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Synchrotron x-ray diffraction study of a charge stripe order in $^{1/8}$-doped La$_{1.875}$Ba$_{0.125}$-$x$Sr$_x$CuO$_4$

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Lattice distortions associated with charge stripe order in $^{1/8}$-doped La$_{1.875}$Ba$_{0.125}$-$x$Sr$_x$CuO$_4$ are studied using synchrotron x-ray diffraction for $x=0.05$ and $x=0.075$. The propagation wave-vector and charge order correlation lengths are determined with a high accuracy, revealing that the oblique charge stripes in orthorhombic $x=0.075$ crystal are more disordered than the aligned stripes in tetragonal $x=0.05$ crystal. The twofold periodicity of lattice modulations along the $c$ axis is explained by long-range Coulomb interactions between holes on neighboring CuO$_2$ planes.

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The interplay between spin and charge correlations in hole-doped CuO$_2$ planes is widely believed to be related to the mechanisms of high-$T_c$ superconductivity. In La$_{2-x}$Ba$_x$CuO$_4$, which is a prototypical high-$T_c$ superconductor, anomalous suppression of superconductivity has been observed at around a specific hole concentration of $x=\frac{1}{8}$, where the low-temperature tetragonal (LTT) crystal phase ($P4_2/nmc$ symmetry) occurs.1,2 Tranquada et al. have found the incommensurate spin and charge orders in the LTT phase of La$_{1.6-x}$Nd$_x$Sr$_x$CuO$_4$ (LNSCO) with $x=0.12$.3−5 The results revealed that a strong relation exists between spin/charge ordering, crystal structure, and the suppression of high-$T_c$ superconductivity. Based on the stripe model,3,6 these relationships can be explained by the pinning of dynamical charge stripe correlations by lattice potentials, resulting in the strong suppression of superconductivity. Recently, a systematic neutron-scattering study of the incommensurate spin/charge order in La$_{1.875}$Ba$_{0.125}$-$x$Sr$_x$CuO$_4$ (LBSCO) with $0.05\leq x \leq 0.085$ has confirmed that charge ordering only occurs in LTT and LTLO (low-temperature less orthorhombic, $Pccn$ symmetry) phases and competes with superconductivity, whereas the robustness of magnetic order depends weakly on crystal structure and $T_c$ suppression compared to charge order.7 Hence an understanding of the microscopic nature of charge order is important for clarifying the relationship between charge correlation and superconductivity.

Although charge order is observed as lattice distortions in neutron scattering, x-ray diffraction can, in principle, directly detect charge distributions, which would provide direct evidence of charge order. A recent synchrotron x-ray-diffraction study of LNSCO at $x=0.12$ has determined the propagation wave vector of the incommensurate charge order, $Q_{ch}=(\pm 2\varepsilon 0 \frac{1}{2})$ with $\varepsilon=0.118$ r.l.u. (reciprocal lattice unit).8 Although the superlattice observed in the x-ray-diffraction study was mainly the result of lattice distortions, precise determination of the wave vector $Q_{ch}$ revealed that the lattice distortions are caused by the formation of charge stripe order. In LBSCO systems, neutron-scattering measurements have found that the in-plane component of $Q_{ch}$ for $x=0.05$ in the LTT structure is different from that for $x=0.075$ in the LTLO structure, which suggests a strong relationship between stripe pattern and crystal symmetry.9 However, detailed information about the three-dimensional correlation of the charge order is not available yet because no synchrotron x-ray-diffraction measurements have been carried out in LBSCO systems.

Synchrotron x-ray-diffraction measurements of LBSCO with $x=0.05$ and $x=0.075$ are conducted to study the nature of charge stripe order in detail, and to examine the relationship between charge correlation and crystal structure.

X-ray-diffraction experiments were performed at the Crystal Structure Analysis Beam Line (BL02B1) (Ref. 10) of SPring-8. X-ray energy was tuned to 30 keV using a sagittally bent Si(311) double monochromator. A double platinum mirror vertically collimates the incident beam and completely eliminates higher-order harmonics. Single crystals of LBSCO with $x=0.05$ and $x=0.075$ were obtained from the same batch as crystals used in previous neutron-scattering studies.9,11 The cylindrical crystals are about 5 mm in diameter with a height of 1 mm. The reciprocal lattice is defined in the $I4/mmm$ symmetry where the two short axes correspond to the distance between the nearest-neighbor Cu atoms along the in-plane Cu-O bond. At the (2 0 0) point, the longitudinal resolution ($\parallel a^* \parallel$ axis) was about 0.014 Å$^{-1}$ and transverse resolutions along the $b^*$ and the $c^*$ axes were $\sim 0.005$ and $\sim 0.046$ Å$^{-1}$, respectively.

