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Spin fluctuations associated with the collapse of the pseudogap in a cuprate superconductor

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Theories of the origin of superconductivity in cuprates are dependent on an understanding of their normal state which exhibits various competing orders. Transport and thermodynamic measurements on $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ show signatures of a quantum critical point, including a peak in the electronic specific heat C versus doping p , near the doping p^* where the pseudogap collapses. The fundamental nature of the fluctuations associated with this peak is unclear. Here we use inelastic neutron scattering to show that close to T_c and near p^* , there are low-energy collective spin excitations with characteristic energies ≈ 5 meV. The correlation length of the spin fluctuations does not diverge in spite of the low energy scale and we conclude that the underlying quantum criticality is not due to antiferromagnetism but most likely to a collapse of the pseudogap. We show that the large specific heat near p^* can be understood in terms of collective spin fluctuations. The spin fluctuations we measure exist across the superconducting phase diagram and may be related to the strange metal behaviour observed in cuprates.

Spin fluctuations can play an important part in determining the low-temperature thermal and quasiparticle properties of strongly-correlated electron systems. Notable examples are heavy-fermion metals [1, 2] such as CeCu_6 and UPt_3 . At low temperatures, these materials show very-large linear heat capacities $\gamma = C/T$ because they form heavy electron quasiparticles incorporating moments of the $4f$ or $5f$ electrons and low-energy ($\lesssim 1$ meV) spin fluctuations develop [3]. While cuprate superconductors do not show the very-large quasiparticle mass m^* observed in heavy-fermions systems, they do show moderate enhancements of γ and m^* up to a factor of ~ 3 with respect to the local-density-approximation (LDA) band structure calculations [4–8] (see Fig. 1b). In this work, we investigate how the spin degrees of freedom contribute to the enhancement of γ and m^* in the cuprates.

It is well known that the high-energy spin excitations persist across the superconducting phase diagram of cuprate superconductors [9]. These excitations can have energies comparable with the exchange constant $J \approx 120$ meV of the parent antiferromagnets. They are strong near the antiferromagnetic zone centre and are believed to cause superconductive pairing [10]. The normal state of cuprate superconductors shows unusual behaviour in transport and thermodynamic properties [9, 11–14] such as the “Planckian” linear T -dependence of the resistivity [9, 13, 15]. These properties are related to excitations with lower energies comparable with $k_B T$ rather than J .

The single-layer cuprate superconductor $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) can be doped across the superconducting phase diagram, where the hole doping

$p = x$. Normal-state heat-capacity measurements have been made at $T \approx T_c$ [16, 17] and also at lower temperatures with superconductivity suppressed by Zn doping [18], a high magnetic field [19], or a high magnetic field and Nd/Eu doping [14]. It is found that the specific heat $\gamma(p)$ shows a peak at $p = p_c \approx 0.22$ (see Fig. 1b). The peak in $\gamma(p)$ and the fan-shaped entropy landscape (Fig. 1a) above T_c resemble systems such as iron-based superconductors [20] and $\text{Sr}_3\text{Ru}_2\text{O}_7$ [21] that display magnetic quantum criticality and where enhancements in the quasiparticle mass have been associated with the presence of low-energy spin fluctuations. Thus, it is natural to ask whether spin fluctuations contribute to the large γ values near p_c in LSCO. The situation in LSCO is subtle because the Fermi energy E_F passes through a van-Hove singularity (VHS) [6, 7] near p_c as the doping is increased and the pseudogap [11] terminates at a critical doping $p^* \approx 0.19$ [13, 19] which is close to p_c .

We have measured the low-energy spin fluctuations in the normal and superconducting states of LSCO for $p = 0.22 \approx p_c$. In the normal state, at $T = T_c = 26$ K, we find incommensurate spin fluctuations with a low energy scale $\hbar\Gamma = 4.6 \pm 0.3$ meV and a correlation length $\xi = 19 \pm 2$ Å. On repeating the measurement at $T = 2$ K (i.e. $T \ll T_c$) with a magnetic field $B = 8.8$ T to partially suppress the superconductivity, we find that the low-energy spin fluctuations are enhanced for all energies below 10 meV. This indicates that, if superconductivity had not intervened, the low-energy spin fluctuations would be substantially stronger. Thus the superconducting dome in LSCO hides a region of coherent low-energy spin fluctuations near p_c .

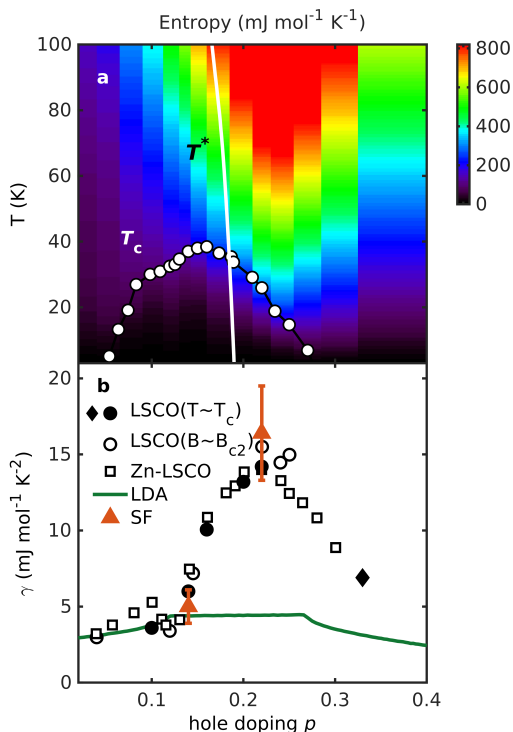


FIG. 1. **Entropy and electronic specific heat in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$.** **a** Entropy as a function of temperature and hole doping $p = x$ for LSCO (derived from data of Ref. [16]). $T_c(p)$ is the superconducting critical temperature (open circles). $T^*(p)$ (solid line) is the pseudogap temperature [11]. **b** Doping dependence of the electronic specific heat coefficient $\gamma(T) = C_{el}/T$ in the normal state, for $T \approx T_c$ (closed circles [17], closed diamond [22]), at high magnetic field $B \approx B_{c2}$ (open circles [19]), or where superconductivity is suppressed by Zn doping (open squares [18]). The green line is the prediction for γ based on the unrenormalised 3D LDA band structure [8]. The solid triangles represent $\gamma(T = T_c)$ of LSCO ($p = 0.14$ and 0.22) calculated using the spin fluctuation theory described in the text.

