MAGNETOOPTICAL STUDY OF SPIN-ORIENTATION PHASE TRANSITION IN $NdFe_3(BO_3)_4$ SINGLE CRYSTAL

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The magnetic field dependences of birefringence in a $NdFe_3(BO_3)_4$ single crystal have been measured in the case where the direction of light propagation coincides with the trigonal crystal axis C_3 ($\mathbf{k}\|C_3$), and the external magnetic field is oriented along the second-order axis C_2 ($\mathbf{H}\|C_2$). In the temperature range, in which an incommensurate phase exists with the formation of a long-period antiferromagnetic helix, the strongly pronounced jumps in the field dependence of birefringence are revealed and identified as a first-order spin-orientation phase transition. The phase transition was accompanied by a hysteresis in the field dependences of birefringence. The H–T phase diagram for a $NdFe_3(BO_3)_4$ single crystal has been plotted in the case where the magnetic field is oriented along the crystal axis C_2 ($\mathbf{H}\|C_2$).

1. Introduction

A $NdFe_3(BO_3)_4$ single crystal with the giant magnetoelectric effect [1] is one of the most intensively studied ferroborates. Unlike the majority of similar compounds, the structural phase transition in this crystal was not observed [2]. The crystal symmetry R32 stays down to helium temperatures. At a temperature of about 30 K, there appear the simultaneous magnetic and electric orderings in the crystal [3]. The $NdFe_3(BO_3)_4$ single crystal is antiferromagnetically ordered, and the magnetic moments of Fe and Nd atoms are parallel to each other in the plane perpendicular to the trigonal axis C_3, being oriented oppositely to the magnetic moments in the neighbor plane [4]. As a result, there appear three types of antiferromagnetic domains. The antiferromagnetic vector $\mathbf{l}$ in each of them is oriented along the corresponding axis of the second order. A further reduction of the crystal temperature down to 13.5 K results in a first-order spin-orientation phase transition from the commensurate phase into the incommensurate one, the latter structurally being a long-period antiferromagnetic helix [4].

It is known that a spin-orientation phase transition takes place in $NdFe_3(BO_3)_4$ crystals under the action of a magnetic field applied along the second-order axis [1, 5]. The hysteresis testifying to a first-order spin-orientation phase transition can be clearly observed in the field dependences of the magnetization. However, this phase transition is strongly expended over the magnetic field. This effect may probably be a result of a polydomain structure of specimens, as well as owing to elastic stresses and defects in them. Therefore, it is rather difficult to determine the magnitude of phase transition field. In such cases, the magnetooptical method often turns out useful. It allows the small non-strained sections of single-crystalline plates a few tens of microns in thickness to be selected and studied. In works [6, 7], this method was applied for the first time to research the spin-orientation phase transition induced by the magnetic field in a $NdFe_3(BO_3)_4$ crystal. The results obtained were confirmed by the data of resonance [8] and acoustic [9] studies of the crystal concerned.

In this work, we report the results of our magnetooptical researches of the spin-orientation phase transition in $NdFe_3(BO_3)_4$ ferroborate. The work is based on the analysis of the data obtained from the field dependences of magnetic birefringence in this single crystal.

2. Experimental Technique

We studied a triangular single-crystalline $NdFe_3(BO_3)_4$ plate, each side of which was equal to 1.5 mm, and the thickness was 70 μm. The developed surface of the crystal was perpendicular to the trigonal C_3 axis. In order to remove elastic stresses which appeared after the mechanical treatment, the specimen was annealed at a temperature of 700 °C for 10 h. The crystal to study was inserted into a solenoid located in an optical helium cryostat. The magnetic field was directed along the C_2

axis. The specimen temperature was measured with a carbon thermometer.

The light birefringence Δn is related to the phase difference δ, which arises between two characteristic linearly polarized waves at the output from a crystal of thickness t, as follows:

$$\Delta n = \delta\lambda/2\pi t, \qquad (1)$$

where λ is the light wavelength. In measurements of δ, we used a quarter-wave plate. The diagram of the experimental setup for δ-measurements is depicted in Fig. 1. Light produced by incandescent lamp (*1*) passed through thermal filter (*2*) and afterward through interference filter (*3*) with the transmittance maximum at the wavelength $\lambda = 633$ nm and a transmission band of 11 nm. Further, light passed through polarizer (*5*) and was focused on specimen (*6*) making use of lens (*4*). The aperture diameter in diaphragm (*9*) located in the image plane of lens (*8*) determined the dimension of the specimen area to study. The diameter of the examined region was about 100 μm. Elliptically polarized light, which came out of the crystal, is transformed by $\lambda/4$ plate (*10*) in linearly polarized one. The polarization plane was rotated at that by the angle $\delta/2$ with respect to the polarization plane of light incident onto the crystal. To measure the angle $\delta/2$, the modulation technique was applied with the modulation of the light polarization plane (modulator (*11*)) and the synchronous detection (amplifier (*14*)). The output signal of the amplifier was applied to an input of personal computer (*15*).

