the binary compounds in the series $Ho_{1-x}Nd_xFe_3(BO_3)_4$ ($x = 0.25, 0.5$) combine ferroelectricity with strong magnetoelectric polarization which can be controlled over wide ranges. The electric polarizations (spontaneous and induced) observed in doped crystals even exceed those for the "parent" compositions $RFe_3(BO_3)_4$ (R = Ho, Nd).

Significant changes in the magnetic properties of binary ferroborates compared to the "pure" ferroborates have been discovered in compounds of the series $Nd_{1-x}Dy_xFe_3(BO_3)_4$.[5–7] In particular, a spontaneous readjustment of the EP magnetic structure of crystalline $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ below the Neel temperature to an EA structure has been detected.[5] The EP → EA reorientation proceeds in a complicated fashion:[6,7] through an intermediate magnetic phase whose structure is not yet fully determined. The behavior of the crystal in an external magnetic field which induces a number of spin-reorientation phase transitions is also complicated. The H-T phase diagrams of this compound[7–9] contained several phase transition lines (and, therefore, magnetic phases). This complicated structure of the phase diagrams appears to be caused by interactions among the several magnetic sublattices of the crystal.

The experimental data accumulated thus far indicate that changing the concentrations of the rare earth ions in the compounds $Nd_{1-x}Dy_xFe_3(BO_3)_4$ has a significant influence on the possibility of spontaneous and induced orientational phase transitions in these materials.

Thus, in $Nd_{0.6}Dy_{0.4}Fe_3(BO_3)_4$ a spontaneous EP → EA reorientation is most likely absent; below the ordering

temperature the system immediately enters an easy-axis magnetic state.[10] However, an external field $\mathbf{H}||C_3$ induces two successive phase transitions over the entire range of temperatures below the Neel point.[10]

In $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$, a spontaneous EP → EA realignment takes place as a result of two phase transitions at $T_{CR1} \approx 25$ K and $T_{CR2} \approx 16$ K—through an intermediate magnetic phase. A sequence of two reorientational phase transitions is also observed in an magnetic field $\mathbf{H}||C_3$, but as opposed to $Nd_{0.6}Dy_{0.4}Fe_3(BO_3)_4$, only at temperatures $T \leq T_{CR2}$.[7,11]

In crystalline $Nd_{0.85}Dy_{0.15}Fe_3(BO_3)_4$ a spontaneous EP → EA reorientation takes place[11] as the result of a single phase transition at $T_{CR} \approx 13$ K. However, in field $\mathbf{H}||C_3$ at temperatures $T \leq T_{CR}$, two successive reorientational phase transitions have been observed, one of which is observed up to $T_{CR}$, while the second is resolvable only to 10 K.

An analysis[12] of magnetic measurements[6] indicates that in $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ and $Nd_{0.85}Dy_{0.15}Fe_3(BO_3)_4$ the magnetic configuration readjusts from an EP to an EA state in a single step (at $T_{CR} \approx 8$ K) as the temperature is lowered in the absence of a field. An external field $\mathbf{H}||C_3$ induces only one spin reorientation phase transition, which is observed for $T \leq T_{CR}$.

In a study of the low-temperature behavior of the elastic characteristics of $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$, we found[13] that a magnetic field directed along the trigonal symmetry axis induces two successive phase transitions, rather than one as had been shown in Ref. 12. We suggested that the readjustment of the magnetic structure under the influence of a magnetic field with this orientation proceeds here by a scenario analogous to that in $Nd_{1-x}Dy_xFe_3(BO_3)_4$ ($x = 0.15, 0.25, 0.4$). But verification of this hypothesis will require more detailed (than in Refs. 6 and 12) studies of the magnetic characteristics of this compound. In addition, the readjustment of the magnetic structure of this compound during temperature changes in the absence of a magnetic field and under the influence of external magnetic fields with other orientations is still an open question.

For this reason, here we carry out a comprehensive investigation of the elastic and magnetic properties of $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ in the vicinity of phase transitions. This paper is divided into several sections for convenience. In the first part we discuss phase transitions in the crystal in external fields $\mathbf{H}||C_3$, as well as with changing temperature in zero magnetic field. The second part is devoted to a study of the behavior of the crystal in magnetic fields oriented in the basal plane, and the third part examines the behavior of the magnetoelastic characteristics of this compound in "oblique" fields. The observed phase transitions are classified and segments of the *H-T* phase diagrams corresponding to different directions of an external magnetic field are constructed.

