Magnetic-Field-Induced Spin Excitations and Renormalized Spin Gap of the Underdoped La$_{1.895}$Sr$_{0.105}$CuO$_4$ Superconductor

J. Chang,[1] A. P. Schnyder,[2] R. Gilardi,[1] H. M. Rønnow,[1,3] S. Pailhes,[1] N. B. Christensen,[1,4] Ch. Niedermayer,[1]
D. F. McMorrow,[5,6] A. Hiess,[7] A. Stunault,[7] M. Enderle,[7] B. Lake,[8] O. Sobolev,[8] N. Momono,[9] M. Oda,[9] M. Ido,[9]
C. Mudry,[2] and J. Mesot[1,*]

[1]*Laboratory for Neutron Scattering, ETH Zurich and PSI Villigen, CH-5232 Villigen PSI, Switzerland*
[2]*Condensed Matter Theory Group, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland*
[3]*Laboratory for Quantum Magnetism, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland*
[4]*Risø National Laboratory, DK-4000 Roskilde, Denmark*
[5]*London Centre for Nanotechnology and Department of Physics and Astronomy, University College London, London, United Kingdom*
[6]*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, United Kingdom*
[7]*Institut Laue-Langevin, BP 156, F-38042 Grenoble, France*
[8]*BENSC Hahn-Meitner-Institut, 14109 Berlin Wannsee, Germany*
[9]*Department of Physics, Hokkaido University-Sapporo 060-0810, Japan*
(Received 7 June 2006; published 15 February 2007)

High-resolution neutron inelastic scattering experiments in applied magnetic fields have been performed on La$_{1.895}$Sr$_{0.105}$CuO$_4$ (LSCO). In zero field, the temperature dependence of the low-energy peak intensity at the incommensurate momentum transfer $\mathbf{Q}_{IC} = (0.5, 0.5 \pm \delta, 0), (0.5 \pm \delta, 0.5, 0)$ exhibits an anomaly at the superconducting T_c which broadens and shifts to lower temperature upon the application of a magnetic field along the c axis. A field-induced enhancement of the spectral weight is observed, but only at finite energy transfers and in an intermediate temperature range. These observations establish the opening of a strongly downward renormalized spin gap in the underdoped regime of LSCO. This behavior contrasts with the observed doping dependence of most electronic energy features.

In studies of high-temperature superconductors (HTSC), one central challenge is to explain the evolution from an antiferromagnetic (AF) Mott insulator to a metallic superconductor upon doping. For example, the intimate interplay between magnetism and superconductivity has been revealed in momentum resolved inelastic neutron scattering (INS) experiments on La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [1–5] and YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) [6,7] that showed the opening of a spin gap (SG) Δ_{SG} at the superconducting (SC) critical temperature (T_c). In contrast to the single-particle SC gap probed by, e.g., angle-resolved photoemission spectroscopy (ARPES) [8,9], the SG scales with T_c in the overdoped to slightly underdoped regime. The situation is more complicated in more underdoped samples. For instance, in YBCO the ratio $\Delta_{SG}/(k_B T_c)$ decreases with strong underdoping [10,11], while for underdoped LSCO ($x < 0.12$) the changes are even more dramatic, since no direct neutron scattering evidence for a SG has been reported so far [12]. This latter fact would seem to indicate that the underdoped regime of LSCO cannot be described in terms of a homogeneous electronic liquid and more exotic scenarios have been suggested, such as the formation of dynamical stripes [13] or presence of a d-density wave (DDW) order [14]. Further insight into the interplay between magnetism and superconductivity in LSCO has been revealed in a number of studies of the effects of an applied magnetic field, which at optimal doping enhances the low-energy magnetic fluctuations and at underdoping induces static antiferromagnetism [15–17].

In this Letter, based on high-resolution INS data as a function of energy $\hbar\omega$, magnetic field H, and temperature T, we identify similarities and differences between the magnetic response in the underdoped and optimal doped regimes of LSCO. Our main result is the observation of a dramatic field-induced enhancement of the response at low energies and the identification, below T_c, of a SG in the underdoped regime ($x = 0.105$), whose characteristic energy is strongly renormalized relative to that measured at optimal doping. We also show that this behavior is not specific to LSCO but corresponds to what is observed in YBCO. Unlike at optimal doping, the opening of the $x = 0.105$ SG is incomplete all the way down to the lowest achievable temperature.

