Spherical Neutron Polarimetry under High Pressure for Multiferroics

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Keywords: high pressure, neutron diffraction, multiferroics.

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The analysis of three dimensional neutron spin polarization vectors in scattering processes, using a technique referred to as spherical neutron polarimetry (SNP), is a very powerful technique of determining complex magnetic structures in magnetic materials.[1] However, the requirement to maintain both incident and scattered neutrons in a highly polarized state has made it difficult to use this technique in conjunction with extreme experimental conditions, such as high pressures. In fact, to date, SNP experiments under high pressures have not been carried out owing to these difficulties.

In the present study, we used a newly developed nonmagnetic Hybrid anvil high-pressure cell (HAC) to overcome the difficulties associated with performing SNP experiments under pressure. The HAC was originally developed by Osakabe for neutron diffraction experiments,[2] and is able to generate hydrostatic pressure up to 10 GPa by using two different materials, WC (with a ferromagnetic Co binder) and sapphire (or silicon carbide, SiC), as anvils.[2] In order to use the HAC for SNP experiments, we employed a combination of a sapphire single crystal and a nonmagnetic diamond composite (with a SiC binder) or WC with a nonmagnetic Ni binder as the anvil materials.

We selected the magnetoelectric (ME) multiferroic compound delafossite CuFeO2 for the first SNP experiment under high pressure. Since the multiferroic ferrite is expected to have various types of magnetic orderings under high pressure, such as collinear spindensity-wave (SDW) and noncollinear spiral structures[3], we considered that it was the best candidate to study the feasibility of the SNP analysis under pressure. Recently, it has been reported that the application of hydrostatic pressure can be used to tune the ferroelectric properties of these materials by changing their magnetic ordering/symmetry.[4]

In this presentation, we will introduce the first SNP experiment under high pressure up to 4.0 GPa in the magnetoelectric multiferroic delafossite CuFeO₂. Although the pressure versus temperature magnetic phase diagram of CuFeO₂ was partially reported in previous unpolarized neutron diffraction study[4] (Fig. 1), the detailed magnetic structures in higher pressure phases have not been determined due to lacking of enough number of observable magnetic reflections indispensable for the proper structure analysis under high pressure. On

the other hand, the SNP analysis can precisely determine the magnetic structural shape by measuring the neutron polarization matrices, even when only a small number of observable Bragg reflections are available. Actually, in this study, we have succeeded in determining the complex magnetic structures up to 4.0 GPa by comparing the observed and calculated polarization matrix.[5]

This study determined the complex spiral magnetic structures in these pressure-induced phases, each of which possesses a different spin orientations and symmetry, by measuring the full neutron polarization matrix. The results presented herein demonstrate that the SNP measurements are feasible in combination with high pressure conditions using this new apparatus, and that this method is a useful approach to studying novel pressure-induced physical phenomena associated with complex magnetic ordering.



Figure 1. Pressure versus temperature phase diagram of multiferroic delafossite CuFeO₂.[5]

Acknowledgments: This work was supported by a JSPS KAKENHI grants (No. 15H05433, 17KK0099).

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