

Magnetic field-induced rotation of the plane of polarization of light in the antiferromagnetic ferroborate $\text{TbFe}_3(\text{BO}_3)_4$

Cite as: *Low Temp. Phys.* **37**, 476 (2011); <https://doi.org/10.1063/1.3614411>
 Published Online: 10 August 2011

V. A. Bedarev, M. I. Pashchenko, A. N. Bludov, S. L. Gnatchenko, L. N. Bezmaternykh, and V. L. Temerov



View Online



Export Citation

ARTICLES YOU MAY BE INTERESTED IN

[Spectroscopic and magneto-optical investigations of spin-reorientation phase transition in \$\text{TbFe}_3\(\text{BO}_3\)_4\$](#)

Low Temperature Physics **37**, 693 (2011); <https://doi.org/10.1063/1.3660219>

[Finite-size scaling relations for a four-dimensional Ising model on Creutz cellular automata](#)

Low Temperature Physics **37**, 470 (2011); <https://doi.org/10.1063/1.3610180>

[Local and quasilocal energy levels of electrons on nanotube surfaces and in rings in magnetic fields](#)

Low Temperature Physics **37**, 506 (2011); <https://doi.org/10.1063/1.3622626>



MONTANA INSTRUMENTS

QUANTUM COMPUTING SPINTRONICS : MOKE DIAMOND NV CENTERS

CLICK HERE
CRYOGENIC
APPLICATION
NOTES

montanainstruments.com/Applications/Application-Notes/

COLD SCIENCE MADE SIMPLE

Magnetic field-induced rotation of the plane of polarization of light in the antiferromagnetic ferroborate $\text{TbFe}_3(\text{BO}_3)_4$

V. A. Bedarev,^{a)} M. I. Pashchenko, A. N. Bludov, and S. L. Gnatchenko

B. I. Verkin Institute of Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, pr. Lenina 47, Kharkov 61103, Ukraine

L. N. Bezmaternykh and V. L. Temerov

L. V. Kirenskii Institute of Physics, Siberian Branch of the Russian Academy of Sciences, Krasnoyarsk 660036, Russia

(Submitted December 14, 2010)

Fiz. Nizk. Temp. **37**, 598–602 (June 2011)

The magnetic field dependences of the rotation of the plane of polarization of light and the magnetization in single crystals of the ferroborate $\text{TbFe}_3(\text{BO}_3)_4$ are studied. The main contribution to the magnetic field-induced rotation of the plane of polarization is found to be from the magnetic subsystem of the terbium ions. © 2011 American Institute of Physics. [doi: [10.1063/1.3614411](https://doi.org/10.1063/1.3614411)]

The rare earth ferroborates $\text{RFe}_3(\text{BO}_3)_4$ are of interest primarily because these compounds have multiferroic properties.^{1,2} In addition, the interaction of the rare-earth and iron subsystems in the ferroborates lead to the appearance of a great variety of magnetic structures, including incommensurate magnetic structures.³ The rare-earth ferroborates have a trigonal structure belonging to the non-centrally symmetric spatial group $R32$. A first order structural phase transition is observed in some of the ferroborates. For example, this sort of phase transition occurs at 156 K in $\text{GdFe}_3(\text{BO}_3)_4$.⁴ X-ray studies have shown that the crystal symmetry decreases from $R32$ to $P3_121$ during this phase transition in $\text{GdFe}_3(\text{BO}_3)_4$, while the point crystal group is unchanged: 32 . It is assumed that this kind of symmetry change also occurs in single crystal $\text{TbFe}_3(\text{BO}_3)_4$ during a first order phase transition at 192 K.

