Investigation of Charge Ordering in $YBa_2Cu_3O_{6+x}$

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Introduction

The formation of stripes of spin and charge degrees of freedom in high-temperature superconductors has attracted considerable scientific interest in the last years. In the case of $YBa_2Cu_3O_{6+x}$, the question whether static stripes do exist in this compound and how they affect superconductivity has not been given a definite answer yet. The existence of a dynamic stripe phase is strongly indicated by charge fluctuations observed in phonon measurements conducted on $YBa_2Cu_3O_{6+x}$ by Mook and Dogan [1], while no static spin- or charge-stripes have been found in these compounds with neutron diffraction experiments. Furthermore, a recent x-ray scattering study of underdoped $YBa_2Cu_3O_{6+x}$ [2] has revealed diffuse features, whose temperature dependence was reported to exhibit an anomaly around the "pseudogap" temperature. This anomaly was interpreted as a signature of electronic stripe formation. In this system, however, superstructures due to oxygen ordering with wave vectors depending sensitively on the oxygen content are also observed [3]. As both phenomena are associated with lattice distortions and are thus expected to be intimately coupled, it is difficult to establish which features of the x-ray data originate in short-range oxygen-vacancy ordering, and which can be attributed to electronic stripe ordering or fluctuations.

In order to give an answer to this question, we have investigated the x-ray diffuse scattering features of optimally doped $YBa_2Cu_3O_{6.92}$ and underdoped $YBa_2Cu_4O_8$ single crystals. The latter compound does not contain any oxygen vacancies, i.e., all oxygen sites along the CuO-chains are full, and therefore any diffuse scattering observed there would be due to electronic stripe formation. Prompted by reports of large isotope effects on various physical properties of high-temperature superconductors [4], we have in addition investigated an optimally doped $YBa_2Cu_3O_{6.92}$ crystal, in which the usual ^{16}O oxygen isotope had been replaced by ^{18}O.

Methods and Materials

The experiment was conducted at the high-energy wiggler beamline 11-ID-C of the Advanced Photon Source at the Argonne National Laboratory. The energy of the x-ray beam was equal to 115 keV. The beamline is optimized for providing as much flux as possible. This is achieved by locating the Si-(3 1 1) monochromator crystal almost at the 1:1 position between beam source (wiggler) and sample, so that the entire 10 mm width of the monochromator is used for focusing on the sample [5]. The investigated samples were mounted on a closed-cycle Displex cryostat allowing temperatures as low as 15K. A Ge detector was used for measuring the intensity of the scattered beam.

The optimally doped $YBa_2Cu_3O_{6.92}$ compound has a superconducting transition temperature of T_c=92.7K. It crystallizes in the orthorhombic *Pmmm* space group, with lattice constants of a=3.8158(1) Å, b=3.8822(1) Å, and c=11.6737(3) Å at T=270 K [6]. $YBa_2Cu_4O_8$ on the other hand has a T_c of 81K and crystallizes in *Ammm* symmetry with lattice parameters: a=3.8410(3) Å, b=3.8720(3) Å, and c=27.231(2) Å [7]. The $YBa_2Cu_3O_{6.92}$ samples (one containing ^{16}O and one ^{18}O) had a crystal volume of approximately 2×2×0.4 mm^3 and were both fully detwinned, while the $YBa_2Cu_4O_8$ sample was approximately 0.5×0.8×0.1 mm^3 large and twinned.

Results

The diffuse scattering intensity of $YBa_2Cu_3O_{6.92}$ was measured along *h* and along *l*, for both low and high temperatures. The diffuse features observed are ~5 orders of magnitude weaker than the main Bragg reflections. The intensity is peaked at *h*=0.25 along the *h*-direction, suggesting a four-unit cell periodicity. The periodicity of the observed modulation along *l* is characteristic of interatomic distances within the unit cell. The integrated intensity of the superstructure peaks decreases smoothly with increasing temperature, with no anomalies observed at any temperatures associated with the onset of electronic instabilities.

