

# Magnetic Excitations and Phase Separation in the Underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ Superconductor Measured by Resonant Inelastic X-Ray Scattering

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(Received 29 October 2009; published 19 February 2010)

We probe the collective magnetic modes of  $\text{La}_2\text{CuO}_4$  and underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) by momentum resolved resonant inelastic x-ray scattering (RIXS) at the Cu  $L_3$  edge. For the undoped antiferromagnetic sample, we show that the single magnon dispersion measured with RIXS coincides with the one determined by inelastic neutron scattering, thus demonstrating that x rays are an alternative to neutrons in this field. In the spin dynamics of LSCO, we find a branch dispersing up to  $\sim 400$  meV coexisting with one at lower energy. The high-energy branch has never been seen before. It indicates that underdoped LSCO is in a dynamic inhomogeneous spin state.

DOI: 10.1103/PhysRevLett.104.077002

PACS numbers: 74.25.Ha, 74.72.-h, 75.50.Ee, 78.70.Ck

**Introduction.**—Since the early days of high  $T_c$  superconductivity, it has been surmised from a theoretical viewpoint that electronic inhomogeneity plays an essential role in the cuprates, possibly in the form of stripes of holes and spin [1]. Later, phase separation was observed experimentally [1,2], revealing the coexistence of patches of distinct electronic phases not only in the form of stripes, but also as charge segregated islands. The central question is whether such spatial inhomogeneity is merely a consequence of static disorder induced by, for instance, the dopants, or if these materials are intrinsically and dynamically phase separated, and extrinsic static potentials just act as pinning centers for the fluctuating charge segregated islands or stripes. We address this question using momentum resolved resonant inelastic x-ray scattering (RIXS) to probe the collective modes of underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO). Given its complete novelty, we first tested our method on  $\text{La}_2\text{CuO}_4$  (LCO), to demonstrate that single magnon dispersion in cuprates can be determined by RIXS, as theoretically proposed by some of us in Ref. [3].

RIXS is a unique spectroscopic technique that provides both momentum and energy resolved information on spin and charge dynamics and is, at the same time, element specific [4]. We use RIXS at the copper  $L_3$  edge, where monochromatic incident radiation ( $\mathbf{k}$ ,  $h\nu_{\text{in}}$ ) excites a  $2p$  core electron resonantly into a Cu  $3d$  empty state. This intermediate state quickly decays again, typically within 1–2 femtoseconds [5]. We measure the energy  $h\nu_{\text{out}}$  and the momentum  $\mathbf{k}'$  of the photons that are emitted. Thus, we determine both the energy  $E = h\nu_{\text{in}} - h\nu_{\text{out}}$  and the momentum  $\mathbf{q} = \mathbf{k} - \mathbf{k}'$  of the excitation left behind in the

sample. The schematics of the experiment is shown in Fig. 1(a); we control the transferred momentum  $q_{\parallel}$  in the  $ab$  basal plane by rotating the sample (angle  $\theta$ ). In the RIXS process, a  $3d$  valence electron can, in principle, flip its spin, using the angular momentum carried by the scat-

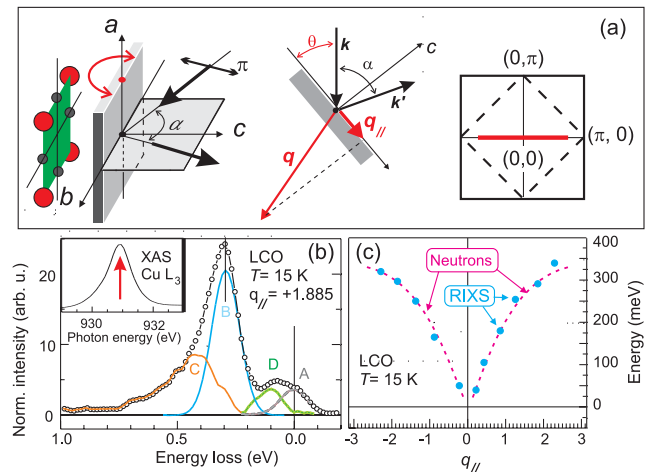


FIG. 1 (color online). Panel (a) layout of the experimental setup and indication of the reciprocal space region [red thick line] spanned inside the first Brillouin zone. Panel (b) decomposition of the LCO spectrum at  $q_{\parallel} = 1.85$ : elastic (A) and single magnon peaks (B); multiple magnon (C) and optical phonons (D) spectral features. In the inset the  $L_3$  absorption spectrum, the red arrow indicates excitation energy. Panel (c) single magnon dispersion determined by RIXS (this work, blue dots) and by inelastic neutron scattering (Ref. [12], dashed purple line).

