DETERMINATION OF Nd-Fe EXCHANGE INTERACTION SIGNS IN EXCITED STATES OF Nd^{3+} IN $NdFe_3(BO_3)_4$ CRYSTAL USING OPTICAL SPECTROSCOPY

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Low temperature optical absorption spectra of neodymium ferroborate crystal $NdFe_3(BO_3)_4$ were studied in the region of optical transition $^4I_{9/2} \rightarrow {}^4G_{5/2}$ in Nd^{3+}. A group-theoretic analysis of transitions between the splitting components of Nd^{3+} Kramers doublets was performed. It was shown that the sign of exchange interaction with the iron subsystem in an excited state of Nd^{3+} can be different from that in the ground state.

$NdFe_3(BO_3)_4$ belongs to the family of trigonal rare-earth ferroborates, which attracts the attention of researchers due to the existence of correlation between magnetic, electrical and elastic subsystems. The crystal structure of $NdFe_3(BO_3)_4$ is described by the space symmetry group $R32$; the rare-earth ion occupies a position with local symmetry D_3. At $T_N \approx 30$ K the crystal orders antiferromagnetically with orientation of Fe^{3+} and Nd^{3+} magnetic moments along one of the C_2 axes in the basal plane. Below $T_{IC} \approx 13.5$ K, the collinear magnetic structure transforms into a long period spiral propagating along the C_3 axis; orientation of the magnetic moments remains parallel to the basal plane.

Experimantal details

We used $NdFe_3(BO_3)_4$ single crystals grown from the melt solution. The samples were made in the form of plane-parallel plates, 0.2–0.7 mm thick, oriented perpendicular to the third-order crystallographic axis. The absorption spectra were recorded for light propagating perpendicular to the C_3 axis, with the direction of the light **E**-vector being parallel (π-spectra) or perpendicular (σ-spectra) to the C_3 axis.

Polarization of electric-dipole transitions in C_2 symmetry

The states of Nd^{3+} ion in the crystal field of D_3 symmetry are Kramers doublets, which are transformed according to the two-valued irreducible representations Γ_4 and $\Gamma_{5,6}$. The lowest level of the ground multiplet has Γ_4 symmetry. At temperatures $T<T_N$, the Kramers doublets having Γ_4 symmetry are split due to the exchange interaction with the iron subsystem; as a result, up to four transitions can be observed.

With easy-plane magnetic ordering of the crystal, the local symmetry of the rare-earth ion decreases. We consider the selection rules for electric-dipole transitions in C_2 symmetry, which is the maximum for easy-plane magnetic ordering.

Polarizations of electric-dipole transitions between Kramers states within the framework of C_2 symmetry. Γ_3, Γ_4 are the one-dimensional irreducible representations of the group C_2.

	Γ_3	Γ_4
Γ_3	p_x σ	p_y, p_z σ,π
Γ_4	p_y, p_z σ,π	p_x σ

Depending on arrangement of the excited doublet sublevels, different transitions can be observed at $T<T_N$ in the π-spectra (Fig. 1).

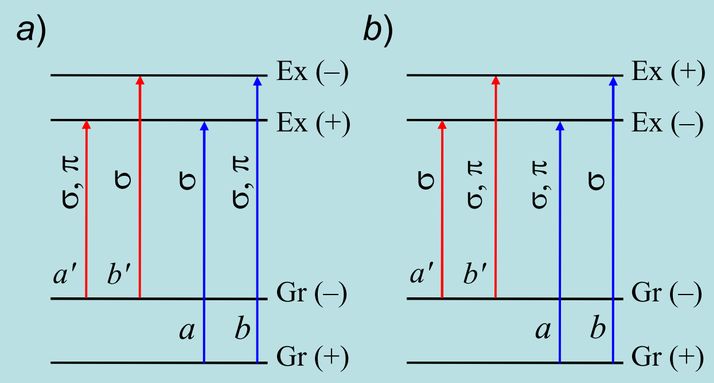

Fig. 1. Schemes of transitions between the components of the ground and excited Kramers doublets and polarization of electric-dipole transitions in C_2 symmetry for different arrangement of the excited doublet sublevels.

Low temperature absorption spectra of $NdFe_3(BO_3)_4$ in the region of the $^4I_{9/2} \rightarrow {}^4G_{5/2}$ transition

The multiplet $^4G_{5/2}$ is split in the crystal field of D_3 symmetry into three Kramers doublets: $^4G_{5/2} \rightarrow 2\Gamma_4 + \Gamma_{5,6}$. In accordance with the selection rules for D_3 symmetry, electric-dipole transitions $\Gamma_4 \rightarrow \Gamma_4$ should be observed in both σ- and π-polarization, while transitions $\Gamma_4 \rightarrow \Gamma_{5,6}$ – only in σ-polarization. The lines D1 and D2 shown in the Fig. 2 correspond to transitions to states with Γ_4 symmetry.