Inelastic neutron scattering (INS) studies [23–31] show that, except for very low dopings, the low-energy, $\hbar\omega \lesssim 30$ meV, spin excitations in cuprate superconductors are strongest at incommensurate wavevectors $\mathbf{Q}_\delta = (0.5 \pm \delta, 0.5)$ and $(0.5, 0.5 \pm \delta)$ and occur throughout the phase diagram of materials such as $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. In the superconducting state, these excitations are suppressed at low energies [24, 32] approximately below the superconducting gap Δ . In the normal state of underdoped cuprates, for example LSCO ($p = 0.14$), the characteristic energy $\hbar\Gamma$ of excitations is strongly temperature-dependent and ω/T scaling has been observed [25]. Previous studies [28–31, 33] of overdoped LSCO have identified low-energy incommensurate magnetic scattering. However, a quantitative characterisation of the magnetic response for $p \approx p_c$ in the normal state has not been attempted.

In this study we use (see Methods) the LET, MERLIN and IN12 spectrometers to map out the \mathbf{Q} - ω dependence of the low-energy spin excitations in LSCO ($p = 0.22$). A complication in this measurement is the presence of phonon scattering. LSCO undergoes a tetragonal (HTT) to orthorhombic (LTO) structural phase transition for $p \lesssim 0.21$. The soft phonon [34] associated with this transition has a reduced wavevector $(0.5, 0.5, 0)$ and an energy of ~ 3 meV in LSCO ($p = 0.22$). The intensity of phonon scattering is proportional to $|\mathbf{Q}|^2$ times a structure factor and in this measurement we minimise the phonon scattering by measuring at small $|\mathbf{Q}|$ near $\mathbf{Q} = (0.5, 0.5, L)$ with $|L| \leq 1$ (See Extended Data Figs. 1–2 for further details). Our samples showed no sign of incommensurate magnetic order at $T = 1.5$ K and $B = 10$ T (See Extended Data Fig. 3) in agreement with recent nuclear magnetic resonance measurements [35] that this only exists for $p < p^*$.

Figure 2a–i show the constant-energy maps of the scattering intensity $S(\mathbf{Q}, \omega)$ at $\hbar\omega = 1.25, 5$ and 7.5 meV measured at $T = 26$ K (T_c) and 2 K at zero field, and $T = 2$ K in a magnetic field of $B = 8.8$ T applied parallel to the c axis. The magnetic response in this energy range is peaked at the four incommensurate wavevectors \mathbf{Q}_δ with $\delta \approx 0.135$ in agreement with a previous study at higher energies [29]. Fig. 3 shows wavevector-dependent \mathbf{Q} cuts of $S(\mathbf{Q}, \omega)$ through the \mathbf{Q}_δ positions along the trajectory shown in Fig. 2b. Fig. 4a–e show the imaginary part of the dynamical spin susceptibility $\chi''(\mathbf{Q}, \omega)$ extracted directly from the measured $S(\mathbf{Q}, \omega)$ by correcting for the bose factor (see Methods) and displayed as $\hbar\omega$ -cuts at $\mathbf{Q} = \mathbf{Q}_\delta$ and \mathbf{Q} -slices at $\hbar\omega = 1.25$ meV.

In Fig. 4a we see the presence of low-energy spin fluctuations in overdoped LSCO near p_c in the normal state at $T = T_c$. On fitting this ω -dependent cut of $\chi''(\mathbf{Q}_\delta, \omega)$ to an overdamped harmonic oscillator response we find a characteristic energy scale $\hbar\Gamma_\delta = 4.6 \pm 0.3$ meV. To put this in context, we note that for underdoped LSCO ($p = 0.14$), where ω/T scaling is observed [25], a larger $\hbar\Gamma_\delta \approx 9.6$ meV is found at $T = 35$ K $\approx T_c$. Lowering the temperature to 2 K (Fig. 4b, closed symbols) causes a suppression of the excitations below ≈ 4 meV due to the opening of the superconducting gap Δ and an increase above this energy due the spin resonance [32]. Applying a modest magnetic field of $B = 8.8$ T at 2 K induces low-energy excitations reminiscent of the normal state (Fig. 4b, open symbols) as vortices are introduced into the system [36]. Fig. 2 and Fig. 3 show the corresponding changes in $S(\mathbf{Q}, \omega)$. For example, it can be seen that $S(\mathbf{Q}, \omega)$ is suppressed at $\hbar\omega = 1.25$ meV on entering the superconducting state (see Fig. 2a and 3d), and a magnetic field induces new excitations (Fig. 2d and 3d).

We can parameterize our normal-state data at $T = T_c$ with a phenomenological susceptibility similar to the

