Observations of Lattice Modulations in an Underdoped $YBa_2Cu_3O_{6+x}$ Superconductor by Using X-ray Diffraction

Z. Islam,[1] S. K. Sinha,[2] D. Haskel,[1] J. C. Lang,[1] G. Srajer,[1]
B. W. Veal,[3] D. R. Haeffner,[1] H. A. Mook[4]
[1]Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.
[2]Department of Physics, University of California, San Diego, CA, U.S.A.
[3]Materials Science Division, Argonne National Laboratory, Argonne, IL, U.S.A.
[4]Oak Ridge National Laboratory, Oak Ridge, TN, U.S.A.

Introduction

There has been much speculation about the microscopic nature of the "pseudogap" phase that appears in the underdoped cuprates in their normal state [1, 2]. One possibility is that this phase is due to ordering of a kind (e.g., "spin/charge stripes") that competes with superconductivity. In underdoped $YBa_2Cu_3O_{6+x}$ compounds, Mook and collaborators [3] reported the existence of incommensurate spin excitations at wave vectors of (±0.1, 0, 0) from the (1/2, 1/2, 0) reciprocal-space point, which they associated with dynamic stripe fluctuations. No static spin or charge density waves were found with neutrons, although Mook and Dogan [4] reported anomalies in phonons of wave vector (±0.2, 0, 0) or the expected wave vector for the corresponding dynamic charge stripes. We have used high-energy synchrotron x-rays to search for such charge stripes in underdoped $YBa_2Cu_3O_{6.63}$ (YBCO) since x-ray scattering integrates over the energies of the fluctuations at the wave vectors looked at. We note that the diffraction from charge stripes is primarily from the lattice distortions associated with them.

Methods and Materials

X-ray scattering experiments were performed at the APS. A set of well-annealed, self-flux-grown crystals was characterized on the Synchrotron Radiation Instrumentation Collabortive Access Team (SRI-CAT) beamline 1-ID by using a Weissenberg camera with 65-keV x-rays. A rectangular, twinned crystal (~1000 × 300 × 70 μm^3) that showed no obvious signs of diffraction peaks from extraneous phases was chosen from the set. The magnetization measurements of this sample (annealed at ~300K for several weeks) revealed T_c to be 60K with a transition width of ~0.5K, suggesting a high degree of compositional homogeneity in the bulk of the sample. Most of the present work was carried out on the SRI-CAT 4-ID beamline by using 36-keV x-rays. The sample was cooled in a closed-cycle He refrigerator. Y-fluorescence was carefully monitored to normalize to the same diffracting volume at every temperature.

Results

Figure 1 shows reciprocal lattice scans (i.e., [H, 0, 0] scans) between (4, 0, 0) and (5, 0, 0) Bragg peaks, respectively, at two different temperatures. Two broad superlattice peaks associated with a modulation vector of $\mathbf{q}_0$ = (~2/5, 0, 0) are clearly observed at both temperatures. There appears to be *no* significant scattering above the diffuse tails near the expected incommensurate charge-fluctuation peaks at (~4.2, 0, 0) and (~4.8, 0, 0) [3]. Extensive searches did not find peaks corresponding to (~0.2, 0, 0), whereas $\mathbf{q}_0$ peaks were observed with intensities on the order of ~500 to 2500 counts/s at 14.3K, some 10^6 to 10^7 orders of magnitude weaker than those of the Bragg peaks. They are due to local lattice distortions primarily along the **a** axis, which, at high temperatures, have been associated with lattice relaxation due to oxygen-vacancy ordering. Figure 2 (top right panel) shows a 2-D mesh revealing the extent of the diffuse scattering in the [H, K, 0] plane. The anisotropy of the widths along **a*** and **b***, respectively, is consistent with the quasi-1-D nature of the underlying distortions.

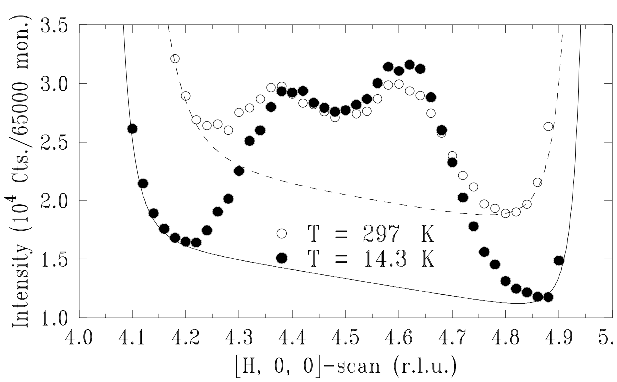

FIG. 1. Comparison of diffuse scattering at room temperature and 14.3K, showing a clear enhancement at low temperature. Solid lines show modeling of the background (see text).

