ESRF	<b>Experiment title:</b> Exploring magnetic transitions in <i>R</i> Fe <sub>3</sub> (BO <sub>3</sub> ) <sub>4</sub> by magnetic-field XMCD measurements	Experiment number: HC-1804
Beamline:	Date of experiment:	Date of report:
ID12	from: 15 July 2015 at 08:00 to: 21 July 2015 at 08:00	21 July 2015
Shifts:	Local contact(s):	<b>Received</b> at
18	Dr. Andrei Rogalev	ESRF:
Names and affiliations of applicants (* indicates experimentalists):		
M.S. Platunov, N.V. Kazak, V.A. Dudnikov		

Kirensky Institute of Physics, Siberia Branch of Russian Academy of Science Akademgorodok 50/38, Krasnoyarsk, 660036 Russia

## **Report:** Introduction:

Rare-earth ferroborates Nd<sub>1-x</sub>Ho<sub>x</sub>Fe<sub>3</sub>(BO<sub>3</sub>)<sub>4</sub> are a particular class of multiferroics. These compounds have rhombohedral structure described by the noncentrosymmetrical trigonal space group R32 (D<sup>7</sup><sub>3</sub>). Nd<sub>1-x</sub>Ho<sub>x</sub>Fe<sub>3</sub>(BO<sub>3</sub>)<sub>4</sub> have complex magnetic structure, phase transitions and magnetic anisotropy. An antiferromagnetic ordering of the Fe<sup>3+</sup> spins exists in the temperature range 38 - 30 K. The mutual orientation of Fe<sup>3+</sup> spins is affected by the paramagnetic subsystem of the rare-earth ions. Depending on the anisotropy of the rare-earth ions, the exchange interaction between the R-Fe ions stabilizes either an "easyplane" (*ab*- plane) magnetic anisotropy, or an "easy-axis" (*c*-axis) anisotropy. In addition, HoFe<sub>3</sub>(BO<sub>3</sub>)<sub>4</sub> has a spontaneous spin-reorientation transition from the "easy-plane" to "easy-axis" state at  $T_{SR} = 4.8$  K [1], while NdFe<sub>3</sub>(BO<sub>3</sub>)<sub>4</sub> has magnetic anisotropy of "easy-plane" over temperature range [2].

We focus our experiment on  $Nd_{0.5}Ho_{0.5}Fe_3(BO_3)_4$  single crystal, for which the spin-reorientation and antiferromagnetic transitions take place at ~ 10 and 32 K, respectively [3]. The complex magnetization behavior with two spin-flop transitions at 1 and 3 T are observed. The puppose of this experiment was found out the nature of spin-flop transitions and to clarify what kind of magnetic sublattices (Ho, Nd or Fe) are responsible for spin-flop transitions.

## **Experimental details**

We have carried out x-ray magnetic circular dichroism (XMCD) measurements at the Nd, Ho  $(L_{3,2})$ , and Fe (K-) edges using total fluorescence yield detection mode in «backscattering» geometry. The temperature was 3 K and a magnetic field was  $\pm 6$  T. The applied magnetic field was along to  $C_3$ -axis as it is reguired for observation the spin-flop transition.

## Results

Figures 1 and 2 are shown the XANES/XMCD spectra recordered at Fe K-(7112 eV), Ho  $L_{3,2}$ -(8071, 8919 eV), and Nd  $L_{3,2}$ -



Fig. 1. Normalized Fe *K*-edge XANES/XMCD spectra recorded at 2 K and in 5 Tesla.

(6208, 6722 eV) absorption edges. The Ho and Nd  $L_{3,2}$ -XANES spectra is dominated by the contributions from the dipole transitions E1  $(2p \rightarrow 5d)$  and quadrupolar transition E2  $(2p \rightarrow 4f)$  at the pre-edge region. The low intensity bifurcated peak observed at the Fe pre-edge can be assigned to  $1s-3d(t_{2g})$  and 1s- $3d(e_g)$  transitions [1]. We have seen the XMCD signal at all measured edges, that confirm the coexistence of three magnetic subsystems of Fe, Ho and Nd ions. The small magnitude of the dichroism at Fe K-edge confirms the antifferomagnetic ordering of Fe<sup>3+</sup> magnetic sublattices as previously presumed from SQUID data.



Fig. 2. Normalized Ho/Nd  $L_{3,2}$ -edges XANES/XMCD spectra recorded at 2 K and in 5 Tesla.

Figure 3 shows the field dependences of XMCD signals at the Fe *K*-edge, Nd and Ho  $L_{3,2}$ - edges. There are several notable features: i) all magnetization curves are undergoing the jump at  $H_{sf} = 1$  T; ii) the

magnetization curves of Fe and Ho display the saturation at ~5 T; iii) the magnetization curve of Nd demonstrates the linear dependence with change of slope above  $H_{sf}$ ; iv) there is weak observable features at the Nd magnetization curve at ~ 0.5 T; v) the magnetization behaviors of Fe and Ho subsystems follow the macroscopic magnetization curve obtained at H parallel  $C_3$ -axis (the inset to fig. 3).

At the field of H = Hsf = 1 T the Ho paramagnetic moment became normal to the applied field and then gradually increase reaching the saturation at 5 T. Under the action of the effective field  $H_{eff} = H + H_{ex}$  the magnetic moments of the iron subsystem gains small cant toward the field direction. The future magnetization process is defined by the Ho<sup>3+</sup> rare-earth subsystem. As result, the contribution from the iron subsystem to magnetization is non-linear and demonstrates the saturation at 5 T. At the low field ( $H \le 1$  T) the Ho ar



Fig. 3. The element-specific magnetization curves of Ho, Nd, and Fe at 2 K. Inset: macroscopic magnetization curve at 2 K. Comparison against the SQUID magnetisation measurements suggests that the actual temperature of the sample was 2 K.

saturation at 5 T. At the low field (H < 1 T) the Ho and Nd magnetization curves demonstrate different of the slopes indicating different strength of *f*-*d* exchange interactions with iron subsystem.

In Nd<sub>0.5</sub>Ho<sub>0.5</sub>Fe<sub>3</sub>(BO<sub>3</sub>)<sub>4</sub> the exchange interaction between the rare-earth ions takes place via oxygen and boron, i.e., R-O-B-O-R and is expected to be weak. The *f-d* superexhange interaction between rare-earth and iron ions through the R-O-Fe determins the magnetic properties of the system. The spin-flop transition at 1 T for all three magnetic subsystems confirms the existence of "easy-axis" anisotropy.

This work has been financed by Council for Grants of the President of the Russian Federation (project nos. NSh-2886.2014.2, SP-938.2015.5), Russian Foundation for Basic Research (project nos. 13-02-00958-a, 13-02-00358-a, 14-02-31051-mol-a). The work of one of coauthors (M.S.P.) was supported by the program of Foundation for promoting the development of small enterprises in scientific and technical sphere ("UMNIK" program). The authors thank BL staff for help during the experiment.

- 1. A. Pankrats, G. Petrakovskii, A. Kartashev, et al., J. Phys.: Condens. Matter 21, 436001 (2009).
- 2. P. Fischer, V. Pomjakushin, D. Sheptyakov, et al. J. Phys.: Condens. Matter 18, 7975 (2006)
- 3. R. P. Chaudhury, F. Yen, B. Lorenz, et al. Phys. Rev. B 80, 104424 (2009).