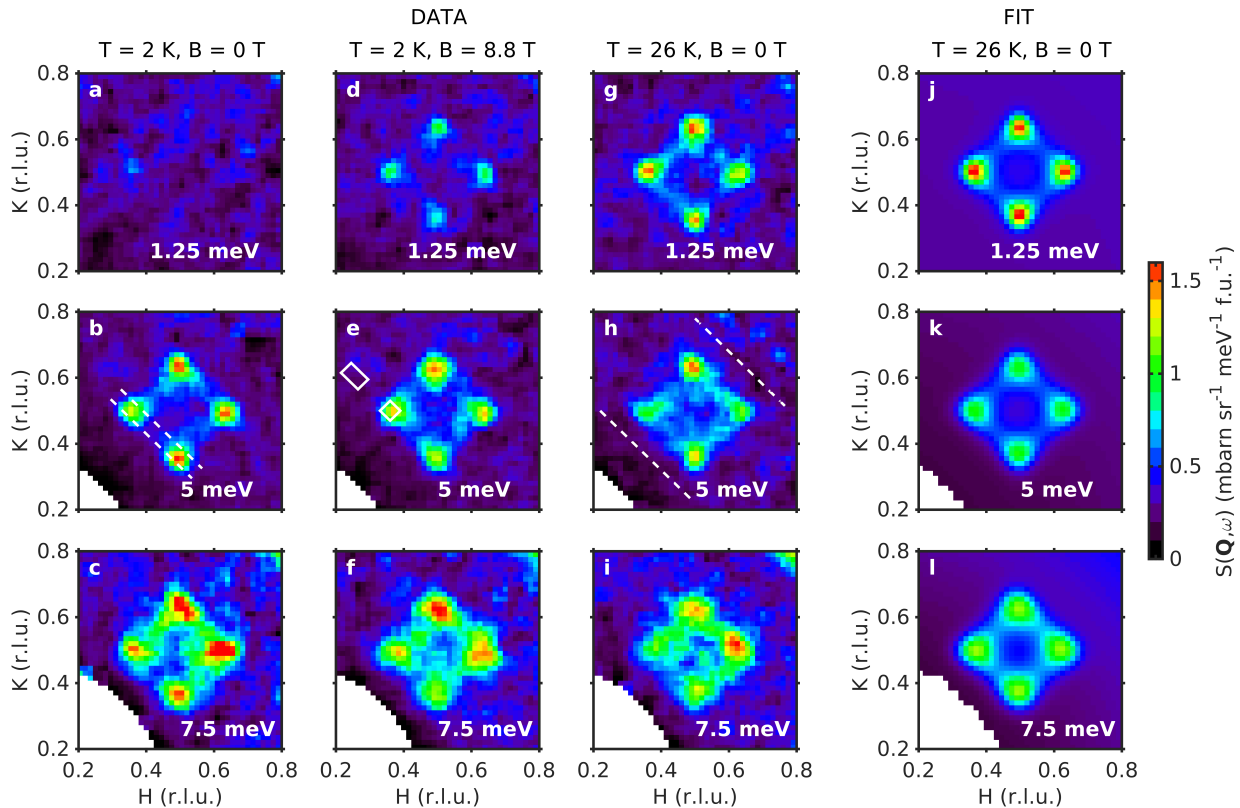


FIG. 2. **Wavevector-dependent maps of low-energy spin fluctuations in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.22$).** Constant-energy maps of $S(\mathbf{Q}, \omega)$ measured at: **a-c** $T = 2$ K, $B = 0$ T; **d-f** $T = 2$ K, $B = 8.8$ T; **g-i** $T = 26$ K (T_c), $B = 0$ T. L is integrated over the range $|L| \leq 1$. White dashed lines in **b** are the range of integration used to produce Fig. 3. White boxes in **e** define integration ranges for signal and background used to produce Fig. 4a-b. **j-l** The result of a global fit of the Eqn. 1 including correction of a magnetic form factor and a $|\mathbf{Q}|^2$ background. Data shown in Figs. 2-4 were collected on LET.

forms used by Pines *et al.* [37] and Aeppli *et al.* [25]

$$\chi''(\mathbf{Q}, \omega) = \frac{\chi_\delta \Gamma_\delta \omega}{\Gamma_\delta^2 [1 + \xi^2 R(\mathbf{Q})]^2 + \omega^2}, \quad (1)$$

where $R(\mathbf{Q})$ is a function which has the symmetry of the 2D Brillouin zone and reproduces the four incommensurate peaks,

$$R(\mathbf{Q}) = \frac{1}{4\delta^2} \left\{ \left[\left(H - \frac{1}{2} \right)^2 + \left(K - \frac{1}{2} \right)^2 - \delta^2 \right]^2 + 4 \left(H - \frac{1}{2} \right)^2 \left(K - \frac{1}{2} \right)^2 \right\}. \quad (2)$$

Near \mathbf{Q}_δ , $R(\mathbf{Q}) \propto |\mathbf{Q} - \mathbf{Q}_\delta|^2$, allowing ξ to be interpreted as a correlation length [25, 37]. Eqn. 1 describes an overdamped harmonic oscillator response with a \mathbf{Q} -dependent relaxation rate

$$\Gamma(\mathbf{Q}) = \Gamma_\delta [1 + \xi^2 R(\mathbf{Q})]. \quad (3)$$

The resulting fitted parameters for $T = 26$ K are shown in Table I (see Methods). Eqn. 1 provides a good description of the excitations in the normal state. For $T = 2$ K in the superconducting state (Fig. 4b), the line shape is

no longer described by the overdamped harmonic oscillator form, however, \mathbf{Q} cuts at a given energy (Fig. 3d-f) can be fitted using the related Eqn. 7 (see Methods).

Landau Fermi liquid theory is used to understand the low-temperature heat capacity of metals. In this picture the one-electron states (e.g. from a LDA band structure) are renormalised by the interaction with excitations such as spin fluctuations. This results in electron quasiparticles with enhanced effective masses and a larger heat capacity γ . For metals with strong antiferromagnetic or ferromagnetic correlations these enhancements can also be understood in terms of the spin excitations. The contribution of the spin fluctuations to the free energy can be estimated using a self-consistent renormalisation spin fluctuation (SF) theory based on a one-loop approximation and the Hubbard model [38–42]. In view of our observation of low-energy excitations near \mathbf{Q}_δ indicating the proximity to magnetic order, here we test whether the large measured γ is due to the spin fluctuations we observe. We can compare the measured magnetic response function $\chi''(\mathbf{Q}, \omega)$ to the SF-theory and hence estimate the heat capacity γ . This approach has been applied to a number of correlated electron systems [43], most recently