### Experimental technique and samples

The single crystals of $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ grown at the Institute of Physics of the Siberian Branch of the Russian Academy of Sciences from a solution-melt based on bismuth trimolybdenate using the technology of Ref. 6 were transparent, greenish hexahedral prisms with heights of 5–10 mm in a direction close to the third order $C_3$ axis of symmetry. Experimental samples for acoustic and magnetic measurements were prepared from these crystals. In order to reduce the effect of errors owing to internal stresses and growth defects, samples of different shapes and sizes were cut from different single crystals. Sample No. 1 (bulk) had dimensions of ~2 mm (along the $C_3$ axis), ~4 mm (along the $C_2$ axis), and ~3 mm ($\perp C_2$ axis). Sample No. 2 (thin) had corresponding dimensions of $0.9 \times 1.2 \times 1$ mm. The samples were oriented using the Laue x-ray technique. The working facets of the samples were polished with corundum powder and their plane-parallelism was checked with an optimeter.

Measurements of the relative velocity changes and acoustic absorption were made on an automatic system.[14] The accuracy of the measurements for sample thicknesses of ~0.5 mm was $\sim 10^{-4}$ for the velocity and ~0.05 dB for the damping. The variations in the velocity and absorption of transverse acoustic modes were studied as a function of temperature in the range of 1.7–120 K and of magnetic field up to 5 T.

The magnetic characteristics of the crystal were studied with an MPMS-XL SQUID magnetometer. The magnetization of the crystal was studied as a function of temperature in the range of 2–200 K and of magnetic field up to 5 T.

### Results and discussion

The temperatures for magnetic ordering ($T_N = 31$ K) and for spontaneous spin-reorientation phase transitions ($T_{CR} = 8$ K) of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ have been determined previously.[6,12] Here we study the behavior of the magnetization in external magnetic fields directed along the trigonal axis and perpendicular to it in the basal plane of the crystal. We believe that the system undergoes a transition from an easy-plane to an easy-axis state below $T_{CR}$. Imposing an external magnetic field $\mathbf{H}||C_3$ when $T \leq T_{CR}$ leads to a spin-flop transition in the magnetic subsystem of the iron ions. The magnetic moments of the rare earth ions then align themselves along the field. The jump observed in the magnetic field behavior of the magnetization at 2, 3, 5, 6, and 7 K has been interpreted[12] as a manifestation of this phase transition. However, with the same orientation of the external field, we have observed[13] two closely spaced features in the magnetic field dependences of the acoustic characteristics of the crystal, rather than one as expected according to Refs. 6 and 12. We believe that these two features are magnetic in nature and correspond to two successive reorientation phase transitions.

In order to test this hypothesis and construct an *H-T* phase diagram ($\mathbf{H}||C_3$) of this compound, we have made a detailed study of its magnetic characteristics. We have also studied the magnetic field behavior of several transverse acoustic waves, which have not been studied before.

Some examples of acoustic mode velocities, $C_{44}$ $(\mathbf{q}||\mathbf{c}, \mathbf{u}||\mathbf{b})$ and $C_{66}(\mathbf{q}||\mathbf{b}, \mathbf{u}||\mathbf{a})$,* at temperatures $T \leq T_{CR}$ as functions of magnetic field are plotted in Fig. 1. These plots

*The following notation is used in the figures: $\Delta s/s$ are the relative changes in the velocities of the acoustic waves ($\mathbf{q}$ is the wave vector and $\mathbf{u}$, the polarization) propagating along the *a*, *b*, and *c* axes of the trigonal crystal ($a||C_2, c||C_3$). The $\Delta s/s(T)$ and $\Delta s/s(H)$ curves in the figures are shifted relative to one another along the ordinate for clarity.

