BL-3A/2012R-58

Uniaxial pressure effects on magnetic and crystal structural phase transitions in a frustrated magnet $CuFe_{1-x}Ga_xO_2$ (x=0.035)

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1 Introduction

A delafossite compound $CuFeO_2$ (CFO) has been extensively investigated as a model material of a triangular lattice antiferromagnet[1]. Owing to the equilateral triangular arrangement of the magnetic Fe^{3+} ions and antiferromagnetic interactions between them, this system has geometrical spin frustration. To lift the degeneracy due to the magnetic frustration, this system exhibits `spin-driven' crystal structural transitions associated with the magnetic phase transitions, at low temperatures[2,3]. Specifically, the crystal structure stretches and contracts along [110] and [1-10] directions, respectively. As a result, the original trigonal crystal structure changes into a monoclinic structure. In this report, we have added the subscript `m' to the monoclinic notation when referring to crystal axes and reciprocal indices. CFO is also known to have high sensitivity to nonmagnetic substitution for the magnetic Fe^{3+} site. While the ground state of this system is a collinear four-sublattice antiferromagnetic state, only a few percent of nonmagnetic Ga^{3+} or Al^{3+} substitution changes it into an incommensurate screw-type magnetic structure[4], which breaks inversion symmetry of the system and accounts for the spin-driven ferroelectricity[5,6].

Quite recently, we have demonstrated that the magnetic phase transitions in CFO can be controlled by application of uniaxial pressure (p), which directly affects the symmetry of the lattice[7]. We have performed neutron diffraction and synchrotron radiation x-ray diffraction measurements on CFO with uniaxial pressure up to 100 MPa applied along [1-10] direction, which is parallel to the a_m axis. As a result, we have found that the magnetic and crystal structural transition temperatures shift toward higher values with increasing p.

In the present study, we have performed synchrotron radiation x-ray diffraction measurements on $CuFe_{1-x}Ga_xO_2$ (x=0.035) with uniaxial pressure up to 100 MPa, in order to elucidate the nonmagnetic impurity effects on the strong spin-lattice coupling in this system.

2 Experiment

A single crystal of $CuFe_{1-x}Ga_xO_2$ (CFGO) with x = 0.035 of nominal composition was grown by floating zone method[8]. The crystal was cut into a rectangular shape with dimensions of 2.5×2.0×5.0 mm^3. The widest surfaces are normal to the hexagonal (110) direction.

The synchrotron radiation x-ray diffraction measurements on CFGO (x=0.035) under applied uniaxial pressure were carried out at the beamline BL-3A in Photon Factory in High Energy Accelerator Research Organization, Tsukuba, Japan. The energy of the incident x-ray was tuned to 14 keV. We have recently developed a uniaxial pressure device, which is loaded into a ^{4}He cryostat. The mechanism to apply the uniaxial pressure is essentially the same as that of the uniaxial pressure devices used in our previous neutron diffraction studies[7,9]. The direction of p was set to be parallel to the [1-10] direction, which is parallel to the a_m axis. By this experimental setup, we mainly observed the p dependence of the lattice constant b_m, which is parallel to the hexagonal [110] direction.

3 Results and Discussion

Figure 1(a) shows the temperature dependence of the diffraction intensities measured by θ-2θscans for the 220 reflection, on cooling in zero uniaxial pressure. The vertical dashed lines show the magnetic phase transition temperatures determined from magnetic susceptibility measurements. As we lower the temperature from the paramagnetic (PM) phase, the system enters a collinear incommensurate magnetic phase, which is referred to as the oblique-partially-disordered (OPD) phase, at T_{N1} = 14 K. We have detected no significant structural anomaly at T_{N1}. With further decreasing temperature, the 220 reflection splits into two peaks around 12.5 K, below which the system exhibits another collinear incommensurate magnetic phase referred to as the partially disordered (PD) phase. This is consistent with the previous x-ray diffraction study on $CuFe_{1-x}Al_xO_2$ with x=0.0155 showing that the monoclinic lattice distortion occurs at the magnetic phase transition from the OPD phase to the PD phase[10]. It should be noted that the 220 reflection actually splits into three reflections as illustrated in Fig. 1(c). These reflections are assigned as 040_m, $-6\text{-}22_m$ and $6\text{-}2\text{-}2_m$ using the monoclinic bases for each domain. Because the $-6\text{-}22_m$ and $6\text{-}2\text{-}2_m$ reflections appear at the same 2θ, there are two peaks in the θ-2θ scan profiles. The splitting of the 220 reflection also indicates that the CFGO sample was in a multi-domain state in zero uniaxial pressure. Around 7.5 K, we found another structural anomaly indicating that the degree of the monoclinic lattice distortion becomes larger. This corresponds to the magnetic phase transition from the PD phase to the ferroelectric incommensurate-magnetic (FE-ICM) phase. Although the phase transition from the PD phase to the FE-ICM phase is identified to be a first-order

