

Magnetic ordering in relation to room-temperature magnetoelectric effect of Z-type hexaferrite

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Recent extensive studies on magnetoelectric multiferroics have revealed that a new class of multiferroics, that is, magnetically-induced ferroelectrics in which ferroelectricity is induced by complex magnetic orders, such as spiral orders, exhibit giant magnetoelectric effects, remarkable changes in electric polarization in response to a magnetic field. Although these magnetically-induced ferroelectrics are attracting great interest in terms of both basic and technological points of view, none of the existing materials show magnetoelectric (ME) effect by applying a low magnetic field at ROOM TEMPERATURE. Recently, however, it is discovered that a Z-type hexaferrite $\text{Sr}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (SCFO) shows the ME effect with a small magnetic field even at room temperature.[1] Although this discovery is important from an application standpoint, the origin of the ME effect of the Z-type hexaferrite is still unclear. To point out a route toward generalization of the promising room-temperature ME effect, it is necessary to clarify the underlying mechanism. For this purpose, we carried out powder neutron diffraction measurements on the Z-type hexaferrites $(\text{Sr},\text{Ba})_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ over wide temperature and magnetic-field ranges.

The first step was to search for T-dependent diffraction peaks in the absence of H. Figure 1 shows neutron powder diffraction (NPD) patterns of SCFO measured at $T=150$ K and 566 K. For the data at 566 K, which is below the magnetic ordering temperature (≈ 670 K), a Rietveld analysis was carried out using the space group $P6_3/mmc$ and the ferrimagnetic structure reported by Tachibana and coworkers[2,3]. As shown in Fig.1, the profile at 566 K is

well explained by these chemical and magnetic structures.

Compared with the NPD profile at 566 K, additional peaks exist evidently at 150 K: $00l_o$ reflections ($l_o=\text{odd}$), which are forbidden in the space group $P6_3/mmc$. In addition, intensities of $00l_e$ Bragg reflections ($l_e=\text{even}$) are enhanced in the profile at 150 K. The $00l_o$ reflections appear below 400 K and increase in intensity with decreasing T. We conclude that they are the magnetic reflections characterized by the magnetic propagation vector of $(0,0,1)$, indicating that $00l_o$ corresponds with $\pm(0,0,1)$ satellite points of $00l_e$.

To elucidate the effect of H on the magnetic structure which develops below ~ 400 K, the magnetic reflections of SCFO were also measured as a function of H at 10 K and 300 K. Significant changes in the neutron intensities were observed by applying H. The intensities of the $10L$ and 008 reflections increase with increasing H and are saturated above 3 T. By contrast, the intensity of the 009 reflection decreases and disappears above 3 T, indicating that the magnetic structure having the $(0,0,1)$ propagation vector exists only below 3 T. Based on these results of the T- and H-dependence of the electric polarization P, we conclude that the magnetic structure characterized by the propagation vector $(0,0,1)$ is the origin of the ME effect in the Z-type hexaferrite system. These magnetic reflections can be ascribed to a transverse conical spin structure which allows finite polarization in terms of the inverse Dzyaloshinskii-Moriya mechanism. Our results provide a natural explanation of the low-field magnetoelectric effect at room temperature in the Z-type hexaferrite.

References

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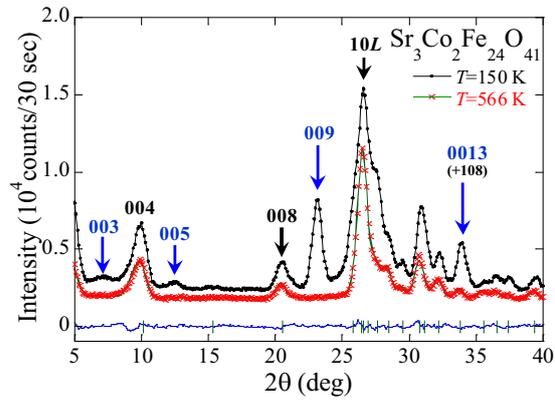


Fig. 1. Neutron powder diffraction patterns of SCFO measured at two temperatures.