Evidence for local lattice fluctuations as a response function of the charge stripe order in the $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ system

N. L. Saini
Unità INFM, Università di Roma "La Sapienza," Piazzale Aldo Moro 2, 00185 Roma, Italy

H. Oyanagi
Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba 305-8568, Japan

A. Lanzara, D. Di Castro, S. Agrestini, and A. Bianconi
Unità INFM and Dipartimento di Fisica, Università di Roma "La Sapienza," Piazzale Aldo Moro 2, 00185 Roma, Italy

F. Nakamura and T. Fujita
Department of Quantum Matter, ADSM, Hiroshima University, Higashi-Hiroshima 739-8526, Japan
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We have used polarized Cu *K*-edge extended x-ray absorption fine structure to investigate the local lattice displacements in the charge-ordered $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ system. The temperature-dependent distance broadening, given by the Debye-Waller factors of the Cu-O bonds measuring local lattice fluctuations, shows a clear steplike increase below the charge stripe order temperature. The results construct a direct evidence for the local lattice fluctuations as a response function of the charge stripe ordering in the cuprate superconductors.

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Advances in the materials science and development of different experimental tools have generated a wealth of information with a significant implication on the high-T_c superconductivity. Indeed an approach based on charge distribution in stripes is making its ground among conventional theories to understand the physics of these superconductors. Several experiments probing different characteristics related with charge, spin, and lattice dynamics are found to be consistent with the striped phases in these materials.[1–20] The stripe phases are now widely observed in the closely related structures as the insulating nickelates[21] and the manganites.[22] In fact, the experiments have suggested that the inhomogeneous electronic ground state with stripe-charge ordering should be one of the right keys to unlock the mystery of high-T_c superconductivity. Recently a systematic work has revealed that the local elastic strain ε (Ref. 23) due to the lattice mismatch between the bcc CuO_2 and the fcc rock-salt sub-lattices plays key role in the stripe formation and their dynamics. The superconducting stripes (superstripes) appear to form for the micro-strain larger than a critical value (ε_c) and the superconductivity shows up near this critical point, the so-called strain critical point.[23]

In this paper we provide a direct evidence for local lattice fluctuations being a response function of the charge stripe ordering in the cuprates. We have exploited polarized Cu *K*-edge extended x-ray absorption fine structure (EXAFS) to measure the temperature dependence of the Cu-O distance broadening given by the correlated Debye-Waller factors of the Cu-O bonds. Here the well known charge stripe ordered phase, $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ system, having the microstrain larger than the critical microstrain ε_c (showing charge stripe ordering temperature $T_{so} \sim 60$ K), has been chosen as a model compound for the study. Thanks to the availability of high quality single crystal samples, technically advanced beamlines equipped with high efficiency fluorescence detection assembly along with high brilliance and polarized x-ray synchrotron radiation sources has made the task easier to obtain high *k*-resolution EXAFS data with high signal to noise ratio. We find that Debye-Waller factor of the Cu-O, measuring local lattice fluctuations, shows an order parameter like change across the charge stripe order temperature. The results provide a direct evidence for the local lattice fluctuations to be a response function of the charge stripe order parameter in the cuprate superconductors.

A well-characterized crystal of $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ (LNSC), grown by the traveling solvent floating zone (TFSZ) method was used for the measurements. The temperature dependent polarized Cu *K*-edge absorption measurements were performed at the beamline BL13B of Photon Factory at High Energy Accelerator Research Organization in Tsukuba. The synchrotron radiation emitted by a 27-pole wiggler source (maximum field B_0 of 1.5 T) inserted in the 2.5 GeV storage ring with a maximum stored current of 360–250 mA was monochromatized by a variable exit beam height double crystal Si(111) monochromator and sagittally focused on the sample. Improved lattice of the storage ring and better monochromator cooling system allowed to get a very stable beam on the sample. The spectra were recorded by detecting the fluorescence yield (FY) using a 19-element Ge x-ray detector array, covering a large solid angle of the x-ray fluorescence emission. The emphasis was given to measure the spectra with a high signal to noise ratio up to a high momentum transfer and for the purpose we have measured several scans, with each scan averaged over 19 channels. The oriented crystal was mounted on a Huber 420 goniometer. The grazing incidence geometry was used for the measurements by keeping the electric vector of the synchrotron light parallel to the Cu-O-Cu bond direction selected by preedge peak intensity representing $1s \rightarrow 3d$ quadrupole transition having minima when the polarization is falling flat

