MANIFESTATION OF SPIRAL MAGNETIC PHASE IN OPTICAL ABSORPTION SPECTRA OF $NdFe_3(BO_3)_4$ CRYSTAL

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Optical absorption spectra of neodymium ferroborate $NdFe_3(BO_3)_4$ in the region of the transition $^4I_{9/2} \rightarrow {}^2H_{11/2}$ are studied in the temperature range 6–32 K. Unusual temperature dependences of integral intensities of absorption lines are found in σ-polarized spectra. We associate such behavior with the transition from collinear magnetic structure to long-period spiral. To describe the temperature dependences of lines intensities we propose a theoretical model which takes into account Dzyaloshinskii–Moriya interaction between Nd^{3+} and Fe^{3+} ions.

$NdFe_3(BO_3)_4$ belongs to the family of trigonal rare-earth ferroborates, which are of interest due to their nontrivial magnetic properties and the correlation between magnetic, electrical and elastic subsystems. The crystal structure of $NdFe_3(BO_3)_4$ is described by the space symmetry group *R*32; 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.

Temperature evolution of optical absorption spectra of $NdFe_3(BO_3)_4$ in the region of magnetic ordering of the crystal

We have studied optical absorption spectra of $NdFe_3(BO_3)_4$ in the region of the transition $^4I_{9/2} \rightarrow {}^2H_{11/2}$ in Nd^{3+} (C-group) in the temperature range 6–32 K. The multiplet $^2H_{11/2}$ is split in the crystal field of D_3 symmetry into six Kramers doublets: $^2H_{11/2} \rightarrow 4\Gamma_4 + 2\Gamma_{5,6}$. 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.

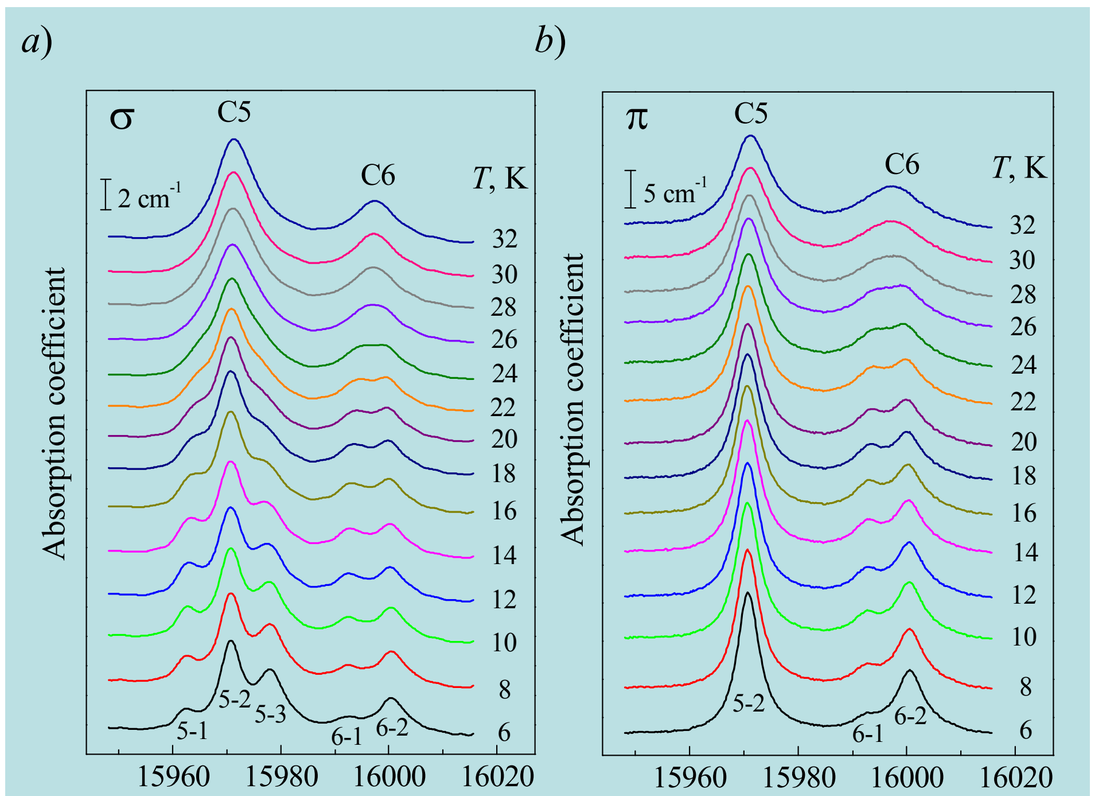

Fig. 1. Absorption spectra of $NdFe_3(BO_3)_4$ in the region of C5 and C6 lines of the optical transition $^4I_{9/2} \rightarrow {}^2H_{11/2}$ (C-group) in σ (a) and π (b) polarizations at different temperatures.

At temperatures $T<T_N$, the Kramers doublets of Nd^{3+} are split due to the exchange Fe-Nd interaction and up to four splitting components can be observed in the spectra. For C5 line, the exchange splitting of the excited state is quite close to the splitting value of the ground doublet (8.8 cm^{-1}); as a result, only three splitting components are observed. In the case of C6 line, the excited state shows practically no splitting in the exchange field. Lines 6-1 and 6-2 are associated with the transitions from the upper and lower sublevels of the ground doublet to the unsplit excited level.