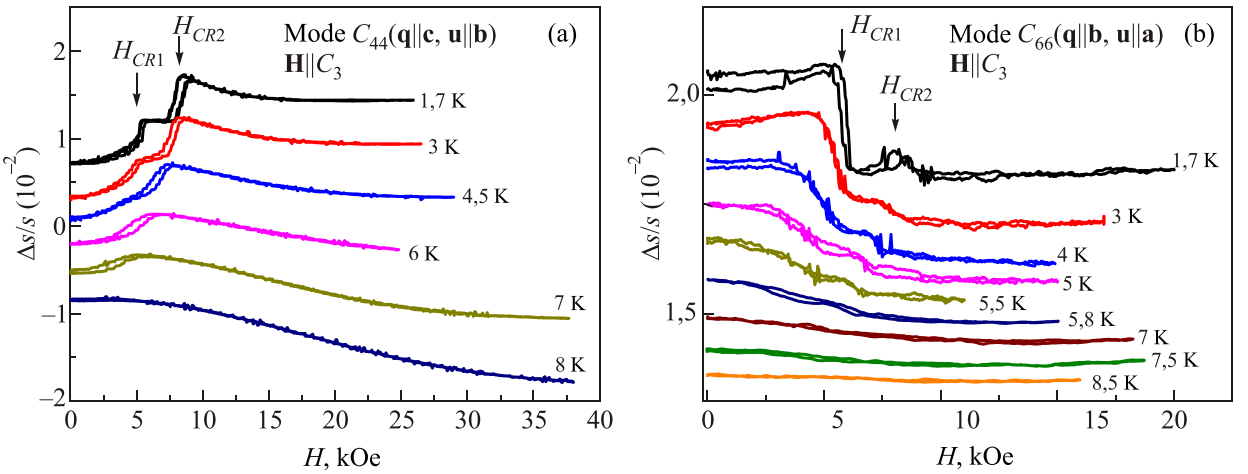


FIG. 1. Magnetic field dependences of $C_{44}$(**q**||**c**, **u**||**b**) (a) and $C_{66}$(**q**||**b**, **u**||**a**) (b) acoustic mode velocities of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ measured at fixed temperatures in the range 1.7 K ≤ T ≤ 8.5 K with **H**||$C_3$.

show that in fields $H_{CR1}$ and $H_{CR2}$ the velocities have anomalies (jumps) of a hysteresis type. An increase in the temperature shifts these features to lower fields. Then the scale of the jumps decreases and they are spread out. We note that the magnitude of the jump at $H_{CR2}$ exceeded that at $H_{CR1}$ for most of the transverse modes (Figs. 1(a) and 2 from Ref. 13). For this reason, we could reliably find the feature at $H_{CR2}$ essentially up to $T_{CR}$, while the anomaly at $H_{CR1}$ could be found only up to 4.5 K. The behavior of the $C_{66}$ mode, which we had not studied previously, was a pleasant exception; for it the jump at $H_{CR1}$ was considerably bigger than the one at $H_{CR2}$ (Fig. 1(b)). This made it possible to extend the range in which an anomaly at $H_{CR1}$ could be observed up to 5.8 K.

The behavior of the magnetization of the crystal in a field **H**||$C_3$ confirms our assumption about the magnetic nature of the acoustic features observed at fields $H_{CR1}$ and $H_{CR2}$. The two-step anomalies were found in plots of the magnetization as a function of magnetic field at temperatures of 2, 3, 4, and 4.5 K (Fig. 2). For higher temperatures ($T \leq 7$ K) only one step, the one at $H_{CR2}$, is still visible. The values of $H_{CR1}$ and $H_{CR2}$ obtained from the acoustic and magnetic measurements are in good agreement with one another (Fig. 3). Note that only one jump, at $H_{CR2}$, was observed in the previous measurements of the magnetization in fields **H**||$C_3$.[6,12]

We can, therefore, conclude that these features of the magnetic field dependences of the acoustic velocities and magnetization are caused by magnetic (spin-reorientation) phase transitions in $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$. These transitions are probably accompanied by the appearance of a magnetically ordered state, as indicated by the hysteresis in the anomalies responsible for these transitions.

It can be seen that the realignment of the magnetic structure of the crystal induced by fields **H**||$C_3$ is also produced in other compounds of the family $Nd_{1-x}Dy_xFe_3(BO_3)_4$ ($x = 0.15, 0.25, 0.4$)[7,9–11] by a sequence of at least two spin-reorientation phase transitions. This means that reducing the dysprosium concentration in the crystal (from 40% to 10%) only lowers the critical magnetic fields $H_{CR1}$ and $H_{CR2}$, while the character of the phase transitions induced by fields **H**||$C_3$ apparently does not change.

We now examine the question of how the spontaneous EP → EA transition takes place in this compound. Does it occur in an intermediate phase? According to experimental data,[11] even in a crystal with 15% dysprosium this kind of reorientation took place as the result of a single phase transition without formation of an intermediate phase. On the other

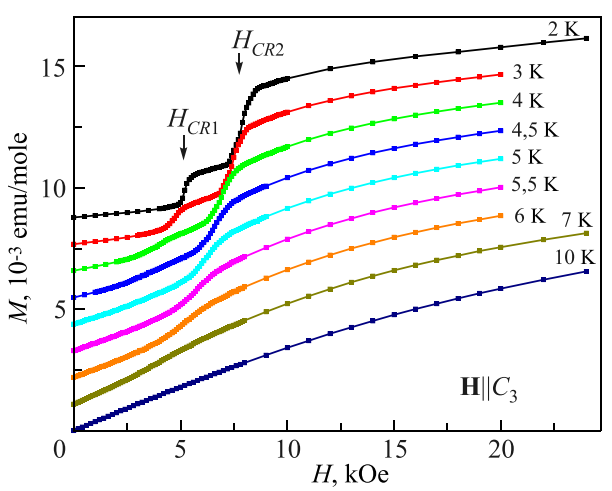


FIG. 2. Magnetic field dependences of the magnetization of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ measured at fixed temperatures in the range $2 \leq T \leq 10$ K, with **H**||$C_3$.