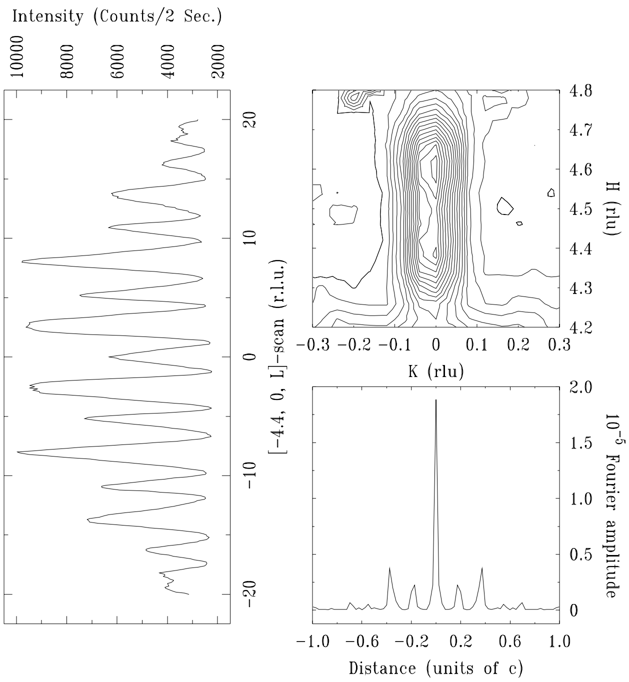

FIG. 2. Top right: 2-D scan showing the extent of diffuse scattering in the [H, K, 0] zone. Left: The modulation of [-4.4, 0, 0] diffuse peak along **c***. Lower right: The Fourier transform showing distances between planes that are correlated. All the data were collected at 14.3K.*

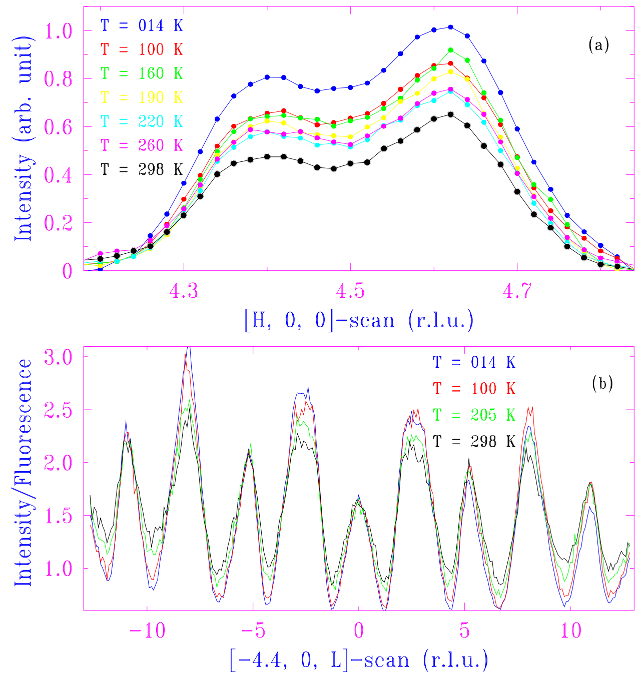

FIG. 3. (a) Diffuse scattering at various temperatures. The background, including the thermal diffuse scattering (TDS), has been removed from the data. (b) L-modulations of (-4.4, 0, 0) diffuse peak at selected temperatures. The intensity modulations are clearly enhanced as T is lowered.

The diffuse peaks after subtracting background (modeled by using two Lorentzian tails and a linear term [Fig. 1]) at various temperatures are shown in Fig. 3(a). The diffuse scattering clearly increases at low *T*. Figure 4(a) summarizes the *T*-dependence of the diffuse scattering. As shown, the intensity gradually increases upon lowering *T* and becomes nearly constant in the 220-260K range. Below T_1 of ~220K, however, the intensity starts to rise beyond the extrapolation (tildes) of the 220-260K intensity, until T_2 is ~160K, below which it seems to level off. Then, as the superconducting state is approached, the intensity gradually starts to rise again and appears to saturate in that phase. The inset shows the intensity after subtracting the 220-260K value.

By scanning along the **c*** axis in reciprocal space through the peaks at 4 + $\mathbf{q}_0$, we see strong modulations as a function of Q_z (see Fig. 2). A direct Fourier transform of this intensity modulation (bottom panel in Fig. 2) reveals two planar distances, z_1 = 0.362 ±0.008 *c* and z_2 = 0.187 ±0.008 *c*, respectively, that are correlated. The z_1 corresponds to the distance between the CuO_2 and CuO_x-chain planes, whereas z_2 is consistent with both the CuO_2-O_{apical} and the chain-Ba distances, respectively. [O_{apical} is the oxygen between Cu(1) and Cu(2) atoms, respectively, along the **c** axis.] Figure 3(b) shows the behavior of the modulations at various temperatures, and Fig. 4(b) displays the temperature-dependence of the correlations. As shown, the Fourier amplitudes of z_1 and z_2 clearly grow at low *T* and appear to saturate in the superconducting state.