Two-dimensional plots of the scattering intensity of $YBa_2Cu_3O_{6.92}$ (^{16}O / ^{18}O – rich) and $YBa_2Cu_4O_8$ are shown in Figure 1. The diffuse scattering patterns of the ^{16}O- and ^{18}O- $YBa_2Cu_3O_{6.92}$ compounds are virtually identical, as Fig. 1a,b demonstrate. While the four-unit cell superstructure of $YBa_2Cu_3O_{6.92}$ is clearly observed in Fig. 1a,b, absolutely no diffuse intensity is observed for $YBa_2Cu_4O_8$, at least down to the intensity level of the diffuse peaks in the optimally doped compounds (Fig. 1c).

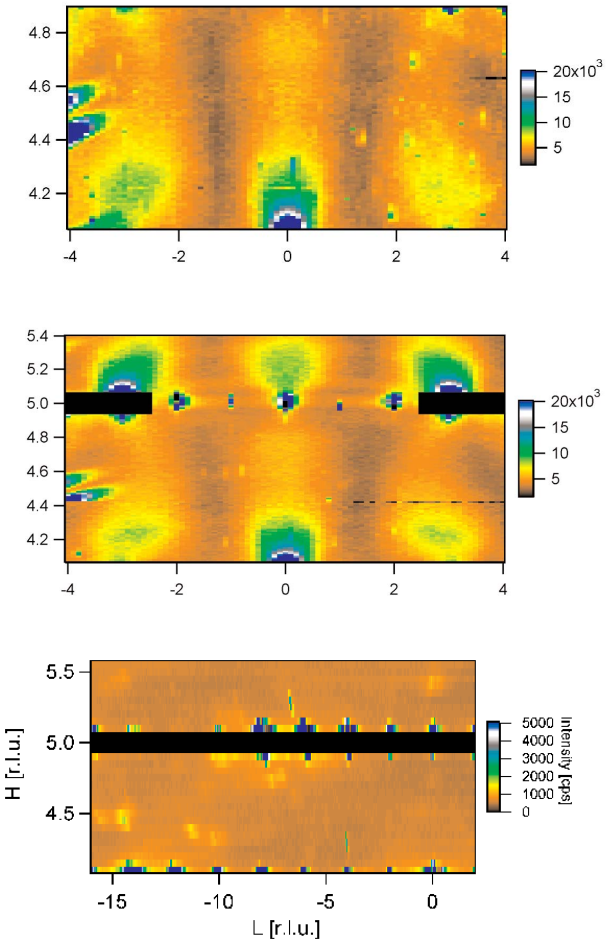

FIG. 1. Contour plots of the diffuse intensity in the (h 0 l) plane of ^{16}O-rich (a) and ^{18}O-rich (b) $YBa_2Cu_3O_{6.92}$, as well of $YBa_2Cu_4O_8$ (c), at T=20K.

Discussion

The total absence of any diffuse features in the scattering pattern of the $YBa_2Cu_4O_8$ compound is a strong indication that the origin of the superstructures observed in the optimally doped samples is related to the ordering of the oxygen vacancies and not to spontaneous electronic stripes being formed in the CuO_2 planes. Since $YBa_2Cu_4O_8$ has all its oxygen sites along the CuO chains occupied and thus does not contain any oxygen vacancies, the superstructures whose origin lies in a periodic ordering of full and empty CuO chains are expected to be absent in this compound, allowing any diffuse signal related to stripe formation to be observed. Yet, no such signal is seen. This means that the diffuse features that are related to stripe order, if existent, must be of very weak intensity, and therefore the easily observable superstructures of the optimally doped compounds are unlikely to be of such origin. Instead, it seems that the oxygen vacancies of the optimally doped compounds are the ones that give rise to the observed diffuse features – a sequence of three full and one empty CuO chain, that is a so-called 'ortho-IV' oxygen-ordered phase, can well be used as a basis for a model explaining the observed pattern.

The persistence of the diffuse scattering reflections of $YBa_2Cu_3O_{6.92}$ up to temperatures well above room temperature provides an additional indication that their origin is not related to electronic stripe formation. Charge density waves or stripe correlations would be expected to fade away already at much lower temperatures

Finally, the fact that the diffuse patterns of the ^{16}O- and the ^{18}O-rich optimally doped compounds are completely identical to each other, suggests that the isotope effects that have been observed in previous works cannot be associated with any variations in oxygen short-range order.

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