

# LOW TEMPERATURE OPTICAL ABSORPTION SPECTRA OF $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$ FERROBORATE

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We study low temperature optical absorption spectra of the  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  crystal in the region of optical transition  $^4I_{9/2} \rightarrow ^2H_{11/2}$  of  $\text{Nd}^{3+}$  in  $\sigma$ - and  $\pi$ -polarizations. The spontaneous spin-reorientation phase transitions at  $T_{\text{cr1}} = 25$  K and  $T_{\text{cr2}} = 16$  K reveal themselves in the absorption spectra. Substantial transformation of the polarized spectra is associated with a change in the intensities of transitions between different splitting components of neodymium Kramers doublets caused by the changes of magnetic structure.

## Introduction

$\text{Nd}_{1-x}\text{Dy}_x\text{Fe}_3(\text{BO}_3)_4$  compounds belong to the family of rare-earth trigonal ferroborates which are of interest due to a wide variety of magnetic properties and multiferroic effects. Substituted ferroborates can exhibit new magnetic reorientation transitions caused by the competition of magnetic anisotropies of different rare-earth ions. In  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  antiferromagnetic ordering occurs at  $T_N = 32$  K. The crystal exhibits two spontaneous spin-reorientation transitions at  $T_{\text{cr1}} = 25$  K and  $T_{\text{cr2}} = 16$  K [1]. It was established that at  $T_{\text{cr1}} < T < T_N$  easy-plane magnetic structure is realized and the range  $T < T_{\text{cr2}}$  corresponds to easy-axis phase [1].

## Experimental details

We used  $\text{Nd}_{1-x}\text{Dy}_x\text{Fe}_3(\text{BO}_3)_4$  single crystal grown from the melt solution. The sample was made in the form of plane-parallel plate with a thickness of 1.35 mm, oriented in such a way that the third- and second-order crystallographic axes lay in the plane of the sample. The absorption spectra were recorded for light propagating perpendicular to the  $C_3$  axis, with the direction of the light  $\mathbf{E}$ -vector being parallel ( $\pi$ -spectra) or perpendicular ( $\sigma$ -spectra) to the  $C_3$  axis.

## Polarized absorption spectra of $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$ in the region of magnetic ordering of the crystal

We studied optical absorption spectra of  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  in the region of the transitions  $^4I_{9/2} \rightarrow ^2H_{11/2}$  in  $\text{Nd}^{3+}$  (C-group) in the temperature range 5.4–27 K. The lines C5 and C6 (Fig. 1) are associated with transitions of the type  $\Gamma_4 \rightarrow \Gamma_4$  in the crystal field  $D_3$ . The C4 line corresponds to the  $\Gamma_4 \rightarrow \Gamma_{5,6}$  transition. For comparison, the spectra of pure  $\text{NdFe}_3(\text{BO}_3)_4$  ferroborate with an easy-plane magnetic structure [2] are presented in Fig. 1. In the region of the easy-plane phase in  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  ( $T \geq 25$  K), the spectra of both crystals practically coincide, except for a slight shift in energy (about 5  $\text{cm}^{-1}$ ).

Substantial transformation of the  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  absorption spectra is observed near the temperatures  $T_{\text{cr1}}$  and  $T_{\text{cr2}}$ , corresponding to the spin-reorientation transitions. They manifest themselves mainly in changes of the intensities of different components of line splitting in the exchange field, which changes its direction.

At  $T < T_{\text{cr2}}$ , the energy intervals between the splitting components of C5 line in  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  are approximately the same as in the pure crystal. This indicates that the reorientation of magnetic moments has a very weak effect on the exchange splitting of the ground and excited Kramers doublets. At the same time, at  $T < T_{\text{cr2}}$  the spectra of pure and substituted ferroborates reveal quite different polarization properties.

For  $\text{NdFe}_3(\text{BO}_3)_4$ , polarization of the C5 line components corresponds to the transition scheme for the easy-plane magnetic ordering (Fig. 2a). We have shown in [3], that the C5 state is characterized by an inverse arrangement of sublevels in the excited doublet regarding to the ground one.