tered photon. Whether this process is allowed depends on the symmetry of the material under consideration and on the absorption edge. For  $K$  edge RIXS, only double-spin-flip scattering is allowed, and on cuprates such as LCO and  $\text{Nd}_2\text{CuO}_4$ , it was indeed observed by Hill and co-workers [6]. In  $L_3$  edge, RIXS on NiO direct spin-flip scattering is allowed and was recently observed [7], whereas for cuprates, the situation is very peculiar: direct spin-flip transitions are possible only if the copper spin has a nonzero projection in the  $xy$  plane [3]. This corresponds to the actual spin structure of layered cuprates, for which  $\text{Cu } L_3$  RIXS thus gives access to the fundamental spin excitations.

In undoped antiferromagnetic LCO, we use  $\text{Cu } L_3$  RIXS to measure the magnon dispersion, which we find to coincide with the one obtained with inelastic neutron scattering. Subsequently, we investigate underdoped superconducting LSCO and discover a high-energy branch in the excitation spectrum that coexists with a less-dispersive branch at lower energy. This signals that LSCO is in a dynamically inhomogeneous state, possibly a stripe liquid with coherent spin dynamics up to 400 meV.

*Experimental method.*—The LCO and the LSCO samples were 100 nm thick films grown by pulsed laser deposition on the (001) surface of  $\text{SrTiO}_3$ . For LSCO, the effective hole density was obtained from the transport measurements that indicated  $T_c = 21.5$  K. The RIXS experiment was performed at the ADDRESS beam line of the Swiss Light Source [8] (Paul Scherrer Institute, Switzerland) using the high-resolution SAXES spectrometer [9]. The incident x rays were tuned at  $\sim 931$  eV corresponding to the maximum of the  $\text{Cu } L_3$  absorption peak. The beam line monochromator and the spectrometer contributed almost equally to the combined energy resolution  $\Delta E \leq 140$  meV. In Fig. 1(a), the experimental layout is shown. The crystallographic axes of the sample are ( $a$ ,  $b$ ,  $c$ ), and  $\pi$  indicates that the incident x rays are linearly polarized parallel to the scattering plane.  $q_{\parallel}$  is the component parallel to the  $ab$  plane of the transferred momentum  $\mathbf{q}$ . As the photon momentum  $\mathbf{k}$  is dictated by the energy of the  $\text{Cu } 2p \rightarrow 3d$  resonant transition, the scattering angle  $\alpha$  determines the maximum reachable  $q_{\parallel}$ . The inset shows the 2D Brillouin zone with the red thick line indicating the region explored in the experiment. Negative transferred momentum  $q_{\parallel}$  corresponds by convention to small values of  $\theta$ , i.e., incidence near grazing. Each spectrum is the result of 30 minutes total accumulation (6 spectra of 5 min). The exact position of the zero on the energy loss scale was determined by measuring for each  $q_{\parallel}$  a non-resonant spectrum from polycrystalline graphite. The spectra were measured with  $\alpha = 90^\circ$  ( $130^\circ$ ) for the smaller (larger)  $|q_{\parallel}|$ .

A typical RIXS spectrum of LCO is presented in Fig. 1(b): the raw spectrum is decomposed into an elastic peak A, a single magnon peak B, a high-energy feature C, and a low-energy ( $\sim 90$  meV) peak D. The latter is assigned to a well-known optical phonon [10], and feature C

is due to higher-order magnetic excitations, namely, bimagnons. We took advantage here of the improvement by a factor of 3 in energy resolution with respect to our previous work [11] that gave the first evidence of dispersing magnetic features in RIXS spectra of LCO. In Ref. [11], the experimental resolution was insufficient to resolve B and C, and the dispersing features had been assigned mainly to bimagnons. The assignment of the dominant peak B to a single magnon excitation is bolstered here by the empirical fact that its dispersion from (0, 0) to  $(\pi, 0)$  is in perfect agreement with neutron scattering data [12], as is shown in Fig. 1(c). These results on the antiferromagnetic system thus validate our experimental method: RIXS has become a true alternative to inelastic neutron scattering (see also Ref. [13]). More details on the LCO results will be published elsewhere later [14].