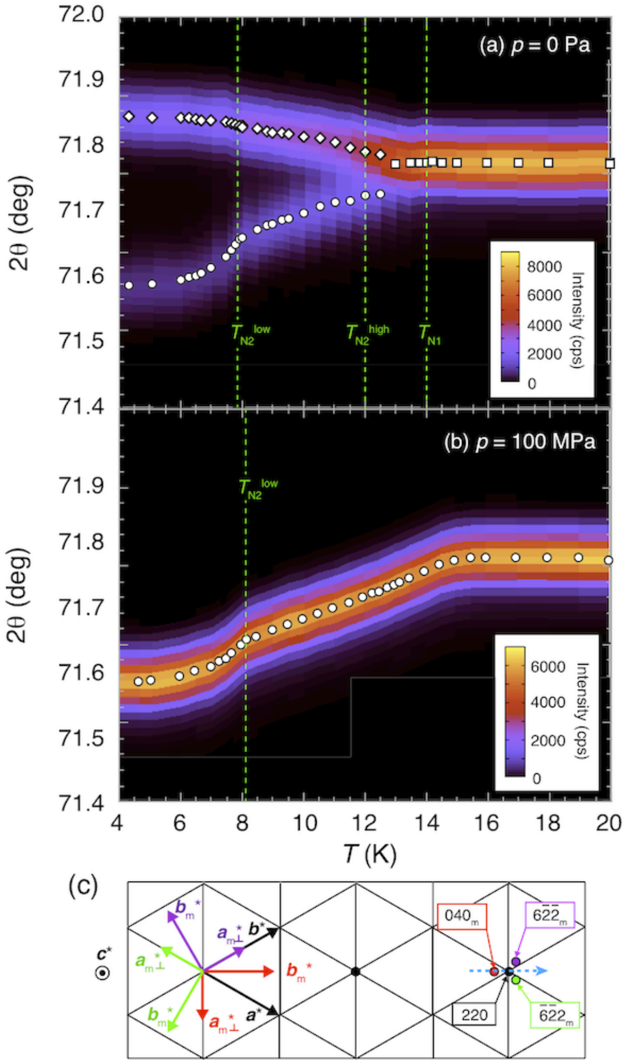

Fig. 1 [(a),(b)] Contour maps showing the temperature dependences of the x-ray diffraction intensities for the 220 reflection of CFGO (x=0.035) measured by the θ-2θ scans on cooling under (a) p = 0 Pa and (b) p = 100 MPa. (c) The reciprocal lattice map of CFGO (qualitatively) showing the splitting of 220 reflection. A horizontal dashed arrow denotes the direction of the θ-2θ scan for the 220 reflection. $a^{*}_{m\perp}$ denotes the c^{*}-plane-projection of monoclinic a axis. b_m denotes the b_m axis.

phase transition[5], we could not observe the discontinuous change in the lattice constant within the resolution of the present x-ray diffraction measurements.

In Fig. 1(b), we show the temperature dependence of the diffraction intensities measured on cooling under p=100 MPa. In contrast to the result for p=0, the splitting of the 220 reflection was not observed under p=100 MPa. This indicates that the application of p results in the single-domain state, as was demonstrated in the previous studies on CFO and CFGO with uniaxial pressure[7,9].

In Fig. 2, we show temperature variations of the lattice constant b_m under p = 0 and 100 MPa. We have found that the trigonal-to-monoclinic structural transition temperature is shifted toward higher value by applying p. We have also measured the p-dependences of b_m at fixed

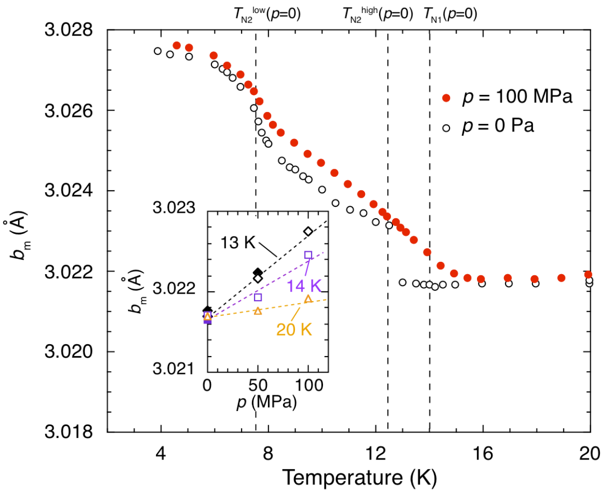

Fig. 2: Temperature dependences of monoclinic b (b_m) on cooling under p = 0 and 100 MPa in CFGO (x = 0.035). Inset shows the p-dependence of b_m measured at fixed temperatures of 13, 14 and 20 K. The open and filled symbols in the inset show the p-increasing and decreasing processes, respectively.

temperatures of 13, 14 and 20 K, revealing that the b_m linearly increases with p, as shown in the inset of Fig. 2. However, we found that the p-induced changes of b_m are relatively small as compared to those in CFO[11]. This implies that the structural instability associated with the trigonal-to-monoclinic structural transition is reduced by the nonmagnetic substitution. More detailed discussions including comparisons with the results of undoped CFO will appear elsewhere[11].

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