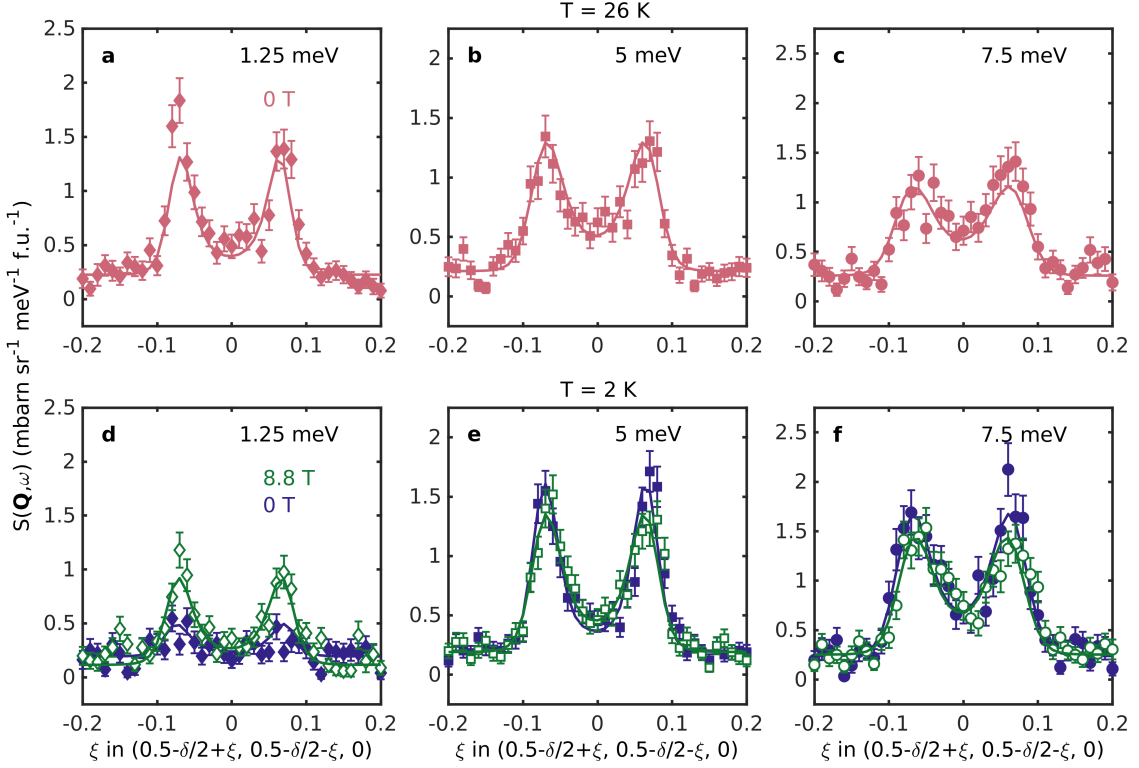


FIG. 3. **Fits of spin-fluctuation model to magnetic excitations.** **a-c** Wavevector-dependent $S(\mathbf{Q}, \omega)$ cuts through \mathbf{Q}_δ for various energies in the normal state. Solid lines are the result of a global fit to the phenomenological spin fluctuation model Eqn. 1. **d-f** Cuts in the superconducting state. Low-energy fluctuations are induced by a magnetic field (panel d). Lines are fits to Eqn. 7 with $\kappa_1(\omega) = 0$ (see Methods). Error bars are determined from Poisson counting statistics or least squares fitting of data and denote one standard deviation.

$\text{Sr}_3\text{Ru}_2\text{O}_7$ [21]. The SF-theory predicts that for $T \rightarrow 0$ (See Refs. [40, 41] and Methods),

$$\gamma_{\text{SF}} = \frac{\pi k_B^2}{\hbar} \left\langle \frac{1}{\Gamma(\mathbf{Q})} \right\rangle_{\text{BZ}}, \quad (4)$$

where $\Gamma(\mathbf{Q})$ is the spin relaxation rate and $\langle \dots \rangle_{\text{BZ}}$ denotes average over the Brillouin zone. We can use our fitted $\Gamma(\mathbf{Q})$ for the normal state at $T = T_c$ (plotted as $\Gamma^{-1}(\mathbf{Q})$ in Fig. 4f) to estimate $\gamma_{\text{SF}}(T_c) = 16.5 \pm 3 \text{ mJ mol}^{-1} \text{ K}^{-2}$ for LSCO ($p = 0.22$), where we have corrected Eqn. 4 for finite temperatures (see Methods). The result agrees reasonably with the measured [17] value $\gamma_{\text{exp}}(T_c) = 14.2 \text{ mJ mol}^{-1} \text{ K}^{-2}$ (see Fig. 1b). We also computed γ_{SF} for slightly underdoped LSCO ($p = 0.14$) where the low-energy spin excitations at T_c have previously been measured and parameterized [25]. Reasonable agreement between γ_{exp} and γ_{SF} is also found (see Fig. 1b and Table I).

In the superconducting state ($T = 2 \text{ K}$, $B = 0 \text{ T}$), the low-energy excitations are suppressed leading to the gaped spectrum in Fig. 4b (blue line) and there is a concomitant reduction of the specific heat [17, 19] with $\gamma_{\text{exp}} \approx 4 \text{ mJ mol}^{-1} \text{ K}^{-2}$. Such a reduction is expected in

the SF-theory on general grounds because of the suppression of the low-energy spin fluctuations. The application of magnetic field $B = 8.8 \text{ T}$ introduces low-energy excitations below the spin gap energy (Fig. 4b, green line) which are associated with the vortices [36]. In this inhomogeneous mixed state, the excitation spectrum is expected to be approximately a superposition of the contribution from the vortices which should be similar to the normal state (Fig. 4a) and the $B = 0$ spectrum in the superconducting state (Fig. 4b, blue line). This is qualitatively consistent with the observed increase in specific heat with $\gamma_{\text{exp}}(T = 2 \text{ K}, B = 8.8 \text{ T}) \approx 8.5 \text{ mJ mol}^{-1} \text{ K}^{-2}$ [19]. The normal-state-like component would continue to grow with increasing magnetic field leading to the large observed $\gamma_{\text{exp}}(T = 2 \text{ K}, B = 34 \text{ T}) \approx 15 \text{ mJ mol}^{-1} \text{ K}^{-2}$ near B_{c2} [19].

Our results are complemented by recent x-ray diffraction measurements [44] of charge-density-wave (CDW) correlations in LSCO ($p = 0.21$) with slightly lower doping. These reveal CDW correlations with a much longer correlation length $\xi_{\text{CDW}} = 75 \pm 5 \text{ \AA}$ and a propagation vector $\mathbf{Q}_{\text{CDW}} = (\delta_{\text{CDW}}, 0)$, where $\delta_{\text{CDW}} \approx 0.236$. As $\delta_{\text{CDW}} \approx 2\delta_{\text{SF}}$ it is likely that the CDW and spin fluctu-

