Large magnetic field-induced spectral weight enhancement of high-energy spin excitations in La$_{1.88}$Sr$_{0.12}$CuO$_4$

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We report electronic Raman scattering experiments on a superconducting La$_{1.88}$Sr$_{0.12}$CuO$_4$ single crystal in a magnetic field. At low temperatures, the spectral weight of the high-energy two-magnon peak increases linearly with field and is amplified by a factor of more than two at 14 T. The effect disappears at elevated temperatures and is not present in undoped La$_2$CuO$_4$. This observation is discussed in terms of an electronically inhomogeneous state in which the field enhances the volume fraction of a phase with local antiferromagnetic order at the expense of the superconducting phase.

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Shortly after the discovery of high temperature superconductivity, antiferromagnetic Mott insulating and superconducting states were shown to be directly adjacent in the phase diagram of the doped copper oxides. This early observation still serves as primary evidence of the prominent role of Coulomb correlations in the mechanism of high temperature superconductivity. Recent neutron scattering [1, 2, 3], nuclear magnetic resonance [4, 5], and muon spin rotation [6] experiments have indicated that the interplay between these two states is more delicate than had long been assumed. In underdoped copper oxides with hole concentrations per copper atom, $x$, around 1/8, antiferromagnetic order and superconductivity in underdoped La$_{2-x}$Sr$_x$CuO$_4$ with $x$ $\sim$ 1/8 is enhanced by a factor of more than two in a magnetic field at 14 T. This profound spectral weight renormalization of the highest-energy, local spin flip excitations cannot be understood by the soft-mode behavior invoked to explain the field dependence of the long-wavelength spin excitations observed in low-energy spectroscopies. It thus requires a revision of our understanding of the interplay between antiferromagnetic order and superconductivity in underdoped copper oxides. Possible implications will be discussed below.

A single crystal of La$_{1.88}$Sr$_{0.12}$CuO$_4$ was grown by the travelling solvent floating zone technique as described previously [1]. Its superconducting transition temperature, $T_c = 34$ K, was determined by monitoring the diamagnetic response in a magnetometer. An undoped La$_2$CuO$_4$ crystal was grown by the same method and annealed under Ar flow at 950°C in order to remove oxygen interstitials. After the annealing procedure, its Néel temperature (also determined by magnetometry) was 320 K. The Raman scattering measurements were performed in quasi-back scattering geometry using a triple monochromator (Dilor $xy$), a charge coupled device (CCD) detector, and a laser wave length of 514.5 nm. The scattered light was collected along the crystallographic c-axis. The magnetic field dependent data were obtained using a superconducting magnet (Oxford Instruments) with a maximum field of 14 T, operated in a temperature range from 4.2 to 300 K. The samples were mounted in a continuous helium flow cryostat installed in the magnet, and the field was applied perpendicular to the CuO$_4$ planes. The beam spot on the sample surface was monitored by a specially designed electronic sensor and a camera installed inside the spectrometer. By adjusting the sample position to compensate for magnetostriction and thermal expansion of the sample mount, it proved possible to keep the beam position fixed during field and temper-
perature changes, thereby minimizing systematic errors due to variations in surface morphology. Further experimental precautions included field-cooling the samples through the superconducting transition, in order to avoid strains due to flux trapping. The laser power was about 10W/cm$^2$. By examining the intensity ratio of the Stokes and anti-Stokes spectra, the temperature of the illuminated region of the sample was estimated to be less than 5 K at low temperature. Spectral corrections were made for the frequency dependence of the collection optics and spectrometers, as well as the detector sensitivity. The spectra were taken in $x'y'$ geometry, that is, with the incident and final photon polarization states at an angle of 45° from the Cu-O bonds. (The small orthorhombic distortion of the copper oxide layers can be ignored.) This yields Raman spectra of $B_{1g}$ symmetry, where the two-magnon peak has the largest amplitude.

