Magnetic-Field-Induced Soft-Mode Quantum Phase Transition in the High-Temperature Superconductor $La_{1.855}Sr_{0.145}CuO_4$: An Inelastic Neutron-Scattering Study

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(Received 21 January 2009; published 30 April 2009)

Inelastic neutron-scattering experiments on the high-temperature superconductor $La_{1.855}Sr_{0.145}CuO_4$ reveal a magnetic excitation gap Δ that decreases continuously upon application of a magnetic field perpendicular to the CuO_2 planes. The gap vanishes at the critical field required to induce long-range incommensurate antiferromagnetic order, providing compelling evidence for a field-induced soft-mode driven quantum phase transition.

DOI: 10.1103/PhysRevLett.102.177006 PACS numbers: 74.72.Dn, 74.25.Nf, 78.70.Nx

Driven by the continued theoretical focus on strong electron correlations and magnetism as a route to unconventional superconductivity [1], the past two decades have seen tremendous efforts invested to characterize the momentum and energy dependence of magnetic fluctuations in cuprate high-T_c superconductors. One of the most remarkable and encouraging results emerging from these studies is that upon entering the superconducting state optimally and overdoped hole [2–6] and electron-doped [7] cuprates develop an excitation gap. This phenomenon manifests itself as a complete suppression of all magnetic fluctuations below a material-dependent energy scale, sometimes referred to as the spin gap, which scales with T_c [7,8]. The correlation between superconductivity and low-energy spin fluctuations is, however, much less clear in underdoped cuprates, where linear scaling between the gap energy and T_c breaks down [5,8,9].

Particularly revealing studies of the excitation gap have involved the application of a magnetic field perpendicular to the CuO_2 planes. In the electron-doped compound $Nd_{1.85}Ce_{0.15}CuO_4$, it has been demonstrated that the excitation gap decreases linearly with increasing magnetic field and extrapolates to zero at H_{c2}—the upper critical field for superconductivity [10]. The situation is more complex in hole-doped $La_{2-x}Sr_xCuO_4$ (LSCO), where application of a magnetic field (i) for $x \gtrsim 0.15$ tends to induce spectral weight below the zero-field gap [11–13] and (ii) for $x \lesssim 0.15$ enhances [14,15] the characteristic zero-field incommensurate (IC) stripe [16] or spin density wave (SDW) order [17] or even induces such order where none was present in zero field [14,18]. The latter observations have found formal expression in a Ginzburg-Landau (GL) model for competing SDW and superconducting orders [19] which predicts the existence of a line of quantum critical points (QCPs) in the $T = 0$ doping-field phase diagram, separating superconducting states with and without coexisting magnetic order (SC + SDW and SC, respectively, in the notation of Ref. [19]). However, one of the key expectations for continuous quantum phase transitions in general [20,21] and of the GL model [19] in particular—the existence of a field-induced soft mode in the spin excitation spectrum—has never been firmly established.

In this Letter, we present an inelastic neutron-scattering study of the low-energy spin fluctuations in $La_{1.855}Sr_{0.145}CuO_4$, which in the absence of a magnetic field is a homogeneous, magnetically disordered superconductor with an excitation gap. We show that the gap decreases as the field-induced transition to a magnetically ordered SC + SDW state is approached and tends to zero at the point where SDW order sets in. Our discovery strongly suggests that the $T = 0$ doping-field phase diagram of LSCO hosts a line of soft-mode driven SDW QCPs terminating, for $H = 0$ T, near $x = 1/8$.

