Kinks and waterfalls as signatures of competing order in angle-resolved photoemission spectra of La$_{2-x}$Sr$_x$CuO$_4$

G. Mazza$^{1,2}$, M. Grilli$^{2,3}$, C. Di Castro$^{2,3}$, and S. Caprara$^{2,3}$

$^1$ International School for Advanced Studies (SISSA), Via Bonomea 265 34136 Trieste, Italy
$^2$ CNISM and Dipartimento di Fisica, Università di Roma “La Sapienza” Piazzale Aldo Moro 5, I-00185 Roma, Italy
$^3$ Consiglio Nazionale delle Ricerche, Istituto dei Sistemi Complessi, via dei Tarmini, I-00185 Roma, Italy

We show that the so-called kinks and waterfalls observed in angle-resolved photoemission spectra of La$_{2-x}$Sr$_x$CuO$_4$, a prototypical high-$T_c$ superconducting cuprate, result from the coupling of quasiparticles with two distinct nearly critical collective modes with finite characteristic wave vectors, typical of charge and spin fluctuations near a stripe instability. Both phonon-like charge and spin collective modes are needed to account for the kinked quasiparticle dispersions. This clarifies the long-standing question whether kinks are due to phonons or spin waves and the nature of the bosonic mediators of the electron-electron effective interaction in La$_{2-x}$Sr$_x$CuO$_4$.

The metallic phase of high-$T_c$ superconducting cuprates evolves remarkably with changing the temperature $T$ and the doping $x$. A normal Fermi-liquid behavior is only found in overdoped samples, with $x$ larger than the optimal value $x_{\text{opt}}$ (where the maximum superconducting critical temperature $T_c$ is achieved). At $x \approx x_{\text{opt}}$ and $T > T_c$ the metallic phase seems to be ruled by the temperature as the only relevant energy scale, a typical signature of quantum criticality. In underdoped samples, with $x < x_{\text{opt}}$, an even stronger anomaly is found, with a pseudogap opening around the Fermi energy, below a doping dependent temperature $T^*(x)$. Whether this is accompanied by the onset of some sort of ordering is still matter of debate. Nonetheless, models with nearly critical collective modes (CMs) coupled to fermion quasiparticles (QPs) may not only explain the anomalous metallic phase, but also provide candidate mediators of a retarded pairing interaction (the so-called glue) $^1$ $^3$, alternative to phonons in ordinary superconductors, and are therefore actively investigated. Various proposals for sources of nearly critical CMs include the antiferromagnetic phase at $x \approx 0$ $^4$, time-reversal-breaking plaquette currents $^5$, order parameters with exotic wave symmetry $^6$ $^7$, or stripe ordering $^8$ $^9$.

In this Letter, we show that the so-called kinks and waterfalls $^{10}$ observed in angle-resolved photoemission spectroscopy (ARPES) identify charge (C) and spin (S) CMs on the verge of a stripe instability as the main source of scattering in La$_{2-x}$Sr$_x$CuO$_4$ (LSCO), solving the long-standing phonon-vs-spin issue, at least in LSCO. The characteristic low-energy scale of the two CMs suggests that a quantum critical point occurs at $x_{\text{QCP}} \approx 0.19$ $^{21}$ $^{23}$, associated to a phase with stripe-like C and S modulation, whose onset occurs via a harmonic incommensurate charge-density wave instability. The other CM, more diffusive and extending to higher energies, is associated with S fluctuations peaked near the wave vector of antiferromagnetic order. The behavior of the characteristic low-energy scale of the two CMs in the overdoped regime is a consequence of the competition between the spin- and the charge-dominated region. At small $x$, S fluctuations are naturally enhanced by incipient antiferromagnetism, whereas in the optimally and overdoped regime C fluctuations dominate.