Figures 1 and 2 show $B_{1g}$ Raman spectra collected for the La$_{1.88}$Sr$_{0.12}$CuO$_4$ crystal at different temperatures and magnetic fields. The data in zero field are in excellent agreement with prior work on similar samples [17, 18]. Upon cooling, they exhibit a gradual evolution from a single broad peak around 3000 cm$^{-1}$ at room temperature to a two-peak profile at base temperature. The two peaks in the low temperature profile are also seen in the electronic Raman spectra of doped nickelates [19], where phases with “stripe” order of spin and charge are well documented. They have thus been ascribed to two-magnon excitations propagating along and perpendicular to the charged domain walls of a “striped” phase [17].

The central result of this paper is the large magnetic field-induced intensity enhancement of the low temperature two-magnon Raman spectrum shown in Fig. 1. While the two-peak line shape is only weakly affected by the field, a field of 14T leads to an intensity increase of more than a factor of two compared to the zero-field data. Within the experimental error, the field dependence of the energy-integrated spectral weight is linear (Fig. 3), in contrast to the sublinear field dependence of the magnetic Bragg reflections observed in some of the neutron diffraction experiments [2, 3].

The profound magnetic field induced renormalization of the high-energy magnon spectrum is unexpected. Several cross checks were performed in order to definitively rule out experimental artefacts. First, the experiment was repeated at T = 50K, 100K, and room temperature, where the spectra were found to be field independent within the experimental error. This is consistent with the temperature dependence of the static magnetic order determined in the neutron diffraction experiments [1, 2, 3]. Second, the intensities of several $A_{1g}$ phonon modes were monitored as a function of magnetic field, and no field dependence was observed within the experimental error.

Third, the experiment was repeated under the same
conditions on an insulating, antiferromagnetically ordered La$_2$CuO$_4$ crystal. Since the magnon bandwidth is much smaller than the Mott-Hubbard gap of 2 eV, the field affects the electrons only via the Zeeman term $g\mu_B H$ in the Hamiltonian. In a 14T field, this term leads to a splitting of the two degenerate magnon branches of order 10 cm$^{-1}$, more than two orders of magnitude lower than the energy of the two-magnon peak. Given the large intrinsic width of this peak, a field-induced intensity or lineshape renormalization should thus not be observable for undoped La$_2$CuO$_4$. The B$_{1g}$ Raman spectra displayed in Fig. 4 demonstrate that this expectation is indeed confirmed by our experiment. Within the experimental error, the two-magnon profile of La$_2$CuO$_4$ (which again agrees very well with prior work on this system [17]) is unaffected by a 14T field. These experimental cross checks provide reassurance that the magnetic field effect we have observed for the two-magnon peak in La$_{1.88}$Sr$_{0.12}$CuO$_4$ is genuine.

The magnetic field dependence of electronic excitations in high temperature superconductors has been the subject of several prior Raman scattering experiments. For instance, recent work on lightly doped, nonsuperconducting La$_{1-x}$Sr$_x$CuO$_4$ with $x \leq 0.03$ has uncovered a field-induced renormalization of low-energy magnons with energies of order 10 cm$^{-1}$ (Ref. [20]). Since this energy scale is comparable to the Zeeman energy, these observations are amenable to an interpretation in the framework of the conventional spin wave theory, at least on a qualitative level [20]. An earlier Raman scattering experiment investigated electronic excitations in highly overdoped, superconducting Tl$_2$Ba$_2$CuO$_{6+\delta}$ in fields exceeding the upper critical field [21]. As a consequence of the suppression of superconductivity, the Raman intensity below the superconducting energy gap, $2\Delta$, was observed to increase with field, while that of the broad density-of-states peak above $2\Delta$ was reduced, in qualitative agreement with the standard BCS theory of superconductivity.

