Magnetic structure of multiferroics RFe$_3$(BO$_3$)$_4$ (R =Ce, Sm)

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Coexistence of magnetic order and electric polarization, *multiferroicity*, has become a top major topic over the past decade in condensed matter physics. In the multiferroic materials, rare-earth ferroborates RFe$_3$(BO$_3$)$_4$ (R = Y and rare-earth metal are a series of new multiferroic compounds containing the R^{3+} ($4f^n$) and Fe^{3+} ($3d^5$ $S = 5/2$) as magnetic ions [1]. The crystal structure is a trigonal and the space group is $R32$. The family of compounds shows diverse magnetoelectric (ME) effects as a function of the R^{3+} ions. In particular, the rare-earth ferroborates having the easy-plane type anisotropy exhibit huge ME effects. Thus we focus on SmFe$_3$(BO$_3$)$_4$ and CeFe$_3$(BO$_3$)$_4$ which have easy-plane type anisotropy. In RFe$_3$(BO$_3$)$_4$, the multiferroicity is explained by the spin-dependent metal-ligand hybridization mechanism, meaning that the local orientation of the magnetic moment plays a key role to determine the multiferroicity. It is important to identify the precise magnetic structure.

We performed neutron diffraction experiment to identify the magnetic structures of the SmFe$_3$(BO$_3$)$_4$ and CeFe$_3$(BO$_3$)$_4$. The single crystal samples were grown by a flux method. 99 % of the natural B and 98.9 % of the natural Sm were enriched by the ^{11}B and ^{154}Sm. The masses of the samples were 26 mg for ^{154}SmFe$_3$(^{11}BO$_3$)$_4$ and 23 mg for CeFe$_3$(^{11}BO$_3$)$_4$. Neutron diffraction was performed by four-circle diffractometer HB-3A installed at ORNL. A vertically focusing silicon (022) monochromator was chosen to obtain the neutrons with the wave length of 1.542 Å. A closed cycle refrigerator was used to achieve 4.1 K as a base temperature.

The nuclear reflections measured at 4.1 K are reasonably refined by the trigonal structure with the space group $R32$ in both compounds, meaning that the symmetry of the crystal structure is retained at the low temperature.

Figure 1(a) shows the temperature evolution of the ω-scans of ^{154}SmFe$_3$(^{11}BO$_3$)$_4$ at $(-1, 0, 0.5)$. An additional peak appears below 33 K, and it is indexed by the propagation vector $\boldsymbol{k} = (0, 0, 1.5)$. This is consistent with the previous powder neutron diffraction measurement [2]. Unlike the NdFe$_3$(BO$_3$)$_4$ [3], any incommensurate magnetic peak is not observed. Analysis of the magnetic peaks using Rietveld refinement, it is found that the magnetic moments of the Fe^{3+} ions form a collinear structure along the crystallographic a axis and they antiferromagnetically propagate along the c axis. The Sm^{3+} moment aligns in the ab plane but it is not parallel to the Fe^{3+} moments.

The temperature evolution of the l-scans of CeFe$_3$(^{11}BO$_3$)$_4$ at $(-1, 0, 0.5)$ is shown in Fig. 1(b). Additional peaks appear around $l = 0.5$ below 30 K, meaning that a magnetic long-range order occurs at 30 K. This is consistent with the previous specific heat and magnetic susceptibility measurements of the polycrystalline sample [4]. The additional peak at $(-1, 0, 0.5)$ is indexed by the commensurate propagation vector $\boldsymbol{k} = (0, 0, 1.5)$, whereas the peaks at $(-1, 0, 0.45)$ and $(-1, 0, 0.55)$ are indexed by the incommensurate vector $\boldsymbol{k} = (0, 0, 1.5 + \delta)$ where $\delta = 0.042$ at 4.1 K. This incommensurability δ decreases with the increase of the temperature as shown in Fig. 1(b). From the measurement of the detailed temperature dependence of the l-scans, it is found that the transition temperature of the commensurate magnetic peaks differs from that of the incommensurate ones. This indicates that the phase separation between the commensurate and incommensurate magnetic structures occurs in the magnetic long-

range order. The detailed analysis of the magnetic structures in both compounds is now in progress.

References

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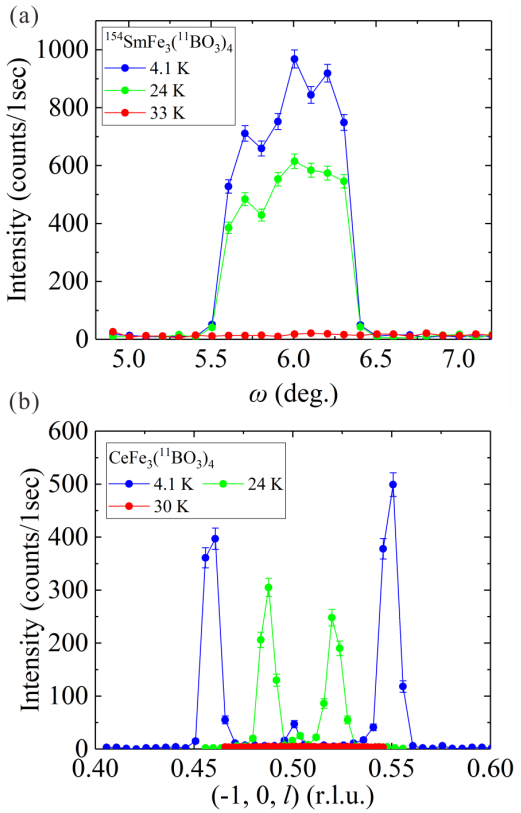

Fig. 1. (a) A temperature evolution of ω-scans of $^{154}SmFe_3(^{11}BO_3)_4$ at $(-1, 0, 0.5)$. (b) A temperature evolution of l-scans of $CeFe_3(^{11}BO_3)_4$ at $(-1, 0, 0.5)$.