Field-induced Lattice Staircase in a Frustrated Antiferromagnet $CuFeO_2$

Let us consider three spins placed on the vertices of a triangle with a nearest neighbor antiferromagnetic interaction, J, which is a system depicted in Fig. 1(a). Two of the three spins form a state of lowest energy when their moments are aligned anti-parallel with each other, but the third one cannot find a stable alignment. This triangular coordination is called "geometrical spin frustration." Although theory predicts that the 2D Ising triangular lattice antiferromagnet (TLA) does not exhibit magnetic order at any finite temperature, real materials with the triangular lattice structure often achieve a magnetic ordering by distorting the lattice symmetry, as illustrated in Fig. 1(b).

$CuFeO_2$ is an archetype TLA. It has the delafossite structure (space group $R\bar{3}m$ at room temperature, displayed in Fig. 1(c). The magnetic structure in zero field is the collinear four-sublattice (4SL) ↑↑↓↓ structure below 11 K [see Fig. 3(b)] [1]. This ground state is realized by distorting the lattice as described above [2,3].

We have performed synchrotron X-ray Bragg diffraction measurements under very high magnetic fields at beamline **BL19LXU**, using a single crystal of $CuFeO_2$ [4]. A pulsed field magnet, up to 40 T, has been developed in collaboration with the ISSP, University of Tokyo and KYOKYGEN, Osaka University [5].

Figure 2 shows the results obtained from our measurements. The lattice constant b_{orth} [with orthorhombic notation shown in Fig. 3(a)] is seen to exhibit stepwise changes with increasing magnetic field in coincidence with multistep magnetization changes. We see that lattice changes scale nicely with magnetization changes.

We discuss the origin of the stepwise changes in the lattice constant b. The nearest neighbor exchange interaction in the basal plane of $CuFeO_2$ is a sum of the direct exchange interaction, which is assumed to be ferromagnetic, and the 90° antiferromagnetic superexchange interaction through an O^{2-} ion [1]. The experimental evidence that the b_{orth} axis elongates at low temperature in zero field [2], implies that the direct exchange interaction diminishes and the antiferromagnetic interaction dominates. In the 4SL phase [see Fig. 3(b)], all spins connected by the nearest neighbor J'_1 bonds along the b_{orth} axis are antiferromagnetically aligned; in other words, the spin arrangement in the J'_1 row is ↑↓↑↓. As shown in Fig. 3(c), in the five-sublattice phase ($13 \leq H \leq 20$ T), one of the five bonds along the J'_1 row in the unit cell accommodates two parallel spins at the ends and costs excess energy. In order to lower this excess in the exchange energy, b_{orth} axis contracts to resume the ferromagnetic direct exchange interaction. Similarly, in the three-sublattice phase ($20 \leq H \leq 34$ T)

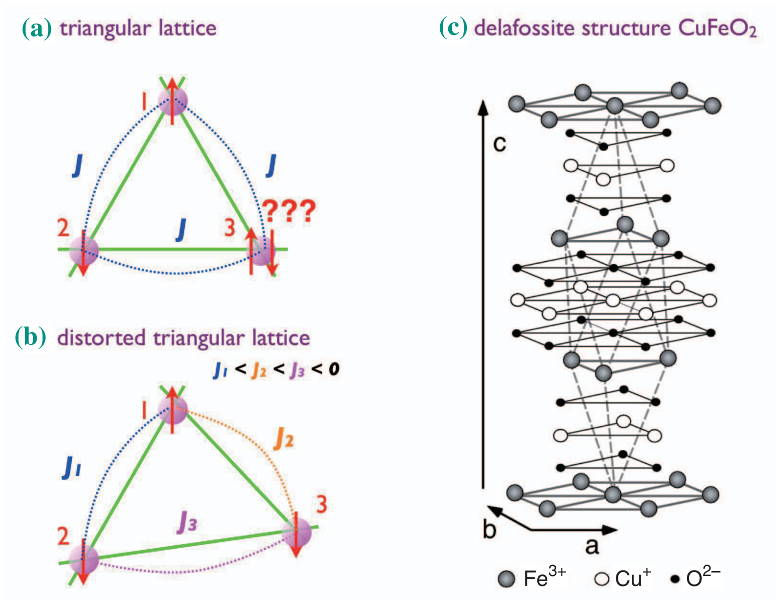

Fig. 1. Three spins placed at the vertices of **(a)** a triangle and **(b)** a distorted triangle resulting in difference in the exchange interactions. **(c)** Delafossite crystal structure of $CuFeO_2$.

