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'I am a senior scientist working at NIMS, Tsukuba, Japan. I am mainly studying pressure effects on strongly correlated electron systems including multiferroic materials. Recently, I developed a non-magnetic Hybrid Anvil Cell for Spherical Neutron Polarimetry experiments, based on collaborations with the ILL and JAEA, Tokai.'

Spherical neutron polarimetry under high pressure for a multiferroic delafossite ferrite

Spin-polarised hot-neutron diffractometer D3

The analysis of three-dimensional neutron spin rotation at the interaction with a sample, using a technique referred to as spherical neutron polarimetry (SNP), is a very powerful means of determining complex magnetic structures in magnetic materials. In the present study, we successfully carried out the first SNP experiment under high pressure up to 4.0 GPa on the D3 instrument, studying the magnetoelectric multiferroic delafossite $CuFeO_2$. The results presented here demonstrate that SNP measurements are feasible under high-pressure conditions, and that this method is a useful approach to studying pressure-induced physical phenomena.

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Since Tasset developed the ILL's CRYOgenic Polarisation Analysis Device (CRYOPAD), which enables three-dimensional neutron polarisation analysis [1], the SNP technique has been used to determine precise spin orientations in complex magnetic structures. In unpolarised neutron diffraction experiments, one needs to collect many magnetic Bragg peaks from different sample positions to perform the standard refinement procedure. In contrast, with the SNP method the so-called polarisation matrix of one Bragg reflection contains an enormous amount of information concerning the magnetic structure; therefore, generally the measurement of just a few reflections yields very precise results. However, since the CRYOPAD requires zero-magnetic field conditions in conjunction with superconducting Meissner screening to avoid neutron depolarisation [1], inside the magnetic vacuum it is necessary to use equipment, such as high-pressure cells, that are made of non-magnetic materials. To date, SNP experiments under high pressures have not been carried out because of this difficulty.

In the present study, we used a newly developed, non-magnetic Hybrid Anvil high-pressure Cell (HAC) (**figure 1, left**) to overcome this challenge. We employed a combination of a single sapphire crystal and a non-magnetic diamond composite (with an SiC binder) or WC with a non-magnetic Ni binder as the anvil materials. We selected the magnetoelectric (ME) multiferroic compound delafossite $CuFeO_2$ 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 spin-density-wave (SDW) and non-collinear spiral structures [2,3], we felt that it was the best candidate for studying the feasibility of SNP analysis under pressure.

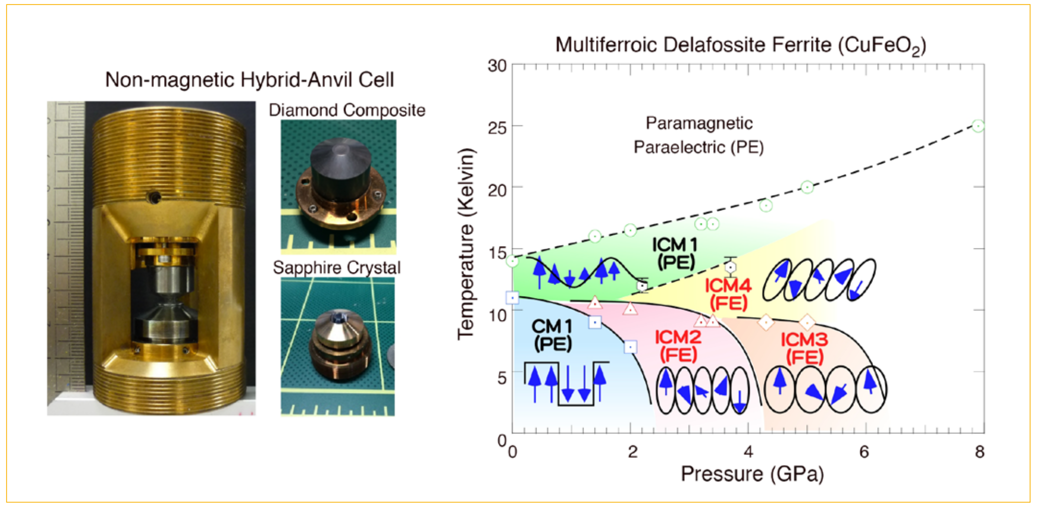

Figure 1

left) Non-magnetic Hybrid Anvil Cell with diamond composite and sapphire anvils.

right) Magnetoelectric phase diagram of multiferroic delafossite ferrite $CuFeO_2$ [4].

