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BOOK OF ABSTRACTS

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Magnetic circular dichroism of f-f transitions in multiferroic antiferromagnet $Ho_{0.75}Nd_{0.25}Fe_3(BO_3)_4$

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We studied absorption and magnetic circular dichroism (MCD) spectra of three electron transitions: $^5I_8 \rightarrow {}^5F_2$, $^5F_3$ and $^5F_5$ of Ho^{3+} ion in antiferromagnet crystal $Ho_{0.75}Nd_{0.25}Fe_3(BO_3)_4$. In Fig. 1 we present results for transition into the state $^5F_2$ (G-band), which is decomposed in the D_3 local symmetry in the following way: $A_1 + E + E$.

In paramagnets, magnetic circular dichroism (MCD) of a transition doublet→singlet in magnetic field directed along the light propagation (parallel to the C_3 axis of the crystal in our case) is given by the equation:

$$\Delta k = k_m c\, \varphi(\omega,\omega_0) + k_m \Delta\omega_0\, \partial\varphi(\omega,\omega_0)/\partial\omega_0 \,. \qquad (1)$$

The first term in (1) is the temperature dependent paramagnetic MCD (*C*-term) and the second one is the diamagnetic effect. Spectrum of the paramagnetic term, $\varphi(\omega,\omega_0)$, coincides with that of absorption. Above T_R=6.9 K the studied crystal transfers from the easy axis state to the easy plane one [1]. So, Figs. 1A and 1B present MCD in the field $H||C_3$ below and above T_R. Below the T_R (T=5 K) shape of the MCD spectrum well corresponds mainly to the diamagnetic term of MCD. Above T_R the MCD spectrum approximately repeats absorption spectrum accurate within signs. This implies that only the paramagnetic MCD remains.

Magnetic field is applied parallel to the trigonal C_3-axis. Consequently, in the easy axis state the magnetic field is directed parallel to the Ho magnetic moments, and energies of the Ho ions in two sublattices change in magnetic field on the values $\pm\mu_B g H$, and MCD acquires diamagnetic shape with some asymmetry relative to y=0 axis because of the paramagnetic component (Fig. 1A).

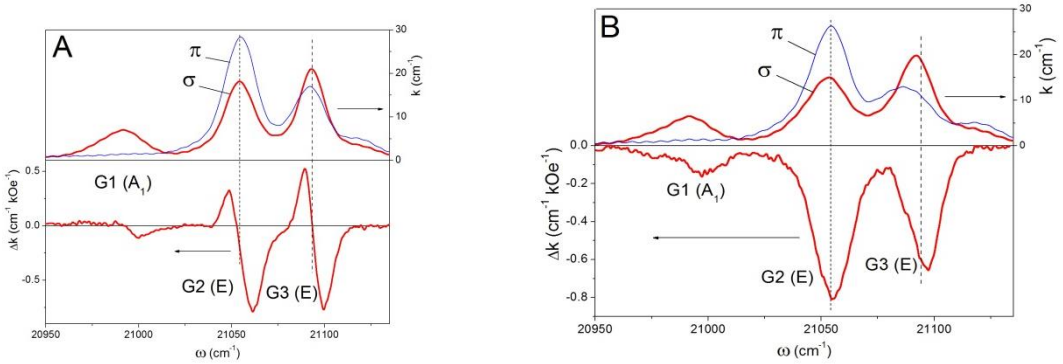

Fig. 1. Polarized absorption (k) and MCD (Δk) spectra at temperatures 5 K (A) and 8 K (B).

Above the reorientation transition the external magnetic field $H||C_3$ is perpendicular to the exchange field H_e from the Fe sublattice. The total effective field is: $H_{ef}=(H_e^2+H^2)^{1/2}$. The change of the Ho ion energy in the magnetic field $H<<H_e$ will be: $\Delta E(H)=\mu_B g(H_{eff}-H_e)\approx \mu_B g H(H/2H_e)$. According to Ref. [2], $H_e\approx$25 kOe in pure Ho ferroborate. If we suppose that in our crystal the

exchange field Fe-Ho is the same, than the splitting in the used magnetic field H=2 kOe will be 25 times decreased. The temperature dependent paramagnetic MCD will be decreased in the same degree. So, both diamagnetic and temperature dependent paramagnetic MCD become not measurable. However a strong effect with the paramagnetic type dispersion is observed (Fig. 1). Consequently, above the reorientation transition we deal with the paramagnetic B-term, or term of mixing.

In the easy axis state eigenfunctions of the Ho Hamiltonian in magnetic field $H||C_3$ coincide with those without magnetic field, when $\boldsymbol{M}_{\mathrm{Ho}},\boldsymbol{M}_{\mathrm{Fe}}||C_3$. Therefore the magnetic field does not mix the wave functions. In the easy plane state eigenfunctions of the Ho Hamiltonian in magnetic field $H||C_3$ do not coincide with those without magnetic field, when $\boldsymbol{M}_{\mathrm{Ho}},\boldsymbol{M}_{\mathrm{Fe}}\perp C_3$, i. e., in the exchange field $H_e\perp C_3$. The latter functions are mixtures of states with opposite projections M_J of Ho moments on the C_3-direction and, therefore, they have diminished magnetic moments. External magnetic field $H||C_3$ switches on the opposite process: it makes states magnetic ones. Thus, B-term appears in crystals due to mixing of states by magnetic field.

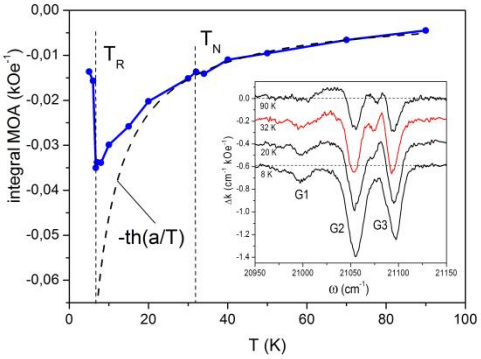

Fig. 2. MOA of ${}^5I_8 \rightarrow {}^5F_2$ transition. Inset: MCD spectrum as a function of temperature.

Fig. 2 demonstrates temperature dependence of integral magneto-optical activity (MOA) of the G-band. Additionally, the theoretical dependence of paramagnetic term c~th(a/T) from (1) is shown on the condition, that it coincides with the experimental MOA at T=70 K. The process of quenching of the temperature dependent paramagnetic MOA is well seen. Transformation of the MCD spectrum with temperature is shown in Fig. 2 inset. The diamagnetic part of MCD is gradually appearing with the temperature increase.

In magnetically ordered crystal, splitting of the ground state and difference of populations of its sublevels is mainly created by the exchange field H_e. Therefore, in the external magnetic field, directed along the exchange field in the easy axis state, we measure only change of the paramagnetic MCD due to change of the difference of the sublevels populations by the external magnetic field:

$$\Delta c(H) = H\frac{\mathrm{dc}}{\mathrm{dH}}(H = H_e) = -(\mu_B g_{\mathrm{CM}}\, H/2\, k_B T)/[\mathrm{ch}^2(\mu_B g_{\mathrm{CM}}\, H_e/2\,\mathrm{kT})].$$

This formula indeed describes the experimental behavior of MCD at $T<T_R$ (Fig. 2).

References

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[2] A.V. Malakhovskii, S.L. Gnatchenko, I.S. Kachur, V.G. Piryatinskaya, I.A. Gudim, Low Temp. Phys. 43, 610 (2017).