The experiments were performed on the cold triple axis spectrometers IN14 at Institut Laue-Langevin, Grenoble, France and PANDA at FRM-II, Munich, Germany. On both instruments, cooled Be filters were inserted after the sample to avoid higher-order contamination of the scattered beam of 5 meV neutrons. The setup gave an energy resolution of 150 μeV (FWHM) or better, ensuring that there is no contribution from elastic scattering at the lowest energy transfers probed by our measurements. The sample consisted of two crystals (total mass ≈ 3.5 g, $T_c \approx 36$ K), cut from the same traveling-solvent floating-zone grown [22] rod, and coaligned to within less than 1°. It was mounted in vertical field cryomagnets with the CuO_2 planes horizontal, allowing access to momentum transfers (Q_h, Q_k, 0). In tetragonal notation ($a \approx b = 3.81$ Å, $c = 13.2$ Å), the low-energy magnetic response of superconducting LSCO peaks at a quartet of wave vectors $\mathbf{Q}_{\rm IC} = (1/2 \pm \delta, 1/2, 0), (1/2, 1/2 \pm \delta, 0)$ as shown in the inset in Fig. 1(b). We present data, recorded at $T \leq 3$ K, as a function of momentum $\mathbf{Q}$, energy transfer $\hbar\omega$, and external magnetic field H. Our results represent ground state properties of $La_{1.855}Sr_{0.145}CuO_4$ since the typical energies $\hbar\omega$ of the spin fluctuations studied are larger than the thermal energy k_BT. This also implies that measured peak amplitudes translate directly into magnetic susceptibility $\chi''(\mathbf{Q}_{\rm IC}, \omega)$ because the thermal population factor $[1 - \exp(-\hbar\omega/k_BT)]^{-1} \approx 1$ is essentially irrelevant.

We start by noting that $La_{1.855}Sr_{0.145}CuO_4$ develops long-range magnetic order with $\delta \approx 0.13$ for $H > H_c = 7 \pm 1$ T [14], as demonstrated by the appearance of sharp Bragg peaks at $\mathbf{Q}_{\rm IC}$ which are absent for $H < H_c \ll H_{c2}$ [see Fig. 1(a)]. Figures 1(b)–1(d) show the variation with field of scans through $\mathbf{Q}_{\rm IC}$ taken at $\hbar\omega = 0.5$, 1, and 2 meV, respectively [23]. For all three energies, the magnetic response is completely suppressed at the lowest fields shown (blue symbols) but becomes finite and peaked at $\mathbf{Q}_{\rm IC}$ upon application of higher fields (red and green symbols). Remarkably, in the case of $\hbar\omega = 2$ meV, shown in Fig. 1(d), a field of just 1 T is sufficient to induce an unambiguous magnetic excitation where none existed in zero field. Increasing the field leads to further enhancement of this signal.

Figures 1(b)–1(d) show that in moderate external fields $La_{1.855}Sr_{0.145}CuO_4$ is nonresponsive at low-energy transfers; i.e., there is a gap in the magnetic excitation spectrum. In Fig. 1(e), we illustrate this point in a different manner, by plotting the $\hbar\omega$ dependence of scans through $\mathbf{Q}_{\rm IC}$, all obtained at one fixed field $H = 2.5$ T. For $\hbar\omega = 1$ meV there is no magnetic response, while the $\hbar\omega = 2$, 4, and 8 meV scans all display incommensurate peaks at $\mathbf{Q}_{\rm IC}$. It is noteworthy that this magnetic signal has a nonmonotonic energy dependence with an intensity minimum between 2 and 8 meV.

Next, in Fig. 2, we show the energy dependence of the spectral weight at $\mathbf{Q}_{\rm IC}$ for $H = 0$, 2.5, and 7 T. In agreement with previous studies near optimal doping [2–4], the zero-field magnetic response is gapped. We define the excitation gap Δ as the energy scale below which no spectral weight can be observed. The $H = 0$ T measurement then yields $\Delta(0\ \mathrm{T}) = 4 \pm 0.5$ meV. In a 2.5 T field, complete suppression of spectral weight takes place only for $\hbar\omega < 1.25$ meV, and hence $\Delta(2.5\ \mathrm{T}) = 1.25 \pm 0.5$ meV. As was clear already from Fig. 1(e), the 2.5 T spectrum displays noticeable local maxima and minima near 2 and 4 meV, respectively. A comparison of the 0 and 2.5 T spectra reveals that the field-induced spectral weight