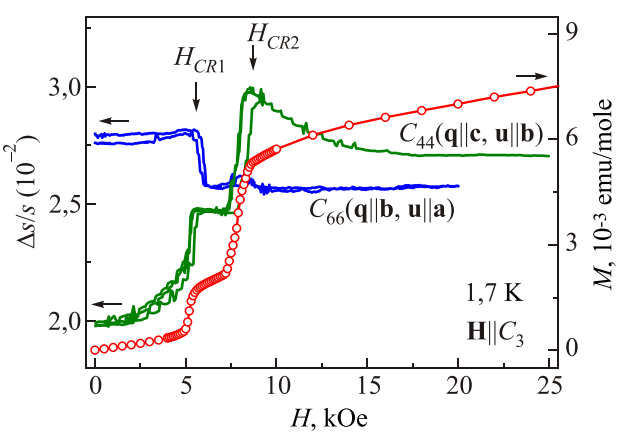


FIG. 3. Magnetic field dependences of the velocities of the acoustic modes $C_{44}$ and $C_{66}$ ($T = 1.7$ K) and of the magnetization ($T = 2$ K), with **H**||$C_3$, of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$.

hand, the possibility of an EP → EA realignment as a result of two phase transitions has been supported theoretically.

We hoped to obtain an answer to this question by studying the behavior of the magnetic and acoustic characteristics of the compound with varying temperature in the absence of an external magnetic field. Like all the representatives of the series $Nd_{1-x}Dy_xFe_3(BO_3)_4$ that have been studied, crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ has no anomalies at the magnetic ordering point in the temperature dependence of the susceptibility $\chi_c$ measured along the $C_3$ axis. A weak feature corresponding to $T_N$ can be seen only in the temperature dependence of $\chi_{\perp c}$ (Fig. 4). $\chi_c$ increases below $T_N$. This behavior is assumed[12] to be characteristic of an easy-plane state. The $\chi_c$ curve reaches a maximum at $T_{CR1} \approx 8$ K. Then there is a sharp drop in $\chi_c(T)$ which was interpreted[12] as a manifestation of a transition into the easy-axis state. Our data on $\chi_{c,\perp c}$ generally confirm the measurements of Refs. 6 and 12. However, in a detailed study of $\chi_c(T)$ below $T_{CR1} \approx 8$ K we found a change in its slope which is difficult to resolve because of the steepness of this section of the curve (inset to Fig. 4). If it is assumed that the EP → EA realignment in $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ takes place (as in $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$) in two stages through an intermediate magnetic phase, then this feature may be related to a second reorientation phase transition.

We now test this assumption using data from the acoustic experiments. Our experience in studying the rare earth ferroborates indicates that the anomalies in their elastic properties in the vicinity of magnetic phase transitions are often more distinct than the anomalies in their magnetic characteristics. For example, the transition into a magnetically ordered state which barely shows up in the susceptibility, can be seen clearly in the temperature dependences of the velocities of all the transverse acoustic modes (Fig. 5). This kind of characteristic behavior in the acoustic velocities near $T_N$ has also been detected in the binary compounds $Nd_{1-x}Dy_xFe_3(BO_3)_4$ ($x = 0.25,\ 0.4$),[7–10] as well as in the "pure" Nd ferroborate (Fig. 7(b)).[15]

At the point where the proposed spontaneous EP → EA spin reorientation takes place ($T_{CR1} \approx 8$ K), the velocities of

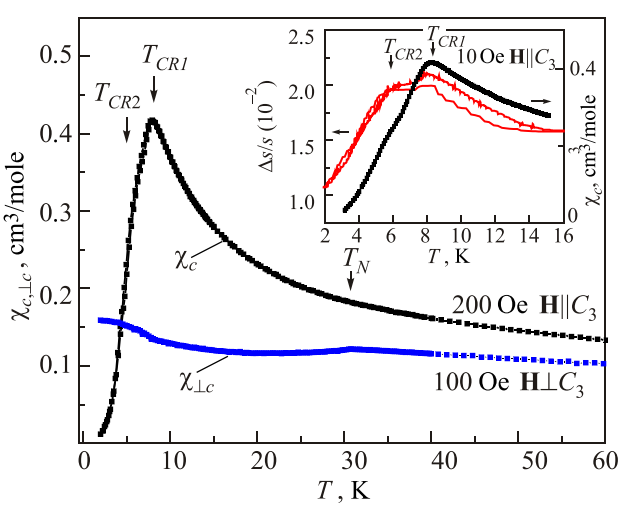


FIG. 4. Temperature dependences of the magnetic susceptibility of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ measured along the direction of the $C_3$ axis, $\chi_c$, and perpendicular to it in the basal plane of the crystal, $\chi_{\perp c}$. The inset shows the temperature dependences of the susceptibility $\chi_c$ and the velocity of the $C_{44}$ mode in the vicinity of the spin-reorientation phase transitions.