The magnetic and magneto-electronic properties of the ferroborates depend strongly on the rare-earth element. The magnetic structure of these compounds is determined by the anisotropy of the rare-earth subsystem, since the iron subsystem is weakly anisotropic. Depending on the rare-earth element, an easy-axis or an easy-plane antiferromagnetic structure is realized. An easy-axis antiferromagnetic structure occurs when $\text{R} = \text{Tb}$.⁵ The iron ion subsystem of this crystal is antiferromagnetically ordered at $T_N = 40$ K, and the magnetic moments are then directed along the trigonal c axis. Because of the (f - d)-interaction the terbium ions are in a magnetized state. The magnetic moments of the terbium ion subsystem are also directed along the c axis. If an external magnetic field is directed along the c axis, then a first order spin-orientational phase transition is observed in crystalline $\text{TbFe}_3(\text{BO}_3)_4$ at temperatures below 40 K.⁵

In a magnetic field the magnetization $M(\mathbf{H})$ of the terbium ferroborate is made up of the projections of the magnetic moments of the terbium, $M_{\text{Tb}}(H)$, and iron, $M_{\text{Fe}}(H)$, ion sublattices in the direction of the magnetic field. If linearly polarized light propagating in the direction of the magnetic field is incident on this crystal, then the plane of polarization

of the light emerging from the crystal will be rotated relative to that of the incident light by an angle

$$\Phi(H) = (A \cdot M_{\text{Fe}} + B \cdot M_{\text{Tb}})t. \quad (1)$$

Here A and B are magneto-optical constants and t is the thickness of the crystal.

This paper is a study of the field dependences of the rotation of the plane of polarization of light. The contributions to this rotation from the terbium and iron ion subsystems in single crystal $\text{TbFe}_3(\text{BO}_3)_4$ are determined.

The single crystal $\text{TbFe}_3(\text{BO}_3)_4$ that was studied was grown from a solution in the alloy.⁵ A wafer perpendicular to the c axis was cut from the single crystal for the magneto-optical studies. The sample was mechanically polished and then annealed at 800 °C for 10 h to eliminate elastic stresses. The prepared sample had a thickness of 110 μm and was quite transparent in the visible.

The sample was fastened to a cold lead and placed under vacuum in an optical helium cryostat. The sample temperature was determined by a thermistor mounted on the cold lead near the sample. The magnetic field created by a superconducting solenoid was parallel to the trigonal c axis of the sample and coincided with the direction of propagation of the light beam used in the measurements. The field dependences of the rotation (of the plane of polarization of the light) were measured by a modulation technique with modulation of the plane of polarization and synchronous detection. The light source was an incandescent lamp with a $\lambda = 633$ nm interference filter placed after it.

The field dependences of the magnetization were measured by a MPMS-XL SQUID magnetometer. In this case the magnetic field was also directed along the trigonal c axis of the crystal.

Figure 1 shows the field dependences of the rotation of the plane of polarization of the light, $\Phi(H)$, measured at temperatures of 8, 10, 15, 20, and 35 K over fields of 0 to 35 kOe. The figure shows that, as the temperature increases, the rotation becomes larger. In addition, at low temperatures

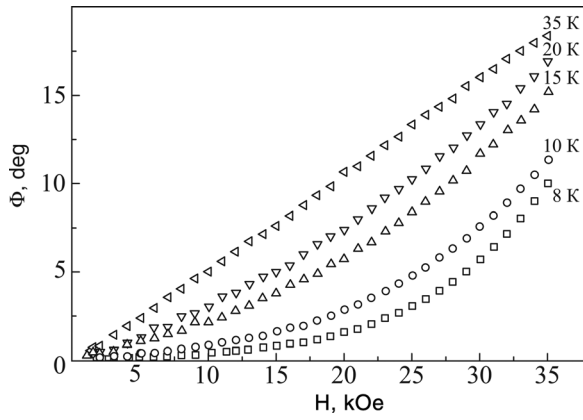


FIG. 1. The angle of rotation of the plane of polarization of light, $\Phi(H)$, as a function of magnetic field in monocrystalline $\text{TbFe}_3(\text{BO}_3)_4$.

$\Phi(H)$ depends nonlinearly on the magnetic field. The deviation from linearity in $\Phi(H)$ gradually becomes smaller with increasing temperature, and at temperatures close to T_N the rotation of the plane of polarization depends almost linearly on the magnetic field.