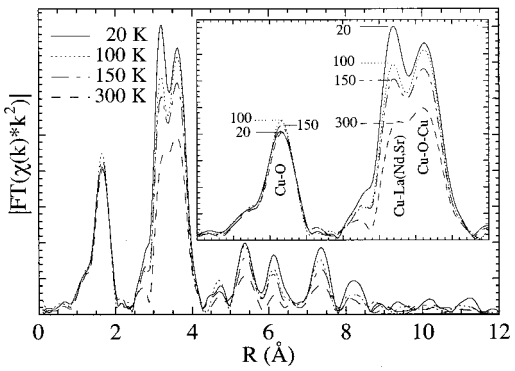

FIG. 1. Fourier transform of the EXAFS spectra (multiplied by $k2$) recorded on the $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ system at several temperatures. The Fourier transforms have been performed between $k=3-17\ \text{Å}^{-1}$ using a Gaussian window and not corrected for the phase shifts. The inset is a zoom over the main peaks representing Cu-O, Cu-La (Nd, Sr), and Cu-O-Cu scatterings.

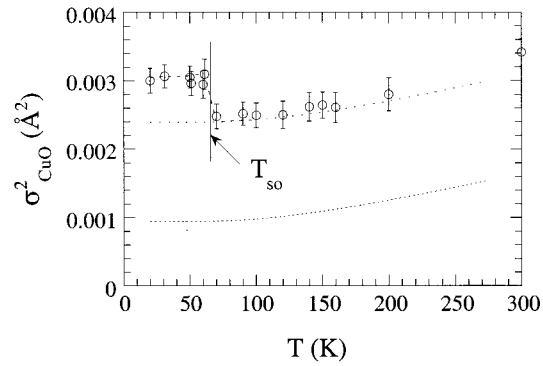

FIG. 2. Temperature dependence of the correlated Debye-Waller factors (symbols) of the Cu-O pairs (σ^2_{CuO}). The expected temperature dependence of the Debye-Waller factor for a fully correlated motion of Cu and O, calculated by Einstein model, is shown by lower dotted line. A constant value of 0.00145 is added to guide the temperature dependence of the experimental Debye-Waller factor (upper dotted line). The dashed line across the charge stripe order temperature (T_{so}) is to guide the eyes.

on the Cu-O bond (i.e., $3d_{x^2-y^2}$ lobes). The sample temperature was controlled and monitored within an accuracy of ± 1 K. A standard procedure was used to extract the EXAFS signal from the absorption spectrum and corrected for the x-ray fluorescence self-absorption before the analysis.[24]

The measurements at several temperatures were repeated at the beamline BM29 of the European Synchrotron Radiation Facility (ESRF), Grenoble. At the BM29 the sample was mounted in a two-stage closed-cycle He cryostat for the measurements. A 13 Ge element solid state detector was used to measure the absorption signal in the normal incidence geometry. The results were found to be highly reproducible in spite of different experimental geometry at two different synchrotron facilities.

The Fourier transforms (FT) of the EXAFS spectra (multiplied by k^2) recorded in the in-plane geometry of the LNSC at some representative temperatures, below the charge stripe ordering ($T<T_{\text{so}}$) and above it ($T>T_{\text{so}}$) are shown in Fig. 1. The FT represents the global atomic distribution of nearest neighbors around the absorbing Cu atom in the system. The FT represents raw experimental data and shows the standard peaks. The main peaks are shown enlarged as inset and denoted by Cu-O, Cu-La(Nd, Sr), and Cu-O-Cu representing respectively the scattering of the ejected photoelectron at the Cu site with the in-plane oxygen atoms (at ~1.88 Å), La(Nd, Sr) atoms (sitting at ~3.2 Å and 45° from the direction of the photoelectron) and a direct multiple scattering with the next Cu atom (at ~3.8 Å), across the in-plane oxygen. The peaks do not represent the real atomic distances and the position should be corrected for the photoelectron backscattering phase shifts to find the quantitative value to the atomic positions with respect to the central Cu.

The FT peaks show interesting temperature dependence. As expected, FT amplitude of the Cu-La(Nd, Sr) and Cu-O-Cu peaks increases with decreasing temperature. We could also see some changes in the peaks above 4 Å however these peaks contain contributions of complex multiple scattering of the photoelectron ejected at the Cu site and out of the present discussion. The Cu-O peak shows the expected increase with lowering the temperature with a decrease across the charge stripe ordering temperature. It is clear from the raw data (Fig. 1) that the amplitude of the Cu-O peak at 20 K is almost equal to the one at 300 K. This indicates anomalous local lattice displacements at the low temperature.