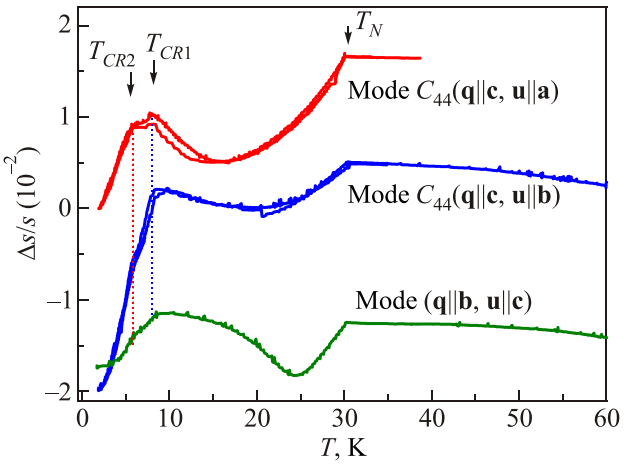


FIG. 5. Temperature dependences of the velocities of some transverse acoustic modes of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ with no external magnetic field.

all the transverse modes also experience rather strong damping (≤2%) which is observed down to the lowest temperature, 1.7 K, in our experiment (Fig. 5). In addition, at $T_{CR2} \approx 6$ K a rather sharp break (or "shoulder") can be seen in all these curves; it shows up most clearly for the $C_{44}(\mathbf{q}||\mathbf{c}, \mathbf{u}||\mathbf{a})$ mode. A comparison of the temperatures at which the acoustic and susceptibility anomalies are observed ($T_{CR1}$ and $T_{CR2}$) shows that they coincide fairly well (inset to Fig. 4). This correlation suggests that these features are of magnetic origin, which is confirmed by the temperature dependences of the acoustic and magnetic characteristics measured in external fields $\mathbf{H}||C_3$. As the field is raised, these features shift to lower temperatures, with the anomalies at $T_{CR1}$ and $T_{CR2}$ observed in fields up to 8 and 5 kOe, respectively.

We have, therefore, discovered that a sequence of two spontaneous magnetic phase transitions take place below the Neel temperature in this compound. This means that the realignment of the magnetic structure at that point can proceed in accordance with a scenario similar to that in $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$. Below $T_N$ the system is ordered with the formation of a collinear antiferromagnetic structure with an EP anisotropy. Then, for $T_{CR2} \le T \le T_{CR1}$ an intermediate "oblique" phase develops and the lowest temperature phase (for $T \le T_{CR2}$) is most likely an easy-axis state. It is possible, however, that at the lowest temperature in our experiments, 1.7 K, the transition to an EA magnetic configuration is still not complete since the temperature dependences of all the acoustic modes have still not "saturated" at this temperature (Fig. 5).

Finally, we discuss the proposition (from Ref. 15) that an incommensurate spiral structure may develop in $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$. This was based on the anomalous increase in the velocities of all the transverse modes below 20 K when there is no external field (Figs. 5 and 6(a)). We ascribed similar behavior in "pure" $NdFe_3(BO_3)_4$ to a phase transition from a collinear EP magnetic phase into an incommensurate spiral phase.[15] According to neutron data,[16] this phase transition took place at $T_{IC} \cong 13$ K in a single crystal sample of $NdFe_3(BO_3)_4$ (in a powder sample, at 19 K (Ref. 17)). In the compound studied here, the amount of neodymium is fairly high, so that at temperatures above the EP → EA spin

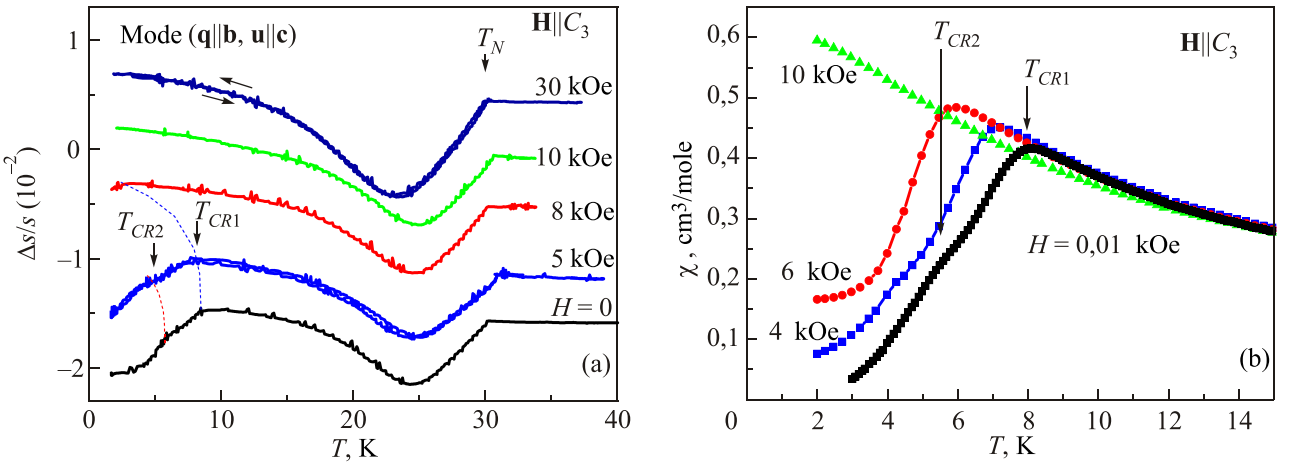


FIG. 6. Temperature dependences of the velocity of the transverse acoustic (**q**||**b**, **u**||**c**) mode (a) and of the magnetic susceptibility $\chi_c$ of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ in different external magnetic fields **H**||$C_3$.