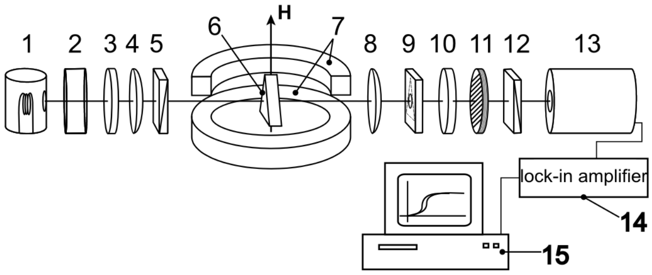

Fig. 1. Diagram of experimental setup: (*1*) incandescent lamp, (*2*) thermal filter, (*3*) interference filter, (*4*) and (*8*) lenses, (*5*) polarizer, (*6*) specimen, (7) superconducting solenoid, (*9*) diaphragm, (*10*) $\lambda/4$-plate, (*11*) modulator, (*12*) analyzer, (*13*) photoelectronic multiplier, (*14*) amplifier, (*15*) personal computer

3. Experimental Results and Their Discussion

In the paramagnetic range, the optical indicatrix of $NdFe_3(BO_3)_4$ crystal, characterized by the point crystal group 32, is an ellipsoid of rotation around the C_3-axis. The occurrence of a magnetic ordering or the application of a magnetic field can reduce the optical class of the crystal to the biaxial one. Let us expand the symmetric part of the dielectric permittivity ${}^S\varepsilon_{ij}$ in a series in the magnetic field H and confine the expansion to the terms quadratic in H:

$${}^S\varepsilon_{ij} = {}^S\varepsilon_{ij}^0 + \Delta^S\varepsilon_{ij} + q_{ijl}H_l + \beta_{ijlk}H_lH_k. \qquad (2)$$

Here, ${}^S\varepsilon_{ij}$ is the symmetric part of the tensor of dielectric permittivity of the crystal in the paramagnetic range, and $\Delta^S\varepsilon_{ij}$ is a variation of the symmetric part of the dielectric permittivity tensor associated with a magnetic and electric ordering. The tensor q_{ijl} describes the birefringence, which is proportional to the magnetic field strength and changes its sign, if the direction of the field is opposite. The tensor q_{ijl} can possess nonzero components only in magnetically ordered crystals [10]. The tensor β_{ijlk} defines the birefringence proportional to H_lH_k. This tensor is symmetric with respect to the (i, j) and (l, k) index pairs and is determined by the point crystal group 32. Thus, the contribution to the birefringence, which is quadratic in H, does not depend on the magnetic symmetry of the crystal, being determined only by the crystal symmetry. In the coordinate system with $z \parallel C_3$ and $x \parallel C_2$, the matrix of β_{ijlk}-coefficients looks like

$$\begin{bmatrix} \beta_{11} & \beta_{12} & \beta_{13} & \beta_{14} & 0 & 0 \\ \beta_{12} & \beta_{11} & \beta_{13} & -\beta_{14} & 0 & 0 \\ \beta_{31} & \beta_{31} & \beta_{33} & 0 & 0 & 0 \\ \beta_{41} & -\beta_{41} & 0 & \beta_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & \beta_{44} & 2\beta_{41} \\ 0 & 0 & 0 & 0 & \beta_{14} & \beta_{11}-\beta_{12} \end{bmatrix}. \qquad (3)$$

To write down this tensor, we used the standard rules of index notation. In the case $\mathbf{H} \parallel x$, the additives $\beta_{xxxx}H_x^2$ and $\beta_{xxyy}H_x^2$ to the tensor ε_{ij} appear in the magnetic field, in accordance with matrix (3). As a result, the cross-section of the optical indicatrix of the crystal in a plane perpendicular to the C_3-axis is an ellipse, and the crystal becomes optically biaxial. One of the principal axes of this ellipse is parallel to the x-axis.

Our researches revealed no spontaneous magnetic birefringence defined by the tensor $\Delta^S\varepsilon_{ij}$ and associated with a transition into the antiferromagnetic state. A spontaneous magnetic birefringence was also not found, when the crystal transformed into the incommensurate magnetic phase. The linear magnetic birefringence was observed only when a magnetic field was applied.