Here, we phenomenologically proceed to analyze the implications of the same two CMs, as derived from Raman experiments, on QP spectra. We consider the general Gaussian form of CM propagator

$$D_{{\lambda}}(\mathbf{q}, \omega_n) = -\frac{1}{T_{{\lambda}}(\mathbf{q} - \mathbf{Q}_0)} + |\omega_n| + \omega_n^2/\Omega_{{\lambda}},$$

(1)

where $\lambda = C, S$, $\omega_n$ is the bosonic Matsubara frequency, $T_{{\lambda}}(\mathbf{q}) = m_{{\lambda}} + \nu_{{\lambda}}[2 - \cos(q_x a) - \cos(q_y a)]$ describes the dispersion of a lattice periodic CM, and reproduces the behavior $T_{{\lambda}}(\mathbf{q} - \mathbf{Q}_0) \approx 0 \approx m_{{\lambda}} + \nu_{{\lambda}}/\Omega_{{\lambda}}(\mathbf{q} - \mathbf{Q}_0)^2$ obtained in different contexts for C $^4$ $^6$ and S $^4$ $^{26}$ CMs. $m_{{\lambda}}$ is proportional to the inverse squared correlation length $\xi_{{\lambda}}^{-2}$, $\nu_{{\lambda}}$ sets the curvature at the bottom of the CM dispersion law, and $a$ is the spacing of the two-dimensional square lattice describing the CuO$_2$ planes of cuprates.
henceforth taken as unit length. The propagator \( \Pi \) is peaked at a characteristic wave vector \( Q_\lambda \), has a diffusive character at low energy, and becomes more propagating above the energy scale \( \overline{\Pi}_\lambda \). The CM dispersion is limited by an energy cutoff \( \Lambda_\lambda \), setting a momentum cutoff \( |\mathbf{q}|_\lambda \approx (\Lambda_\lambda/\nu_\lambda)^{1/2} \). The values of the characteristic wave vectors, \( Q_C \) and \( Q_S \), are extracted from neutron scattering experiments: The incommensurability of the \( S \) density modulation, half of the incommensurability of the \( C \) density modulation when observed at \( x = 1/8 \) \([30, 31]\), saturates for \( x > 1/8 \), yielding \( Q_C = \pi (\pm \frac{1}{2}, 0), \pi (0, \pm \frac{1}{2}) \), \( Q_S = \pi (1 \pm \frac{1}{2}, 1), \pi (1, 1 \pm \frac{1}{2}) \).

We adopt for the fermion QPs on the CuO\(_2\) planes of LSCO a tight-binding dispersion law including nearest \((t = 400 \text{ meV})\) and next-to-nearest \((t' = -0.21t)\) neighbor hopping terms,

\[
\epsilon_\mathbf{k} = -2t (\cos k_x + \cos k_y) - 4t' \cos k_x \cos k_y - \mu, \quad (2)
\]

where \( \mu \) is the chemical potential. Similarly to the electron-phonon coupling, QPs are here coupled to CMs through dimensional coupling constants \( g_\lambda \). The survey of Raman data on LSCO \([17]\) yielded the doping evolutions of \( m, \Lambda, \overline{\Pi}, \) and \( \kappa \equiv g^2/\nu \), reported in Tab. I. Our aim is to fit ARPES data with the same CM parameters, although it should be borne in mind that Raman response accounts for the suppression of the CM-QP coupling away from the lowest-order QP self-energy \( \Sigma(\mathbf{k}, \omega) \), and therefore the value of \( \overline{\Pi}_\lambda \) is not fully constrained. We adjust \( \overline{\Pi} \) as a fitting parameter \([27]\), which in turn fixes \( \nu = \Lambda/\overline{\Pi}^2 \) and \( g = \sqrt{kT\nu} \). The obtained values (see Tab. I), yield four peaks in the CM dispersion, consistent with the observation of four separate peaks for the \( S \) CM in neutron scattering experiments \([31, 32]\).

The effect of CMs on QP spectra is captured computing the lowest-order QP self-energy \( \Sigma(k, \omega) = \Sigma_C(k, \omega) + \Sigma_S(k, \omega) \), and the single-particle spectral density

\[
A(k, \omega) = \frac{1}{\pi} \frac{\text{Im} \Sigma(k, \omega)}{| \omega - \epsilon_\mathbf{k} - \text{Re} \Sigma(k, \omega) |^2 + |\text{Im} \Sigma(k, \omega)|^2}. \tag{3}
\]

The imaginary part of the self-energy is

\[
\text{Im} \Sigma_\lambda(k, \omega) = g_\lambda^2 \int_{BZ} \frac{d^2 \mathbf{q}}{(2\pi)^2} \gamma_\lambda(q - Q_\lambda) \times [\gamma_\lambda(q - Q_\lambda) - (\omega - \epsilon_{k-q})^2/\overline{\Omega}_\lambda^2]^2 + (\omega - \epsilon_{k-q})^2, \tag{4}
\]

where a sum over the star of equivalent wave vectors \( Q_\lambda \) is understood, \( f_{\pm}(\epsilon_{k-q} - \omega) \), and the smooth cutoff function \( \gamma_\lambda(q) = \exp[-(2 - \cos q_x - \cos q_y)/Q^2] \) accounts for the suppression of the CM-QP coupling away from \( Q_\lambda \). As an order-of-magnitude estimate, \( Q_\lambda \approx |\overline{q}|_\lambda \). For any given \( k \) and \( \omega \), we numerically integrate Eq. (4) and obtain \( \text{Re} \Sigma \) via Kramers-Kronig transformation.

