

**Low-energy antiferromagnetic spin fluctuations limit the coherent superconducting gap in cuprates**Yangmu Li,<sup>1</sup> Ruidan Zhong,<sup>1,\*</sup> M. B. Stone,<sup>2</sup> A. I. Kolesnikov,<sup>2</sup> G. D. Gu,<sup>1</sup> I. A. Zaliznyak,<sup>1</sup> and J. M. Tranquada<sup>1,†</sup><sup>1</sup>*Condensed Matter Physics and Materials Science Division, Brookhaven National Laboratory, Upton, New York 11973, USA*<sup>2</sup>*Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

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Motivated by recent attention to a potential antiferromagnetic quantum critical point at  $x_c \sim 0.19$ , we have used inelastic neutron scattering to investigate the low-energy spin excitations in crystals of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  bracketing  $x_c$ . We observe a peak in the normal-state spin-fluctuation weight at  $\sim 20$  meV for both  $x = 0.21$  and  $0.17$ , inconsistent with quantum critical behavior. The presence of the peak raises the question of whether low-energy spin fluctuations limit the onset of superconducting order. Empirically evaluating the spin gap  $\Delta_{\text{spin}}$  in the superconducting state, we find that  $\Delta_{\text{spin}}$  is equal to the coherent superconducting gap  $\Delta_c$  determined by electronic spectroscopies. To test whether this is a general result for other cuprate families, we have checked through the literature and find that  $\Delta_c \leq \Delta_{\text{spin}}$  for cuprates with uniform  $d$ -wave superconductivity. We discuss the implications of this result.

DOI: [10.1103/PhysRevB.98.224508](https://doi.org/10.1103/PhysRevB.98.224508)**I. INTRODUCTION**

Layered copper oxide compounds continue to be intriguing because they combine exotic ordered states, such as high-temperature superconductivity, with a challenging electronic environment that defies simple description [1]. The parent compounds are charge-transfer Mott insulators, for which charge excitations face a gap of  $\sim 2$  eV [2], while the only low-energy excitations are the antiferromagnetic spin waves associated with the magnetic moments localized on Cu [3]. By chemical substitution or addition of oxygens in the spacer layers, it is possible to introduce holes into the  $\text{CuO}_2$  planes. The holes would like to delocalize to minimize their kinetic energy, but this competes with the local superexchange interactions, resulting in complex inhomogeneous correlations [4,5], with a significant modification of the spin excitations from the antiferromagnetic state [6]. Another consequence is that, at low concentrations, only the doped holes contribute to the low-temperature transport properties; with increasing doping, the effective carrier density begins to rise faster than the dopant density [7,8]. It rises quite rapidly as the doped-hole density  $p$  approaches a putative pseudogap critical point at  $p_c \sim 0.19$  [9,10].

Given the prominence of antiferromagnetic spin fluctuations, it is commonly (but not universally) believed that they play a role in the hole pairing that is essential for superconductivity [5,11,12]. It has also motivated proposals that the pseudogap critical point might be associated with an antiferromagnetic quantum critical point [10,13]. We set out to test this possibility by using inelastic neutron scattering to measure the spin fluctuations in the system  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) at doping levels  $p = x = 0.17$  and  $0.21$ , superconducting compositions close to but bracketing  $p_c$ . If critical fluctuations

are important, then we might expect the normal state to exhibit spin fluctuations spread over a substantial energy range with no characteristic energy scale. Instead, we found an effective normal-state spin gap of similar magnitude in both samples. Furthermore, the spin gap  $\Delta_{\text{spin}}$  identified by the shift in magnetic spectral weight on cooling below the superconducting transition temperature  $T_c$  is approximately equal to the coherent superconducting gap  $\Delta_c$  that has been determined previously by measurements such as Andreev reflection [14] and electronic Raman scattering [15]; it corresponds to the magnitude of the  $d$ -wave gap at the wave vectors at the ends of the normal-state Fermi arcs (delimited by the pseudogap) as determined by angle-resolved photoemission spectroscopy (ARPES) [16,17] and scanning tunneling spectroscopy (STS) [18–20] and indicated in Fig. 1(a).

Is this behavior unique to LSCO? To check, we looked through the literature to identify neutron scattering measurements of  $\Delta_{\text{spin}}$  and corresponding Raman scattering results for  $\Delta_c$ . We found that, for all cuprate families studied, these data satisfy the relation  $\Delta_c \leq \Delta_{\text{spin}}$  in the regime where uniform  $d$ -wave superconductivity occurs. It is intriguing that the correlation is with  $\Delta_c$  rather than  $2\Delta_c$ , as one might have expected from models that assume proximity to a Fermi-liquid state [21]. In fact, we argue that the gap relationship indicates the incompatibility of Bogoliubov quasiparticles of a spatially uniform  $d$ -wave superconductor with the antiferromagnetic spin excitations of local Cu moments [22].

The rest of the paper is organized as follows. We describe the experimental methods in the next section. The analysis of our data, as well as a detailed comparison with results from the literature, is presented in Sec. III. This is followed by a discussion of the results and a comparison with theoretical analyses. We conclude with a brief summary.

**II. EXPERIMENTAL METHODS**

Cylindrical crystals of LSCO 7 mm in diameter were grown for  $x = 0.17$  and  $0.21$  by the traveling-solvent

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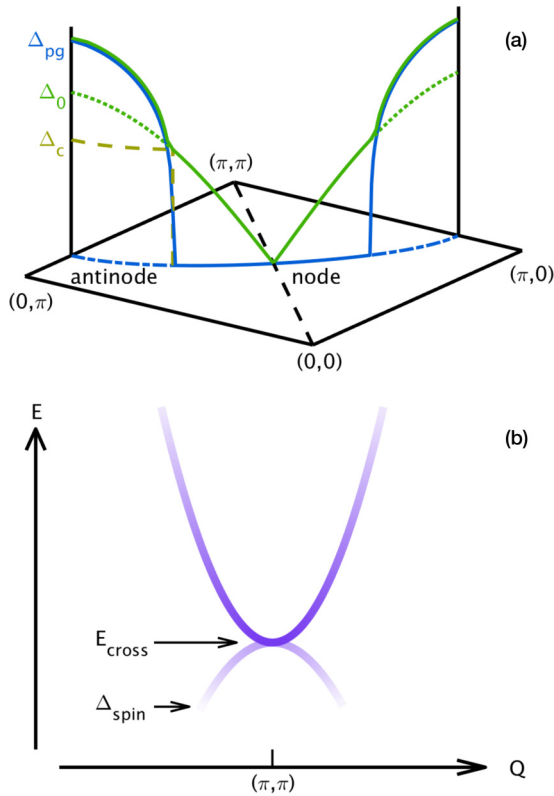


FIG. 1. (a) Schematic of electronic gaps around a quadrant of reciprocal space for a square  $\text{CuO}_2$  plane with lattice constant  $a = 1$ . Green line: below  $T_c$ , a  $d$ -wave gap is found near the node, extrapolating to  $\Delta_0$ , but switches to the pseudogap in the antinodal region, rising to  $\Delta_{pg}$ . Blue line: above  $T_c$ , the coherent gap (with maximum energy of  $\Delta_c$ ) closes to form a Fermi arc. (b) Schematic spin excitation spectrum as a function momentum transfer  $\mathbf{Q}$ , defining the spin gap  $\Delta_{\text{spin}}$  and  $E_{\text{cross}}$ .

floating-zone method. For each composition, a single feed rod with a length of 20–25 cm was used; after growth, the initial several centimeters of the crystal rod were removed and discarded, while the remainder was annealed in flowing  $\text{O}_2$  at  $980^\circ\text{C}$  for 1 week. The superconducting transition temperatures, 37 and 30 K, were determined by dc magnetization measurements with a field of 10 G applied (after cooling in zero field). Figure 2 shows the measurements for small pieces (0.1–0.3 g) taken from the bottom and the top of each crystal rod. (The orientation of crystal axes and crystal shape with respect to the field was arbitrary; differences in orientation contribute to the apparent variation in magnitude of the diamagnetic response. No correction was made for shape anisotropy, which is likely responsible for the susceptibility exceeding the full-shielding response of  $\chi_v = -1$ .) The variation of  $T_c$  across each sample rod is less than 1 K.