Figures 1(a) and (b) show $H$-scan profiles of superlattice peaks around $Q=(2−2e 0 0.5)$ for $x=0.05$ and $x=0.075$, respectively, after adjusting against a background measurement. Raw data are shown in the inset of each figure. As can be seen in the figures, the superlattice peaks for both $x=0.05$ and $x=0.075$ crystals are clearly located at $H=1.76$ r.l.u. Thus the incommensurability $2\varepsilon$ is exactly $0.240\pm 0.001$ r.l.u. In a previous neutron-scattering study,8 the incommensurability of elastic magnetic peaks of $x=0.05$ samples was found to be $\varepsilon=0.120\pm 0.001$ r.l.u., indicating that the superlattice peaks observed in the present study indeed correspond to second-order harmonics of magnetic order. The linewidths are clearly broadened with respect to instrument resolution (indicated by bold horizontal lines in the figures), which gives finite in-plane correlation.
lengths along the $a$ axis ($\xi_a$) of $130 \pm 20$ Å and $120 \pm 30$ Å for $x = 0.05$ and $x = 0.075$, respectively.

$K$-scan profiles of superlattice peaks at around $Q = (2 - 2e 0 0.5)$ are shown in Figs. 2(a) and (b) after background correction. Figure 2(c) shows a trajectory of the $q$ scan and the locations of superlattice peaks in reciprocal-lattice space. Note that the $K$ direction is perpendicular to the propagation wave vector $Q_{ch}$. The peak for the $x = 0.05$ crystal is almost at $K = 0$ (indicated by a dashed line in the figure), whereas for the $x = 0.075$ crystal, the peak is clearly shifted away from $K = 0$. The amplitude of the peak shift was found to be $0.007 \pm 0.001$ r.l.u, the same as found in a previous neutron-scattering study. In addition, the $x = 0.075$ crystal used in the x-ray diffraction was composed of a single domain unlike the crystal from the neutron-scattering study, which contained a twin due to the orthorhombic symmetry of $Pcnc$. Hence the shift of the superlattice peak in the $x = 0.075$ crystal is clearly not an artifact, with the quartet of superlattice peaks forming a regular rectangular shape in reciprocal lattice space, as shown by the open diamonds in Fig. 2(c).

This arrangement of peaks satisfies the orthorhombic symmetry of the LTLO phase, not the tetragonal symmetry of the LTT phase, indicating that the pattern of charge order is closely related to crystal structure. The in-plane correlation length along the $b$ axis ($\xi_b$) for the tetragonal $x = 0.05$ crystal was $110 \pm 10$ Å, and similar for $\xi_a$. In comparison, $\xi_b$ of the orthorhombic $x = 0.075$ ($=70 \pm 8$ Å) is clearly shorter than $\xi_a$ and also shorter than the $\xi_b$ of the $x = 0.05$ crystal.

Figure 3 shows the $L$-dependence of the superlattice peaks around $Q = (2 - 2e 0 \pm 0.5)$ for (a) $x = 0.05$ and (b) $x = 0.075$, corresponding to out-of-plane correlations. The plots show the difference between data at $T = 11$ and $45$ K.
Raw data for the $x=0.05$ crystal at each temperature are plotted in the inset of Fig. 3~\textit{a}. Intensities of both the samples modulate sinusoidally and exhibit broad maxima at \( L = 0.5 \text{ r.l.u.} \), indicative of a twofold periodicity along the \( c \) axis. The linewidth is much broader than instrument resolution. Thus a reasonably short out-of-plane correlation length \( \xi_c \) of \( \approx 9 \text{ Å} \) was obtained for both the \( x=0.05 \) and \( 0.075 \) crystals, which is shorter than that of the next-nearest-neighbor (NNN) distance between CuO$_2$ planes. The large anisotropy between \( \xi_a \) and \( \xi_b \) suggests two-dimensional charge correlations. Solid curves in Fig. 3 denote fits to the equation \[ u F(L)^2 \{ 1 - e^{-2 \pi L} \}^2 \sin(\pi L). \] The good agreement of this equation with the data indicates that there is an antiphase relationship between NNN CuO$_2$ layers, which are separated by a distance \( c \), which can be explained by a long-range Coulomb interaction between doped holes on the CuO$_2$ planes. The integrated intensity along \( L \) of the superlattice peak is \( \approx 10^7 \) times weaker than that of the fundamental \( 200 \) Bragg reflection of intensity \( \approx 10^8 \) cps. In addition, the relative intensity of superlattice peak to the fundamental peak is \( \approx 10 \) times weaker than found in the neutron-scattering study. These results show that lattice distortions are the main contributor to the superlattice intensity and that the relative intensity is qualitatively consistent with a model in which the largest atomic displacement resulting from charge order is oxygen. The amplitude of oxygen displacement along the \( a \) axis can be estimated to be less than \( 10^{-3} \text{ Å} \) by a simple calculation based on the stripe model and using the measured relative intensities.