The signal of Cu-O bonds is well separated from the higher shells of other neighboring atoms and could be separately analyzed. Here the Cu-O EXAFS signal is due to single scattering and probes the pair correlation function between Cu and in-plane oxygen. The EXAFS spectra due to the Cu-O were simulated by a standard least square approach using curved wave theory.[24] The starting parameters were taken from the diffraction studies. The number of parameters which may be determined by EXAFS is limited by the number of independent data points: $N_{\text{ind}}\sim(2\Delta k\Delta R)/\pi$, where Δk and ΔR are respectively the ranges in k and R space over which the data are analyzed. In our case $\Delta k=14\ \text{Å}^{-1}$ and $\Delta R=1$ Å give $N_{\text{ind}}\sim 9$ for the first shell EXAFS.[24]

Temperature dependence of the Debye-Waller factor of the Cu-O pairs, σ^2_{CuO}, that takes into account both static and dynamic distortions of the CuO_2 plane has been determined by the analysis of the Cu-O EXAFS and shown in Fig. 2. The σ^2_{CuO} measures the correlated displacements between Cu and O and not the same as one observed in the diffraction experiments where the Debye-Waller factor accounts for the mean-square deviation of a given atom from its average site in the crystal. The σ^2_{CuO} shows an anomalous upturn at ~60 K where the charge stripe ordering is known to occur as shown by several experimental techniques.[6,11,14] Indeed appearance of any charge density wave like instability gives an anomalous change in the Debye-Waller factor, known from studies of density wave systems.[25] Therefore we conclude that the anomalous upturn in the correlated Debye-Waller factor, σ^2_{CuO} is due to a charge instability giving charge stripe ordering in this system.

The anomalous upturn of σ^2_{CuO} is related to a particular local structural distortion rather than a long-range structural transition. This anomaly occurs when the distribution of

Cu-O bonds gets larger than thermal broadening as shown earlier for the $La_{1.85}Sr_{0.15}CuO_4$ (LSCO) system.[2] Indeed, the change in σ^2_{CuO} is similar to the one observed at around 100 K in LSCO, and other related systems that do not have any long-range structural transition at the anomalous temperature. It has been found that the anomalous upturn in σ^2_{CuO} is due to a particular local lattice distortion (the Q_2 type of rhombic distortion) in the CuO_2 plane related with the charge stripe ordering.[2,26] Therefore the present results, obtained on a model system, provide a clear indication of local lattice distortions in the CuO_2 plane that could be considered a response function of the charge stripe order parameter in the cuprates. Very recently similar conclusions have been drawn by pair distribution function analysis of neutron diffraction, showing increased Cu-O bond distribution, measured by the mean-square displacements, across the charge stripe ordering temperature in the $La_{2-x}(Ba, Sr)CuO_4$.[5]

The jump across the T_{so} is found to be smaller than that for the LSCO system, however, the value of σ^2_{CuO} above the T_{so} is much larger than for the LSCO system.[26] In the LSCO system the σ^2_{CuO} above the T_{so} remains nearly the same as the one expected from the Einstein model (dotted line) while for the present case the σ^2_{CuO} is larger (by $\sim$0.00145) even if it follows the expected temperature dependence at higher temperature (Fig. 2). The higher value of σ^2_{CuO} appears to be due to the temperature independent disorder in the CuO_2 plane, introduced by the presence of the Nd. In fact, the presence of Nd is believed to be a reason for more static character of the charge stripes in the LNSC system[6,11,14] and hence observable by slower techniques. Here we should recall that EXAFS measures instantaneous displacements with a characteristic time scale $\sim 10^{-15}$ s and the charge stripe ordering temperature measured by this technique may differ from the one measured by slower techniques (e.g., Ref. 6) for the systems where the charge stripe ordering has more dynamic character.

To get further insight into the local lattice fluctuations we have determined the pair distribution function (PDF) of local Cu-O bond lengths. A standard procedure was used to draw PDF from analysis of the EXAFS oscillations only due to the Cu-O distances. In this procedure the Cu-O EXAFS is simulated by the least squares fit with an input containing a distribution of several distances (where the N_{tot} is fixed to the nominal value and σ^2_{CuO} for each distance is given by the Einstein model for a correlated Cu-O distribution). The only variable parameter is the relative probability of different distances with a constraint of N_{tot} to be fixed. Within experimental uncertainties the results remain the same while two distances model (with variable parameters to be the relative probabilities, the two distances and the two σ^2_{CuO}) was considered. The feasibility of the methods has been shown earlier for the case of cuprates[2,26] manganites[27] and other complex systems.[28,29]

Figure 3 shows the Cu-O PDF at two temperatures along with the difference between the two across the T_{so}· Cu-O PDF for the LSCO at a temperature $T<T_{so}$ (Ref. 26) is also shown for a comparison. The Cu-O distribution in the title compound displays an asymmetric peak, either at a