For the  $x = 0.17$  sample, three crystals, with a total mass of  $\sim 35$  g, were coaligned by x-ray Laue diffraction with a tetragonal [110] axis in the vertical direction. For the  $x = 0.21$  sample, four crystals, with a total mass of 25.5 g, were coaligned with a tetragonal [100] in the vertical direction using the neutron alignment station CG-1B at the High Flux Isotope Reactor, Oak Ridge National Laboratory (ORNL). To describe the scattering data, we will use a unit cell based

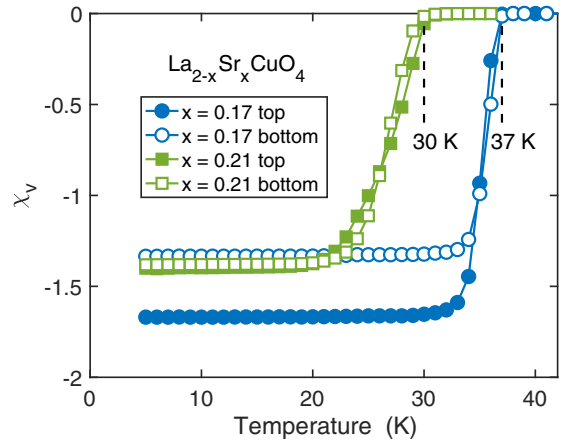


FIG. 2. Volume susceptibility measured for samples of LSCO with  $x = 0.17$  and  $0.21$ . For  $x = 0.17$ , bottom (top) was 4.5 cm (15 cm) from the start of growth; for  $x = 0.21$ , bottom (top) was 6 cm (14 cm) from the start of growth.

on the low-temperature orthorhombic phase with  $a \approx b \approx 5.35 \text{ \AA}$ ,  $c \approx 13.2 \text{ \AA}$  [23], and wave vectors  $\mathbf{Q} = (h, k, l)$  in units of  $(2\pi/a, 2\pi/b, 2\pi/c)$ . The  $x = 0.17$  crystals were coaligned with the [100] and [001] axes in the horizontal plane; for  $x = 0.21$ , [110] and [001] were in the plane.

The inelastic neutron scattering measurements were performed at the SEQUOIA time-of-flight spectrometer (BL-17) of the Spallation Neutron Source, ORNL [24]. For each composition, the sample was mounted in a closed-cycle helium refrigerator for temperature control. Measurements were performed with an incident energy of either 30 or 60 meV, using the high-resolution or high-flux chopper, respectively, operating at a frequency of 420 Hz, for sample temperatures of base (4 or 5 K), 36 K (near  $T_c$ ), and 300 K. For each condition, data were collected with the in-plane sample orientation relative to the incident beam rotated in  $1^\circ$  steps over a range of at least  $120^\circ$ . The initial data reduction was performed with MANTID [25]. Reference measurements on a vanadium standard were used to convert the intensity data to absolute units, and we extracted the imaginary part of the dynamical susceptibility  $\chi''(\mathbf{Q}, \omega)$  in the conventional fashion [26], making use of the magnetic form factor for  $\text{Cu}^{2+}$  hybridized with four in-plane O neighbors [27].

In choosing to measure at 36 K, we intended to be as close as possible to  $T_c$  for  $x = 0.17$ , so that we would not be excessively above  $T_c$  for  $x = 0.21$ . Working without the susceptibility data in hand, we chose a measurement temperature that is just slightly below  $T_c$  for  $x = 0.17$ ; however, given the finite width of the transition seen in Fig. 2, 36 K is at the start of the transition, and we are confident that had the measurements been performed at 37 K, the results would have been virtually the same.

### III. ANALYSIS

#### A. Data reduction

Although phonon scattering is relatively weak at small  $Q$ , so is the magnetic scattering. Figure 3 shows a plot of scattered intensity along  $\mathbf{Q} = (H, 0, 0)$ ; the phonon contributions

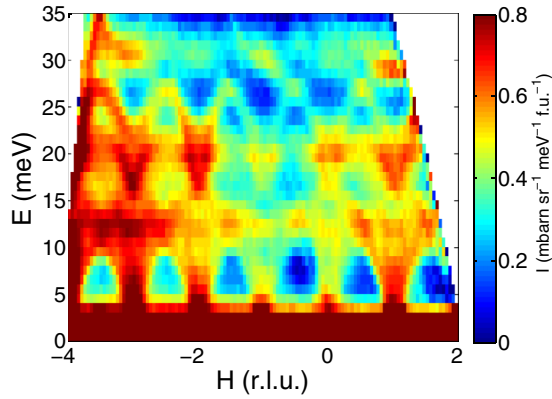


FIG. 3. Scattered intensity as function of energy and  $\mathbf{Q} = (H, 0, 0)$  for LSCO  $x = 0.17$  at 36 K. The intensity has been integrated over  $-0.2 \leq K \leq 0.2$  and over all measured  $L$  within the range  $-2 \leq L \leq 5$ . Antiferromagnetic scattering is allowed around  $H = 1, -1, -3$ . The range of sample orientations measured was selected so that, for  $H = -1$ , the  $L$  range sampled was within  $-2 \leq L \leq 2$ ; the  $L$  ranges at other  $H$  values vary considerably. The difference in phonon intensity at  $H = -1$  and 1 is due to different sampled  $L$  ranges.

are obviously quite significant even at  $H = -1$ , where the  $L$  range has been optimized to emphasize the magnetic response. The neutron scattering cross section is proportional to

$$S(\mathbf{Q}, \omega) \sim \chi''(\mathbf{Q}, \omega)/(1 - e^{-kT/\hbar\omega}), \quad (1)$$

where  $S(\mathbf{Q}, \omega)$  is the dynamic structure factor and  $\chi''(\mathbf{Q}, \omega)$  is the imaginary part of the dynamic susceptibility. Previous work has shown that the magnetic contribution to  $\chi''$ , at least for  $\hbar\omega \lesssim 20$  meV, decreases with increasing temperature [28], whereas the phonon part is essentially constant with temperature. At 300 K, the magnetic contribution to  $\chi''$  is weak relative to the phonon part; hence, we will subtract the measured  $\chi''$  at 300 K from the low-temperature data to largely remove the phonon contribution.

Figure 4 shows constant-energy slices of the low-temperature magnetic  $\chi''$  at excitation energies of 8, 18, and 30 meV as a function of the in-plane wave vector, where the antiferromagnetic wave vector corresponds to  $\mathbf{Q}_{\text{AF}} = (1, 0, 0)$ . (We have integrated over momentum transfer perpendicular to the plane, covering  $-2 < L < 5$ .) The usual incommensurate peaks are seen at low energy, consistent with previous studies [29,30]. By averaging the magnetic response over the first Brillouin zone, we obtain the  $\mathbf{Q}$ -integrated response  $\chi''(\omega)$ . Figures 5(a) and 5(b) show the results for  $\chi''(\mathbf{Q})$  at  $T = 36$  K  $\sim T_c$  and at  $T \ll T_c$ . For each sample, there is a peak at  $\sim 20$  meV, although the response is clearly weaker for the  $x = 0.21$  sample.

The underlying cause of this peak in the magnetic spectrum has not been fully established, but a coupling between phonons and superexchange has been proposed [31,32]. The key observation here is that the peak is still clearly resolved in the normal state of a sample with  $p > p_c$ . The presence of a clear energy scale is inconsistent with expectations for critical magnetic fluctuations. Hence, our results provide evidence against a generic antiferromagnetic quantum critical point in

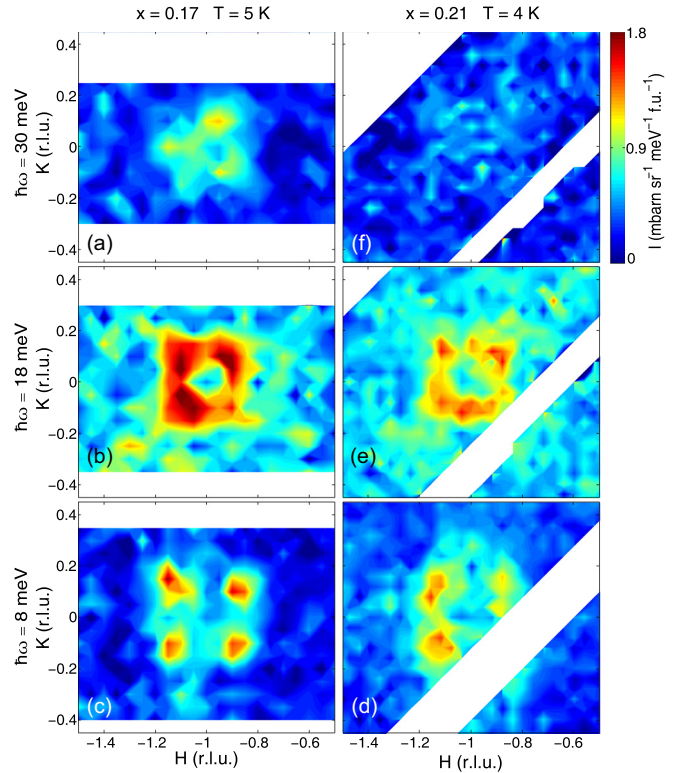


FIG. 4. Constant-energy slices of magnetic scattering intensity (after subtraction of the phonon contribution) as a function of the in-plane wave vector for the  $x = 0.17$  (left) and  $0.21$  (right) samples at base temperature. Excitation energies are (a) and (d) 8, (b) and (e) 18, (c) and (f) 30 meV; energy width is 1 meV and incident energy is 60 meV. White areas correspond to detector gaps.

cuprates. Instead, evidence points to a crossover with  $p$  from stronger to weaker correlations [6,33].