[see Fig. 3(d)], one of the three bonds along the J'_1 row in the unit cell accommodates two parallel spins at the ends, so that an additional lattice contraction along the b_{orth} is needed. In these field-induced lattice changes, we expect that a uniform distortion will occur rather than a local distortion at the "wrong" bonds. This explains qualitatively the stepwise lattice contractions in b_{orth} associated with the multistep magnetization changes in $CuFeO_2$.

Our unique apparatus, combining strong magnetic field and synchrotron X-rays, opens a new field in materials science research. Many interesting phenomena are expected to occur in magnetic materials as well as superconductors at high magnetic fields. The duration of the pulsed magnetic field is 5 - 25 milli-seconds and consequently, we need strong X-rays to obtain reliable data. BL19LXU is best suited for this purpose, since the World's strongest high energy X-rays are available at this beamline.

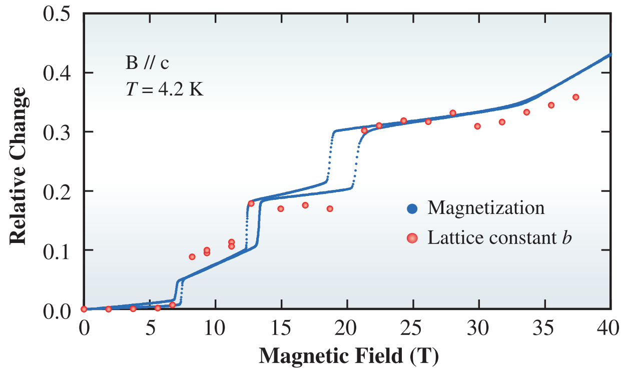

Fig. 2. Magnetic field dependence of the relative changes of the magnetization normalized with the saturation moment 5 μB of Fe^{3+} (blue circles) and the fractional change of the lattice constant (red circles), $\alpha[b_{orth}(B = 0\ T) - b_{orth}(B)] / b_{orth}(B = 0\ T)$, where α is a numerical factor. The measurements were done at $T = 4.2$ K.

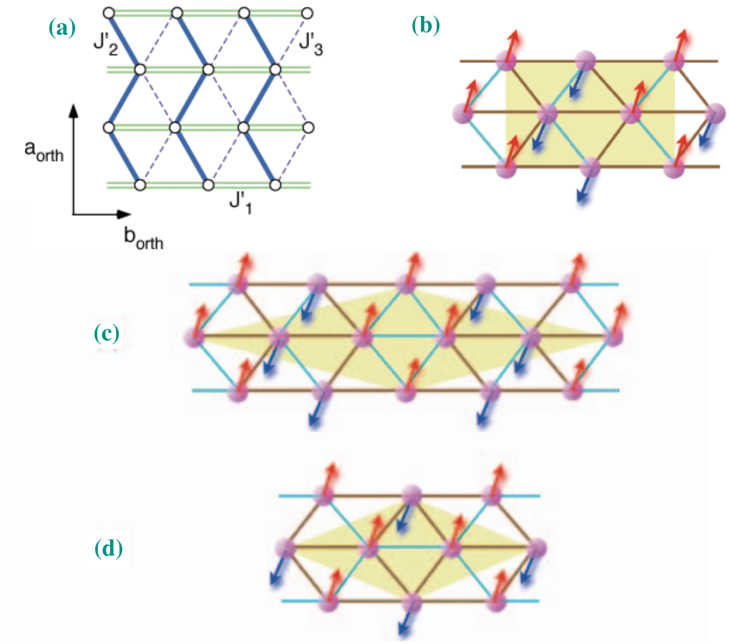

Fig. 3. **(a)** The exchange interaction paths in the basal plane. The spin structures in the basal plane of **(b)** the four-sublattice ↑↑↓↓ state, **(c)** the five-sublattice ↑↑↑↓↓ state and **(d)** the three-sublattice ↑↑↓ state. Blue and brown bonds denote ferromagnetic and antiferromagnetic arrangements, respectively. The magnetic unit cells in the respective phases are marked with yellow.

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References

[1] M. Mekata *et al.*: J. Phys. Soc. Jpn. **62** (1993) 4474.
[2] N. Terada *et al.*: J. Phys. Soc. Jpn. **75** (2006) 023602.
[3] N. Terada *et al.*: J. Phys. Soc. Jpn. **75** (2006) 113702.
[4] *N. Terada, Y. Narumi, K. Katsumata, U. Staub, K. Kindo, M. Hagiwara, Y. Tanaka, A. Kikkawa, H. Toyokawa, T. Fukui, R. Kanmuri, T. Ishikawa and H. Kitamura: Phys. Rev. B* ***74*** *(2006) 180404(R).*
[5] Y. Narumi *et al.*: J. Synchrotron Rad. **13** (2006) 271.