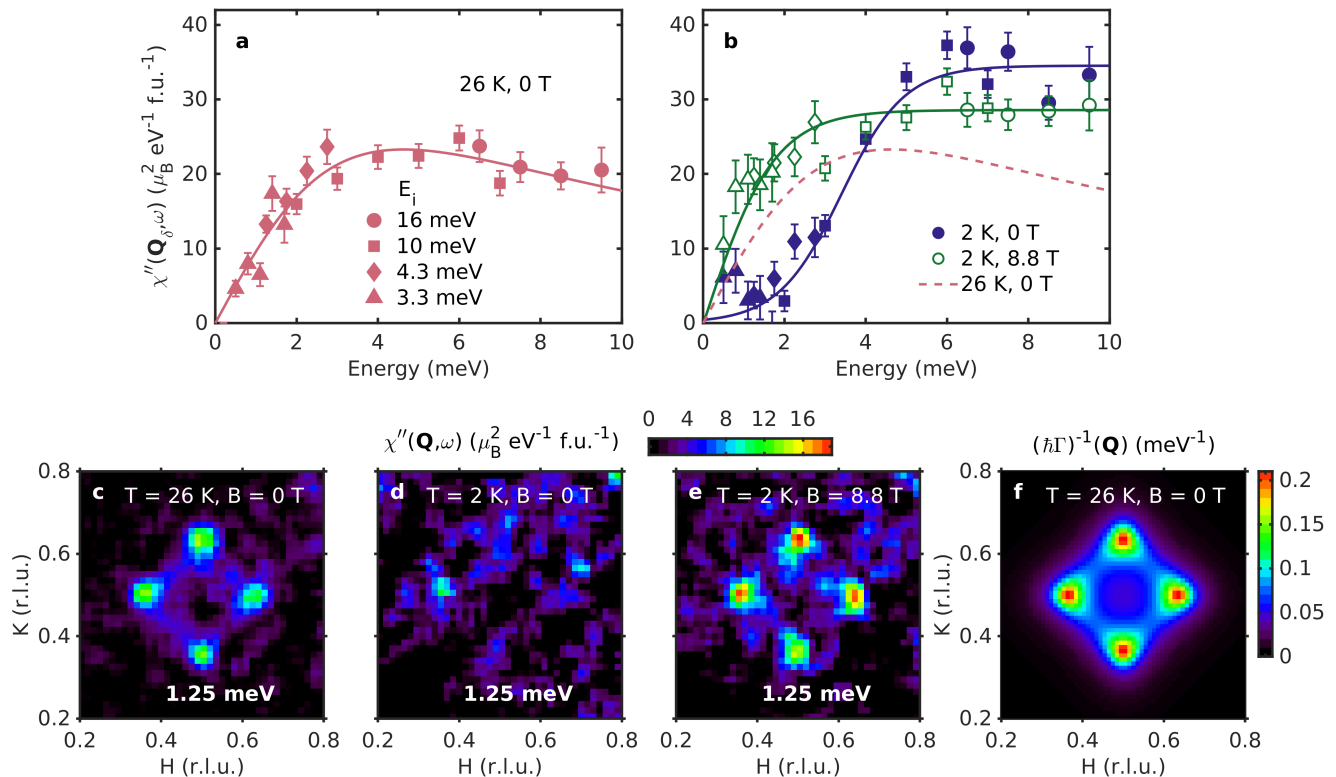


FIG. 4. **Magnetic excitations with a low energy scale in the normal and superconducting states of LSCO ($p = 0.22$).** Energy dependence of the dynamical spin susceptibility $\chi''(\mathbf{Q}, \omega)$ at $\mathbf{Q} = \mathbf{Q}_\delta$ in the **a** normal state showing the low energy scale, and **b** superconducting state showing the suppression of $\chi''(\mathbf{Q}_\delta, \omega)$ for $B = 0$ (closed symbols) and induced response for $B = 8.8$ T (open symbols). The solid line in **a** is a fit to the overdamped harmonic oscillator response Eqn. 8. Solid lines in **b** are guides to the eye. Symbols indicate incident energy E_i . **c-e** Maps of low-energy $\chi''(\mathbf{Q}, \omega)$ at $T \approx T_c$ and at $T = 2$ K with $B = 0, 8.8$ T. **f** Wavevector-dependence of the relaxation parameter $\Gamma^{-1}(\mathbf{Q})$ in the normal state obtained from a global fit.

ations are coupled or derive from a common origin such as stripes. However, the long CDW correlation length $\xi_{\text{CDW}} \approx 4\xi_{\text{SF}}$ means that the low-energy collective charge and spin fluctuations are very different and presumably the CDW-fluctuations do not make a large contribution to the specific heat.

We have demonstrated the presence of low-energy spin fluctuations in the normal state of overdoped LSCO ($p = 0.22$) and shown, using SF-theory, how these can account for the large electronic specific heat observed for this composition. We now discuss how this result relates to other phenomenologies used to understand the peak in $\gamma(p)$, these are: the presence of a van-Hove singularity near E_F [8], antiferromagnetic quantum criticality [45] or the collapse of the pseudogap [11, 14]. LDA-band structure calculations predict that E_F will pass through a VHS in the electron density of states with increasing p . The green solid line in Fig. 1b shows $\gamma(p)$ calculated from the 3D LDA band structure of Ref. [8] in the absence of quasiparticle mass enhancement. It is well known that the quasiparticle mass m^* and Fermi velocity v_F are renor-

malised [8] in cuprates from ARPES [6, 7] and quantum oscillation measurements [4, 5]. We can see from the specific heat data in Fig. 1b that a renormalisation of m^* and v_F by ≈ 3 is required near p_c .

The peak in $\gamma(p)$ is suggestive of a quantum critical point (QCP) related to antiferromagnetism or the disappearance of the pseudogap. In standard scenarios of 2D antiferromagnetic quantum criticality with the dynamic critical exponent $z = 2$ [45] we would expect an increase of the correlation length ξ as we approach the critical doping p_c with $\xi \propto 1/\sqrt{\Gamma}$. Comparing previous measurements on underdoped LSCO for $T \approx T_c$ with our data, we find (see Table I) that between $p = 0.14$ and $p = 0.22$, ξ decreases from 27 ± 4 to 19 ± 2 Å in spite of $\hbar\Gamma$ decreasing from 9.6 ± 0.8 to 4.6 ± 0.3 meV. Thus we conclude that the peak in $\gamma(p)$ is not due to proximity to a simple antiferromagnetic QCP rather additional ingredients which introduce low-energy spin excitations are required. It has been emphasised [14] that in the related systems $\text{La}_{2-x-y}(\text{Eu}, \text{Nd})_y\text{Sr}_x\text{CuO}_4$, p_c is very close to the critical doping p^* where the pseudogap disappears so this

| Doping p | T (K) | B (T) | χ_δ (μ_B^2 meV $^{-1}$ f.u. $^{-1}$) | $\hbar\Gamma_\delta$ (meV) | ξ^{-1} (\AA^{-1}) | δ (r.l.u.) | γ_{exp} (mJ mole $^{-1}$ K $^{-2}$) | γ_{SF} |
|-------------|----------|----------|---|-------------------------------|-------------------------------------|----------------------|---|----------------------|
| 0.14 | 35 | 0 | 376(16) [25] | 9.6(8) [25] | 0.037(6) [25] | 0.123 [25] | 6.0 [24] | 5(1) |
| 0.22 | 26 | 0 | 71(2) | 4.6(3) | 0.053(8) | 0.134(4) | 14.2 [17] | 16.5(30) |
| 0.22 | 2 | 0 | | | | | 4.0 [17, 19] | |
| 0.22 | 2 | 8.8 | | | 0.057(9) | 0.135(4) | 8.5 [19] | |
| 0.22 | 2 | 34 | | | | | 15.0 [19] | |

TABLE I. **Parameterization the magnetic response and the heat capacity of LSCO.** Columns 4-7 show the parameterization of $\chi''(\mathbf{Q}, \omega)$ using Eqn. 1 (normal state) and Eqn. 7 (superconducting state). Column 8 is the measured low-temperature linear heat capacity $\gamma = C/T$. Column 9 is γ calculated from SF-theory using parameters in columns 5-7.

would be the obvious candidate. For dopings $p < p^*$, the pseudogap removes the low-energy electron quasiparticle states near E_F and their spin degrees of freedom [11]. Thus as these are restored near p^* they bring entropy and low-energy spin fluctuations.