reorientation point, a transition from a collinear EP magnetic structure to a spiral structure is entirely possible.

We have also found that the anomalous rise in the velocity of the $C_{44}$ (**q**||**c**, **u**||**a**) mode is accompanied by temperature hysteresis (Figs. 5 and 7(a)). As in $NdFe_3(BO_3)_4$, the size of the hysteresis loop depended on the shape and size of the sample, as well as on the local stresses inevitably introduced by gluing (Fig. 7(a), for the case of $H = 0$). This may be caused by the formation of an antiferromagnetic domain structure that is nonuniform over the volume of the sample. It is also possible that ferroelectric domains may be created, as in the multiferroic $MnWO_4$,[18] since the formation of a spiral magnetic structure and electric polarization are interrelated in neodymium ferroborate.[15]

It was surprising that applying a magnetic field **H**||$C_3$ enhanced the hysteresis observed in the $C_{44}$(**q**||**c**, **u**||**a**) mode (Fig. 7(a)). At the same time, in "pure" $NdFe_3(BO_3)_4$ we observed no effect of fields **H**||$C_3$ on the size of the hysteresis loop for a mode in the same configuration (Fig. 7(b)). Recall that the incommensurate structure in $NdFe_3(BO_3)_4$ is a long-period antiferromagnetic spiral for which the magnetic moments of the iron and neodymium lie in the basal plane of the crystal. Thus, the absence of a "reaction" to a field ($\leq$50 kOe) directed perpendicular to this plane may mean that its energy is not sufficient to overcome the energy of the easy-plane magnetic anisotropy and of the change in the magnetic configuration that develops in the crystal, as well as of the realignment of the resulting domain structure.

The specifics of the reaction of the elastic characteristics to an external magnetic field **H**||$C_3$ observed in $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$, as opposed to $NdFe_3(BO_3)_4$, are most likely caused by the presence of dysprosium ions, which have an easy-axis single-ion anisotropy. The existence of yet another pair of magnetic sublattices makes the mechanism of the interaction between the sublattices of the crystal more complicated and, thereby, leads to the appearance of new magnetic (possibly, ferroelectric) phases. This is also indicated by the fact that the hysteresis behavior, even without a field, occurs only for the $C_{44}$ (**q**||**c**, **u**||**a**) mode (Figs. 5 and 8), while we observed hysteresis in other modes in neodymium ferroborate.

Several possible scenarios for the realignment of the magnetic structure can be proposed. For example, for $T \leq 20$ K, the magnetic moments of neodymium and iron form a plane spiral. The magnetic moments of dysprosium (which can, in principle, be at an angle to the magnetic moments of iron and neodymium) do not change their position in the plane. For temperatures below $T_{CR1}$ and $T_{CR2}$, respectively, the dysprosium and then the neodymium moment emerge from the plane. A field $H \geq H_{CR2}$ along the