In order to compare the field dependence of the rotation $\Phi(H)$ and that of the magnetization $M(H)$, these dependences were measured at the same temperatures and over the same range of magnetic fields. The field dependences of the magnetization shown in Fig. 2 are similar to those of the rotation. At low temperatures the $M(H)$ curves vary nonlinearly with the magnetic field and are essentially linear for temperatures approaching T_N , as in the case of $\Phi(H)$.

The nonlinear dependences of the magnetization and rotation on magnetic field indicate that, besides a contribution that is linear in the magnetic field \mathbf{H} , there is a contribution to these dependences from higher powers of \mathbf{H} . We now consider what sort of terms, other than linear in the magnetic field, can exist in the field dependences of the magnetization and of the rotation in terbium ferroborate. To do this, we expand the magnetization $M(\mathbf{H})$ and the antisymmetric part of the dielectric constant $\epsilon_{ij}^a(\mathbf{H})$ in \mathbf{H} , i.e.,

$$M = \chi_{ij}H_j + \chi_{ijk}H_jH_k + \chi_{ijkl}H_jH_kH_l + \dots, \quad (2)$$

$$\epsilon_{ij}^a = \tau_{ij\alpha}H_\alpha + \tau_{ija\beta}H_\alpha H_\beta + \tau_{ija\beta\gamma}H_\alpha H_\beta H_\gamma + \dots \quad (3)$$

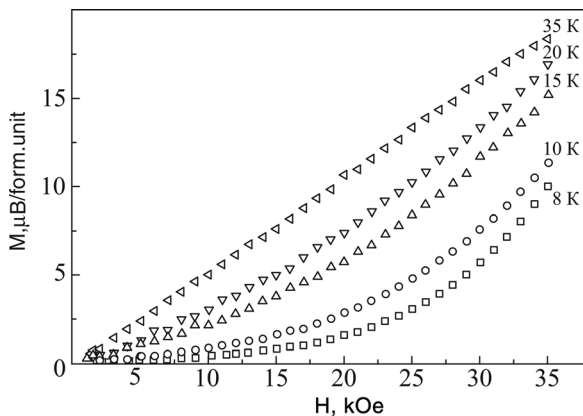


FIG. 2. The magnetization $M(H)$ of the ferroborate $\text{TbFe}_3(\text{BO}_3)_4$ as a function of magnetic field: data points (experiment), smooth curves (calculated field dependences of the magnetization of the terbium subsystem).

Here the axial third rank c -tensor χ_{ijk} , which is symmetric in all its indices, and the polar fourth rank c -tensor $\tau_{ija\beta}$, which is antisymmetric with respect to the first and symmetric with respect to the second pair of indices, describe the contributions to the magnetization and rotation, respectively, which are quadratic in the magnetic field. Both of these contributions are allowed within the same point magnetic groups.⁶ The linear and cubic field contributions to the magnetization and rotation are allowed for all point magnetic groups.

Therefore, we have to establish whether terbium ferroborate belongs to a point magnetic group within which contributions quadratic in the magnetic field are allowed for the magnetization and for the rotation of the plane of polarization of light. After magnetic ordering terbium ferroborate can undergo a transition from the 32 point crystallographic group to the 32, 32', 3 point magnetic groups.⁷ As opposed to the 32 point magnetic group, in the 32' and 3 point magnetic groups a weakly ferromagnetic moment and a magnetization that is quadratic in the field are allowed. Given this, we now try to understand into which particular magnetic group terbium ferroborate may transform after antiferromagnetic ordering. It is known that the magnetic cell is doubled with respect to the crystallographic cell in the ferroborates $\text{GdFe}_3(\text{BO}_3)_4$ and $\text{NdFe}_3(\text{BO}_3)_4$.^{1,2} This kind of doubling appears to exist in $\text{TbFe}_3(\text{BO}_3)_4$, since a first order spin-orientational phase transition is observed when a magnetic field is applied along the c axis. This kind of phase transition can only be observed when the magnetic cell is doubled with respect to the crystallographic cell. This is because, if the magnetic and crystallographic cells coincide, then there will be 9 Fe^{3+} ions in the cell and an antiferromagnetic structure, for which a first order spin-orientational phase transition in a magnetic field could be observed, cannot develop in this kind of cell. Since this sort of phase transition is observed experimentally, the magnetic cell is doubled with respect to the crystallographic cell in $\text{TbFe}_3(\text{BO}_3)_4$. From the standpoint of symmetry, this means that the magnetic symmetry of the crystal contains a symmetry element of an antitranslation type, which prevents the existence of a weakly antiferromagnetic moment. Of the possible magnetic groups for terbium ferroborate, a weakly ferromagnetic moment is forbidden only in magnetic group