For  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$ , polarization of C5 line components corresponds to the case of easy-axis magnetic ordering (Fig. 2b), where transitions between states of the same type are polarized along the antiferromagnetism vector ( $C_3$  axis), and transitions between states of different types are polarized in the plane perpendicular to this axis. So, the spectral data are fully consistent with the statement of the work [1] concerning the transition of the crystal to the easy-axis state at  $T < T_{\text{cr2}}$ .

As for the evolution of the absorption spectra in the temperature range  $T_{\text{cr2}} < T < T_{\text{cr1}}$ , it requires further, more detailed study.

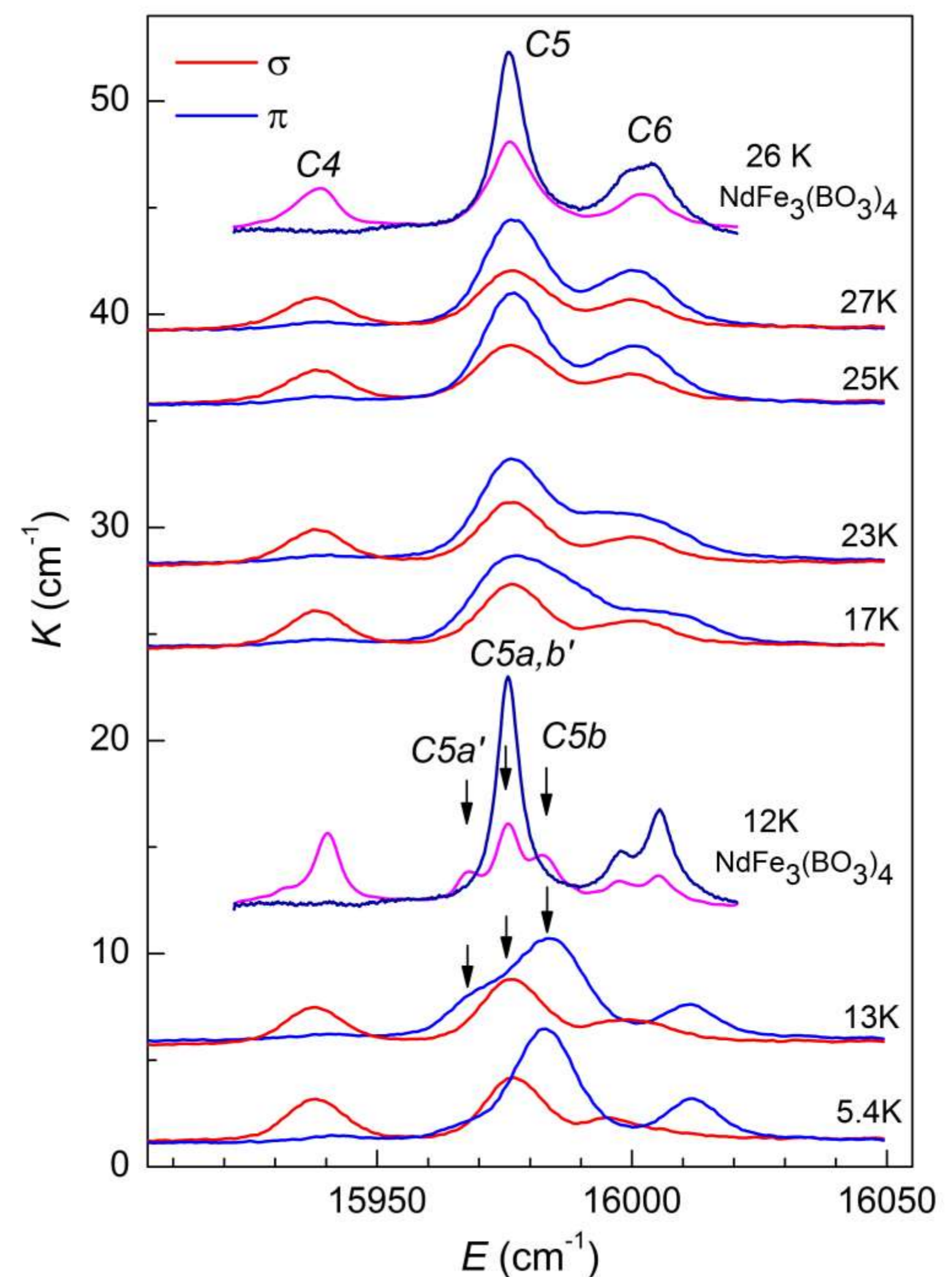


Fig. 1. Absorption spectra of  $\text{Nd}_{0.75}\text{Dy}_{0.25}\text{Fe}_3(\text{BO}_3)_4$  in the region of C4, C5 and C6 lines of the