Our experiments have been performed on a large cylindrical single crystal of La$_{2-x}$Sr$_x$CuO$_4$ ($m \sim 3.86$ g, $x = 0.105$) grown by the traveling solvent floating zone (TSFZ) method [18]. The SC volume fraction was estimated from measurements of the specific heat C. At $T_c = 30$ K, we observed a jump of $\Delta C/T_c = 3$ mJ/(K^2 mol). Below T_c the electronic contribution was estimated to be $C_{el}^s/T = \gamma_{el}^s = 0.5$ mJ/(K^2 mol) while in the normal state we found $C_{el}^n/T = \gamma_{el}^n = 5.5$ mJ/(K^2 mol). This indicates that the SC volume fraction $1 - \gamma_{el}^s/\gamma_{el}^n$ is larger than 90%. Together with a sharp SC transition ($\Delta T_c = 1.5$ K), this demonstrates the high quality of the crystal. The structural transition from the high-temperature tetragonal to the low-temperature orthorhombic phase occurs at 290 K, which confirms $x = 0.105 \pm 0.005$. The magnetic response is

peaked in the reciprocal space at the incommensurate wave vector $\mathbf{Q}_{IC} = (0.5 \pm \delta, 0.5, 0)$, $(0.5, 0.5 \pm \delta, 0)$ (in tetragonal units of $2\pi/a = 1.65$ Å^{-1} used throughout this Letter). The value $\delta = 0.097 \pm 0.005$ is in line with a Sr concentration of $x = 0.105 \pm 0.005$ [19]. The sample has been characterized by magnetization and ac-susceptibility measurements in magnetic fields H up to 8 T along the crystallographic c axis [20], as well as small-angle neutron scattering (SANS) and μSR [21]. All measurements indicate that in the underdoped regime of LSCO, the vortex liquid phase dominates the magnetic phase diagram, while a quasi-long-range ordered vortex lattice can only be observed at low fields or temperatures [21]. This contrasts with the well-ordered vortex lattice observed at all fields in the low-temperature regime of optimally doped LSCO [22]. The melting line of the vortex lattice is shown in Fig. 1(a).

INS measurements of the spin excitations were performed on the cold neutron spectrometer IN14 at the Institut Laue-Langevin, Grenoble, France. We used a vertically curved graphite monochromator and a horizontally curved graphite analyzer with fixed final energy E_f in the range 3.5–5 meV that gave an energy resolution of about 60–150 μeV. A cooled beryllium filter removed higher-order contamination from the scattered beam. The sample was cut into two pieces which were coaligned within less than a degree and mounted in a 15 T vertical cryomagnet
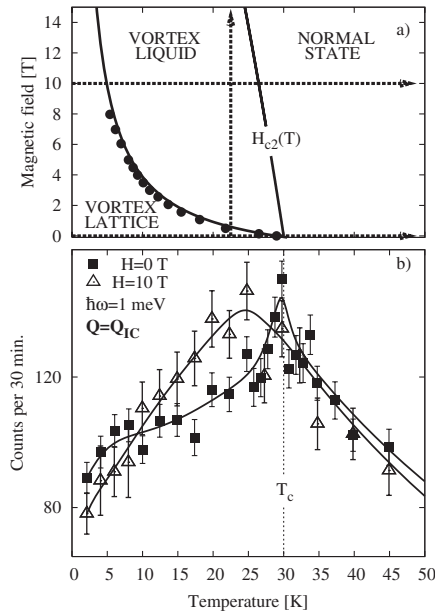
such that momentum transfers $\mathbf{Q} = (Q_h, Q_k, 0)$ were accessible. All measurements were performed after field cooling from above T_c. With a similar setup the field dependence of the elastic signal at the incommensurate wave vectors was measured on the FLEX spectrometer at Hahn-Meitner Institute.

