Low Temperature structural phase transition in $BaCuSi_2O_6$

E. C. Samulon[1], Z. Islam[2], S. E. Sebastian[1], P. B. Brooks[3], M. K. McCourt Jr.[3], J. Ilavsky[2], I. R. Fisher[1]
[1] *Geballe Laboratory for Advanced Materials and Department of Applied Physics, Stanford University, Stanford, CA 94305*
[2] *Advanced Photon Source, Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, IL 60439 and*
[3] *Department of Physics, Stanford University, Stanford, CA 94305*

$BaCuSi_2O_6$ is a quasi-2D compound composed of $Cu_2Si_4O_{12}$ layers in which pairs of Cu^{2+} (s $=\frac{1}{2}$) ions form vertical spin dimers [1, 2]. In zero magnetic field, the material has a singlet ground state, with a significant gap to the triplet states [3]. Magnetic fields exceeding $H_{c1} \sim$ 23 T close the spin gap, resulting in long range canted antiferromagnetic order at low temperature [4]. Here we report results of x-ray scattering experiments performed at the Advanced Photon Source that reveal a weak structural phase transition at approximately 100 K [5]. We discuss implications for the high-field ordered state.

At room temperature, charge Bragg peaks of $BaCuSi_2O_6$ were observed consistent with the $I4_1/acd$ tetragonal structure previously reported [2]. However, at 16 K, the even-order (H 0 0) peaks were found to have split along the longitudinal direction (Fig. 1). A careful set of measurements revealed that the splitting was consistent with transformation twins of an orthorhombic (or weakly monoclinic) lattice. The structural phase transition was accompanied by an incommensurate (IC) lattice modulation characterized by a reduced wavevector $\mathbf{q}_{IC} = (0, 0.129 \pm 0.001, 0)$, referred to the orthorhombic latttice, and its 2^{nd} and 3^{rd} harmonics [5]. The temperature dependence of the IC peaks indicated a first order phase transition with $T_c \sim 103$K for warming cycles (Fig. 2). Further experiments are in progress to obtain a model

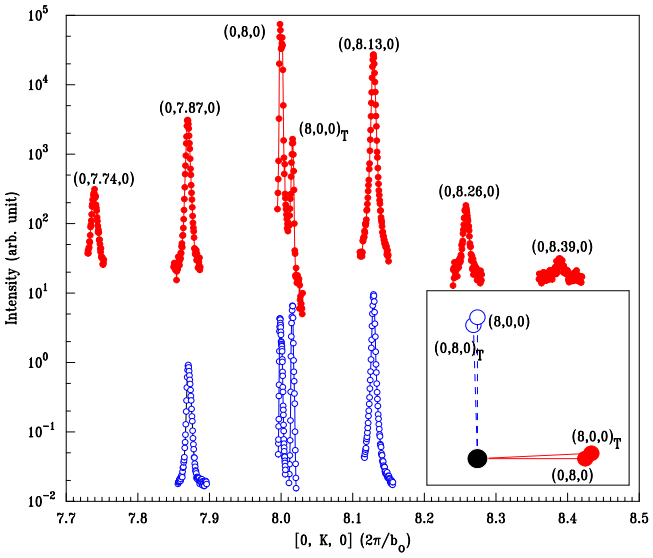

FIG. 1: Reciprocal lattice scans showing the orthorhombic splitting of the (8 0 0) Bragg peak, as well as the splitting of the transformation twin, at 16 K. In addition, associated IC peaks and harmonics are shown, multiplied by $\sim 10^4$ to appear on the same scale as the central peaks [5].

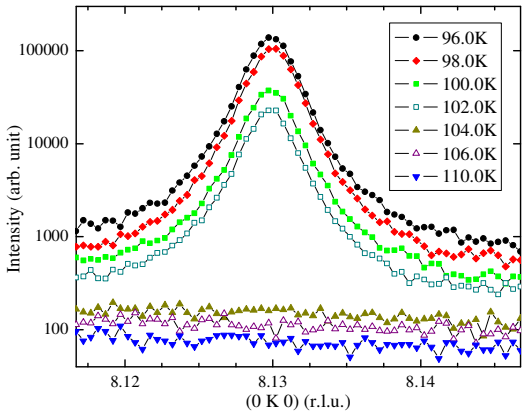

FIG. 2: Temperature dependence of the superlattice peak (0 8.129 0) referenced to the low temperature orthorhombic structure, on heating between 95 K and 110 K. Scans are displaced vertically for clarity.

for the IC modulation, which is likely associated with rotation of SiO_4 tetrahedra in the $Cu_2Si_4O_{12}$ layers.

These observations imply that the spin Hamiltonian describing this system is more complex than originally thought [4]. The incommensurate modulation will likely lead to inequivalent dimers, and the lower crystal symmetry means that anisotropic effects (including Dzyaloshinski-Moriya antisymmetric exchange and anisotropic exchange constants J') are not precluded. However, modifications to the spin Hamiltonian must be subtle given that BEC critical scaling exponents are observed for temperatures down to 30 mK [6].

The use of the APS is supported by the U.S. DOE, Office of Science, under Contract No. W-31-109-ENG-38. This work is supported by the National Science Foundation, DMR-0134613.

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