The chemical potential \( \mu \) is fixed imposing that

\[
2 \int_{BZ} \frac{d^2 \mathbf{k}}{(2\pi)^2} \int_{-\infty}^{+\infty} d\omega A(k, \omega) f_{+}(\omega) = 1 - x.
\]

\[
\begin{array}{|c|c|c|c|c|c|c|c|}
\hline
\chi & m & \Lambda & \overline{\Pi} & \kappa & \overline{q} & \nu & Q \\
\hline
0.15 & 2.5 & 248.0 & 25.0 & 5.5 & 0.9 & 300.0 & 0.6 \\
0.17 & 4.35 & 248.0 & 25.0 & 8.0 & 0.9 & 300.0 & 0.6 \\
0.20 & 8.7 & 310.0 & 25.0 & 11.7 & 1.0 & 300.0 & 0.5 \\
0.25 & 9.9 & 186.0 & 41.3 & 13.7 & 0.9 & 240.0 & 0.5 \\
0.26 & 7.55 & 300.0 & 41.3 & 17.5 & 1.0 & 280.0 & 0.55 \\
\hline
\end{array}
\]

**TABLE I.** Parameters of the \( C \) and \( S \) CMs: \( m, \Lambda, \overline{\Pi}, \) and \( \kappa \) are extracted from Raman data \([17]\). We calculate the ARPES intensity convoluting \( A(k, \omega) \) with a Gaussian of width \( \approx 10 \text{ meV} \), mimicking energy resolution, and considering only the occupied states.

![FIG. 1.](image_url)

We track the QP dispersions along the cuts of the BZ reported in Fig. 2 for \( x = 0.15 \) and \( T = 40 \text{K} \). Dots represent the maxima of the MDCs.

The main features of ARPES data are determined by the dynamical structure of the CMs of Eq. (1). In Fig. 3 we report the spectra along the cuts A-F of Fig. 2 for \( x = 0.15 \) and \( T = 40 \text{K} \). We track the QP disper-
FIG. 2. (Color online) Fermi surface of LSCO at $x = 0.15$ (black solid line). The dashed (red online) and solid (blue online) lines mark the C and S hot lines, respectively. The shaded circles mark the loci of the waterfalls (from Ref. [16]). The spectra in Figs. 1B are calculated along the cuts A-F.

FIG. 3. (Color online) Same as in Fig. 1 but on a narrower energy range. Although the structure of the kinks is due to both S and C CMs, the arrows mark the kinks due to (mostly) C or S scattering.

The strong scattering near the hot lines, reminiscent of the Bragg scattering occurring when some ordering takes place at specific $Q_\lambda$, gives instead rise to the high-energy waterfall features in Fig. 1.

We obtain waterfalls that compare fairly well with the experiments [10, 15, 16], although our perturbative scheme underestimates their binding energy and broadening. In particular, our hot lines reproduce the loci of the BZ where the waterfalls are observed [16] (shaded circles in Fig. 2). The waterfalls along the cuts A-C (at binding energies $\approx 600$ meV, $\approx 300$ meV, and $\approx 250$ meV, respectively) correspond to the nearly cross-shaped accumulation of the loci well inside the BZ in Fig. 2. In our scheme, these are due to C incipient order, which also produces additional waterfalls along a square contour surrounding the $\Gamma$ point of the BZ. These are visible in panels B and C of Fig. 1 at approximately $700 - 800$ meV. Their presence cannot be ascertained in Ref. [16], where the data at higher binding energy are not reported. A reanalysis of the data is required to check for the presence of these additional waterfalls.