In Fig. 1(b), we show the effect of H on the T dependence of the intensity at $\mathbf{Q}_{IC}$ with an energy transfer of $\hbar\omega = 1$ meV. These data demonstrate that the intensity is enhanced by application of $H = 10$ T in an intermediate temperature range 10 K $< T < T_c$, corresponding roughly to the liquid region of the vortex phase diagram, see Fig. 1(a). A similar behavior was previously reported for LSCO samples close to optimal doping [4,16]. This was interpreted in terms of induced excitations inside the vortex core [16]. In effect, H shifts and broadens the cusplike anomaly at T_c to a lower $T_c(H)$. The shift corresponds qualitatively to the shift in T_c as seen in Fig. 1(a). We note that the lower limit on the field effect might be related to the onset at $T_f = 10$ K of Cu moment freezing observed by μSR on the same sample [23].

Next, we plot in Fig. 2 the H dependence of the normalized intensity at $\mathbf{Q}_{IC}$. The *elastic* magnetic signal at $T = 2$ K exhibits a large enhancement in agreement with previous reports on samples of similar doping levels [17], and is well fitted by the equation $I \sim (H/H_{c2}) \ln(H_{c2}/H)$ predicted by Demler *et al.* [15]. Remarkably, the *inelastic* response at $T = 23$ K exhibits a field dependence at low-energy transfers similar to the *elastic* signal. The dependence on $\hbar\omega$ and $\mathbf{Q}$ of the intensity, taken at different temperatures and fields, is shown in Fig. 3. In Fig. 3(a) the intensity at $\mathbf{Q}_{IC}$ and at $H = 0$ T shows a smooth dependence on $\hbar\omega$ above T_c. At $T = 2$ K, an anomaly is exhibited at $\hbar\omega = 1.5$ meV below which intensity is reduced relative to that in the normal state. Figure 3(b) shows
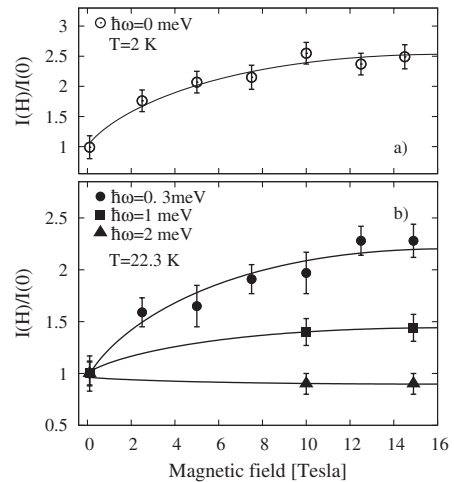

FIG. 1. (a) Phase diagram of underdoped LSCO ($x = 0.105$) as a function of a magnetic field H and temperature T [20]. ● are the measured values for the melting line of the vortex lattice [20]. The straight line is the position of T_c as a function of H. The dashed arrows indicate the T and H scans that are described in this Letter. (b) Two T scans at $\mathbf{Q}_{IC}$ and $\hbar\omega = 1$ meV transfer. △ (■) denote the measured intensity at $H = 10$ T ($H = 0$ T); solid lines are guides to the eye.

FIG. 2. Magnetic field dependence of the response at $\mathbf{Q}_{IC}$ for LSCO ($x = 0.105$). (a) Elastic response with $T = 2$ K. The solid line is a theoretical prediction [15]. (b) Inelastic response with $T = 22.3$ K; solid lines are guides to the eye.
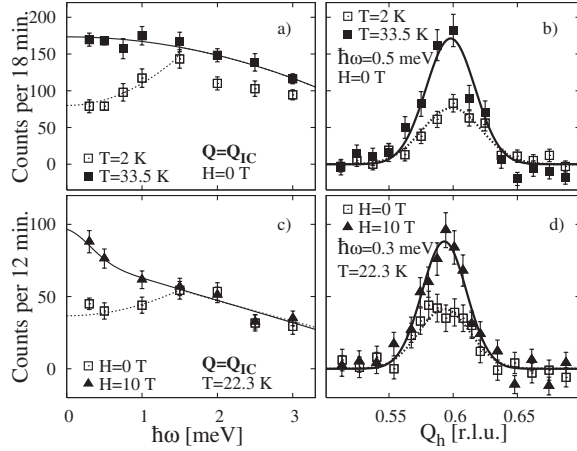
FIG. 3.   Energy and **Q** dependence of magnetic scattering from LSCO ($x = 0.105$). Energy scans are taken at $\mathbf{Q}_{IC}$: (a) comparison of the response in the SC phase with that of the normal state in zero field. (c) Effect of a 10 T field at $T = 22.3$ K. Q scans through $\mathbf{Q}_{IC}$: (b) comparison of the response in the SC phase with that of the normal state using $\hbar\omega = 0.5$ meV. (d) The effect of a 10 T field at 22.3 K, $\hbar\omega = 0.3$ meV, $E_f = 3.5$ meV. In this configuration the resolution (HWHM) was 0.03 meV. The data have been background subtracted and lines are guide to the eye.