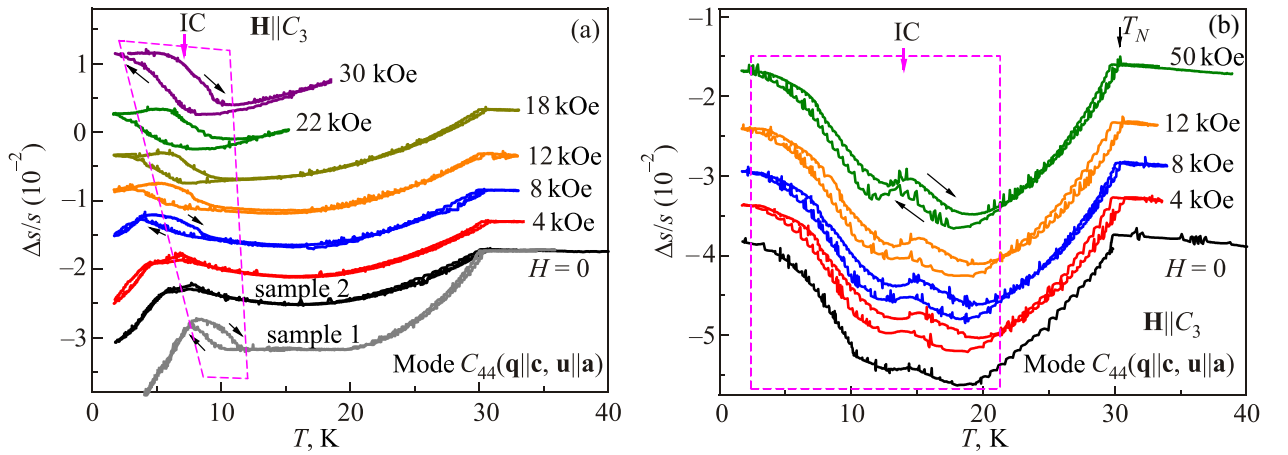


FIG. 7. Temperature dependences of the velocity of the acoustic mode $C_{44}$(**q**||**c**, **u**||**a**) measured for different external magnetic fields H||$C_3$ in crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ (a) and $NdFe_3(BO_3)_4$ (b). The dashed box indicates the region in which the proposed incommensurate spiral phase (IC) exists.

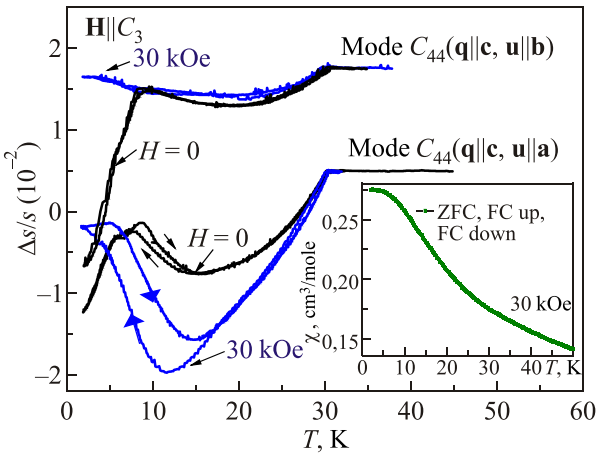


FIG. 8. Temperature dependences of the velocity of the $C_{44}$ acoustic mode excited by $(\mathbf{q}||\mathbf{c}, \mathbf{u}||\mathbf{a})$ and $(\mathbf{q}||\mathbf{c}, \mathbf{u}||\mathbf{b})$ waves, measured with zero magnetic field and in a magnetic field $H = 30$ kOe $(\mathbf{H}||C_3)$ in crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$. The inset shows the temperature dependence of the susceptibility $\chi_c$ in a field $H = 30$ kOe.

$C_3$ axis returns the magnetic moments of Nd to the plane (conserving the spiral configuration), while it turns the magnetic moments of Dy in the direction of the field.

It is also possible that the spiral in a magnetic field $\mathbf{H}||C_3$ may become conical out of the plane as, for example, in $Eu_{0.55}Y_{0.45}MnO_3$.[19] We note that the deformation of noncollinear magnetic structures by a magnetic field can also be accompanied by the appearance, disappearance, or changes in the magnitude and direction of the electric polarization, because, for example, of the development of magnetoelectric domains.

Another argument in favor of the appearance of a spiral structure in this crystal may be the fact that hysteresis shows up in fields $H \geq 8$ kOe applied to the basal plane of the crystal (Fig. 9). This may be related to a transition of the magnetic subsystem of the crystal from a spiral phase into a flop phase similar to that observed in $NdFe_3(BO_3)_4$.[15]

We note that measurements of the magnetic susceptibility in very low magnetic fields (10 Oe) and in a field $\mathbf{H}||C_3 = 30$ kOe with cooling and heating revealed no anomalies in the $\chi_c(t)$ temperature curves within the range where the proposed spiral phase exists (see the insets to Figs. 4 and 8). This is not surprising, since the transition

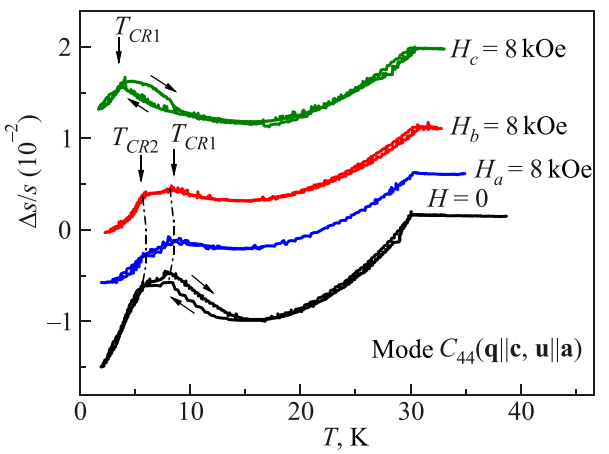


FIG. 9. Temperature dependences of the velocity of the $C_{44}(\mathbf{q}||\mathbf{c}, \mathbf{u}||\mathbf{a})$ acoustic mode measured with zero magnetic field and in magnetic fields $H = 8$ kOe $(\mathbf{H}||C_3)$, $(\mathbf{H}||C_2)$, and $(\mathbf{H}\perp C_3 \perp C_2)$ in crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$.