While subtracting the high-temperature phonon contribution does a reasonably good job of isolating the magnetic response, the differences between  $\chi''(\omega)$  below and near  $T_c$ , as shown in Figs. 5(a) and 5(b), are small. To determine the change in  $\chi''$  across  $T_c$  while minimizing the uncertainty, we take the difference in the uncorrected  $\chi''$ , in which case the phonon signal cancels out directly, and fit the results. Examples of such differences for the  $x = 0.17$  sample are shown in Fig. 6. [Note that the background should be temperature independent, but converting the measured intensity to  $\chi''$  scales the background in a temperature-dependent fashion. To account for this, we fit a constant background term when evaluating  $\Delta\chi''$ ; this background shift has already been subtracted in Figs. 6(c) and 6(d).]

## B. Identifying the spin gap

The change in  $\chi''(\omega)$  between base temperature and 36 K is shown in Figs. 5(c) and 5(d), demonstrating the development of a spin gap in the superconducting state and a shift of weight from below to above the gap. To be explicit, we choose to define the spin gap  $\Delta_{\text{spin}}$  to be the energy at which the temperature difference in  $\chi''(\omega)$  crosses zero, which corresponds to  $9 \pm 1$  meV for both samples.



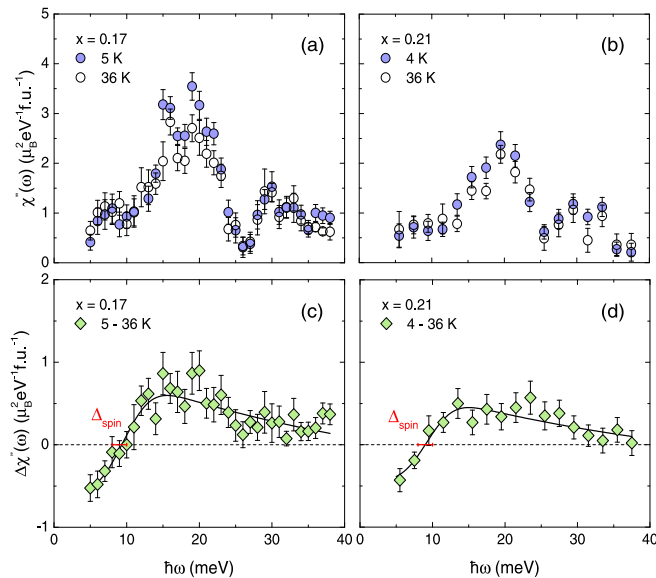


FIG. 5.  $\mathbf{Q}$ -integrated  $\chi''(\omega)$  for (a)  $x = 0.17$  and (b)  $x = 0.21$  at base temperature and  $T = 36$  K; the energy bin width is twice as large for the  $x = 0.21$  results to compensate for the different sample orientation with respect to the spectrometer. Difference in  $\chi''(\omega)$  between base temperature and 36 K for (c)  $x = 0.17$  and (d)  $x = 0.21$ . (The differences were taken using data without phonon correction, as the phonon contribution cancels almost completely.) In the latter, the experimental  $\Delta_{\text{spin}}$ , discussed in the text, is indicated in red.

Our definition of the spin gap is different from what has been used previously. In two early studies of LSCO  $x = 0.15$ , the gap was defined as the energy below which the magnetic signal is essentially zero, yielding a gap of 3.5 meV [34,35]. In another pair of studies on LSCO  $x = 0.14$  [36] and  $x = 0.163$

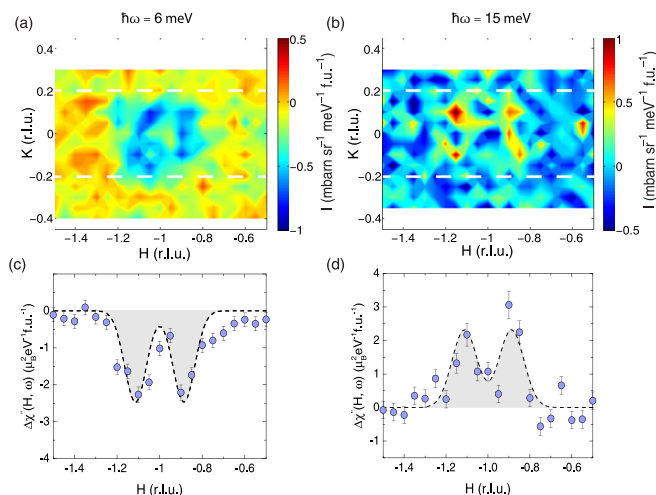


FIG. 6. Analysis of  $\Delta\chi'' = \chi''(5\text{ K}) - \chi''(36\text{ K})$  for LSCO  $x = 0.17$ . Constant-energy slices of  $\Delta\chi''$  for (a)  $\hbar\omega = 6$  meV and (b) 15 meV. (c) and (d) The corresponding cuts obtained after integrating over  $K$  within the range denoted by the dashed white lines in (a) and (b); a constant background difference has been subtracted. The dashed lines in (c) and (d) are fitted Gaussian peaks used to evaluate the magnetic contribution to  $\Delta\chi''(\omega)$ .

[37], the measured  $\chi''$  was fit with formulas having parallels with electronic spectroscopy, yielding gaps of 6 and 6.7 meV, respectively. Given the current state of theory for the cuprates, we are not aware of any generally accepted definition of the spin gap. We believe that our choice of definition is appropriate for comparison with results taken from electronic spectroscopies (see below). Applying our definition to the data in [37] yields  $\Delta_{\text{spin}} \approx 9$  meV.

Comparing with  $\chi''(\omega)$  at 36 K, we note that the amount of spectral weight that shifts through the superconducting transition is small (consistent with previous observations of limited weight in the “resonance” peak [38]), and  $\Delta_{\text{spin}}$  occurs in a range where the spectral weight in the normal state is already weak. This raises the question as to whether the energy cost of gapping low-energy spin fluctuations limits the coherent superconducting gap  $\Delta_c$ .

### C. Comparing with the coherent superconducting gap

To test this idea, we need to compare  $\Delta_{\text{spin}}$  with measurements of  $\Delta_c$ . While ARPES studies have provided evidence for a coherent gap scale associated with the normal-state Fermi arc [16,17], Raman scattering data are available for a larger variety of cuprates. In particular, Raman spectra with  $B_{2g}$  symmetry probe electronic states near the  $d$ -wave gap node, and measurements below  $T_c$  yield an intensity peak at  $\Omega(B_{2g}) = 2\Delta_c$ . Raman studies on LSCO find that  $\Omega(B_{2g})$  is essentially independent of doping for a significant range of  $x$  and corresponds to  $\Delta_c \approx 8$  meV for  $x = 0.15$  and 0.17 [39,40]. (We will assume an uncertainty of  $\pm 15\%$  in the values of  $\Delta_c$  estimated from Raman results, as the peak at  $\Omega_{B_{2g}}$  tends to have significant width.) Andreev reflection data suggest that this trend extends out to  $x = 0.2$  [41]. Hence, we find that  $\Delta_c \approx \Delta_{\text{spin}}$ , within the experimental uncertainties, for our LSCO samples.

This result suggests that low-energy ungapped spin fluctuations may limit the coherent superconducting gap. If this is true for LSCO, it ought to be true for other cuprates as well. We evaluate the latter next.

### D. Testing the relationship on other cuprates

We have gone through the literature and identified studies of various cuprates that allow a reasonable estimate for  $\Delta_{\text{spin}}$ . The results are listed in Table I, where we also report values for  $E_{\text{cross}}$ , as defined in Fig. 1(b). We note that, in all cases,  $\chi''(\omega)$  is weak in the normal state at and below  $\hbar\omega = \Delta_{\text{spin}}$ .

Raman scattering studies on  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ ,  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , and  $\text{HgBa}_2\text{CuO}_{4+\delta}$  have found that  $2\Delta_c \approx 6kT_c$  [55,56], which allows us to estimate  $\Delta_c$  from  $T_c$ . For  $\text{HgBa}_2\text{CuO}_{4+\delta}$ , we chose to interpolate the results of Li *et al.* [57], while for  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ , Blumberg *et al.* [58] found  $2\Delta_c = 4.4kT_c$ .

We plot  $\Delta_c$  and  $E_{\text{cross}}$  vs  $\Delta_{\text{spin}}$  in Fig. 7. We find that  $\Delta_c \leq \Delta_{\text{spin}}$  for all studied cuprates, whereas  $E_{\text{cross}}$  does not show a simple universal relationship with either  $\Delta_{\text{spin}}$  or  $\Delta_c$ . (The fly in the ointment is LSCO, where  $E_{\text{cross}}$  is comparable to that of other cuprates but  $\Delta_c$  and  $\Delta_{\text{spin}}$  are much smaller.) Where there is a spin gap present in the normal state that is already larger than  $\Delta_c$ , as in the case of underdoped  $\text{HgBa}_2\text{CuO}_{4+\delta}$

TABLE I. Results for the spin gap and  $E_{\text{cross}}$  for a variety of cuprates;  $p/n$  corresponds to an estimate of the doped hole/electron concentration. Values in parentheses are uncertainties in (meV).