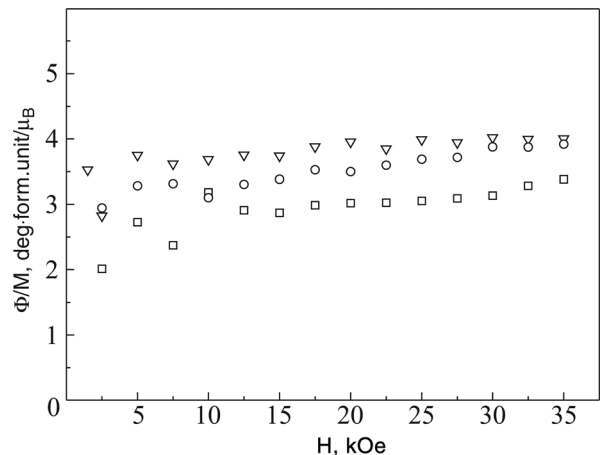


FIG. 3. Φ/M as a function of magnetic field for T (K) = 8 (\square), 15 (\circ), and 35 (∇).

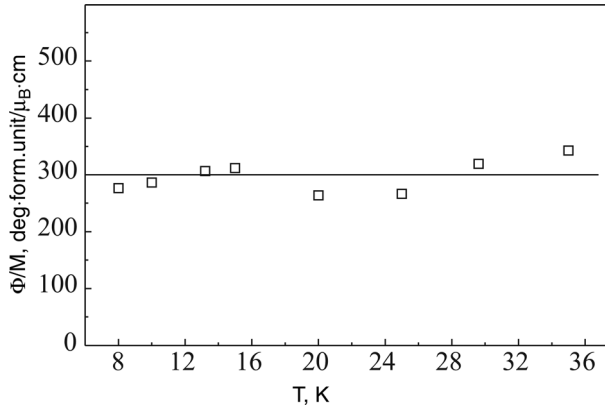


FIG. 4. Φ/M as a function of temperature normalized to the sample thickness.

32. Thus, the contributions to the magnetization and rotation in crystalline $\text{TbFe}_3(\text{BO}_3)_4$ which are nonlinear in the magnetic field are associated with terms that are cubic in the magnetic field, since the quadratic terms are forbidden in magnetic group 32.

The terbium and iron ion subsystems can both contribute to the rotation of the plane of polarization of light in terbium ferrobortate. In order to determine whether both magnetic subsystems or only one of them contributes to the rotation, we plot the field dependence of Φ/M . As an example, these curves for temperatures of 8, 15, and 35 K are shown in Fig. 3. It is clear that Φ/M is almost independent of the magnetic field. Thus, the ratio Φ/M is constant at a given temperature. Using the field dependences of the rotation and of the magnetization, we found the value of Φ/M for different temperatures, so it was possible to plot Φ/M as a function of temperature, normalized to a sample thickness of $t = 110 \mu\text{m}$, in Fig. 4. It can be seen that over temperatures of 8 to 35 K, Φ/M varies little and has an average value of about $300 \text{ deg} \cdot \text{form.unit}/\mu_B \cdot \text{cm}$. The fact that the ratio Φ/M is constant suggests that the contribution to the rotation originates from only one of the magnetic subsystems of terbium ferrobortate—the iron ion subsystem or the terbium ion subsystem.

In order to determine which of these subsystems predominates in the rotation, we now estimate the contributions of the two subsystems to the magnetization.