We obtained the temperature dependence of the ratio *C* of the integral intensities of "hot" and "cold" transitions with the same symmetry, such as the components 5-1 and 5-3 of the line C5 and the components 6-1 and 6-2 of the line C6 (Fig. 2). The ratio of intensities of the "hot" and "cold" transitions should be determined only by the ratio of populations of the upper and lower sublevels of the ground doublet ($\exp(-\Delta E(T)/k_B T)$, where $\Delta E(T)$ is the splitting of the ground doublet). But only for π-polarized components of line C6 the dependence $C(T)$ can be described in terms of the thermoactivation model. As for σ-polarization, for both lines C5 and C6, the $C(T)$ dependences change their character just in the temperature range that corresponds to the reorientation of magnetic moments from the spiral to collinear structure.

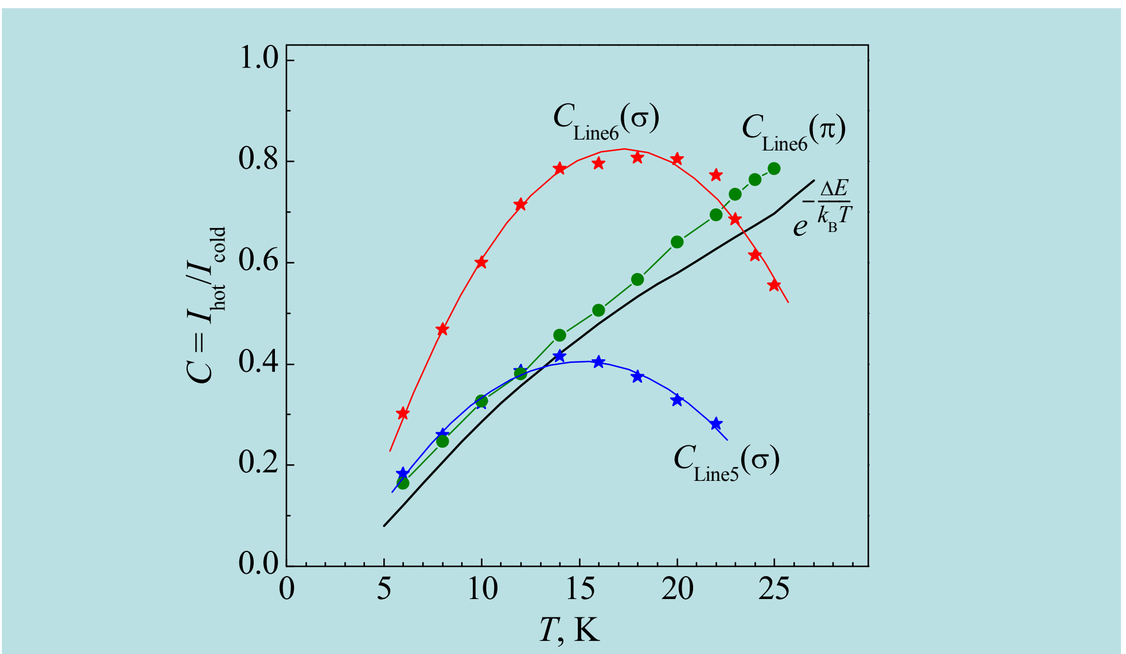

Fig. 2. The ratio of integral intensities of "hot" and "cold" components of absorption lines C5 and C6 depending on the temperature. The black solid line corresponds to thermoactivation model. Solid red and blue curves are the results of fitting the experimental data by the theoretic model.

Theoretical model

The Hamiltonian of the system has the form:

$$\hat{H} = \hat{H}_{Nd} + \hat{H}_{Fe} + \hat{H}_{Fe\text{-}Nd} \qquad (1)$$

where $\hat{H}_{Nd}$ describes the interaction of Nd^{3+} ion with crystal field; $\hat{H}_{Fe}$ describes the interaction of N_{nb}=6 Fe^{3+} ions, surrounding the Nd^{3+} ion, with the rest iron ions (in mean-field approximation). $\hat{H}_{Fe\text{-}Nd}$ is the interaction between the Nd^{3+} ion and Fe^{3+} ions:

$$\hat{H}_{Fe-Nd} = N_{nb}\left\{J_{xx}\left(\hat{\mathbf{J}}^x_{Fe}\hat{\mathbf{J}}^x_{Nd} + \hat{\mathbf{J}}^y_{Fe}\hat{\mathbf{J}}^y_{Nd}\right) + J_{zz}\hat{\mathbf{J}}^z_{Fe}\hat{\mathbf{J}}^z_{Nd} + J_{DM}\left(\hat{\mathbf{J}}^x_{Fe}\hat{\mathbf{J}}^y_{Nd} - \hat{\mathbf{J}}^y_{Fe}\hat{\mathbf{J}}^x_{Nd}\right)\right\}$$

The last term describes Dzyaloshinskii–Moriya (DM) interaction between the Nd^{3+} and Fe^{3+} ions. The eigenvalues and the eigenvectors of the Hamiltonian (1) corresponding to the ground and excited states were obtained numerically. As the result we obtained the temperature dependences of the angles between mean value of $\boldsymbol{J}$ projection on the basal plane and *a*-axis for the ground ($\varphi_{1,2}$) and excited ($\varphi'_{1,2}$) doublets. The ratio of intensities of "hot" and "cold" components is:

$$C = \frac{I_{hot}}{I_{cold}} \sim \left(\frac{\cos(\varphi_2 - \varphi'_1)}{\cos(\varphi_1 - \varphi'_2)}\right)^2 e^{-\frac{\Delta E}{k_B T}}$$

The results of fitting the experimental dependencies $C_{Line\ 5,6}(T)$ are presented in Fig. 2. The calculations show good agreement between the experimental and theoretical data. It should be noted that without taking into account the DM interaction, it is impossible to describe the experimental dependences even qualitatively. Thus, we have shown that it is DM interaction that is responsible for the unusual temperature dependences of the light absorption coefficient.