*RIXS response of LSCO.*—Having demonstrated the power of RIXS, we now apply it to the underdoped high  $T_c$  superconductor LSCO with a hole concentration  $p = 0.08$ . The momentum resolved spectra in Fig. 2 cover a large part of the Brillouin zone along the  $(-\pi, 0) \rightarrow (\pi, 0)$  direction, namely, the  $q_{\parallel} = [-2.29, 2.29]$  range. The peaks at high-energy loss are  $dd$  excitations, which are very similar to those of LCO. At lower energy, on the contrary, the magnon region of the spectrum is markedly changed by doping.

Already, the three-dimensional intensity plot of Fig. 2(a) clearly shows that, when approaching the Brillouin zone boundary, the magnetic excitation spectrum contains multiple *dispersing* peaks. One should note that, after normalization to the  $dd$  peak, the spectra are less intense for  $q_{\parallel} > 0$  than for  $q_{\parallel} < 0$ . This asymmetry is also present in LCO and fully agrees with the theoretical cross section of Ref. [3] for pure spin excitations. It is thus an additional and independent indication for the magnetic character of

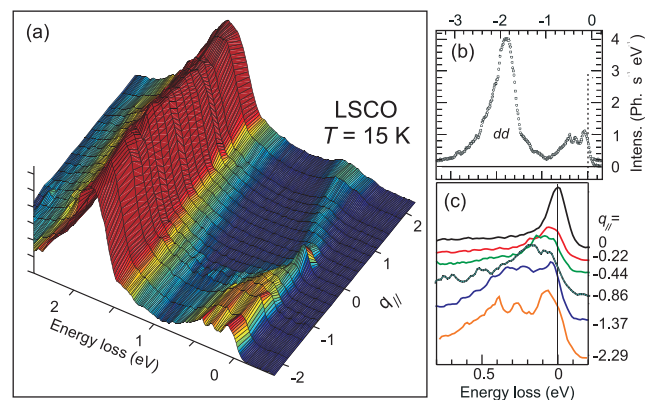


FIG. 2 (color online). Overview of the RIXS results for LSCO at  $T = 15$  K. Panel (a) 3D representation of the whole dataset, where the intensity (normalized to the integral of the  $dd$  excitations) is plotted as function of the energy loss and of the transferred momentum  $q_{\parallel}$ . Panel (b) example of one spectrum including the  $dd$  excitations ( $q_{\parallel} = -2.29$ ), Panel (c) selected spectra in the low energy spectral region.

the dispersing modes. Therefore, also LSCO spectra are dominated by spin dynamics. Although unavoidably contributing to the LSCO spectra to some extent, charge (electron-hole) excitations play a secondary role in the underdoped case. To fully exploit the RIXS information on spin dynamics, we made a detailed decomposition of each spectrum. Our data analysis is illustrated in Fig. 3(a) for  $q_{\parallel} = -2.29$ . After subtracting, in analogy to LCO, the elastic line and phonon losses, the resulting magnetic part of the spectrum is characterized by two separate peaks. The double peak structure of the magnetic spectrum is observed at low temperature (15 K in the present experiment) while the peaks amalgamate at room temperature [15], [see Fig. 3(b)]. For  $|q_{\parallel}| \geq 0.44$ , the two contributions to the magnetic scattering intensity are discernible. To highlight the doping-induced changes in the spin dynamics, in Fig. 3(c), the RIXS spectra of LSCO and LCO are compared at  $q_{\parallel} = -1.26$ . It is apparent that upon hole doping, the magnon peak of LCO loses intensity and a lower energy feature appears while also the higher-energy part changes. The resulting dispersion of the two magnetic branches is shown in Fig. 4(a). The upper branch, reaching 400 meV, is very close to the magnon dispersion of the parent compound LCO. A detailed analysis of the spectra shows that within  $\sim 25$  meV, the two branches are symmetric in  $q_{\parallel}$ , whereas the assignment of two separate