As noted above, the iron ion subsystem acquires an antiferromagnetic ordering at temperatures below 40 K, while the magnetic moments of the Fe^{3+} ions are directed along the trigonal c axis by magnetic anisotropy forces created by the terbium ion subsystem. If we limit ourselves to a two-lattice model, then the magnetization of the iron ion subsystem per formula unit in a magnetic field $\mathbf{H} \parallel c$ can be estimated using the well known formula⁸

$$M_{\text{Fe}}(H) = 3g_{\text{Fe}}\mu_B S(B_S(y^+) + B_S(y^-)), \quad (4)$$

where

$$y^\pm = \frac{|H_{\text{Fe-Fe}}^\pm + H| S g_{\text{Fe}} \mu_B}{kT}. \quad (5)$$

TABLE I. Magnetizations of the iron and terbium ion subsystems per formula unit at different temperatures.

T , K	$\mu_B/\text{form.unit}$	kOe	$\mu_B/\text{form.unit}$
8	$1.347 \cdot 10^{-5}$	39.3	3.067
10	$1.127 \cdot 10^{-4}$	38.4	3.476
15	$1.737 \cdot 10^{-3}$	38	3.774
20	$6.361 \cdot 10^{-3}$	36.1	4.024
35	$3.1 \cdot 10^{-2}$	21	4.206

Here $g_{\text{Fe}} \approx 2$ is the g -factor of the Fe^{3+} ion, μ_B is the Bohr magneton, $S = 5/2$ is the spin of the Fe^{3+} and $H_{\text{Fe-Fe}} = 705 \text{ kOe}$ is the exchange field for the Fe-Fe antiferromagnetic interaction.⁵ Equations (4) and (5) can be used to calculate the magnetization of the iron ion subsystem per formula unit at temperatures $T = 8, 10, 15, 20,$ and 35 K and for a magnetic field $H = 35 \text{ kOe}$. These results are listed in Table I.

For comparison with the magnetization of the iron ion subsystem, we calculate the magnetization of the terbium ion subsystem for the same temperatures $T = 8, 10, 15, 20,$ and 35 K and magnetic field $H = 35 \text{ kOe}$. It has already been pointed out above that, as opposed to the iron ion subsystem, the Tb-Tb exchange interaction is small, while the terbium ion subsystem is under the influence of the exchange field $\mathbf{H}_{\text{Tb-Fe}}$. For opposite directions of the magnetic moments of Tb, the fields $\mathbf{H}_{\text{Tb-Fe}}$ are also directed oppositely. Optical studies⁹⁻¹¹ have shown that the ground state of the Tb ion is a quasidoublet. This quasidoublet is split by the exchange field $\mathbf{H}_{\text{Tb-Fe}}$. The maximum value Δ of this splitting at low temperatures is about 32 cm^{-1} , and the splitting decreases with rising temperature, going to zero at T_N . The minimum difference in the energies of the ground and excited states is about 190 cm^{-1} in the Tb ion.⁹ If the ground state of Tb is a quasidoublet that is well separated from the excited states, then the magnetization of the terbium ion subsystem in a magnetic field \mathbf{H} will be given by²

$$M_{\text{Tb}}(H) = \frac{g_{\text{eff}}\mu_B}{2} \cdot \left[\text{th} \left(\frac{g_{\text{eff}}\mu_B(H + H_{\text{Tb-Fe}})}{2kT} \right) + \text{th} \left(\frac{g_{\text{eff}}\mu_B(H - H_{\text{Tb-Fe}})}{2kT} \right) \right], \quad (6)$$

where $g_{\text{eff}} = 17.8$ is the effective g -factor for the quasidoublet ground state of the Tb^{3+} ion¹⁰ and μ_B is the Bohr magneton. In order to estimate the magnetization of the terbium ion subsystem at temperatures of 8, 10, 15, 20, and 35 K, it was necessary initially to determine $H_{\text{Tb-Fe}}$ for these temperatures. It is known that the exchange field $H_{\text{Tb-Fe}} = \Delta/g_{\text{eff}}\mu_B$. Thus, using the temperature dependence of the splitting Δ of the quasidoublet ground state it is easy to calculate $H_{\text{Tb-Fe}}$ (see Table I). Now, knowing $H_{\text{Tb-Fe}}$ it is possible to calculate the magnetization of the Terbium ion subsystem per formula unit in a magnetic field $H = 35 \text{ kOe}$ using Eq. (6). These values of the magnetization are also listed in Table I. These estimates show that at any temperature, the magnetization of the terbium ion subsystem exceeds that of the iron ion subsystem by at least two