Compound	$p/n$	$T_c$ (K)	$\Delta_{\text{spin}}$ (meV)	$E_{\text{cross}}$ (meV)	Refs.
YBa <sub>2</sub> Cu <sub>3</sub> O <sub>6+x</sub>	0.10	59	15(5)	32.5	[42]
	0.11	62.7	20(5)	34	[43]
	0.16	93	28(5)	41	[44]
	0.17	92.5	27(5)	41	[45]
Bi <sub>2</sub> Sr <sub>2</sub> CaCu <sub>2</sub> O <sub>8+δ</sub>	0.16	93	32(3)	43	[46]
	0.18	91	30(5)	40	[47]
	0.19	87	32(5)	42	[48]
	0.20	83	30(4)	38	[49]
	0.21	70	24(5)	34	[50]
HgBa <sub>2</sub> CuO <sub>4+δ</sub>	0.10	71	28(5)	50	[51]
	0.13	88	40(5)	59	[52]
La <sub>2-x</sub> Sr <sub>x</sub> CuO <sub>4</sub>	0.16	38.5	8(1)	45	[29,37]
	0.17	37	9(1)	45	this work
	0.21	30	9(1)	45	this work
Nd <sub>2-x</sub> Ce <sub>x</sub> CuO <sub>4</sub>	0.15	25	4.5(1)		[53,54]

[51,52], the spin fluctuations do not limit the development of superconducting coherence, and there is no significant shift in magnetic spectral weight across  $T_c$ .

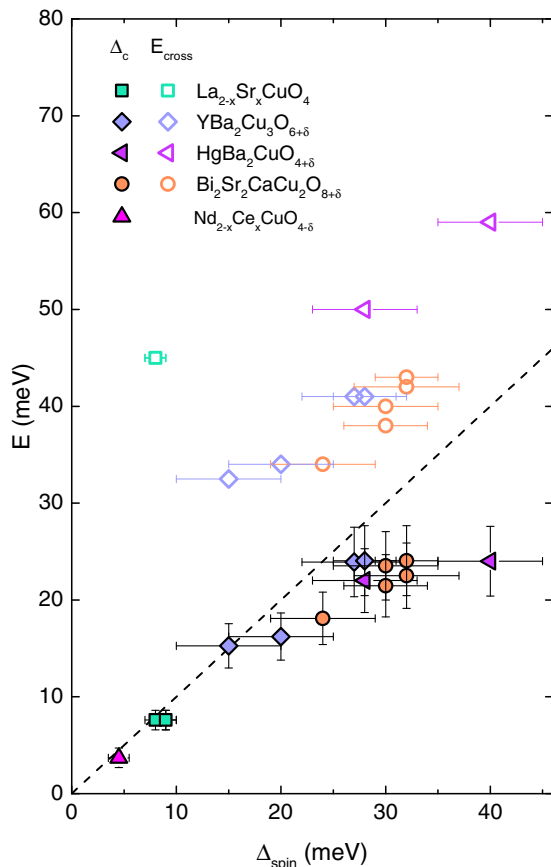


FIG. 7. Plot of  $\Delta_c$  and  $E_{\text{cross}}$  vs  $\Delta_{\text{spin}}$ , corresponding to the data and references in Table I.

We note that analysis of in-plane tunneling measurements on YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> with  $T_c \approx 90$  K yield  $\Delta_c = 28(3)$  meV [59]. While this value of  $\Delta_c$  is slightly larger than the value of 23 meV estimated from  $T_c$ , it is still compatible with  $\Delta_c \leq \Delta_{\text{spin}}$  given the  $\Delta_{\text{spin}}$  values listed in Table I and taking uncertainties into account.

#### IV. DISCUSSION

Our focus on the spin gap and its relation to the coherent superconducting gap differs from more commonly discussed relationships, so we will try to put it in perspective. Many early theoretical analyses of magnetic excitations in metallic cuprates started from a weak-coupling approach, analyzing the spin response in terms of the excitation of free electrons across the Fermi surface, with interactions treated in the random-phase approximation [60–63]. When the superconducting gap opens, the interactions can pull spin excitations below  $2\Delta_0$  (the lower bound for spin excitations of noninteracting electrons), resulting in a spin resonance peak [21,64–66]. Such a model appeared to give a reasonable description of initial experimental results on the enhancement of commensurate magnetic excitations below  $T_c$  in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> [67–69] and Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub> [46,49]. Of course, these early studies only probed commensurate scattering near  $E_{\text{cross}}$ ; later studies established the dispersion away from  $E_{\text{cross}}$  and the presence of that dispersion in the normal state [42,43,47,51,70–73]. Furthermore, it has been demonstrated that the dispersion evolves continuously from the antiferromagnetic-insulator phase, where one has local Cu moments coupled by superexchange [6,74,75]. Recent numerical studies of dynamical correlations in Hubbard models with on-site Coulomb repulsion comparable to the bandwidth support this perspective [76,77]. Thus, there is good reason to believe that the antiferromagnetic excitations detected by neutron scattering across the phase diagram are predominantly from local Cu moments coupled by superexchange [78].

As noted by Anderson [22], the superexchange interaction does not involve quasiparticles. Hence, the interaction of quasiparticles with the spin correlations is inevitably strong. Non-Fermi-liquid behavior can result from charge carriers scattering off of spin fluctuations [79]. The impact of such interactions is evident in the transport properties of cuprates. For example, deviations from  $\rho \sim T$  in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> correspond to the onset of the gap in antiferromagnetic spin fluctuations observed by nuclear magnetic resonance [80–82].

Millis *et al.* [83] showed for a  $d$ -wave superconductor that spin fluctuations with energies below a critical threshold are pair breaking. Dahm and Scalapino [84] recently analyzed the dependence of the superconducting gap and transition temperature on  $\chi''(\mathbf{Q}, \omega)$  for underdoped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub>, using experimentally determined spin-fluctuation data [85]. They found that the spin excitations that disperse upwards from  $E_{\text{cross}}$  enhance  $T_c$ , while those below  $E_{\text{cross}}$  tend to be bad for superconductivity.

Those theoretical results are consistent with the empirical evidence that the presence of low-energy spin fluctuations correlates with a reduced  $T_c$ ; however, they do not directly address the relationship between  $\Delta_{\text{spin}}$  and  $\Delta_c$ . To understand that, we note that when a spin gap is present, quasiparticles

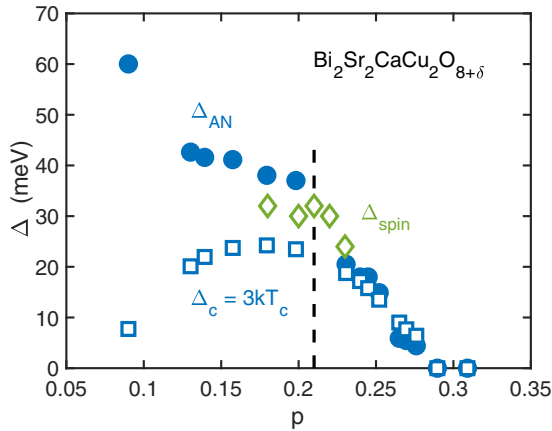


FIG. 8. Comparison of the antinodal quasiparticle energy at  $T < T_c$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  [93] (solid circles),  $\Delta_c$  estimated from  $T_c$  (open squares), and the reported values of  $\Delta_{\text{spin}}$  from Table I (open diamonds). Note that we have used the values of  $p$  reported in [93], which were determined from the size of the Fermi surface, resulting in an increase in  $p$  by 0.02 from typical estimates based on  $T_c(p)$  [96]. To put the spin gap results on the same scale, we have increased the corresponding values of  $p$  listed in Table I by 0.02. The vertical dashed line indicates the adjusted  $p$  value at which an STS study [20] found a crossover in the Bogoliubov quasiparticle distribution from a finite arc to the full Fermi surface.

with energies below the gap can propagate without interacting with the spins, leaving them sharply defined in the near-nodal regime, as observed by ARPES. Towards the antinodal region, quasiparticles with energies above  $\Delta_{\text{spin}}$  can decay by creating spin fluctuations. That contribution to the electronic self-energy can make it unfavorable for those electronic states to participate in the coherent superconducting state. This perspective is consistent with the idea that the pseudogap detected by ARPES is a consequence of antiferromagnetic (AF) correlations [86]. Furthermore, all of the cuprates for which quantum oscillations have been observed in high magnetic fields [87–89] have large spin gaps [90], consistent with sharp quasiparticles at energies close to the chemical potential. In  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ , the effective mass diverges as  $p$  is reduced towards 0.085 [91], which correlates with the closing of the spin gap [74,82,92].