The field-induced enhancement of the high-energy Raman intensity in La$_{1.88}$Sr$_{0.12}$CuO$_4$ defies a description in terms of such conventional models. As already pointed out above, the two-magnon peak energy is more than two orders of magnitude larger than the Zeeman energy in a 14T field. The mechanism invoked to explain the field-induced renormalization of low-energy magnons in lightly doped La$_{1-x}$Sr$_x$CuO$_4$ (Ref. [20]) is therefore not applicable to our system. Likewise, our results cannot be explained by the BCS theory, because the energy of the two-magnon Raman peak is much larger than that of the superconductivity-induced $2\Delta$ peak [22, 23]. Further, we have shown that its amplitude increases with field, in contrast to the decrease predicted by the BCS theory and observed in Tl$_2$Ba$_2$CuO$_{6+\delta}$ [21].

Since the two-magnon peak is a signature of local antiferromagnetic interactions, our data are qualitatively consistent with the notion of a magnetic field-induced enhancement of antiferromagnetic spin correlations developed on the basis of low-energy spectroscopy experiments [17, 24]. However, they extend the energy range probed by the earlier experiments by more than an order of magnitude and demonstrate that a field of 14T profoundly affects the magnon spectrum over its entire band width up to the highest-energy, local spin-flip excitations.

This surprising observation is difficult to reconcile
with theories based on soft-mode behavior controlled by a nearby quantum critical point \[16\]. The fact that the characteristic two-peak lineshape of the two-magnon peak is only weakly affected by the field, while its amplitude increases linearly, rather suggests a much simpler picture based on the coexistence of two phases with very different electronic structures: a phase with localized electrons and well-developed local antiferromagnetic order that gives rise to the two-magnon peak (such as a “striped” phase); and a phase that gives a much weaker contribution to the high-energy Raman spectrum (such as a phase dominated by fermionic quasiparticles that also sustains superconductivity). The intensity enhancement of the two-magnon peak then simply reflects a magnetic field-induced increase of the volume fraction of the former phase. As the temperature is increased above the critical point for spontaneous phase separation, the system enters a homogeneous phase without static magnetic order. The magnetic field effect is hence expected to disappear, as experimentally observed (Fig. 2). This is not inconsistent with scenarios in which antiferromagnetic order is nucleated by vortices \[8, 9, 10, 11, 12, 13, 14, 15\]. Note, however, that vortices do not seem to be \textit{required} for antiferromagnetic order, as manifestations of static magnetic order are present even in zero field \[1, 21, 22\]. This suggests that the primary effect of the magnetic field is to shift the thermodynamic balance of the antiferromagnetic and superconducting phases \[16\], not to create the vortices. Our data are also consistent with a coexistence of superconducting and magnetically ordered phases on a mesoscopic scale, as observed for instance for metallic and charge-ordered insulating phases in some manganites \[20\]. While our Raman experiments are sensitive predominantly to regions with magnetic order, recent infrared experiments on the Josephson plasma resonance in La\(_{1.875}\)Sr\(_{0.125}\)CuO\(_4\) have provided complementary evidence of a spatially inhomogeneous \textit{superconducting} state \[27\]. A very recent high-field magnetoresistance study \[28\] has come to a similar conclusion.

In conclusion, we have reported the discovery of a large magnetic field-induced spectral weight enhancement of the two-magnon Raman peak in superconducting La\(_{1.88}\)Sr\(_{0.12}\)CuO\(_4\). These data are most naturally explained in a two-phase coexistence scenario, where the magnetic field enhances the volume fraction of a phase with local magnetic order at the expense of the superconducting phase. This indicates that these two phases are separated by a first-order transition, and not by a quantum critical point.