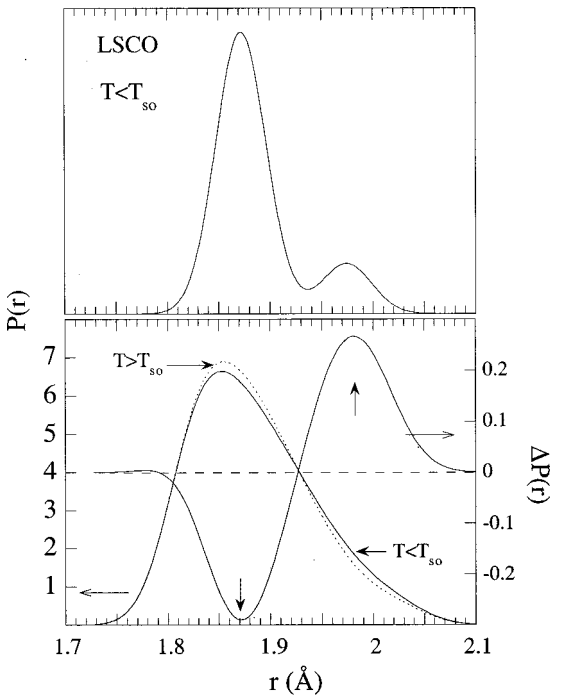

FIG. 3. Pair distribution function (PDF) of the Cu-O bonds determined by the polarized Cu-O EXAFS for the $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ system below ($T<T_{so}$) and above ($T>T_{so}$) the charge stripe order temperature. The difference across the T_{so} is also shown. Cu-O PDF for the $La_{1.85}Sr_{0.15}CuO_4$ at $T<T_{so}$ is shown (Ref. 26).

temperature lower than the charge stripe order, T_{so} or above it. There is a small redistribution of Cu-O bonds across the T_{so} that could be seen in the difference. In the charge stripe ordered state ($T<T_{so}$) there is an elongation of a part of the Cu-O bonds. This elongation of the Cu-O bonds is similar to the one observed in the LSCO system and associated with appearance of the rhombic distortion of the CuO_2 plane resulting longer Cu-O bonds. Here the amplitude of the local distortion, defined by the separation between the long and short Cu-O bonds, remains the same (Fig. 3) in the two systems indicating intrinsic similarity of the local lattice displacements across the T_{so} in these materials. However, while the bond distribution gets two-peak function at low temperature, from a single peak function above the T_{so} in the LSCO system,[26] it remains asymmetric in the LNSC, with a probability weight transfer across the T_{so}. This further indicates that charge stripe ordering in the cuprates is associated with dynamic local lattice fluctuations rather than static disorder. The superconductivity survives with charge stripe ordering associated to a large contribution of dynamic local lattice displacements as the case of LSCO, while it gets anomalously suppressed with charge stripe ordering associated with less dynamic character of local lattice displacements.

It has been reported that magnetic order plays important role in the superconductivity of the cuprates while the charge stripe ordering could coexist with the superconductivity.[14] Indeed the superconductivity is found to be suppressed when the magnetic order appears.[14] Our finding suggests that the superconductivity disappears when the local lattice fluctua-

tions, associated with the charge stripe ordering, get less dynamic. Comparing the present results with the earlier results on the magnetic order it appears that the local lattice fluctuations are less dynamic in presence of magnetic ordering. Therefore the two results compliments each other. Thus the local lattice fluctuations are more static in the absence of superconductivity, while magnetic ordering is present (as shown by other experiments[14]); on the other hand the dynamic contribution to the local lattice fluctuations increased while the system is superconducting with suppressed magnetic order.

In summary, we have used polarized Cu *K*-edge EXAFS spectroscopy to explore local lattice fluctuations associated charge stripe ordering in the cuprates with $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ (LNSC) as a model system. We find an order parameter like change in the instantaneous local lattice fluctuations across the charge stripe ordering temperature, revealed by an anomalous increase in the correlated Debye Waller factor of the Cu-O pairs. We have argued that the anomalous change, associated with appearance of a dynamic rhombic distortion of the CuO_2 plane, could be considered as a response function of the charge stripe ordering in these materials. From the temperature dependent Cu-O bond distribution we find that amplitude of the dynamic rhombic distortion of the CuO_2 plane in the LNSC remains similar to the one found in the superconducting LSCO system, however, contribution (probability) of this distortion to the charge stripe ordering in LNSC is smaller. This appears to indicate that the contribution of dynamic local lattice displacements to the charge stripe ordering is vital for the system to be superconducting. In conclusion, the results suggest that a particular local lattice distortion could be a response function of the charge stripe order in the cuprates. A large isotope effect on the charge stripe ordering[30] and pseudogap temperature[31] further suggests that electron-lattice interactions are important ingredients for the high-T_c in the doped cuprates and there appears an intimate relationship between the charge stripe ordering, local lattice fluctuations and high-T_c superconductivity. Here, the local strain in the electronically active CuO_2 plane ε, that controls the local lattice fluctuations, seems to be a key parameter to resolve mystery of the high-T_c superconductivity.[23]

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