Recent ARPES results covering a broad range of doping in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212) appear to be quite consistent with our analysis [93–95]. For under- and optimally doped samples, the quasiparticle peak near the antinodal (AN) wave vector is broad and has an energy of 30 meV or greater. As the doping increases beyond  $p_c$  and  $T_c$  decreases below 70 K, the AN peak suddenly sharpens, and its energy drops below 20 meV. From Table I, we see that  $\Delta_{\text{spin}} \approx 24$  meV for overdoped Bi2212 with  $T_c = 70$  K. To illustrate this, we have plotted in Fig. 8 the AN peak energy from [93] as a function of  $p$ , together with  $\Delta_c$  estimated from  $T_c$  and the results for  $\Delta_{\text{spin}}$  listed in Table I. It appears that the superconducting gap becomes coherent around the entire Fermi surface when  $\Delta_0$  drops below  $\Delta_{\text{spin}}$ . This interpretation is consistent with the observation of a crossover of Bogoliubov quasiparticles from an arc to a full Fermi surface at  $p_c$  by STM [20]. (We note that Ref. [94] interprets the change in AN electronic self-energy in

terms of electron-phonon coupling. While we do not dispute that  $e$ -ph coupling may play some role, we argue that the dominant effect is associated with spin fluctuations.)

Evidence for strong damping of quasiparticles interacting with spin correlations also comes from measurements on samples with no spin gap, such as LSCO with  $x < 0.13$  [97,98]. For example, an ARPES study on LSCO  $x = 0.08$  found a broad spectral function at the node (compared to  $x = 0.145$ ) with a peak well below the Fermi energy even for  $T < T_c$  [99]. This feature was interpreted as a nodal superconducting gap. We suggest that this is a signature of strong damping due to the presence of gapless AF spin fluctuations. Indeed, Loder *et al.* [100] found a gap in Hartree-Fock calculations of a pair-density-wave state, including spin-stripe order. Similar behavior is also seen in Bi2212 in the most underdoped, yet superconducting, samples [101]. The absence of coherent quasiparticles in underdoped LSCO is also evident from the large magnitude of the in-plane resistivity [102,103] and the fact that the low-temperature resistivity shows insulating behavior when the superconductivity is suppressed by a strong magnetic field [104].

Of course, while sharply defined quasiparticles are the starting point for the BCS model [105], they are not a general prerequisite for coherent superconductivity [106]. In cuprates, there is reason to believe that this occurs by Josephson coupling between local regions with strong hole pairing, just as has been proposed for the most extreme case of stripe ordered  $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$  [107–110]. While it may seem surprising that superconductivity could occur without normal-state quasiparticles, we note that this is exactly what happens along the  $c$  axis in most underdoped cuprates, where transport is incoherent and Josephson coupling is essential [111].

## V. SUMMARY

Our neutron scattering investigation of low-energy spin fluctuations in LSCO near  $x_c \sim 0.19$  has led to two significant conclusions. First, the presence of structure in the energy dependence of magnetic spectral weight in the normal state is inconsistent with antiferromagnetic critical behavior. Second, looking at the shift in the magnetic spectral weight across  $T_c$  leads to the proposal that low-energy spin fluctuations limit the superconducting order. Comparisons with the coherent superconducting gap defined by electronic spectroscopies lead to the relation  $\Delta_c \leq \Delta_{\text{spin}}$ . This relation is closely connected to the crossover from superconducting coherence on a finite arc ( $\Delta_c < \Delta_0$ ) for  $p < p_c$  to coherence all around the nominal Fermi surface ( $\Delta_c = \Delta_0$ ) for  $p > p_c$ .

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- [1] B. Keimer, S. A. Kivelson, M. R. Norman, S. Uchida, and J. Zaanen, From quantum matter to high-temperature superconductivity in copper oxides, *Nature (London)* **518**, 179 (2015).
- [2] D. N. Basov and T. Timusk, Electrodynamics of high- $T_c$  superconductors, *Rev. Mod. Phys.* **77**, 721 (2005).
- [3] M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Magnetic, transport, and optical properties of monolayer copper oxides, *Rev. Mod. Phys.* **70**, 897 (1998).
- [4] J. Orenstein and A. J. Millis, Advances in the physics of high-temperature superconductivity, *Science* **288**, 468 (2000).
- [5] P. A. Lee, N. Nagaosa, and X.-G. Wen, Doping a Mott insulator: Physics of high-temperature superconductivity, *Rev. Mod. Phys.* **78**, 17 (2006).
- [6] M. Fujita, H. Hiraka, M. Matsuda, M. Matsuura, J. M. Tranquada, S. Wakimoto, G. Xu, and K. Yamada, Progress in neutron scattering studies of spin excitations in high- $T_c$  cuprates, *J. Phys. Soc. Jpn.* **81**, 011007 (2012).
- [7] W. J. Padilla, Y. S. Lee, M. Dumm, G. Blumberg, S. Ono, K. Segawa, S. Komiyama, Y. Ando, and D. N. Basov, Constant effective mass across the phase diagram of high- $T_c$  cuprates, *Phys. Rev. B* **72**, 060511 (2005).
- [8] L. P. Gor'kov and G. B. Teitel'baum, Interplay of Externally Doped and Thermally Activated Holes in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and Their Impact on the Pseudogap Crossover, *Phys. Rev. Lett.* **97**, 247003 (2006).
- [9] J. L. Tallon and J. W. Loram, The doping dependence of  $T^*$ —What is the real high- $T_c$  phase diagram? *Physica C (Amsterdam, Neth.)* **349**, 53 (2001).
- [10] S. Badoux, W. Tabis, F. Laliberté, G. Grissonnanche, B. Vignolle, D. Vignolles, J. Béard, D. A. Bonn, W. N. Hardy, R. Liang, N. Doiron-Leyraud, L. Taillefer, and C. Proust, Change of carrier density at the pseudogap critical point of a cuprate superconductor, *Nature (London)* **531**, 210 (2016).
- [11] S. A. Kivelson and E. Fradkin, How optimal inhomogeneity produces high temperature superconductivity, in *Handbook of High-Temperature Superconductivity*, edited by J. R. Schrieffer and J. S. Brooks (Springer, New York, 2007), pp. 570–596.
- [12] D. J. Scalapino, A common thread: The pairing interaction for unconventional superconductors, *Rev. Mod. Phys.* **84**, 1383 (2012).
- [13] A. Eberlein, W. Metzner, S. Sachdev, and H. Yamase, Fermi Surface Reconstruction and Drop in the Hall Number due to Spiral Antiferromagnetism in High- $T_c$  Cuprates, *Phys. Rev. Lett.* **117**, 187001 (2016).
- [14] G. Deutscher, Coherence and single-particle excitations in the high-temperature superconductors, *Nature (London)* **397**, 410 (1999).
- [15] T. P. Devereaux and R. Hackl, Inelastic light scattering from correlated electrons, *Rev. Mod. Phys.* **79**, 175 (2007).
- [16] W. S. Lee, I. M. Vishik, K. Tanaka, D. H. Lu, T. Sasagawa, N. Nagaosa, T. P. Devereaux, Z. Hussain, and Z. X. Shen, Abrupt onset of a second energy gap at the superconducting transition of underdoped  $\text{Bi}2212$ , *Nature (London)* **450**, 81 (2007).
- [17] A. Kaminski, T. Kondo, T. Takeuchi, and G. Gu, Pairing, pseudogap and Fermi arcs in cuprates, *Philos. Mag.* **95**, 453 (2015).
- [18] A. Pushp, C. V. Parker, A. N. Pasupathy, K. K. Gomes, S. Ono, J. Wen, Z. Xu, G. Gu, and A. Yazdani, Extending universal nodal excitations optimizes superconductivity in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , *Science* **324**, 1689 (2009).
- [19] T. Kurosawa, T. Yoneyama, Y. Takano, M. Hagiwara, R. Inoue, N. Hagiwara, K. Kurusu, K. Takeyama, N. Momono, M. Oda, and M. Ido, Large pseudogap and nodal superconducting gap in  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$  and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ : Scanning tunneling microscopy and spectroscopy, *Phys. Rev. B* **81**, 094519 (2010).
- [20] K. Fujita, C. K. Kim, I. Lee, J. Lee, M. H. Hamidian, I. A. Firmo, S. Mukhopadhyay, H. Eisaki, S. Uchida, M. J. Lawler, E.-A. Kim, and J. C. Davis, Simultaneous transitions in cuprate momentum-space topology and electronic symmetry breaking, *Science* **344**, 612 (2014).
- [21] M. Eschrig, The effect of collective spin-1 excitations on electronic spectra in high- $T_c$  superconductors, *Adv. Phys.* **55**, 47 (2006).
- [22] P. W. Anderson, A re-examination of concepts in magnetic metals: The 'nearly antiferromagnetic Fermi liquid,' *Adv. Phys.* **46**, 3 (1997).
- [23] P. G. Radaelli, D. G. Hinks, A. W. Mitchell, B. A. Hunter, J. L. Wagner, B. Dabrowski, K. G. Vandervoort, H. K. Viswanathan, and J. D. Jorgensen, Structural and superconducting properties of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  as a function of Sr content, *Phys. Rev. B* **49**, 4163 (1994).
- [24] G. E. Granroth, A. I. Kolesnikov, T. E. Sherline, J. P. Clancy, K. A. Ross, J. P. C. Ruff, B. D. Gaulin, and S. E. Nagler, SEQUOIA: A Newly Operating Chopper Spectrometer at the SNS, *J. Phys.: Conf. Ser.* **251**, 012058 (2010).
- [25] O. Arnold, J. C. Bilheux, J. M. Borreguero, A. Buts, S. I. Campbell, L. Chapon, M. Doucet, N. Draper, R. Ferraz Leal, M. A. Gigg, V. E. Lynch, A. Markvardsen, D. J. Mikkelsen, R. L. Mikkelsen, R. Miller, K. Palmen, P. Parker, G. Passos, T. G. Perring, P. F. Peterson, S. Ren, M. A. Reuter, A. T. Savici, J. W. Taylor, R. J. Taylor, R. Tolchenov, W. Zhou, and J. Zikovsky, Mantid—Data analysis and visualization package for neutron scattering and  $\mu\text{SR}$  experiments, *Nucl. Instrum. Methods Phys. Res., Sect. A* **764**, 156 (2014).
- [26] G. Xu, Z. Xu, and J. M. Tranquada, Absolute cross-section normalization of magnetic neutron scattering data, *Rev. Sci. Instrum.* **84**, 083906 (2013).
- [27] A. C. Walters, T. G. Perring, J.-S. Caux, A. T. Savici, G. D. Gu, C.-C. Lee, W. Ku, and I. A. Zaliznyak, Effect of covalent bonding on magnetism and the missing neutron intensity in copper oxide compounds, *Nat. Phys.* **5**, 867 (2009).
- [28] G. Aeppli, T. E. Mason, S. M. Hayden, H. A. Mook, and J. Kulda, Nearly singular magnetic fluctuations in the normal state of a high- $T_c$  cuprate superconductor, *Science* **278**, 1432 (1997).
- [29] B. Vignolle, S. M. Hayden, D. F. McMorrow, H. M. Rønnow, B. Lake, C. D. Frost, and T. G. Perring, Two energy scales in the spin excitations of the high-temperature superconductor  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , *Nat. Phys.* **3**, 163 (2007).
- [30] O. J. Lipscombe, S. M. Hayden, B. Vignolle, D. F. McMorrow, and T. G. Perring, Persistence of High-Frequency Spin Fluctuations in Overdoped Superconducting  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $x = 0.22$ ), *Phys. Rev. Lett.* **99**, 067002 (2007).
- [31] J. J. Wagman, D. Parshall, M. B. Stone, A. T. Savici, Y. Zhao, H. A. Dabkowska, and B. D. Gaulin, Quasi-two-dimensional