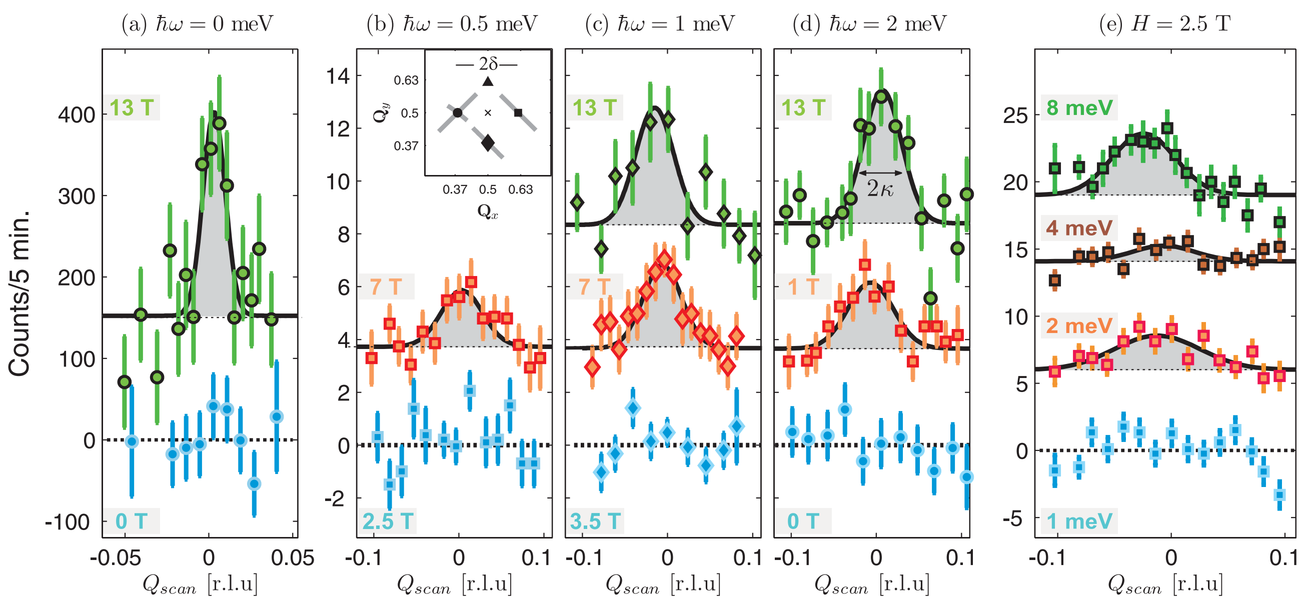

FIG. 1 (color online). (a)–(e) Elastic and inelastic $\mathbf{Q}$ scans through $\mathbf{Q}_{\rm IC} = (1/2 \pm \delta, 1/2, 0), (1/2, 1/2 \pm \delta, 0)$ as indicated in the inset of (b) [23]. We use $\mathbf{Q} = \mathbf{Q}_{\rm IC} + \mathbf{Q}_{\rm scan}$. (a) Elastic $\mathbf{Q}$ scans at external magnetic fields 0 and 13 T. (b)–(d) Magnetic field dependence of $\mathbf{Q}$ scans recorded at fixed energy transfers $\hbar\omega = 0.5$, 1, and 2 meV, respectively. (e) Energy dependence of $\mathbf{Q}$ scans obtained with $H = 2.5$ T. For clarity, the scans in (a)–(e) have been offset vertically with respect to each other. The dashed lines are guides to the eye, and the solid lines are Gaussian fits with a linear background. At 13 T, we find elastic and inelastic correlation lengths $\xi(0\ \mathrm{meV}) \sim 150$ Å and $\xi(2\ \mathrm{meV}) \sim 48$ Å obtained from the half width at half maximum $1/\kappa$ of the fitted peak.

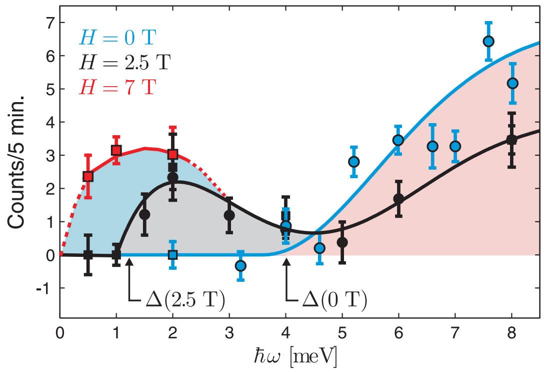

FIG. 2 (color online). Inelastic neutron-scattering response at $\mathbf{Q}_{\mathrm{IC}}$ as a function of energy transfer $\hbar\omega$ for $H = 0$ T (blue line), $H = 2.5$ T (black line), and $H = 7$ T (red line). The square points are deduced from Gaussian fits to $\mathbf{Q}$ scans as shown in Fig. 1, while circular points are from three-point (background-$\mathbf{Q}_{\mathrm{IC}}$-background) scans. All lines are guides to the eye.