orders of magnitude. We may, therefore, conclude that the contribution of the iron ion subsystem to the total magnetization of terbium ferroborate is very small and that the contribution from the terbium ion subsystem is dominant. To confirm this, the experimental field dependences of the magnetization were compared with the calculated dependences $M(H)_{\text{Tb}}$ for the terbium ion subsystem neglecting the magnetization of the iron ions. Figure 2 shows that the experimental and theoretical curves agree well for 8, 10, and 15 K. At 20 and 35 K, the calculated dependences begin to deviate from the experimental curves and are slightly lower. It appears that, at high temperatures, it is no longer possible to consider only the quasidoublet ground state of the terbium ion and excited states of the terbium ion must be taken into account.

We have shown that the main contribution to the magnetization of the ferroborate $\text{TbFe}_3(\text{BO}_3)_4$ within the range of temperatures and magnetic fields studied here is from the terbium ion magnetic subsystem. Since the ratio Φ/M is constant, the major contribution to the magnetic field-induced rotation of the plane of polarization of light is also from the terbium ion magnetic subsystem. Thus, the constant $300 \text{ deg} \cdot \text{form.unit}/\mu_B \cdot \text{cm}$ derived from the temperature dependence of Φ/M corresponds to the magneto-optical constant B of Eq. (1).

^{a)}Email: bedarev@ilt.kharkov.us

-
- ¹A. K. Zvezdin, S. S. Krotov, A. M. Kadomtseva, G. P. Vorob'ev, Yu. F. Popov, A. P. Pyatakov, L. N. Bezmaternykh, and E. N. Popova, *Pis'ma Zh. Eksp. Teor. Fiz.* **81**, 335 (2005).
²A. K. Zvezdin, G. P. Vorob'ev, A. M. Kadomtseva, Yu. F. Popov, A. P. Pyatakov, L. N. Bezmaternykh, A. V. Kuvardin, and E. N. Popova, *Pis'ma Zh. Eksp. Teor. Fiz.* **83**, 600 (2006).
³M. Janoschek, P. Fischer, J. Schefer, B. Roessli, V. Pomjakushin, M. Meven, V. Petricek, G. Petrakovskii, and L. Bezmaternikh, *Phys. Rev. B* **81**, 094429 (2010).
⁴S. A. Klimin, D. Fausti, A. Meetsma, L. N. Bezmaternykh, P. H. M. van Loosdrecht, and T. T. M. Palstra, *Acta Crystallogr. B* **61**, 481 (2005).
⁵C. Ritter, A. Bulaev, A. Vorotynov, G. Petrakovskii, D. Velikanov, V. Temerov, and I. Gudim, *J. Phys. Condens. Matter* **19**, 196227 (2007).
⁶N. F. Kharchenko, A. V. Bibik, and V. V. Eremenko, *Pis'ma Zh. Eksp. Teor. Fiz.* **42**, 447 (1985).
⁷B. A. Tavger and V. M. Zaitsev, *Zh. Eksp. Teor. Fiz.* **30**, 564 (1956).
⁸S. V. Vonsovskii, *Magnetism* (Nauka, Moscow, 1971) [in Russian].
⁹M. N. Popova, E. P. Chukalina, T. N. Stanislavchuk, and L. N. Bezmaternykh, *Izv. RAN, Ser. fiz.* **70**, 1652 (2006).
¹⁰M. N. Popova, *J. Magn. Magn. Mater.* **321**, 716 (2009).
¹¹E. A. Popova, D. V. Volkov, A. N. Vasiliev, A. A. Demidov, N. P. Kolmakova, I. A. Gudim, L. N. Bezmaternykh, N. Tristan, Yu. Skourski, B. Buechner, C. Hess, and R. Klingeler, *Phys. Rev. B* **75**, 224413 (2007).

Translated by D. H. McNeill