- spin and phonon excitations in  $\text{La}_{1.965}\text{Ba}_{0.035}\text{CuO}_4$ , *Phys. Rev. B* **91**, 224404 (2015).
- [32] J. J. Wagman, J. P. Carlo, J. Gaudet, G. Van Gastel, D. L. Abernathy, M. B. Stone, G. E. Granroth, A. I. Kolesnikov, A. T. Savici, Y. J. Kim, H. Zhang, D. Ellis, Y. Zhao, L. Clark, A. B. Kallin, E. Mazurek, H. A. Dabkowska, and B. D. Gaulin, Neutron scattering studies of spin-phonon hybridization and superconducting spin gaps in the high-temperature superconductor  $\text{La}_{2-x}(\text{Sr}, \text{Ba})_x\text{CuO}_4$ , *Phys. Rev. B* **93**, 094416 (2016).
- [33] N. E. Hussey, J. Buhot, and S. Licciardello, A tale of two metals: Contrasting criticalities in the pnictides and hole-doped cuprates, *Rep. Prog. Phys.* **81**, 052501 (2018).
- [34] K. Yamada, S. Wakimoto, G. Shirane, C. H. Lee, M. A. Kastner, S. Hosoya, M. Greven, Y. Endoh, and R. J. Birgeneau, Direct Observation of a Magnetic Gap in Superconducting  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  ( $T_c = 37.3$  K), *Phys. Rev. Lett.* **75**, 1626 (1995).
- [35] S. Petit, A. H. Moudden, B. Hennion, A. Vietkin, and A. Revcolevschi, Spin dynamics study of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  by inelastic neutron scattering, *Physica B (Amsterdam, Neth.)* **234-236**, 800 (1997).
- [36] T. E. Mason, G. Aeppli, and H. A. Mook, Magnetic Dynamics of Superconducting  $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$ , *Phys. Rev. Lett.* **68**, 1414 (1992).
- [37] B. Lake, G. Aeppli, T. E. Mason, A. Schröder, D. F. McMorrow, K. Lefmann, M. Isshiki, M. Nohara, H. Takagi, and S. M. Hayden, Spin gap and magnetic coherence in a clean high-temperature superconductor, *Nature (London)* **400**, 43 (1999).
- [38] H.-Y. Kee, S. A. Kivelson, and G. Aeppli, Spin-1 Neutron Resonance Peak Cannot Account for Electronic Anomalies in the Cuprate Superconductors, *Phys. Rev. Lett.* **88**, 257002 (2002).
- [39] B. Muschler, W. Prestel, L. Tassini, R. Hackl, M. Lambacher, A. Erb, S. Komiyama, Y. Ando, D. C. Peets, W. N. Hardy, R. Liang, and D. A. Bonn, Electron interactions and charge ordering in  $\text{CuO}_2$  compounds, *Eur. Phys. J. Spec. Top.* **188**, 131 (2010).
- [40] S. Sugai, Y. Takayanagi, N. Hayamizu, T. Muroi, R. Shiozaki, J. Nohara, K. Takenaka, and K. Okazaki, Superconducting pairing and the pseudogap in the nematic dynamical stripe phase of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , *J. Phys.: Condens. Matter* **25**, 475701 (2013).
- [41] R. S. Gonnelli, A. Calzolari, D. Daghero, L. Natale, G. A. Ummarino, V. A. Stepanov, and M. Ferretti, Evidence for pseudogap and phase-coherence gap separation by Andreev reflection experiments in  $\text{Au/La}_{2-x}\text{Sr}_x\text{CuO}_4$  point-contact junctions, *Eur. Phys. J. B* **22**, 411 (2001).
- [42] C. Stock, W. J. L. Buyers, R. A. Cowley, P. S. Clegg, R. Coldea, C. D. Frost, R. Liang, D. Peets, D. Bonn, W. N. Hardy, and R. J. Birgeneau, From incommensurate to dispersive spin-fluctuations: The high-energy inelastic spectrum in superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ , *Phys. Rev. B* **71**, 024522 (2005).
- [43] S. M. Hayden, H. A. Mook, P. Dai, T. G. Perring, and F. Doğan, The structure of the high-energy spin excitations in a high-transition-temperature superconductor, *Nature (London)* **429**, 531 (2004).
- [44] D. Reznik, J.-P. Ismer, I. Eremin, L. Pintschovius, T. Wolf, M. Arai, Y. Endoh, T. Masui, and S. Tajima, Local-moment fluctuations in the optimally doped high- $T_c$  superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ , *Phys. Rev. B* **78**, 132503 (2008).
- [45] H. Woo, P. C. Dai, S. M. Hayden, H. A. Mook, T. Dahm, D. J. Scalapino, T. G. Perring, and F. Doğan, Magnetic energy change available to superconducting condensation in optimally doped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ , *Nat. Phys.* **2**, 600 (2006).
- [46] H. F. Fong, P. Bourges, Y. Sidis, L. P. Regnault, A. Ivanov, G. D. Gu, N. Koshizuka, and B. Keimer, Neutron scattering from magnetic excitations in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , *Nature (London)* **398**, 588 (1999).
- [47] G. Xu, G. D. Gu, M. Hucker, B. Fauque, T. G. Perring, L. P. Regnault, and J. M. Tranquada, Testing the itinerancy of spin dynamics in superconducting  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , *Nat. Phys.* **5**, 642 (2009).
- [48] B. Fauqué, Y. Sidis, L. Capogna, A. Ivanov, K. Hradil, C. Ulrich, A. I. Rykov, B. Keimer, and P. Bourges, Dispersion of the odd magnetic resonant mode in near-optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , *Phys. Rev. B* **76**, 214512 (2007).
- [49] H. He, Y. Sidis, P. Bourges, G. D. Gu, A. Ivanov, N. Koshizuka, B. Liang, C. T. Lin, L. P. Regnault, E. Schoenher, and B. Keimer, Resonant Spin Excitation in an Overdoped High Temperature Superconductor, *Phys. Rev. Lett.* **86**, 1610 (2001).
- [50] L. Capogna, B. Fauque, Y. Sidis, C. Ulrich, P. Bourges, S. Pailhes, A. Ivanov, J. L. Tallon, B. Liang, C. T. Lin, A. I. Rykov, and B. Keimer, Odd and even magnetic resonant modes in highly overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , *Phys. Rev. B* **75**, 060502 (2007).
- [51] M. K. Chan, C. J. Dorow, L. Mangin-Thro, Y. Tang, Y. Ge, M. J. Veit, G. Yu, X. Zhao, A. D. Christianson, J. T. Park, Y. Sidis, P. Steffens, D. L. Abernathy, P. Bourges, and M. Greven, Commensurate antiferromagnetic excitations as a signature of the pseudogap in the tetragonal high- $T_c$  cuprate  $\text{HgBa}_2\text{CuO}_{4+\delta}$ , *Nat. Commun.* **7**, 10819 (2016).
- [52] M. K. Chan, Y. Tang, C. J. Dorow, J. Jeong, L. Mangin-Thro, M. J. Veit, Y. Ge, D. L. Abernathy, Y. Sidis, P. Bourges, and M. Greven, Hourglass Dispersion and Resonance of Magnetic Excitations in the Superconducting State of the Single-Layer Cuprate  $\text{HgBa}_2\text{CuO}_{4+\delta}$  near Optimal Doping, *Phys. Rev. Lett.* **117**, 277002 (2016).
- [53] K. Yamada, K. Kurahashi, T. Uefuji, M. Fujita, S. Park, S.-H. Lee, and Y. Endoh, Commensurate Spin Dynamics in the Superconducting State of an Electron-Doped Cuprate Superconductor, *Phys. Rev. Lett.* **90**, 137004 (2003).
- [54] J. Zhao, P. Dai, S. Li, P. G. Freeman, Y. Onose, and Y. Tokura, Neutron-Spin Resonance in the Optimally Electron-Doped Superconductor  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$ , *Phys. Rev. Lett.* **99**, 017001 (2007).
- [55] N. Munnikes, B. Muschler, F. Venturini, L. Tassini, W. Prestel, S. Ono, Y. Ando, D. C. Peets, W. N. Hardy, R. Liang, D. A. Bonn, A. Damascelli, H. Eisaki, M. Greven, A. Erb, and R. Hackl, Pair breaking versus symmetry breaking: Origin of the Raman modes in superconducting cuprates, *Phys. Rev. B* **84**, 144523 (2011).
- [56] W. Guyard, A. Sacuto, M. Cazayous, Y. Gallais, M. Le Tacon, D. Colson, and A. Forget, Temperature Dependence of the Gap Size near the Brillouin-Zone Nodes of  $\text{HgBa}_2\text{CuO}_{4+\delta}$  Superconductors, *Phys. Rev. Lett.* **101**, 097003 (2008).
- [57] Y. Li, M. Le Tacon, Y. Matiks, A. V. Boris, T. Loew, C. T. Lin, L. Chen, M. K. Chan, C. Dorow, L. Ji, N. Barišić, X. Zhao,