Discussion

In summary, our experiments showed that the diffuse peaks corresponding to $\mathbf{q}_0$ = (~2/5, 0, 0) originate from mutually coupled, primarily longitudinal atomic displacements ($\delta\mathbf{u}\cdot\mathbf{a}$) of the CuO_2, CuO_x-chain, and O_{apical} and/or Ba, respectively. The primary result of our study is the significant increase of this diffuse scattering below ~220K, which roughly corresponds to the temperature of entry into the “pseudogap” phase in this material. Unless the activated behavior becomes entirely different in the low-temperature regime, there is no possibility of observing low-temperature changes associated with spontaneous oxygen ordering, since, at this temperature, the projected relaxation time is more than 1000 years. We propose that the lattice modulations observed in underdoped YBCO become strongly correlated with instabilities (i.e., charge fluctuations) in the CuO_2 planes

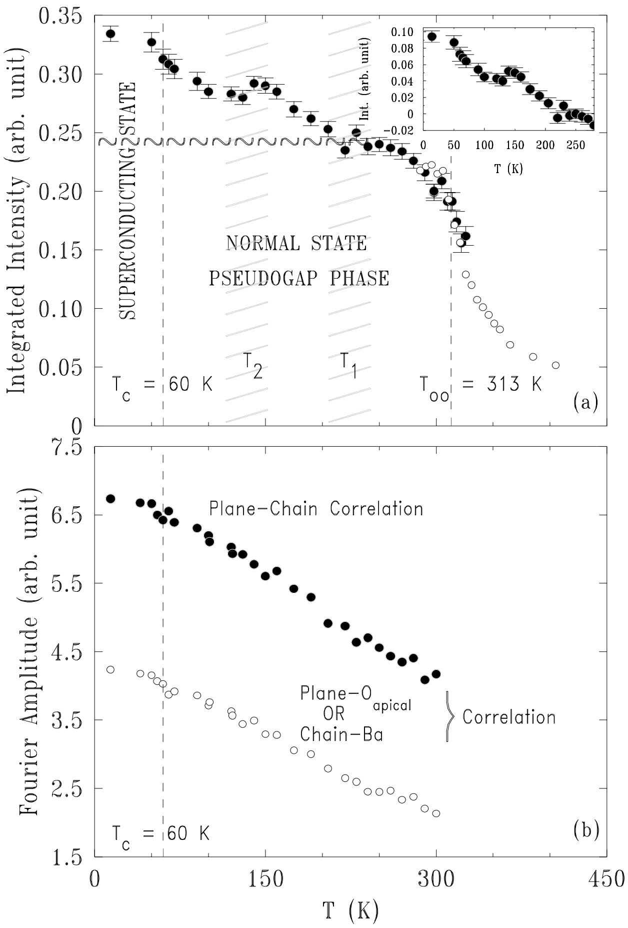

FIG. 4. (a) T-dependence of the diffuse scattering at (4.4, 0, 0). (○) are data from Ref. 5 normalized to overlap with our data for 300-328K. (~) depict low-T extrapolation of the intensity between ~220 and 260K. Hatched areas identify T regions where breaks in the intensity are observed. Inset: "Order parameter" of the charge stripes. (b) T-dependence of the correlations.

that grow into the pseudogap phase. The charge fluctuations or stripes use oxygen-ordered superstructure as a spatial template. The Fig. 4 inset shows how the "order parameter" of such a stripe phase may evolve as the pseudogap phase and the superconducting state, respectively, are entered. In this connection, we note that recent elastic neutron diffraction work on a single crystal YBCO sample [6] showed a peak at $\mathbf{q}_{spin}$ = (~0.2, 0, 0). The absence of this peak in the x-ray scattering leads us to speculate that, in fact, this peak results from the magnetic stripe corresponding to our observed scattering at twice this wave vector, $\mathbf{q}_0 = 2\mathbf{q}_{spin}$, and it is also commensurate with the oxygen ordering in the chain planes.

Acknowledgments

Use of the APS was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. We thank J. Almer for his help with the Weissenberg camera. We have benefitted from discussions with S. C. Moss, D. de Fontaine, and A. Paulikas.

References

[1] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
[2] B. Batlogg and C. M. Varma, Phys. World **13(**2) (2000).
[3] H. A. Mook et al., Nature **395**, 145 (1998); ibid. **404**, 729 (2000).
[4] H. A. Mook and F. Dogan, Nature **401**, 145 (1999).
[5] N. H. Andersen et al., Physica C **317**-**318**, 259 (1999).
[6] M. Arai et al., Intl. J. Mod. Phys. B **14**, 3312 (2000).