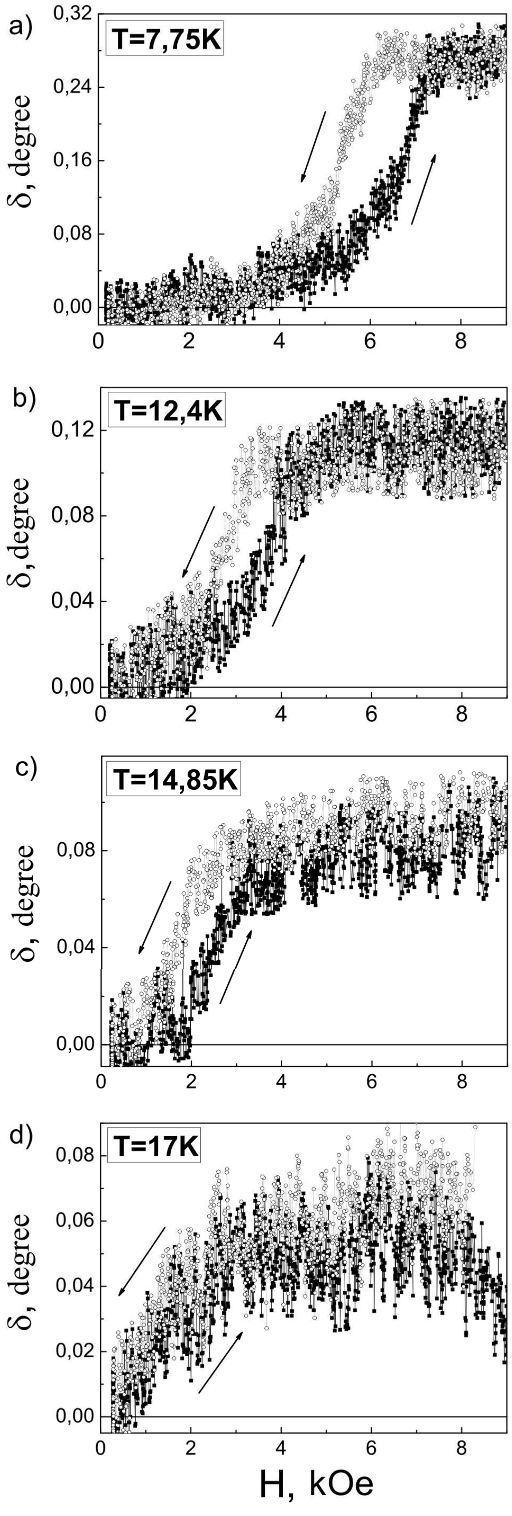

Fig. 2. Field dependences of the light birefringence in a $NdFe_3(BO_3)_4$ single crystal measured in the geometry $\mathbf{k} \parallel C_3$ and $\mathbf{H} \parallel C_2$ at various temperatures $T = 7.75$ (*a*), 12.4 (*b*), 14.85 (*c*), and 17 K (*d*)

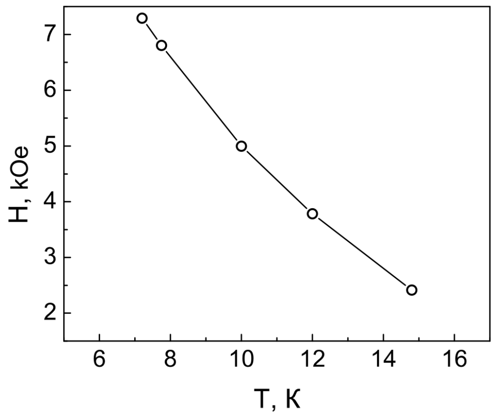

Fig. 3. Magnetic phase diagram H–T of a $NdFe_3(BO_3)_4$ single crystal for the magnetic field orientation $\mathbf{H} \parallel C_2$

In Fig. 2, the experimental field dependences of the linear magnetic birefringence of light, $\delta(H)$, measured at various temperatures from 7.5 to 17 K in the geometry $\mathbf{H} \parallel x$ are shown. If the direction of the magnetic field is inverted, the dependence $\delta(H)$ does not change its sign. The birefringence magnitude is identical for magnetic fields H equal by the value, but opposite by the direction. Therefore, the observed magnetic birefringence quadratically depends on the magnetic field, and the linear magnetic birefringence described by the tensor q_{ijl} makes no contribution to those dependences. In contrast to the field dependences of the magnetization $M(H)$ [1], our experimental curves demonstrate a pronounced jump $\delta(H)$, which corresponds to the first-order spin-orientation phase transition. Similarly to the dependences $M(H)$ [1], our experimental curves have a hysteresis in the phase transition interval at low temperatures. In fields that exceed the transition fields, the birefringence weakly depends on H. The magnitude of $\delta(H)$ and the hysteresis width decrease with the temperature growth. At a temperature of about 17 K, the first-order phase transition induced by the magnetic field is not observed any more.