the effect of cooling through T_c in the **Q** scan through $\mathbf{Q}_{IC}$ at $H = 0$ T and with $\hbar\omega = 0.5$ meV. Cooling from 33.5 K to 2 K results in a twofold reduction of the intensity at $\mathbf{Q}_{IC}$. Figures 3(c) and 3(d) show the change in the dependence on $\hbar\omega$ and **Q** of the intensity at fixed $T = 22.3$ K upon applying $H = 10$ T, respectively. The effect of the applied field, Fig. 3(c), is to remove the zero-field anomaly at $\hbar\omega = 1.5$ meV, and is similar to the effect of raising the temperature above T_c, Fig. 3(a). Note that here the magnetic-field-induced redistribution of spectral weight is limited to frequencies smaller than 1.5 meV.

To better appreciate the features of the excitation spectrum described above, a comparison with the SG signatures observed in optimally doped LSCO is relevant. (i) Although the details vary from experiment to experiment [4,16,24], magnetic field-induced excitations for $\hbar\omega < \Delta_{SG}$ and $T < T_c$ remain a common observation. (ii) For $\hbar\omega < \Delta_{SG}$, beside the suppression of intensity at low temperatures, an anomaly was reported at T_c that marks the closing of the spin gap as the temperature crosses T_c [25]. (iii) A strong suppression of the intensity below $\hbar\omega \approx \Delta_{SG}$ is observed in the SC state but not in the normal state [26]. In our underdoped LSCO sample, we infer from Figs. 1(b) and 3 similar spin gap features as in optimally doped LSCO. (i) An applied magnetic field acts to redistribute the spectral weight for 10 K $< T < T_c$ at $\hbar\omega < 1.5$ meV [Fig. 1(b) and 3(c)]. (ii) There is the cusp-like anomaly at T_c scanning the temperature for fixed energy below 1.5 meV. (iii) Energy scans reveal an anomaly at $\hbar\omega = 1.5$ meV which disappears in the normal state.

The field effect (i) alone provides compelling evidence for a spin gap at 1.5 meV. This is further supported by the cusp at T_c (ii), and consistent with the energy dependence (iii). We also find that the intensity at $\mathbf{Q}_{IC}$ displays the same functional dependence as that reported near optimal doping [27] as a function of $T > T_c$ or $\hbar\omega > 1.5$ meV.

We plot in Fig. 4(a) the dependence of the SG on T_c for LSCO and YBCO samples. Vertical and horizontal axes have been normalized by the values of the SG and SC critical temperature at optimal doping, respectively. The ratio $\Delta_{SG}/(k_B T_c)$ exhibits a strong downward multiplicative renormalization in the underdoped regime. This strong renormalization was also observed in YBCO [10,11], in which context it was attributed to inhomogeneities induced by oxygen doping [11]. We must, however, rule out this explanation for our $x = 0.105$ LSCO sample in view of the sharpness of the SC transition ($\Delta T_c = 1.5$ K). Instead, we believe that the proper interpretation of Fig. 4 is that of a universal doping dependence of Δ_{SG}/Δ_{SG}(Opt) with a linear regime that defines the approach to optimal doping and an underdoped regime whose defining property is a doping dependent $\Delta_{SG}/(k_B T_c)$.