The collective spin fluctuations we observed near $p = p^*$ can account for the large low-temperature electronic heat capacity. An increase in the correlation time of the fluctuations $\tau = \Gamma^{-1}$ with respect to smaller doping is observed but there is no concomitant increase in the correlation length ξ . This suggests that the low energy scale is not associated with standard antiferromagnetic quantum criticality [45] but rather with a change in the electronic properties such as the collapse of the pseudogap. The overdoped cuprate $\text{Ti}_2\text{Ba}_2\text{CuO}_{6+\delta}$ shows large quasiparticle mass enhancements of ~ 3 for dopings $p \geq 0.27$ [4] indicating that the co-existence of low-energy spin fluctuations [28] and large quasiparticle mass persists at large p . The existence of spin fluctuations with an energy scale comparable to temperature $\hbar\Gamma \approx k_B T$ may be related to the strange metal or T -linear resistivity behaviour observed in overdoped cuprates. This has been described in terms of ‘‘Planckian dissipation’’ [9, 13, 15] where the quasiparticle lifetime varies as $\tau \approx \hbar/(k_B T)$. T -dependent measurements of the spin fluctuations could establish such a connection.

METHODS

Single crystal sample growth and characterisation. Single crystals of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.22$) were grown by the traveling-solvent floating-zone method. The crystals were annealed in 1 bar of flowing oxygen at 800 °C for 6 weeks. The Sr concentration was determined by SEM-EDX and ICP-AES to be $x = 0.215 \pm 0.005$. SQUID magnetometry measurements show that $T_{c,\text{onset}} = 26$ K. Previous INS measurements on these crystals [29] have shown a double-peak structure (~ 10 and 120 meV) in the local susceptibility $\chi''(\omega)$ in the superconducting state.

Inelastic neutron scattering. Inelastic and elastic neutron scattering experiments were performed us-

ing the direct-geometry time-of-flight spectrometers LET and MERLIN at ISIS Facility and the IN12 triple-axis spectrometer at the Institut Laue-Langevin. Co-aligned single crystals with a total masses of 29.8, 32.7 and 5.9 grams were used at LET, MERLIN and IN12, respectively. At LET, the c axis was mounted vertically and the data were collected by rotating the samples in 1 degree steps using incident neutron energies $E_i = 3.3, 4.3, 10$ and 16 meV, in the high-flux mode with the resolution chopper frequency set to 120 Hz, giving rise to elastic energy resolutions of $\sim 0.12, 0.18, 0.6$ and 1.2 meV, respectively. A vertical magnetic field up to 8.8 T was applied along the c axis. At MERLIN, we used $E_i = 30$ meV and the chopper frequency 150 Hz, giving rise to an elastic energy resolution of ~ 1.8 meV, and the samples were oriented with [110] and [001] directions in the horizontal scattering plane. At IN12, a vertical magnetic field up to 10 T was applied along the c axis. The data were collected using a fixed final energy $E_f = 4.7$ meV, collimation of open-80'-open-open, a horizontally focused PG analyser and a velocity selector in the incident beam. Our initial observation of the low-energy spin fluctuations and the field-induced response was made on IN12 (See Extended Data Fig. 4).

LSCO ($x = 0.22$) has a tetragonal structure with $a = b \approx 3.77$ Å and $c \approx 13.18$ Å. We label the reciprocal space as $\mathbf{Q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^* \equiv (H, K, L)$. The scattered intensity has been scaled to absolute units using the incoherent scattering of vanadium. The scattering cross-section is related to the scattering function $S(\mathbf{Q}, \omega)$ and energy- and wavevector-dependent magnetic response function $\chi''(\mathbf{Q}, \omega)$ by the fluctuation-dissipation theorem

$$\frac{k_i}{k_f} \frac{d^2\sigma}{d\Omega dE} = S(\mathbf{Q}, \omega) \quad (5)$$

$$= \frac{2(\gamma r_e)^2}{\pi g^2 \mu_B^2} |F(\mathbf{Q})|^2 \frac{\chi''(\mathbf{Q}, \omega)}{1 - \exp(-\hbar\omega/k_B T)}, \quad (6)$$

where $(\gamma r_e)^2 = 0.2905$ barn sr $^{-1}$, and $F(\mathbf{Q})$ the magnetic form factor. The data were fitted using Eqn. 7 and convoluted with the instrumental resolution using the Horace package [46].

Data fitting. In order to obtain a global fit of Eqn. 1

to a set of constant-energy cuts through the normal-state data at $T = T_c$ such as those in Extended Data Fig. 5, we fit the individual cuts to the form

$$\chi''(\mathbf{Q}, \omega) = \frac{\chi''(\mathbf{Q}_\delta, \omega)[\xi^{-4} + \kappa_1^4(\omega)]}{[\xi^{-2} + R(\mathbf{Q})]^2 + \kappa_1^4(\omega)}, \quad (7)$$

where $\chi''(\mathbf{Q}_\delta, \omega)$ and $\kappa_1^2(\omega)$ vary as

$$\chi''(\mathbf{Q}_\delta, \omega) = \frac{\chi_\delta(\omega/\Gamma_\delta)}{1 + (\omega^2/\Gamma_\delta^2)} \quad (8)$$