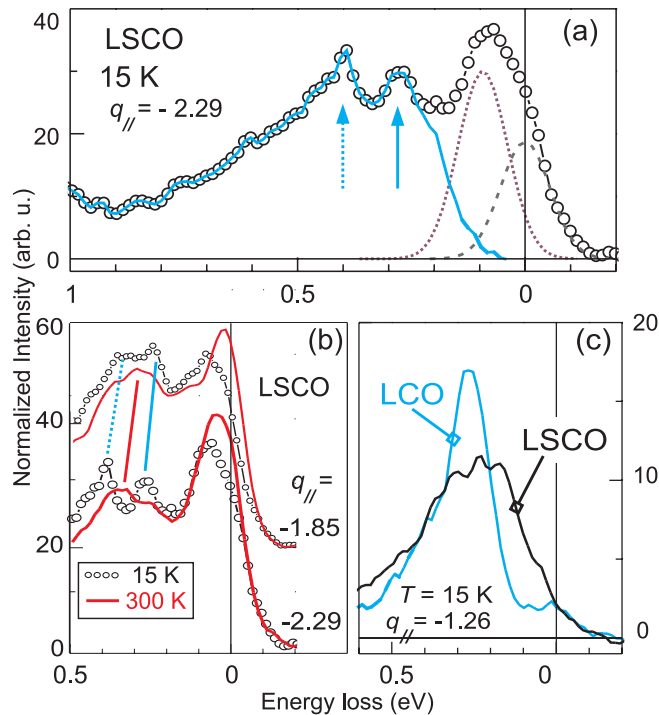


FIG. 3 (color online). Panel (a) example of RIXS spectrum decomposition for underdoped LSCO in the low energy region: elastic peak (grey dashed line), phonon peak (purple dotted line), and magnetic excitations (blue solid line). Panel (b) temperature effects for  $q_{\parallel} = -2.29$  and  $-1.85$ : the single peak structure at room  $T$  splits into two peaks at low  $T$ . Panel (c) comparison between LCO and LSCO for  $q_{\parallel} = -1.26$  at low temperature.

branches becomes ambiguous for  $|q_{\parallel}| < 0.44$ . At room temperature, the two peaks do not separate and the dispersion of the resulting magnetic feature lies in between the two low temperature branches, as shown in Fig. 4(b).

The lower energy magnetic branch also appears in recent neutron data by Lipscombe *et al.* [16] on underdoped LSCO with the same hole concentration ( $p = 0.085$ ). The branch disperses up to 200 meV, see Fig. 4(c), which indeed corresponds to our lower magnetic RIXS branch. The dispersion of the magnetic RIXS branch is also precisely in between the one determined with neutron scattering on LSCO for  $p = 0.05$  [17] and the one for  $p = 0.16$  by Vignolle *et al.* [18]. At smaller  $q_{\parallel}$ , we observe a magnetic RIXS signal within the limits of our resolving power near the neck of the famous “hour glass” dispersion seen by neutron scattering [2,18–20]. One must keep in mind, though, that in RIXS, we are measuring momenta away from  $(0, 0)$  and the neutron data are taken around  $(\pi, \pi)$ . These are strictly equivalent points in the Brillouin zone only in the presence of long-range antiferromagnetic order. Irrespective of this, the characteristics of the lower branch of the magnetic excitation spectrum that we observe in RIXS nicely agree with the inelastic neutron scattering branch observed in the same system around  $(\pi, \pi)$ . In contrast, we have discovered a higher-energy magnetic branch dispersing up to  $\sim 400$  meV in a doped cuprate superconductor. As indicated above, this branch is very close to the magnon dispersion of the LCO showing that high-energy spin dynamics is present also in underdoped LSCO, a fact not observed with neutrons yet. Observing in underdoped LSCO both a low- and high-energy magnetic branch suggests the presence of an in-