optical transition  $^4I_{9/2} \rightarrow ^2H_{11/2}$  (C-group) of  $\text{Nd}^{3+}$  in  $\sigma$ - and  $\pi$ - polarizations at different temperatures. For comparison, the spectra of  $\text{NdFe}_3(\text{BO}_3)_4$  at 12 and 26 K [2] are shown, shifted along the energy axis by 5  $\text{cm}^{-1}$ .

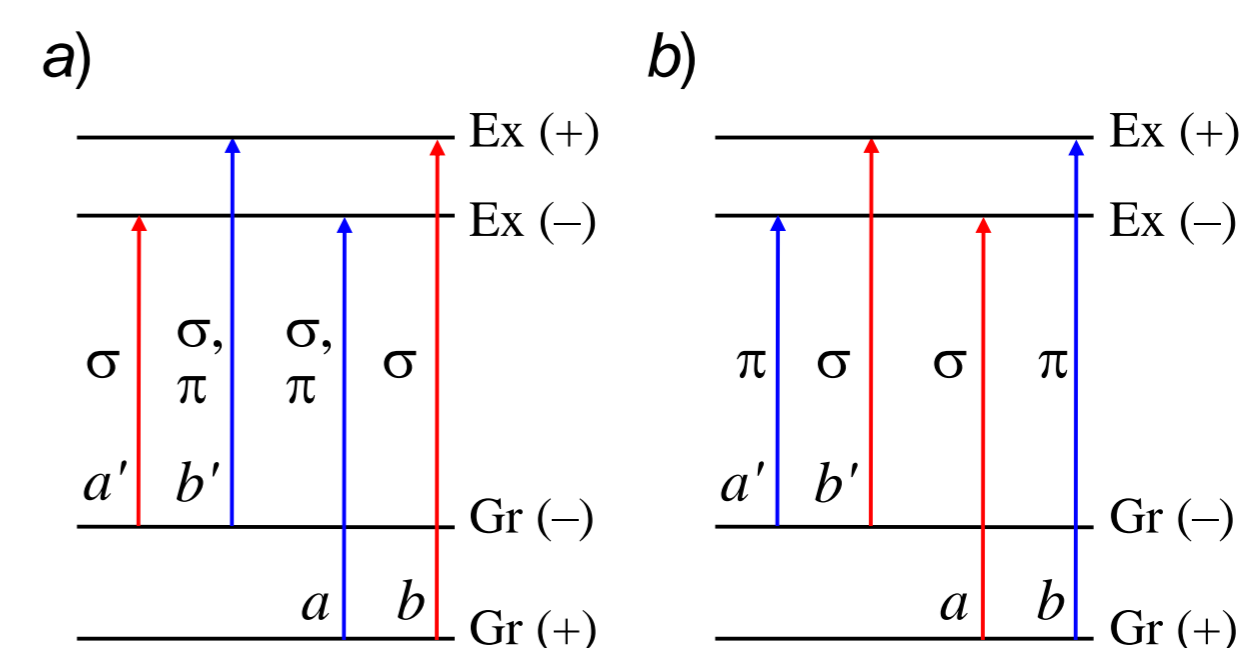


Fig. 2. Schemes of transitions between the components of the ground and excited Kramers doublets and polarization of electric-dipole transitions for easy-plane (a) and easy-axis (b) magnetic ordering.

[1] G.A. Zvyagina, K.R. Zhekov, A.A. Zvyagin, I.A. Gudim, and I.V. Bilych, Low Temp. Phys. **38**, 446 (2012).

[2] I.S. Kachur, V.S. Kurnosov, V.G. Piryatinskaya, A.V. Yeremenko, and I.A. Gudim, Low Temp. Phys. **43**, 590 (2017).

[3] V.V. Slavin, I.S. Kachur, V.S. Kurnosov, and V.G. Piryatinskaya, Low Temp. Phys. **51**, 1062 (2025).