and

$$\kappa_1^2(\omega) = \frac{\omega}{\Gamma_\delta} \xi^{-2} \quad (9)$$

to reproduce Eqn. 1 [43]. We obtain $\xi^{-1} = 0.053 \pm 0.008 \text{ \AA}^{-1}$ from fitting the $\hbar\omega = 1.25 \text{ meV}$ data. The solid lines in Extended Data Fig. 5a-c show fits of Eqn. 7 to the constant-energy cuts of $S(\mathbf{Q}, \omega)$. Extended Data Fig. 5d-f show the fitting parameters $\chi''(\mathbf{Q}_\delta, \omega)$, $\kappa_1^2(\omega)$, and δ as a function of energy. The fitted values of $\chi''(\mathbf{Q}_\delta, \omega)$ are well described by Eqn. 8, with $\hbar\Gamma_\delta = 4.3 \pm 0.3 \text{ meV}$ and $\chi_\delta = 71 \pm 2 \mu_B^2 \text{ eV}^{-1} \text{ f.u.}^{-1}$. The value of Γ_δ agrees with that obtained directly from the raw data (Fig. 4a). The gradient of Fig. 5e yields $6.4 \times 10^{-4} \text{ \AA}^{-2} \text{ meV}^{-1}$ close to the expected value $\xi^{-2}/(\hbar\Gamma_\delta) = 6.1 \times 10^{-4} \text{ \AA}^{-2} \text{ meV}^{-1}$ according to Eqn. 9. Thus, the fitting procedure is self-consistent demonstrating that the magnetic response in the normal state of LSCO ($p = 0.22$) at T_c is well described by Eqns. 1 and 2.

In the superconducting state, the line shape of the energy-dependent magnetic response at $\mathbf{Q} = \mathbf{Q}_\delta$ (Fig. 4b) is no longer described by the overdamped harmonic oscillator form. The constant-energy cuts (Fig. 3d-f) were fitted by a model used by Aeppli *et al.* in Ref. [25] which is equivalent to Eqn. 7 with $\kappa_1(\omega) = 0$.

Spin fluctuation heat capacity model. The heat capacity of nearly antiferromagnetic metals can be understood within the self-consistent renormalisation spin fluctuation theory (SF-theory) based on a one-loop approximation and the Hubbard model [38–42]. The low-temperature free energy F can be expressed as

$$F = \sum_{\mathbf{Q}} \int_0^{\omega_c} d\omega F_{\text{osc}}(\omega, T) \frac{3}{\pi} \frac{\Gamma(\mathbf{Q})}{\omega^2 + \Gamma^2(\mathbf{Q})}, \quad (10)$$

where $F_{\text{osc}}(\omega_0, T) = \hbar\omega_0/2 + k_B T \ln[1 - \exp(-\hbar\omega_0/k_B T)]$ is the free energy of a harmonic oscillator with frequency ω_0 and $\Gamma(\mathbf{Q})$ is the relaxation rate of a spin fluctuation mode of wavevector \mathbf{Q} . Eqn. 10 may be used to obtain an approximate expression [40, 41] for the linear coefficient of the specific heat γ ,

$$\gamma = \frac{C}{T} = -\frac{\partial^2 F}{\partial T^2} \quad (11)$$

$$= \sum_{\mathbf{Q}} \int_0^{\omega_c} d\omega \frac{C_{\text{osc}}(\omega, T)}{T} \frac{3}{\pi} \frac{\Gamma(\mathbf{Q})}{\Gamma(\mathbf{Q})^2 + \omega^2}. \quad (12)$$

This is the sum of the specific heat C_{osc} of harmonic oscillators with a frequency distribution of ω_0 corresponding to the response function $\chi''(\mathbf{Q}, \omega)/\omega$, where

$$C_{\text{osc}}(\omega_0, T) = \frac{\hbar^2 \omega_0^2}{k_B T^2} \frac{e^{\hbar\omega_0/k_B T}}{(e^{\hbar\omega_0/k_B T} - 1)^2}. \quad (13)$$

The low-temperature limit of Eqn. 12 is Eqn 4. At finite temperatures, Eqn. 12 can be evaluated numerically if $\Gamma(\mathbf{Q})$ is known as in the present case.