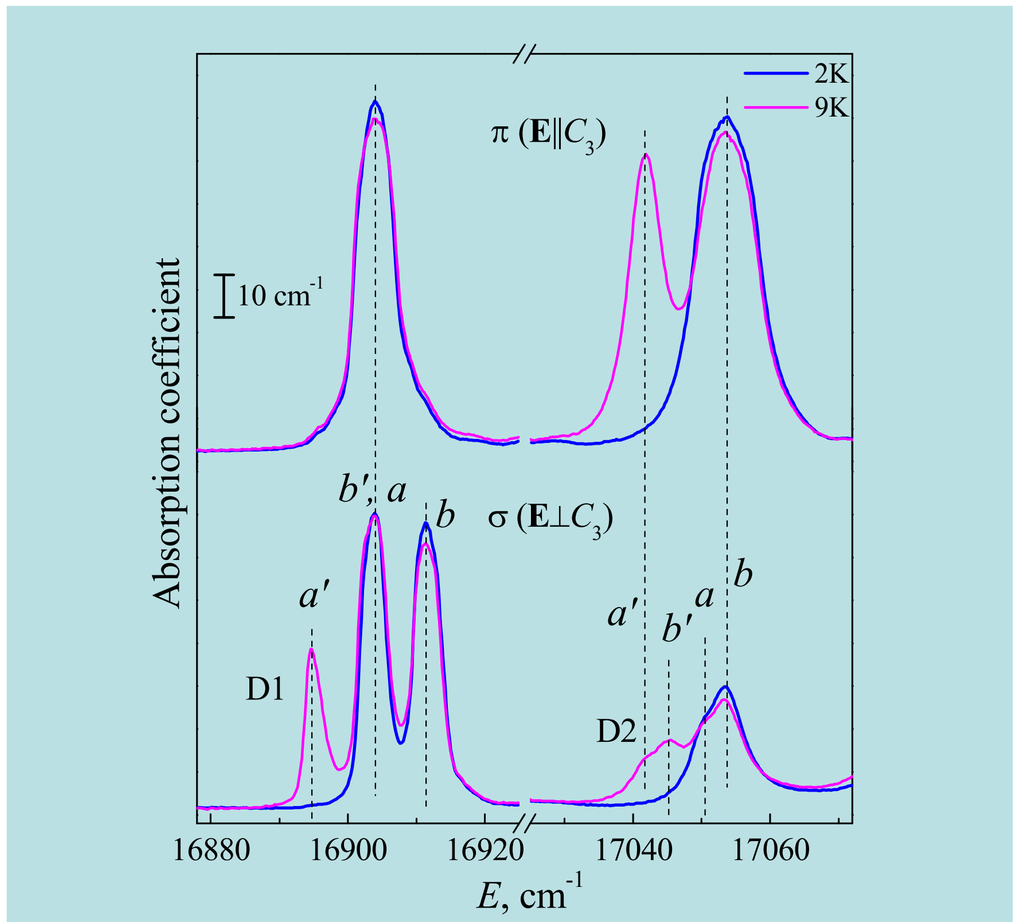

Fig. 2. Fragment of the absorption spectra of $NdFe_3(BO_3)_4$ in the region of optical transition $^4I_{9/2} \rightarrow {}^4G_{5/2}$ in π and σ-polarizations at different temperatures.

At temperatures $T<T_N$, the doublet states D1 and D2 are split due to the exchange Fe-Nd interaction. At a temperature of 2 K, only the transitions *a* and *b* from the lowest sublevel of the ground doublet (Fig. 1) can be observed. As the temperature increases, the transitions *a*' and *b*' from the excited sublevel of the ground doublet also appear.

In the case of the D1 line, the splitting values of the ground and excited states are quite close. That is why transitions *a* and *b*' have very close energies, and only three splitting components can be observed.

The splitting components of the absorption lines D1 and D2 demonstrate quite different polarization properties. In the case of D2 line, only transitions *a*' and *b* are observed in the π-spectra. This corresponds to the scheme in Fig. 1 (a) where the order of levels in ground and excited doublets is the same.

For D1 line, transitions *a* and *b*' appear in π-polarization, in accordance with the scheme in Fig. 1 (b) where the order of levels in excited state is reverse with respect to the ground state. Such order of levels means a different sign of the exchange interaction with the iron subsystem in the ground and excited states of neodymium.

Thus, we studied polarization properties of the optical absorption spectra of $NdFe_3(BO_3)_4$ in the magnetically ordered state of the crystal. Using a group-theoretical analysis, it was established that for some excited states of neodymium, the Nd–Fe exchange interaction has the opposite sign with respect to the exchange in the ground state.