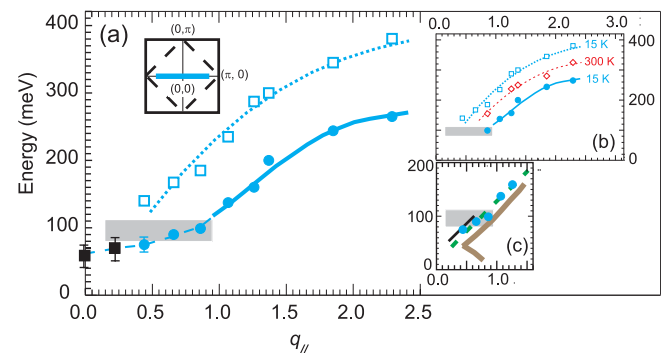


FIG. 4 (color online). Panel (a) dispersion of the two magnetic excitations in underdoped LSCO at  $T = 15$  K: one higher-energy LCO-like branch (open blue squares) and one lower energy branch (full blue circles). At small  $q_{\parallel}$  (black squares), the separation into subcomponents, if any, is not resolvable. In the shaded area, phonon scattering influences the data analysis. The dotted blue line is the LCO dispersion taken from Fig. 1(c) and multiplied by 1.09. The solid blue line is a guide to the eye. Panel (b) dispersion at room  $T$  (red diamonds) and low  $T$  (blue circles and squares). Panel (c) low energy branch measured with RIXS near  $(0, 0)$  (blue circles) and with neutrons close to  $(\pi, \pi)$  (thin black line for  $p = 0.05$ , green dashed line for  $p = 0.085$ , brown thick line for  $p = 0.16$ ).

homogeneous magnetic state: although the energy is similar, the spectral shape of the higher-energy feature in LSCO is different than in LCO, indicating that the phase separation takes place at a microscopic scale.

A propagating coherent LCO-like magnetic excitation dispersing down to  $q_{\parallel} \approx 0.44$  (corresponding to a wavelength of  $\sim 3.5$  lattice parameters) requires a magnetic patch having a linear extent considerably longer than 15 Å. This extent can be associated with the dimensions of the patches (either stripes or islands) on the 1–2 fs time scale of  $L$  edge RIXS. This is consistent with the notion that RIXS is a very fast probe whereas slower methods are expected to observe smaller island sizes. Indeed, using nuclear quadrupole resonance measurements Singer *et al.* [21] report magnetic patch size of 30 Å in LSCO. Also with scanning tunneling microscopy, another very slow probe, Kohsaka *et al.* [22] observe in the related cuprate superconductor  $\text{Ca}_{2-x}\text{Na}_x\text{CuO}_2\text{Cl}_2$  ( $x$  around 0.08) separate metallic and insulating areas of approximately 20 Å in diameter. Our data are compatible with a model of stripe patches fluctuating along the crystallographic axis of a twinned sample (i.e., a kind of stripe liquid). According to the Hubbard model calculations by Seibold and Lorenzana [23] for static stripes, there are two magnetic branches separated by about 100 meV, of which the higher branch is a remnant of LCO as it corresponds to magnetic excitations parallel to the stripe direction. The presence of these two excitations is consistent with our data. However, we do not observe static stripe formation, which suggests that at low temperature, underdoped LSCO is a stripe liquid in which the directions perpendicular and parallel to the stripes are macroscopically indistinguishable. An additional indication for the dynamic nature of this inhomogeneous magnetic state is that high-energy van Hove singularities at  $q_{\parallel} = 0$ , which are typical of static stripes [23], are absent; instead we observe a rather flat distribution of spectral intensity up to about 0.9 eV [see in Fig. 2(c) the spectra at  $q_{\parallel} = 0$  and  $-0.22$ ].

**Conclusions.**—The present finding of a high-energy branch in the excitation spectrum of an underdoped high  $T_c$  superconductor, coexisting with a less-dispersive branch at lower energy is a strong evidence that our LSCO sample is in a dynamically inhomogeneous state, possibly a stripe liquid with spin dynamics up to 400 meV. In more general terms, our experimental findings demonstrate the power of newest generation high-resolution momentum dependent RIXS to probe the spin dynamics of high  $T_c$  superconductors. This paves the way to further clarify the interplay of the observed magnetically inhomogeneous state with superconductivity. To do so, further cross-fertilization between RIXS and neutron spectroscopy will be of paramount importance.

This work was performed at the ADRESS beam line of the SLS (PSI) using the SAXES spectrometer developed

jointly by Politecnico di Milano, SLS, and EPFL. The authors acknowledge T. Devereaux, M. Grioni, S. Kivelson, G. Sawatzky, Z.-X. Shen, and J. Zaanen for important exchanges of ideas; and M. Radovic and F. Miletto Granozio for providing the LCO sample. This work is supported by the Office of Science, U.S. Department of Energy (DOE), Contract No. DE-FG02-08ER4650 (CMSN).

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