It is remarkable that $\Delta_{SG}/(k_B T_c)$ decreases in the underdoped regime, since this is opposite to the doping dependence of most energy features (maximum SC gap or pseudogap) which are found, from ARPES measurements, to increase with underdoping [8,9]. In a phenomenological Fermi-liquid (PFL) picture, which is relatively successful at optimal doping, the SG is directly related to the SC gap. Although its maximum increases with underdoping, the slope of the SC d-wave gap has been measured to soften at the nodes with underdoping [28]. This means that the ratio $\Delta_{SG}/(k_B T_c)$ will decrease with underdoping [29]. In this picture, the anomaly at T_c in the temperature dependence displayed in Fig. 1(b) is caused by the closing of the SG upon entering the normal state. In Fig. 1(b) we show the calculated dependence on temperature of the zero-field
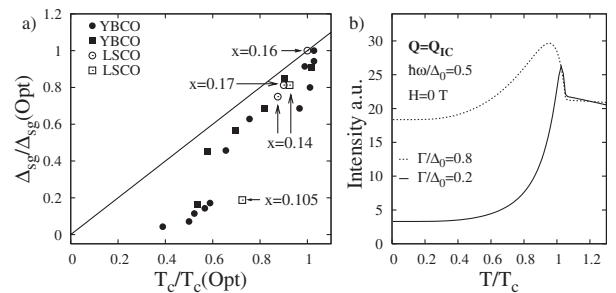

FIG. 4.   (a) Normalized Δ_{SG} vs normalized T_c for LSCO (open symbols) and YBCO ($\bullet$ [10] and $\blacksquare$ [11]). $\bigcirc$ are taken from previously published data, $x = 0.14$, $x = 0.16$, and $x = 0.17$ while $\square$ are from this work ($x = 0.105$) and Ref. [23] ($x = 0.14$). (b) Intensity at $\mathbf{Q}_{IC}$, at the energy transfer $\hbar\omega = 0.5\Delta_0$, and at $H = 0$ T vs T calculated within a phenomenological Fermi-liquid (PFL) approach. The solid (dashed) line uses the inverse lifetime $\Gamma = 0.2\Delta_0$ ($\Gamma = 0.8\Delta_0$).

neutron intensity at $\mathbf{Q}_{IC}$ and energy transfer $\hbar\omega = 0.5\Delta_0$ using the PFL parameters chosen to reproduce the T dependence at optimal doping [30]. A cusplike anomaly at T_c separates the normal regime from a d-wave SC regime with a decreasing residual intensity due to a phenomenological inverse lifetime of $\Gamma = 0.2\Delta_0$. In this spirit, to reproduce the measured residual spectral weight at low T in $x = 0.105$ LSCO would require $\Gamma \approx \Delta_{SG}$ resulting in strong broadening of the peak at T_c as shown in Fig. 4(b). Thus, the naïve PFL approach is unable to reconcile a sharp anomaly at T_c (Δ_{SG}) for small enough $\hbar\omega$ (T) and the large, compared to optimal doping, residual intensity in the low-T (low-$\hbar\omega$) tail of Fig. 1(b) [Fig. 3(a)].

One way to explain the large residual intensity at low temperatures and energies in strongly underdoped HTSC could be in terms of a two-component model. The first component would correspond to the gapped response seen around optimal doping. The second (quasielastic) component could be related to the slowing down of the magnetic signal observed by μSR [31], and the central mode recently reported in YBCO ($T_c = 18$ K) [32]. The fact that both components develop well-defined peaks at $\mathbf{Q}_{IC}$ suggests an intertwinning of the two components on a nanometer length scale. This could possibly be explained within a picture of static and fluctuating stripes, although it remains a challenge to quantify this picture to describe the detailed field and temperature effects of the magnetic response reported here.

In summary, we have reported a dramatic field-induced enhancement of the INS response for low-energy transfers in a restricted temperature range. The field dependence suggests that the approach developed [15] to explain the previously observed field-induced static magnetic order [16] (see also Ref. [17]) may also be applicable to the dynamics. The field-induced excitation may be more difficult to capture in a PFL approach. Furthermore, we have identified a SG, $\Delta_{SG} = 1.5$ meV, in underdoped LSCO $x = 0.105$. The strong renormalization of the SG as a function of underdoping appears to be a generic feature of HTSC and might signal a profound modification of the underlying electronic excitations as the system approaches its Mott insulating phase. In particular, we are not dealing with a simple damping or smearing of the gap. The cusp upon heating through T_c implies that the gap remains well defined as it closes in energy, which also indicates that the SG is directly coupled to the SC-order parameter.