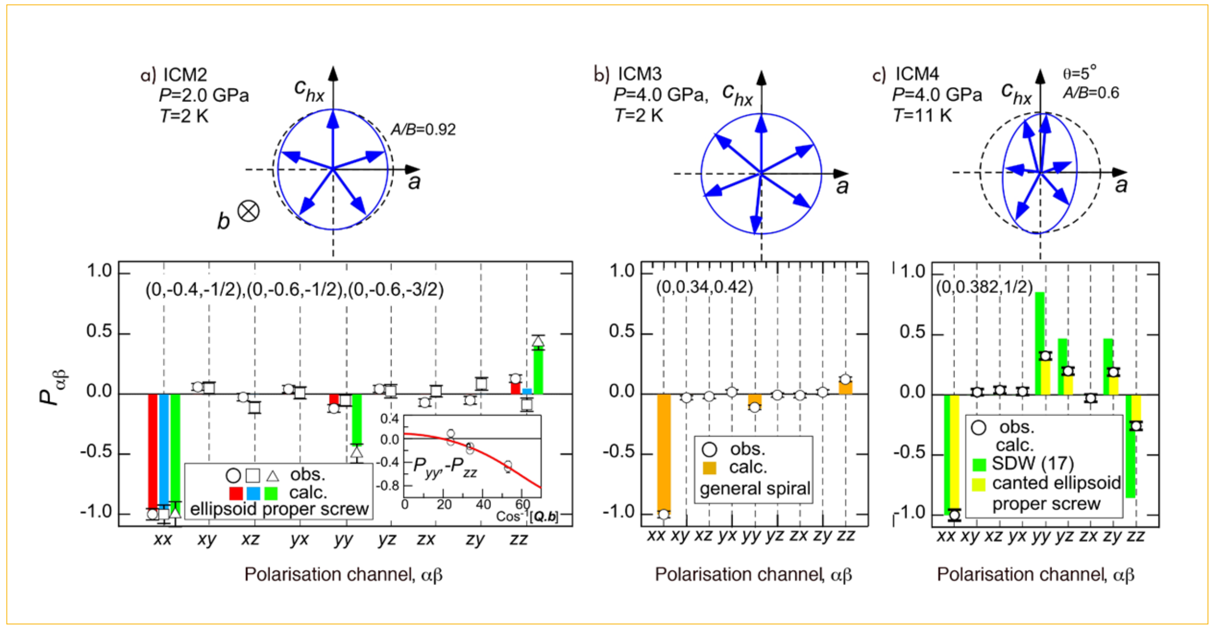

Figure 2

Summary of schematic illustrations of magnetic structure and comparison between observed and calculated values of neutron polarisation matrix elements for the pressure-induced ferroelectric phases in multiferroic $CuFeO_2$ [4].

First, we confirmed the collinear magnetic structure in the ICM1 and CM1 phases, and found precise spin-canting angles of 17±2 ° and 5±2 ° for the ICM1 and CM1 phases, respectively [4]. The magnetic structure parameters are consistent with previously reported values determined by unpolarised neutron diffraction studies. These results therefore demonstrate that SNP measurements are feasible under high-pressure conditions using a non-magnetic HAC.

Secondly, we succeeded in determining the magnetic structures of the pressure-induced ferroelectric phases in $CuFeO_2$ [4]. The magnetic structures and comparison between experimental and calculated values for the polarisation matrix elements are summarised in **figure 2**. In the case of the ICM2 phase, an ellipsoidal proper screw structure with magnetic point group 21′ was determined with an ellipsoidicity ratio of 0.92 ± 0.05. This magnetic symmetry is consistent with observed electric polarisation. We also ascertained the magnetic structure in the ICM3 phase, and found a spiral structure with spins confined to the *ac*-plane. Since the k-vector is of triclinic symmetry [k = (0, 0.34, 0.42)], this magnetic structure (termed the general spiral) possesses both a cycloidal modulation along the *c*-axis and a proper screw modulation along the *b*-axis in the ICM3 phase, which has point group 11′, allowing electric polarisation along a general direction. This study also identified the existence of a phase transition between the ICM1 and ICM4 phases in the intermediate temperature region by measuring the temperature dependence of the polarisation matrix elements at several pressures. One possible magnetic structure in the ICM4 phase is presented as a canted ellipsoidal proper screw with point group 21′.

In conclusion, the present study provides evidence that SNP measurements are viable even in high-pressure conditions [4]. It is our hope that the present technique will allow researchers to elucidate pressure-induced physical phenomena associated with complex magnetic ordering.