into the spiral phase also does not show up in the corresponding susceptibility for "pure" crystalline $NdFe_3(BO_3)_4$.[20] Here we point out, also, that in the magnetically ordered phase the degeneracy of the $C_{44}$ mode is eliminated (Fig. 8, $H = 0$). This means that in the magnetic group of the crystal, the third order axis of symmetry is lost and the magnetic structure that appears below $T_N$ cannot be an easy-axis state (with an easy-axis direction coinciding with the $C_3$ axis).

Based on these magnetic and acoustic experiments, we have constructed the low-temperature segment of the *H-T* phase diagram of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ for $\mathbf{H}||C_3$ (Fig. 10). This diagram, as in the case of $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$, is characterized by several phase transition curves and, therefore, by several magnetic phases. In principle, the phase diagrams for these compounds are similar because of the closeness of their chemical compositions. The differences lie in the coordinate boundaries of the magnetic phases, which are determined, of course, by the dysprosium ion concentrations. In addition, the large amount of neodymium compared to $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ extends the domain of existence of the easy-plane collinear phase and, apparently, makes a transition into a spiral structure possible.

The question of the existence of a transition to an incommensurate magnetic phase in $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$, as well as an exact determination of the magnetic configuration of each phase in such a crystal, can be answered uniquely by, for example, neutron scattering experiments similar to those of Ref. 16. In addition, we believe it is equally important to study the behavior of the magnetoelectric polarization of this compound, as well as to carry out magneto-optical studies similar to those of Ref. 18, which might aid in visualizing the domain structure that develops in the crystal.

In conclusion, we note that the comparative analysis of the magnetic and elastic characteristics of crystalline $Nd_{0.75}Dy_{0.25}Fe_3(BO_3)_4$ and $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ with varying temperature and in external magnetic fields $\mathbf{H}||C_3$ indicate, with a high degree of probability, that a spontaneous EP → EA transition occurs in the related compound $Nd_{0.85}Dy_{0.15}Fe_3(BO_3)_4$ as a result of two phase transitions via an intermediate magnetic phase.

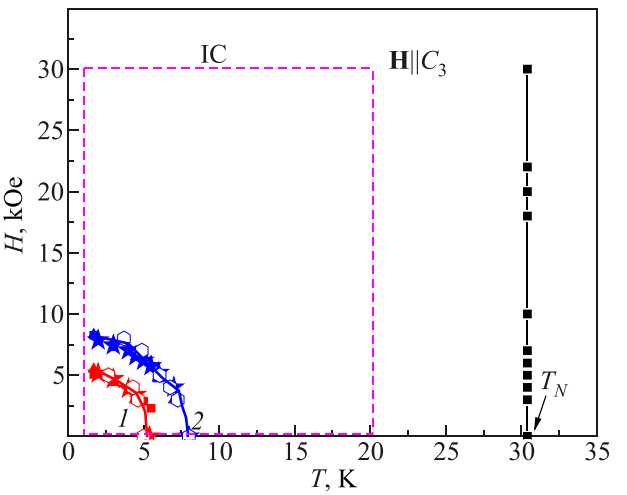


FIG. 10. The low-temperature segment of the *H-T* phase diagram of crystalline $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$, $(\mathbf{H}||C_3)$. The region in which the proposed incommensurate structure occurs is indicated by the dashed box. Curves *1* and *2* denote the boundaries of the magnetic phases.

To summarize, in this paper we have studied the behavior of the elastic and magnetic characteristics of single crystal $Nd_{0.9}Dy_{0.1}Fe_3(BO_3)_4$ at low temperatures and in external magnetic fields $\mathbf{H}||C_3$. It has been shown that a magnetic field directed along the trigonal symmetry axis induces two successive phase transitions for temperatures $T \leq 6\,\mathrm{K}$ $(T_{CR2})$, and only one phase transition in the temperature range $6\,\mathrm{K}(T_{CR2}) \leq T \leq 8\,\mathrm{K}$ $(T_{CR1})$. A transition of the magnetic subsystem into a magnetically ordered state shows up in the temperature dependences of the acoustic mode velocities and of the magnetic susceptibility. It has been found that a spontaneous EP → EA reorientation takes place in this crystal as a result of two spin-reorientation phase transitions. For the first time an *H-T* phase diagram of this compound for $(\mathbf{H}||C_3)$ has been constructed. It has several phase transition curves and, therefore, several magnetic phases, which may be caused by interactions among various magnetic sublattices of the crystal. The possibility of a spontaneous transition into an incommensurate (spiral) magnetic phase in this compound has been discussed.

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