*Electronic address: joel.mesot@psi.ch
[1] S. Wakimoto *et al.*, Phys. Rev. Lett. **92**, 217004 (2004).
[2] T. E. Mason *et al.*, Phys. Rev. Lett. **68**, 1414 (1992).
[3] K. Yamada *et al.*, Phys. Rev. Lett. **75**, 1626 (1995).
[4] R. Gilardi *et al.*, Europhys. Lett. **66**, 840 (2004).
[5] B. Lake *et al.*, Nature (London) **400**, 43 (1999).
[6] P. Bourges, *Neutron Scattering in Novel Materials* (World Scientific, Singapore, 2000).
[7] P. Dai *et al.*, Science **284**, 1344 (1999).
[8] J. C. Campuzano *et al.*, *Physics of Superconductors* (Springer, New York, 2004).
[9] A. Damascelli *et al.*, Rev. Mod. Phys. **75**, 473 (2003).
[10] P. Bourges *et al.*, Physica (Amsterdam) **B215**, 30 (1995).
[11] P. Dai *et al.*, Phys. Rev. B **63**, 054525 (2001).
[12] C.-H. Lee *et al.*, J. Phys. Soc. Jpn. **69**, 1170 (2000).
[13] S. A. Kivelson *et al.*, Rev. Mod. Phys. **75**, 1201 (2003).
[14] S. Chakravarty *et al.*, Phys. Rev. B **63**, 094503 (2001).
[15] E. Demler *et al.*, Phys. Rev. Lett. **87**, 067202 (2001).
[16] B. Lake *et al.*, Science **291**, 1759 (2001).
[17] B. Lake *et al.*, Nature (London) **415**, 299 (2002).
[18] T. Nakano *et al.*, J. Phys. Soc. Jpn. **67**, 2622 (1998).
[19] K. Yamada *et al.*, Phys. Rev. B **57**, 6165 (1998).
[20] R. Gilardi *et al.*, Eur. Phys. J. B **47**, 231 (2005).
[21] U. K. Divakar *et al.*, Phys. Rev. Lett. **92**, 237004 (2004).
[22] R. Gilardi *et al.*, Phys. Rev. Lett. **88**, 217003 (2002).
[23] J. Chang *et al.* (to be published).
[24] J. M. Tranquada *et al.*, Phys. Rev. B **69**, 174507 (2004).
[25] T. E. Mason *et al.*, Phys. Rev. Lett. **77**, 1604 (1996); T. E. Mason *et al.*, Physica (Amsterdam) **B199**, 284 (1994).
[26] N. B. Christensen *et al.*, Phys. Rev. Lett. **93**, 147002 (2004).
[27] G. Aeppli *et al.*, Science **278**, 1432 (1997).
[28] J. Mesot *et al.*, Phys. Rev. Lett. **83**, 840 (1999); S. V. Borisenko *et al.*, Phys. Rev. B **66**, 140509(R) (2002).
[29] A. Schnyder *et al.*, Phys. Rev. B **70**, 214511 (2004).
[30] The imaginary part of the magnetic susceptibility of a weakly interacting gas of SC quasiparticles is calculated within the random phase approximation. The fermiology parameters are chosen to reproduce the T dependence of the INS response at optimal doping in LSCO (see Ref. [28]), with the ansatz $\Delta(\mathbf{Q}, T) = \Delta_0(\mathbf{Q}) \times \sqrt{1 - (T/T_c)^4}$ and $\Delta_0(0, \pi) = 10$ meV for the T dependence of the d-wave SC gap.
[31] Ch. Niedermayer *et al.*, Phys. Rev. Lett. **80**, 3843 (1998).
[32] C. Stock *et al.*, Phys. Rev. B **73**, 100504(R) (2006).