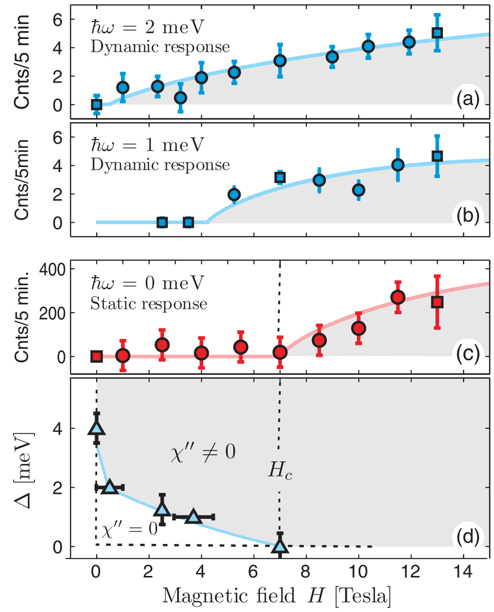

FIG. 3 (color online). (a),(b) Magnetic field dependence of the inelastic neutron response at $\mathbf{Q}_{\mathrm{IC}}$ with $\hbar\omega = 2$ and 1 meV, respectively. (c) Elastic neutron response at $\mathbf{Q}_{\mathrm{IC}}$ as a function magnetic field. In (a)–(c) the square points are deduced from Gaussian fits to $\mathbf{Q}$ scans as shown in Fig. 1, while circular points are from three-point (background-$\mathbf{Q}_{\mathrm{IC}}$-background) scans. All lines are guides to the eye. (d) Field dependence of the excitation gap Δ.

in the range $1.25 \lesssim \hbar\omega \lesssim 4$ meV has been transferred from energies above $\Delta(0\ \mathrm{T})$. Finally, at 7 T, which is near the onset field for static SDW order, spin excitations are observed even at the lowest energy transfers probed [see also Fig. 1(b)]. This indicates that the gap has completely collapsed: $\Delta(7\ \mathrm{T}) = 0 \pm 0.5$ meV.

The field dependencies of the dynamic response at $\mathbf{Q}_{\mathrm{IC}}$ for $\hbar\omega = 2$ and 1 meV are shown in Fig. 3(a) and 3(b), respectively. For $\hbar\omega = 2$ meV field-induced spin excitations are observed already for $H = 0.5 \pm 0.5$ T [see also Fig. 1(d)], and the magnetic signal increases continuously with increasing H. By contrast, for $\hbar\omega = 1$ meV the response is completely suppressed for $H \lesssim 3.7 \pm 0.8$ T, and excitations appear only at larger fields. For comparison, Fig. 3(c) shows that the onset field for long-range static SDW order is $H_c = 7 \pm 1$ T [14].

We have now arrived at the main result of this Letter. Figure 3(d) displays the field dependence of the excitation gap Δ, obtained by combining the data in Figs. 2, 3(a), and 3(b). The gap is extremely sensitive to the application of a magnetic field: Following an initial dramatic drop, Δ subsequently softens more slowly and finally vanishes at the critical field $H_c \ll H_{c2}$ which marks the onset of long-range incommensurate magnetic order.

We start our discussion by pointing out a strong resemblance of the field-induced spectral evolution in $La_{1.855}Sr_{0.145}CuO_4$ to that which takes place when the SC to SC + SDW transition is approached through changes in chemical composition. Increasing levels of Zn replacing Cu in $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$, which for $y = 0$ has a well-developed gap, leads to suppression of T_c and to a gradual shift of Δ [24]. Eventually, for $y = 0.017$, long-range SDW order coexisting with SC sets in, and no excitation gap can be resolved [24]. Similarly, as a function of decreasing Sr content, $La_{2-x}Sr_xCuO_4$ evolves from a SC state with $\Delta \sim 4$ meV, for $x > 0.13$, to a SC + SDW state with significant peaked spectral weight below this energy scale [25]. What we have discovered is that the same spectral evolution can be accomplished continuously by application of a magnetic field and with no variation in chemical disorder.