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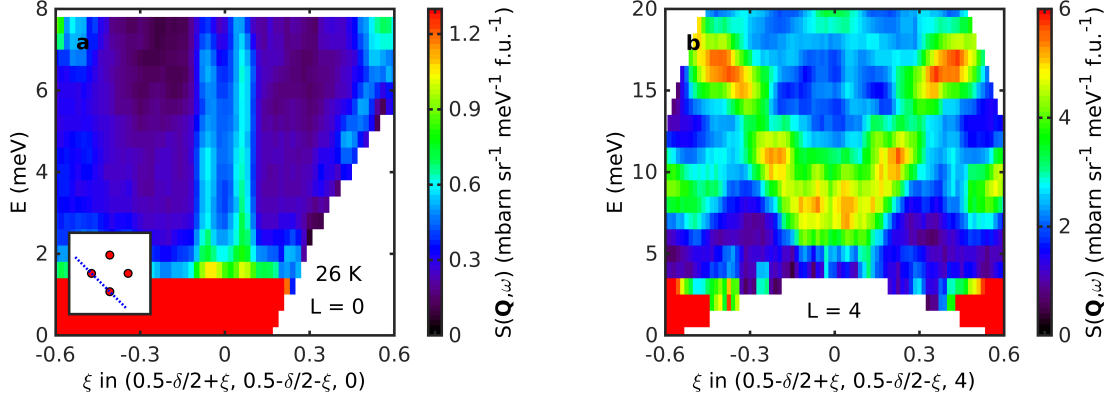
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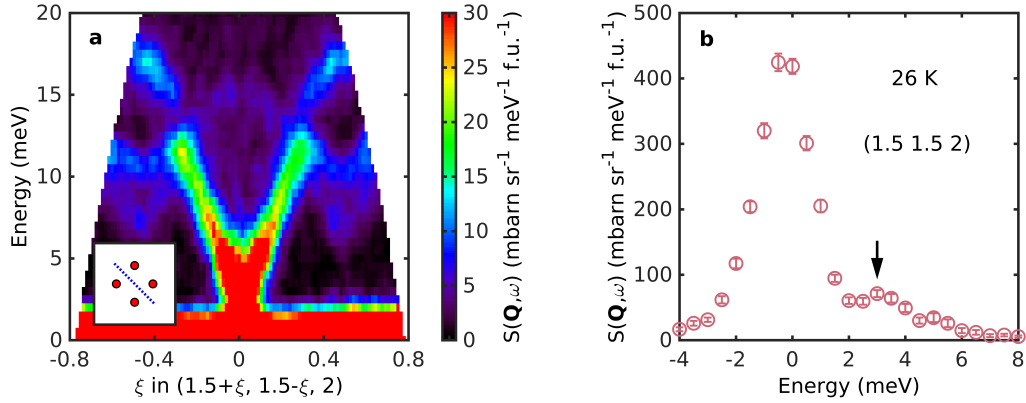
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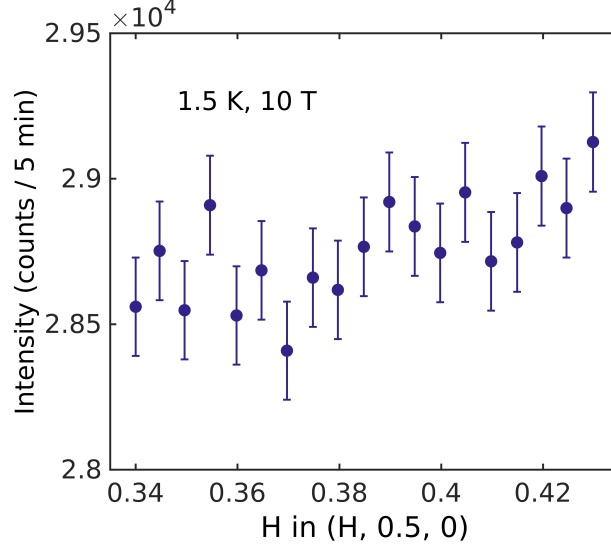
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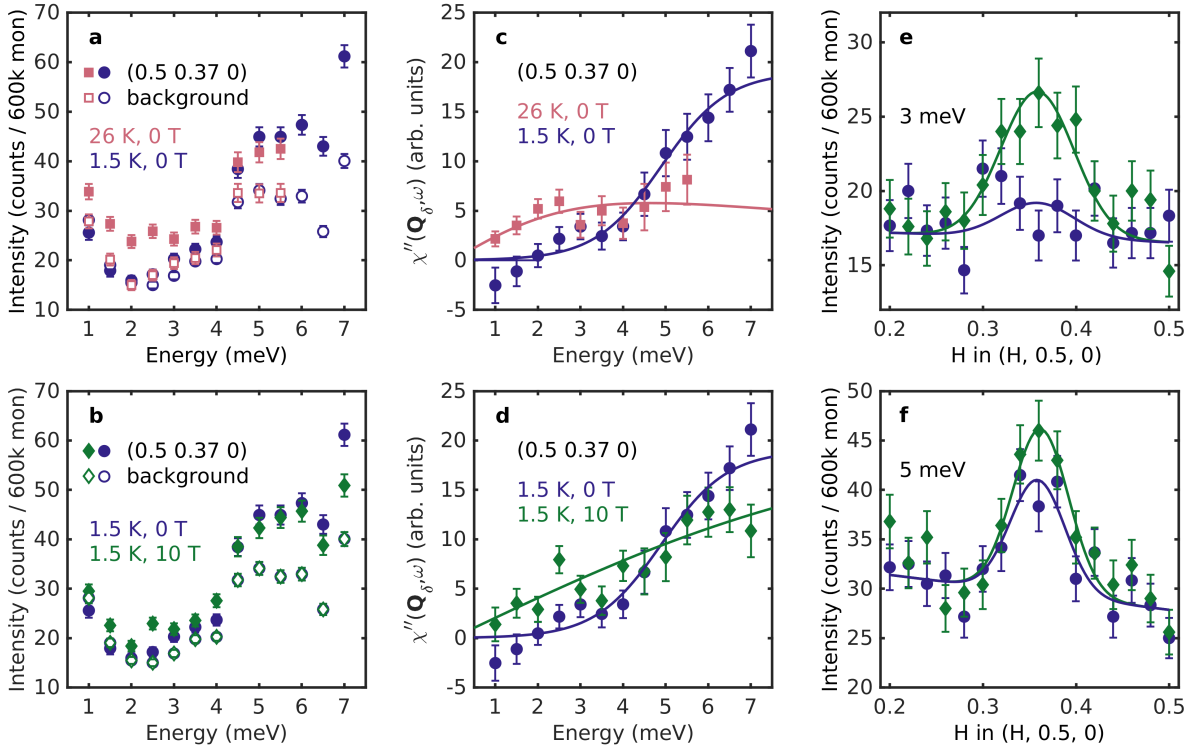
Extended Data Figure 1. **Spin fluctuations and phonons in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.22$) near \mathbf{Q}_δ .** $S(\mathbf{Q}, \omega)$ as a function of energy and wavevector along a trajectory through two incommensurate wave vectors $\mathbf{Q}_\delta = (0.5-\delta, 0.5, L)$ and $(0.5, 0.5-\delta, L)$ (see inset to panel **a**). Integration ranges are **a** $L \in [-1, 1]$ and **b** $L \in [3.8, 4.2]$. Strong phonons are observed (panel **b**) for $L \approx 4$, but these are not visible near $L = 0$ (panel **a**) where spin fluctuations are seen. Data were collected on LET (panel **a**) and MERLIN (panel **b**).



Extended Data Figure 2. **Phonons in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.22$) near $(1.5, 1.5, 2)$.** **a** $S(\mathbf{Q}, \omega)$ as a function of energy and wavevector across $(1.5, 1.5, 2)$ with $L \in [1.8, 2.2]$ at $T = 26$ K. **b** Energy dependence of $S(\mathbf{Q}, \omega)$ at $(1.5, 1.5, 2)$. The arrow denotes a phonon at ~ 3 meV corresponding to the rotation of the CuO_6 octahedra. These features are quite different from the scattering we observe near $(0.5, 0.5, 0)$ identified as magnetic scattering. Data were collected on MERLIN.



Extended Data Figure 3. **The absence of field-induced spin density wave order in LSCO ($p = 0.22$).** Elastic scan through the $\mathbf{Q}_\delta = (0.5 - \delta, 0.5, 0)$ position collected on IN12 with $E_f = 4.7$ meV shows no evidence of spin density wave order at $T = 1.5$ K and $B = 10$ T.



Extended Data Figure 4. **Low-energy spin fluctuations measured by IN12 cold neutron triple-axis spectrometer.** **a-b** Measurements made at $\mathbf{Q}_\delta = (0.5, 0.37, 0)$ (closed symbols) and a background estimated from the average of $(0.56, 0.31, 0)$ and $(0.44, 0.43, 0)$ (open symbols). **c-d** Signal isolated from the data in **a-b** and corrected by a bose factor. Data are consistent with the LET data and show low-energy spin fluctuations in the normal state (panel **c**) and a field-induced signal in the superconducting state (panels **d**). **e-f** Constant-energy scans across \mathbf{Q}_δ at $T = 1.5$ K, $B = 0$ and 10 T.