- M. Greven, and B. Keimer, Doping-Dependent Photon Scattering Resonance in the Model High-Temperature Superconductor  $\text{HgBa}_2\text{CuO}_{4+\delta}$  Revealed by Raman Scattering and Optical Ellipsometry, *Phys. Rev. Lett.* **111**, 187001 (2013).
- [58] G. Blumberg, A. Koitzsch, A. Gozar, B. S. Dennis, C. A. Kendziora, P. Fournier, and R. L. Greene, Nonmonotonic  $d_{x^2-y^2}$  Superconducting Order Parameter in  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ , *Phys. Rev. Lett.* **88**, 107002 (2002).
- [59] J. Y. T. Wei, N.-C. Yeh, D. F. Garrigus, and M. Strasik, Directional Tunneling and Andreev Reflection on  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  Single Crystals: Predominance of  $d$ -Wave Pairing Symmetry Verified with the Generalized Blonder, Tinkham, and Klapwijk Theory, *Phys. Rev. Lett.* **81**, 2542 (1998).
- [60] J. Ruvalds, C. T. Rieck, J. Zhang, and A. Virosztek, Spin susceptibility scaling in high-temperature superconductors, *Science* **256**, 1664 (1992).
- [61] J. P. Lu, Neutron Scattering as a Probe for Unconventional Superconducting States, *Phys. Rev. Lett.* **68**, 125 (1992).
- [62] N. Bulut and D. J. Scalapino, Weak-coupling analysis of the neutron-scattering spectral weight, *Phys. Rev. B* **47**, 3419 (1993).
- [63] Q. Si, Y. Zha, K. Levin, and J. P. Lu, Comparison of spin dynamics in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ : Effects of Fermi-surface geometry, *Phys. Rev. B* **47**, 9055 (1993).
- [64] D. Z. Liu, Y. Zha, and K. Levin, Theory of Neutron Scattering in the Normal and Superconducting States of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ , *Phys. Rev. Lett.* **75**, 4130 (1995).
- [65] F. Onufrieva and P. Pfeuty, Spin dynamics of a two-dimensional metal in a superconducting state: Application to the high- $T_c$  cuprates, *Phys. Rev. B* **65**, 054515 (2002).
- [66] M. R. Norman, Linear response theory and the universal nature of the magnetic excitation spectrum of the cuprates, *Phys. Rev. B* **75**, 184514 (2007).
- [67] J. Rossat-Mignod, L. P. Regnault, C. Vettier, P. Bourges, P. Bulet, J. Bossy, J. Y. Henry, and G. Lapertot, Spin dynamics in the high- $T_c$  system  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ , *Phys. B (Amsterdam, Neth.)* **180-181**, 383 (1992).
- [68] H. A. Mook, M. Yethiraj, G. Aeppli, T. E. Mason, and T. Armstrong, Polarized Neutron Determination of the Magnetic Excitations in  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , *Phys. Rev. Lett.* **70**, 3490 (1993).
- [69] H. F. Fong, B. Keimer, D. Reznik, D. L. Milius, and I. A. Aksay, Polarized and unpolarized neutron-scattering study of the dynamical spin susceptibility of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , *Phys. Rev. B* **54**, 6708 (1996).
- [70] M. Arai, T. Nishijima, Y. Endoh, T. Egami, S. Tajima, K. Tomimoto, Y. Shiohara, M. Takahashi, A. Garrett, and S. M. Bennington, Incommensurate Spin Dynamics of Underdoped Superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{6.7}$ , *Phys. Rev. Lett.* **83**, 608 (1999).
- [71] P. Bourges, Y. Sidis, H. F. Fong, L. P. Regnault, J. Bossy, A. Ivanov, and B. Keimer, The spin excitation spectrum in superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6.85}$ , *Science* **288**, 1234 (2000).
- [72] J. M. Tranquada, H. Woo, T. G. Perring, H. Goka, G. D. Gu, G. Xu, M. Fujita, and K. Yamada, Quantum magnetic excitations from stripes in copper oxide superconductors, *Nature (London)* **429**, 534 (2004).
- [73] V. Hinkov, P. Bourges, S. Pailhes, Y. Sidis, A. Ivanov, C. D. Frost, T. G. Perring, C. T. Lin, D. P. Chen, and B. Keimer, Spin dynamics in the pseudogap state of a high-temperature superconductor, *Nat. Phys.* **3**, 780 (2007).
- [74] P. Dai, H. A. Mook, R. D. Hunt, and F. Doğan, Evolution of the resonance and incommensurate spin fluctuations in superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ , *Phys. Rev. B* **63**, 054525 (2001).
- [75] R. J. Birgeneau, C. Stock, J. M. Tranquada, and K. Yamada, Magnetic neutron scattering in hole-doped cuprate superconductors, *J. Phys. Soc. Jpn.* **75**, 111003 (2006).
- [76] E. W. Huang, C. B. Mendl, S. Liu, S. Johnston, H.-C. Jiang, B. Moritz, and T. P. Devereaux, Numerical evidence of fluctuating stripes in the normal state of high- $T_c$  cuprate superconductors, *Science* **358**, 1161 (2017).
- [77] A. Nocera, Y. Wang, N. D. Patel, G. Alvarez, T. A. Maier, E. Dagotto, and S. Johnston, Doping evolution of charge and spin excitations in two-leg Hubbard ladders: Comparing DMRG and FLEX results, *Phys. Rev. B* **97**, 195156 (2018).
- [78] M. Hücker, G. D. Gu, and J. M. Tranquada, Spin susceptibility of underdoped cuprate superconductors: Insights from a stripe-ordered crystal, *Phys. Rev. B* **78**, 214507 (2008).
- [79] I. Vekhter and A. V. Chubukov, Non-Fermi-Liquid Behavior in Itinerant Antiferromagnets, *Phys. Rev. Lett.* **93**, 016405 (2004).
- [80] T. Ito, K. Takenaka, and S. Uchida, Systematic Deviation from  $T$ -linear Behavior in the In-Plane Resistivity of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ : Evidence for Dominant Spin Scattering, *Phys. Rev. Lett.* **70**, 3995 (1993).
- [81] M. Takigawa, A. P. Reyes, P. C. Hammel, J. D. Thompson, R. H. Heffner, Z. Fisk, K. C. Ott, Cu and O NMR studies of the magnetic properties of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.63}$  ( $t_c = 62$  K), *Phys. Rev. B* **43**, 247 (1991).
- [82] S.-H. Baek, T. Loew, V. Hinkov, C. T. Lin, B. Keimer, B. Büchner, and H.-J. Grafe, Evidence of a critical hole concentration in underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_y$  single crystals revealed by  $^{63}\text{Cu}$  NMR, *Phys. Rev. B* **86**, 220504 (2012).
- [83] A. J. Millis, S. Sachdev, and C. M. Varma, Inelastic scattering and pair breaking in anisotropic and isotropic superconductors, *Phys. Rev. B* **37**, 4975 (1988).
- [84] T. Dahm and D. J. Scalapino, Dependence of  $T_c$  on the  $q - \omega$  structure of the spin-fluctuation spectrum, *Phys. Rev. B* **97**, 184504 (2018).
- [85] T. Dahm, V. Hinkov, S. V. Borisenko, A. A. Kordyuk, V. B. Zabolotnyy, J. Fink, B. Büchner, D. J. Scalapino, W. Hanke, and B. Keimer, Strength of the spin-fluctuation-mediated pairing interaction in a high-temperature superconductor, *Nat. Phys.* **5**, 217 (2009).
- [86] N. Zaki, H.-B. Yang, J. D. Rameau, P. D. Johnson, H. Claus, and D. G. Hinks, Cuprate phase diagram and the influence of nanoscale inhomogeneities, *Phys. Rev. B* **96**, 195163 (2017).
- [87] L. Taillefer, Fermi surface reconstruction in high- $T_c$  superconductors, *J. Phys.: Condens. Matter* **21**, 164212 (2009).
- [88] S. E. Sebastian and C. Proust, Quantum oscillations in hole-doped cuprates, *Annu. Rev. Condens. Matter Phys.* **6**, 411 (2015).
- [89] N. Barišić, S. Badoux, M. K. Chan, C. Dorow, W. Tabis, B. Vignolle, G. Yu, J. Béard, X. Zhao, C. Proust, and M. Greven, Universal quantum oscillations in the underdoped cuprate superconductors, *Nat. Phys.* **9**, 761 (2013).
- [90] M. Hücker, N. B. Christensen, A. T. Holmes, E. Blackburn, E. M. Forgan, R. Liang, D. A. Bonn, W. N. Hardy, O. Gutowski, M. v. Zimmermann, S. M. Hayden, and J. Chang, Competing charge, spin, and superconducting orders in underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_y$ , *Phys. Rev. B* **90**, 054514 (2014).

