

X-ray Scattering Investigation of Magnetism and Charge-Density Waves in Insulating and Superconducting Copper Oxides

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Introduction

We performed resonant and nonresonant x-ray diffraction experiments on single-crystal samples of copper-oxide compounds to search for unusual features that could help us understand the complex electronic state that is responsible for high- T_C superconductivity. Specifically, we studied two systems: $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ (an antiferromagnetic insulator) and $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ (a high- T_C superconductor with $T_C = 30\text{K}$). With regard to the former, we succeeded in measuring the magnetic x-ray scattering signal of the ordered Cu moments. The good signal-to-background ratio of this measurement helped calibrate the sensitivity of our experimental setup for further measurements of small-amplitude spin-density waves (SDWs) and charge-density waves (CDWs). With regard to the $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ compound, we performed searches for CDW order [1] and for a time-reversal, symmetry-breaking phase that was predicted theoretically [2]

Methods, Materials, and Results

It is extremely challenging to detect the scattering from small magnetic moments in low-dimensional systems with x-rays. Notable work has been accomplished by J.P. Hill and coworkers, who succeeded in observing copper magnetism in $\text{PrBa}_2\text{Cu}_3\text{O}_{6.92}$ at the European Synchrotron Radiation Facility [3]. However, the observed count rates were small: about 6 counts per second. By using high-brilliance beamline 4-ID at the APS, we recently made another step forward. Magnetic Bragg peaks due to the ordering of Cu moments were observed in $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ (a prototypical $S = 1/2$ 2-D square-lattice antiferromagnet). The data were taken with the incident photons tuned to the Cu resonance. The resulting count rate was as high as 300 counts per second, as shown in Fig. 1. This large signal was on a par with the signal achievable with neutron scattering on a large single crystal. Since the x-ray beam spot size was about $150 \times 150 \mu\text{m}$, small single crystals were sufficient for these studies. This work has opened the door for further studies of small-amplitude SDWs and CDWs (such as stripes) on small samples by using x-rays.

We next studied a high-quality, single-crystal sample of $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ grown by the traveling-solvent floating zone technique. (The mosaic measured with x-rays was as

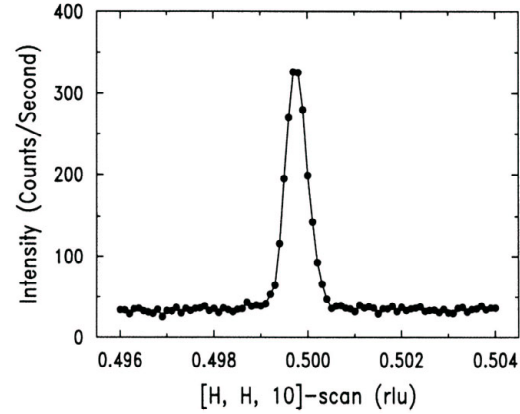


FIG. 1. Magnetic x-ray scattering from ordered Cu moments in $\text{Sr}_2\text{CuO}_2\text{Cl}_2$. The scan was taken at $T = 8.4\text{K}$ through a 3D antiferromagnetic Bragg position.

narrow as 0.03° FWHM (full width at half-maximum [FWHM]). This composition was chosen since it is known that SDW order exists in the sample, as demonstrated by neutron scattering experiments [4]. It has been believed that CDW order should accompany SDW order, especially if a “stripe phase” is responsible for the spin ordering. We performed extensive searches for CDW order in this system, using both nonresonant x-rays and photons tuned to the Cu resonance. No peaks were found at the reciprocal space positions that would be expected for the CDW order that corresponds to a stripe phase. In addition, an exhaustive search was performed to observe signatures of a time-reversal, symmetry-breaking phase by using resonant x-ray diffraction. Again, no peaks were found, even with the high sensitivity that can be achieved on the 4-ID beamline.

On the other hand, in our $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ sample, we succeeded in observing new superlattice peaks at (110) and (330) (labeled in orthorhombic notation). A sharp Bragg peak at these locations would indicate that the crystal has the low-temperature tetragonal (LTT) structure. This is unexpected, however, since it is known that the $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ composition is orthorhombic at low temperatures. The peak that we observed near (330) was broad and weak, as shown in Fig. 2. In particular, the peak was broadest along the (1 -1 0) direction. This

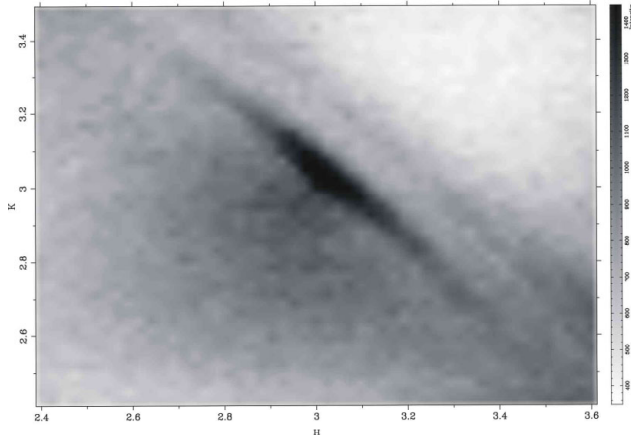


FIG. 2. Intensity contour plot in the vicinity of the low-temperature tetragonal superlattice peak at (330) in $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$. The scan was taken at $T = 11.5\text{K}$.

intensity may indicate the presence of dynamic fluctuations toward the LTT structure for this composition. Another possibility may be that this scattering originated from the twin boundaries of the predominant low-temperature orthorhombic (LTO) phase. So far, CDW order has been observed only in La_2CuO_4 -based materials with the LTT structure. Here, the weakness of the LTT peak in $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ may explain the lack of observable CDW peaks.

Acknowledgments

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