One of the hallmarks of a continuous quantum phase transition is the existence of a mode in the excitation spectrum which responds to changes in an experimentally tunable parameter (such as magnetic field, pressure, or doping) by softening towards zero, causing qualitative changes in the ground state wave function once the $\hbar\omega = 0$ limit is reached [20,21]. Quantum disordered spin dimer systems have proven to be a fertile ground for studies of this canonical behavior. For example, in the case of $TlCuCl_3$, the Zeeman splitting of excited triplet states in a magnetic field leads to a linear reduction of the singlet-triplet gap and to Bose-Einstein condensation of magnons and gapless spin excitations above the critical field required to fully close the gap [26]. By analogy, we interpret our observation of a gradual reduction of Δ and closure of this gap at the onset field for magnetic order as evidence that the field-induced SC to SC + SDW transition is a continuous quantum phase transition. An early demonstration of nearly singular magnetic fluctuations and scaling [27] in a sample of almost identical composition to ours provides further support for this interpretation.

The observed field dependence of Δ is in qualitative agreement with a key prediction of GL theory for SC to SC + SDW quantum phase transitions [19], namely, the appearance of a field-induced soft precursor mode (a $S = 1$

collective "spin resonance," associated with oscillations of the SDW order parameter about zero [19]) centered, for $H \leq H_c \leq H_{c2}$, at energy $\epsilon(H) = \epsilon(0) + C_1(H/H_{c2}) \times \log(H_{c2}/H)$, with $\epsilon(0)$ and C_1 being constants. To make the connection to GL theory, we must interpret Δ as marking the low-energy tail of the soft mode. In turn, this suggests to view the low-energy peak in the complex spectral line shape at 2.5 T (see Fig. 2) as the soft mode. The existence of a field-induced spectral peak was first reported in LSCO $x = 0.16$ [11]. Subsequent experiments on $x = 0.17$ [12] and $x = 0.18$ [13] samples gave no indications of a well-defined mode, although in-gap spectral weight was reported in both cases. Our results reaffirm the existence of a field-induced mode in the SC phase of LSCO and suggest, as would be implied by the conditions of validity of the GL model [19], that it is most easily observed when in close proximity to the line of continuous SC to SC + SDW quantum phase transitions.

More generally, closure of the excitation gap appears to be a universal phenomenon in cuprates hosting quantum phases in which superconductivity can coexist with magnetic order. In $YBa_2Cu_3O_{6+p}$ (YBCO), the gap collapses abruptly for $p \approx 0.5$ [8], and a robust SC phase with incommensurate, quasistatic fluctuations (an electronic nematic) emerges at lower doping levels [28]. On the other hand, in $Nd_{1.85}Ce_{0.15}CuO_4$ where SC and magnetism do not coexist, the excitation gap extrapolates to zero only at H_{c2} [10].

These considerations raise the possibility that, when coexistence of superconductivity and magnetism is an issue, two separate energy scales need to be considered: (i) the zero-field gap $\Delta(0\ \mathrm{T})$ related to superconductivity and (ii) a gap related to magnetic order. Further, these scales may display distinct field dependencies. Related ideas about spectral separation were presented in Ref. [25] in the context of spatial phase separation between SC and magnetically ordered regions. By not requiring $\epsilon(0) = \Delta(0\ \mathrm{T})$, GL theory has spectral separation built in with no need for phase separation beyond what is implied by the soft mode being stabilized near vortices [19]. Interestingly, quantum Monte Carlo computations of the excitation spectrum of a mixture of magnetically ordered and disordered patches, intended to model the effect of SDW order pinned by vortices or impurities, does produce a low-energy peak below the excitation gap of the fully disordered system [29]. Further experimental and theoretical work is needed to clarify these issue.

In summary, we have discovered that, under the influence of a magnetic field applied perpendicular to the CuO_2 planes, the gap to magnetic excitations in $La_{1.855}Sr_{0.145}CuO_4$ decreases gradually and vanishes at the onset field for long-range static SDW order. We have argued that these observations, which follow the expectations for a continuous soft-mode driven quantum phase transition, suggest the existence of a line of such transitions in the doping-field phase diagram of LSCO. Further work is needed to clarify precisely how the closure of an excitation gap and appearance of IC magnetic order relate to reconstruction of the Fermi surface into small pockets as observed in YBCO with $p \sim 0.5$ [30].

This work was supported by the Swiss NSF (through NCCR, MaNEP, and Grants No. 200020-105151 and No. PBEZP2-122855) and by the Ministry of Education and Science of Japan. We gratefully acknowledge discussions with C. Mudry, B. M. Andersen, M. Kenzelmann, A.-M. Tremblay, B. Keimer, L. Taillefer, and A. T. Boothroyd.

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