The obtained experimental data allowed us to determine the phase transition fields at various temperatures. The phase transition field was determined as follows. First, at a fixed temperature, we determined the field values corresponding to the middle points in the intervals, where the dependence $\delta(H)$ changed drastically, at both the increasing and decreasing external magnetic fields. The phase transition field was calculated as an average value for those two quantities. The obtained data were used to plot the phase diagram, which is exhibited in Fig. 3.

It is of interest that the first-order phase transition induced by a magnetic field was observed only in the incommensurate phase, in which the long-period antiferromagnetic helix is formed. At temperatures above 15 K, one could expect that a spin-flop phase transition would have taken place in the antiferromagnetic commensurate phase in the case $\mathbf{H} \parallel C_2$. However, it was not observed in our experiment, although this phase transition was observed in another easy-plane antiferromagnetic ferroborate, $GdFe_3(BO_3)_4$ [11, 12]. The absence of spin-flop phase transition in our case can be associated with a deviation of the magnetic field orientation from the axis of easy magnetization by an angle that exceeds a certain critical value ψ_c. The spin-flop transition is known to disappear in this case, and the rotation of the antiferromagnetism vector occurs smoothly [13–15]. Neglecting the influence of demagnetizing fields, it is possible to assert [13–15] that the ψ_c-value is of the order of the ratio H_a/H_e between the effective fields of magnetic anisotropy and exchange antiferromagnetic interaction in the crystal. Ferroborate $NdFe_3(BO_3)_4$ is known to have the high effective exchange field $H_e = 580$ kOe [5] and the effective field of magnetic anisotropy H_a, which is very insignificant in the basic plane (it amounts to about 60 Oe according to resonance researches [8] and to 12 Oe according to magnetic ones [5]). The estimation of ψ_c gives a magnitude of $0.3'$. Such a small value of the critical angle may probably result in that we did not observe the first-order phase transition in the antiferromagnetic phase of $NdFe_3(BO_3)_4$ crystal. On the other hand, the reason for why the spin-flop transition is distinctly observed in ferroborate $GdFe_3(BO_3)_4$, for which the ratio H_a/H_e is rather close to ours, may be the influence of demagnetizing fields in bulk specimens, which favor the increase of ψ_c [13–15]. We studied an $NdFe_3(BO_3)_4$ plate about 70 μm in thickness, and the magnetic field was oriented in the plate plane. Therefore, the arising demagnetizing fields were much lower in our case than the demagnetizing fields appearing in a bulk $GdFe_3(BO_3)_4$ specimen [11, 12]. Hence, they do not affect the critical angle in our experiment.

4. Conclusions

To summarize, while studying the field dependences of the magnetic birefringence in a $NdFe_3(BO_3)_4$ single crystal, we found that the magnetic field $\mathbf{H} \parallel C_2$ induces a first-order spin-orientation phase transition only in the temperature range, where the incommensurate phase exists and the antiferromagnetic helix is realized. In the temperature range, where the commensurate antiferromagnetic phase exists, the first-order phase transition in the magnetic field $\mathbf{H} \| C_2$ is not observed. We associate this fact with the smallness of the critical angle ψ_c for the magnetic field deviation from the axis of easy magnetization. The field dependences of the magnetic birefringence were used to determine the fields of the first-order spin-orientation phase transition at various temperatures. The corresponding magnetic phase diagram H–T was plotted.

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МАГНІТООПТИЧНЕ ДОСЛІДЖЕННЯ ІНДУКОВАНОГО МАГНІТНИМ ПОЛЕМ СПІН-ОРІЄНТАЦІЙНОГО ФАЗОВОГО ПЕРЕХОДУ У МОНОКРИСТАЛІ $NdFe_3(BO_3)_4$

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Р е з ю м е

Проведено дослідження польових залежностей магнітного двозаломлення світла у монокристалі $NdFe_3(BO_3)_4$ у випадку, коли напрямок поширення світла збігається з тригональною віссю кристала C_3 ($\mathbf{k}\|C_3$), а зовнішнє магнітне поле орієнтоване вздовж осі другого порядку C_2 ($\mathbf{H}\|C_2$). У температурному проміжку існування неспіврозмірної фази з утворенням довгоперіодичної антиферомагнітної спіралі виявлено чітко виражені скачки на польових залежностях двозаломлення, які ідентифіковані як спін-орієнтаційний фазовий перехід першого роду. Фазовий перехід супроводжувався гістерезісом на польових залежностях двозаломлення. Побудовано магнітну фазову H–T діаграму $NdFe_3(BO_3)_4$ для орієнтації магнітного поля вздовж осі другого порядку кристала $\mathbf{H}\|C_2$.