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[1] S. Katano, M. Sato, K. Yamada, T. Suzuki, and T. Fukase, Phys. Rev. B \textbf{62}, R14677 (2000).
[2] B. Lake \textit{et al.}, Nature \textbf{415}, 299 (2002).
[3] B. Khaykovich \textit{et al.}, Phys. Rev. B \textbf{66}, 014528 (2002).
[4] V.I. Mitrovic \textit{et al.}, Nature \textbf{413}, 501 (2002).
[5] K. Kakuyanagi, K. Kumagai, Y. Matsuda, and M. Hasegawa, Phys. Rev. Lett. \textbf{90}, 197003 (2003).
[6] R.I. Miller \textit{et al.}, Phys. Rev. Lett. \textbf{88}, 137002 (2002).
[7] B. Lake \textit{et al.}, Science \textbf{291}, 1759 (2001).
[8] D.P. Arovas, A. J. Berlinsky, C. Kallin, and S.C. Zhang, Phys. Rev. Lett. \textbf{79}, 2871 (1997).
[9] A. Himeida and M. Ogata, Phys. Rev. B \textbf{60}, R9935 (1999).
[10] J.X. Zhu and C.S. Ting, Phys. Rev. Lett. \textbf{87}, 147002 (2001).
[11] H.D. Chen, J.P. Hu, S. Capponi, E. Arrigoni, and S.C. Zhang, Phys. Rev. Lett. \textbf{89}, 137004 (2002).
[12] M. Franz, D.E. Sheehy, and Z. Tesanovic, Phys. Rev. Lett. \textbf{88}, 257005 (2001).
[13] A. Ghosal, C. Kallin, and A.J. Berlinsky, Phys. Rev. B \textbf{66}, 214502 (2002).
[14] J.X. Zhu, I. Martin, and A.R. Bishop, Phys. Rev. Lett. \textbf{89}, 067003 (2003).
[15] B.M. Andersen, P. Hedegard, and H. Bruus, Phys. Rev. B \textbf{67}, 134528 (2003).
[16] E. Demler, S. Sachdev, and Y. Zhang, Phys. Rev. Lett. \textbf{87}, 067202 (2001); Y. Zhang, E. Demler, and S. Sachdev, Phys. Rev. B \textbf{66}, 094501 (2002); S. Sachdev and E. Demler, Phys. Rev. B \textbf{69}, 144504 (2004).
[17] S. Sugai and N. Hayamizu, J. Phys. Chem. Solids \textbf{62}, 177 (2001), and references therein.
[18] J.G. Naeini, X. K. Chen, J. C. Irwin, M. Okuya, T. Kimura, and K. Kishio, Phys. Rev. B \textbf{59}, 9642 (1999).
[19] G. Blumberg, M.V. Klein, and S.-W. Cheong, Phys. Rev. Lett. \textbf{80}, 564 (1998).
[20] A. Gozar, B.S. Dennis, G. Blumberg, S. Komiya, and Y. Ando, Phys. Rev. Lett. \textbf{93}, 027001 (2004).
[21] G. Blumberg, M. Kang, and M.V. Klein, Phys. Rev. Lett. \textbf{78}, 2461 (1997).
[22] X.K. Chen, J.C. Irwin, H.J. Trodahl, T. Kimura, and K. Kishio, Phys. Rev. Lett. \textbf{73}, 3290 (1994).
[23] S. Sugai and T. Hosokawa, Phys. Rev. Lett. \textbf{85}, 1112 (2000).
[24] T. Suzuki, T. Goto, K. Chiba, T. Shinoda, T. Fukase, H. Kimura, K. Yamada, M. Ohashi, and Y. Yamaguchi, Phys. Rev. B \textbf{57}, R3229 (1998).
[25] S. Ohsugi, Y. Kitaoka, H. Yamanaka, K. Ishida, and K. Asayama, J. Phys. Soc. Jpn. \textbf{63}, 2057 (1999).
[26] For a review, see A. Moreo, S. Yunoki, and E. Dagotto, Science \textbf{283}, 2034 (1999).
[27] S.V. Dordevic, S. Komiya, Y. Ando, and D.N. Basov, Phys. Rev. Lett. \textbf{91}, 167401 (2003).
[28] S. Komiya and Y. Ando, Phys. Rev. B \textbf{70}, 060503(R) (2004).