- [91] S. E. Sebastian, N. Harrison, and G. G. Lonzarich, Towards resolution of the Fermi surface in underdoped high- $T_c$  superconductors, *Rep. Prog. Phys.* **75**, 102501 (2012).
- [92] V. Hinkov, D. Haug, B. Fauqué, P. Bourges, Y. Sidis, A. Ivanov, C. Bernhard, C. T. Lin, and B. Keimer, Electronic liquid crystal state in superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6.45}$ , *Science* **319**, 597 (2008).
- [93] I. K. Drozdov, I. Pletikosić, C.-K. Kim, K. Fujita, G. D. Gu, J. C. S. Davis, P. D. Johnson, I. Božović, and T. Valla, Phase diagram of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  revisited, *Nat. Commun.* **9**, 5201 (2018).
- [94] Y. He, M. Hashimoto, D. Song, S.-D. Chen, J. He, I. M. Vishik, B. Moritz, D.-H. Lee, N. Nagaosa, J. Zaanen, T. P. Devereaux, Y. Yoshida, H. Eisaki, D. H. Lu, and Z.-X. Shen, Rapid change of superconductivity and electron-phonon coupling through critical doping in Bi-2212, *Science* **362**, 62 (2018).
- [95] Y.-G. Zhong, J.-Y. Guan, X. Shi, J. Zhao, Z.-C. Rao, C.-Y. Tang, H.-J. Liu, Z. Y. Weng, Z. Q. Wang, G. D. Gu, T. Qian, Y.-J. Sun, and H. Ding, Continuous doping of a cuprate surface: Insights from *in situ* angle-resolved photoemission, *Phys. Rev. B* **98**, 140507(R) (2018).
- [96] S. D. Obertelli, J. R. Cooper, and J. L. Tallon, Systematics in the thermoelectric power of high- $T_c$  oxides, *Phys. Rev. B* **46**, 14928 (1992).
- [97] J. Chang, Ch. Niedermayer, R. Gilardi, N. B. Christensen, H. M. Ronnow, D. F. McMorrow, M. Ay, J. Stahn, O. Sobolev, A. Hiess, S. Pailhes, C. Baines, N. Momono, M. Oda, M. Ido, and J. Mesot, Tuning competing orders in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  cuprate superconductors by the application of an external magnetic field, *Phys. Rev. B* **78**, 104525 (2008).
- [98] H. Jacobsen, I. A. Zaliznyak, A. T. Savici, B. L. Winn, S. Chang, M. Hücker, G. D. Gu, and J. M. Tranquada, Neutron scattering study of spin ordering and stripe pinning in superconducting  $\text{La}_{1.93}\text{Sr}_{0.07}\text{CuO}_4$ , *Phys. Rev. B* **92**, 174525 (2015).
- [99] E. Razzoli, G. Drachuck, A. Keren, M. Radovic, N. C. Plumb, J. Chang, Y.-B. Huang, H. Ding, J. Mesot, and M. Shi, Evolution from a Nodeless Gap to  $d_{x^2-y^2}$ -Wave in Underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , *Phys. Rev. Lett.* **110**, 047004 (2013).
- [100] F. Loder, S. Graser, A. P. Kampf, and T. Kopp, Mean-Field Pairing Theory for the Charge-Stripe Phase of High-Temperature Cuprate Superconductors, *Phys. Rev. Lett.* **107**, 187001 (2011).
- [101] I. M. Vishik, M. Hashimoto, R.-H. He, W.-S. Lee, F. Schmitt, D. Lu, R. G. Moore, C. Zhang, W. Meevasana, T. Sasagawa, S. Uchida, K. Fujita, S. Ishida, M. Ishikado, Y. Yoshida, H. Eisaki, Z. Hussain, T. P. Devereaux, and Z.-X. Shen, Phase competition in trisected superconducting dome, *Proc. Natl. Acad. Sci. USA* **109**, 18332 (2012).
- [102] Y. Ando, A. N. Lavrov, S. Komiya, K. Segawa, and X. F. Sun, Mobility of the Doped Holes and the Antiferromagnetic Correlations in Underdoped High- $T_c$  Cuprates, *Phys. Rev. Lett.* **87**, 017001 (2001).
- [103] V. J. Emery and S. A. Kivelson, Superconductivity in Bad Metals, *Phys. Rev. Lett.* **74**, 3253 (1995).
- [104] Y. Ando, G. S. Boebinger, A. Passner, T. Kimura, and K. Kishio, Logarithmic Divergence of Both In-Plane and Out-of-Plane Normal-State Resistivities of Superconducting  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  in the Zero-Temperature Limit, *Phys. Rev. Lett.* **75**, 4662 (1995).
- [105] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Theory of superconductivity, *Phys. Rev.* **108**, 1175 (1957).
- [106] A. A. Patel, M. J. Lawler, and E.-A. Kim, Coherent Superconductivity with a Large Gap Ratio from Incoherent Metals, *Phys. Rev. Lett.* **121**, 187001 (2018).
- [107] Q. Li, M. Hücker, G. D. Gu, A. M. Tsvelik, and J. M. Tranquada, Two-Dimensional Superconducting Fluctuations in Stripe-Ordered  $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ , *Phys. Rev. Lett.* **99**, 067001 (2007).
- [108] E. Berg, E. Fradkin, E.-A. Kim, S. A. Kivelson, V. Oganesyan, J. M. Tranquada, and S. C. Zhang, Dynamical Layer Decoupling in a Stripe-Ordered High- $T_c$  Superconductor, *Phys. Rev. Lett.* **99**, 127003 (2007).
- [109] E. Fradkin, S. A. Kivelson, and J. M. Tranquada, Colloquium: Theory of intertwined orders in high temperature superconductors, *Rev. Mod. Phys.* **87**, 457 (2015).
- [110] V. J. Emery, S. A. Kivelson, and O. Zachar, Spin-gap proximity effect mechanism of high-temperature superconductivity, *Phys. Rev. B* **56**, 6120 (1997).
- [111] S. O. Katterwe, A. Rydh, and V. M. Krasnov, Doping-Induced Change in the Interlayer Transport Mechanism of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  near the Superconducting Transition Temperature, *Phys. Rev. Lett.* **101**, 087003 (2008).