The temperature dependence of superlattice peak intensity and of the \( 300 \) reflection, which corresponds to the order parameter of structural phase transition into the LTT or LTLO phase, were measured. Results are shown in Figs. 4~\textit{a} and \textit{b} for the \( x=0.05 \) and \( 0.075 \) crystals, respectively. Structural phase transition temperatures \( T_{d2} \) are indicated in the figures at the point where the \( 300 \) superlattice intensity diminishes. The \( T_{d2} \) transition temperatures for the \( x=0.05 \) and \( x=0.075 \) crystals were thus estimated to be 38 and 34 K, respectively, almost identical to those obtained by neutron scattering.$^7$ Remarkably, the temperature dependence of the superlattice peak intensity (closed circles) is almost identical to that of the order parameter for the LTT/LTLO phase (open circles), suggesting that the ordering process of charge order is closely related to that of LTT/LTLO structural phase transition. These results are quite a contrast to the LNSCO system, where the superlattice peak evolves gradually as temperature decreases whereas the LTT order parameter exhibits a first-order phase transition.$^4,8$

High-\( Q \) resolution as well as the high statistics of the present x-ray-diffraction study have provided precise propagation wave vectors of the superlattice peaks associated with charge order, giving \( Q_{d2} = ( \pm 0.24 \mp \eta \frac{1}{2} ) \) with $\eta=0$ and
that mensurability with the lattice is essentially important for stabilizing charge order. In this case, one can easily imagine that $e$ should have a commensurate value of $\frac{1}{4}$, as predicted theoretically.\textsuperscript{12} Tranquada $et$ $al.$ have noted that the incommensurate value of $e$ can be regarded as a disordered stripe in which there is the mixture of distinct stripe periods of $4a$ and $5a$.\textsuperscript{14} In scattering intensities calculated under this assumption, the charge order peak is broadened whereas the magnetic order peak remains sharp. In fact, in our LBSCO system, the intrinsic linewidth of the superlattice peak along the $H$ direction is considerably broader than that of resolution-limited magnetic peaks observed by neutron scattering.\textsuperscript{7,11} These results imply that charge stripe order in cuprates is intrinsically disordered in comparison with the aligned stripe, consistent with the approval of the Japan Synchrotron Radiation Research Institute (Proposal Nos. 1998A0157, 1999A0234, and 2000B0321).

Line broadening of the superlattice peak is seen along both the $H$ direction and the $K$ direction. In particular, these systematic experiments using single domain crystals have revealed that linewidths along the $K$ axis for orthorhombic $x=0.075$ crystals are much broader than for tetragonal $x=0.05$ crystals. Based on the stripe model, the linewidth along $K$ corresponds to the mosaicity of the charge stripe. In addition, the orthorhombic symmetry of superlattice peaks in $x=0.075$ crystals suggests that the charge stripes are oblique. As Fujita $et$ $al.$ have noted,\textsuperscript{9} a corrugated pattern in the CuO$_2$ plane in LTLO phase can easily produce steps or kinks in the stripes, giving rise to the oblique of charge stripe. In this point of view, more oblique stripes could introduce the steps or kinks more randomly, which yields charge stripe mosaicity. Therefore oblique stripe order becomes more disordered or smectic in comparison with the aligned stripe, consistent with the present results.

In conclusion, the propagation wave vector and three-dimensional correlation of charge order in LBSCO systems were determined accurately using high-intensity synchrotron x-ray diffraction. Despite the $\frac{1}{4}$-hole doping, the incommensurability of the superlattice peak ($e=0.12$ r.l.u.) is clearly shifted away from the commensurate value of $\frac{1}{4}$, indicating that charge stripe order in cuprates is intrinsically disordered. The orthorhombic $x=0.075$ crystal provided detailed information about the peak shift as well as the linewidth of the superlattice peak, indicating that the oblique stripes in $x=0.075$ crystal are more disordered than the aligned stripes in $x=0.05$ crystal. The charge order was also found to be $2c$ periodic and two dimensional in nature. A proper determination of the atomic displacement pattern associated with the charge order is required to fully understand the essential nature of the (disordered) charge stripe order.

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