On the other hand, both the C and S scattering are responsible for the dense occurrence of waterfalls near the $M$ points (cuts D-F in Fig. 2). However, as it is clear from panels D-F in Fig. 1, the waterfalls are shifted to lower binding energy in this region of the BZ and merge with the kinks. Moreover, approaching the hot spots, the waterfall evolves into a rounding of the QP dispersions, with a spectral intensity vanishing as $\sqrt{\omega}$ [4, 34, 35]. This rounding is reminiscent of the additional low-energy kinks observed in BSCCO [36, 37], but not in LSCO, possibly due to a lower resolution.

To follow the evolution of the kinks, we show in Fig. 3 the low-energy spectra, along the same cuts of Fig. 1. In the nodal (\textit{Γ}X) direction (cut A), the waterfall is well separated from the low-energy kink, which is more closely inspected in Fig. 4 (a). Here we report the kinked dispersion separately due to C [(red) squares] and S [(green) diamonds] CMs, and to the combination of both [(blue) circles]. The binding energy of the kink [(blue) arrow in Fig. 4 (a)] is clearly set by the C CM [(red) arrow], at $\approx 60 - 70$ meV, in good agreement with the experimental dispersion for LSCO at the same doping and temperature [38]. The markedly propagating character of the C CM (phonon-like away from the hot lines [27, 28, 35]) makes its contribution to the kink rather sharp.

We emphasize that the characteristic energy of the C CM is extracted from Raman experiments (Tab. I) and...
is not adjusted here by introducing additional modes at suitably chosen phonon frequencies. On the other hand, the more diffusive S CM does not fix an energy scale and rather renormalizes the bare QP dispersion over a broader energy range, affecting the QP velocities far from the kink and making the kink more pronounced. Both C and S CMs must be simultaneously taken into account in order to quantitatively explain the kink. The analysis of Raman spectra shows that the interaction mechanism switches from S to C CM with increasing x \[ 0 \leq x \leq 0.15 \]. This characterizes the doping evolution of the QP dispersion in the range \[ x = 0.15 - 0.26 \]. In Fig. 4 (b), one sees that the S-vs-C switching produces no appreciable effect on the low-energy dispersion, which is determined cooperatively by the two CMs, so that the slope remains quite constant in the doping considered range [dashed line in Fig. 4(b)]. On the other hand, the rather broad and moderately coupled phonons should keep their effects (most pronounced around the nodal regions) even in the superconducting phase.

In conclusion, the salient aspects of ARPES experiments in LSCO are well reproduced by the same two (C and S) CMs previously obtained to fit Raman experiments. This solves for LSCO the long-standing issue whether the kinks are due to phonons or spin fluctuations: we reach the Solomonic conclusion that both play a role. By the interplay of the two CMs, we can explain the highly non-trivial doping evolution of the low- and high-energy QP velocity along the nodal (ΓX) direction, with \( v_F \) almost doping independent and \( v_{HE} \) decreasing with increasing doping (along with the suppression of the coupling with the S CM). We predict the presence of multiple kinks (actually, a kink and a low-energy rounding, analogous to those observed in BSCCO \[ 36, 37 \]). We also predict additional waterfalls at high binding energy, along a square contour around the Γ point of the BZ.

The C-S cooperative behavior might be specific of LSCO, where the tendency to charge ordering seems to be more pronounced than, e.g., in YBCO, where the kinks are more rounded. We also stress that our analysis only holds above \( T_c \). Below \( T_c \) the S CM changes and displays the peculiar resonance at \( (\pi, \pi) \), which alters the shape of the kinks, producing a characteristic \( s \)-shaped dispersion in the antinodal regions \[ 14, 38 \]. On the other hand, the rather broad and moderately coupled phonons should keep their effects (most pronounced around the nodal regions) even in the superconducting phase.

Since, our analysis demonstrates the presence of two CMs, with characteristic wave vectors representative of stripe-like textures, our phenomenological model substantiates the presence of a competing C and S quasi-ordered phase compatible with fluctuating stripes. The assessed relevant role of C and S CMs in LSCO also identifies them as candidate mediators of the pairing glue in these systems.

S.C., C. D.C., and M.G. acknowledge financial support from “University Research Project” of the “Sapienza” University n. C26A115HTN.
For a review, see, e.g., D. R. Garcia and A. Lanzara, X. J. Zhou, T. Cuk, T. Devereaux, N. Nagaosa, T. Dahm, V. Hinkov, S. V. Borisenko, A. A. Kordyuk, A. V. Chubukov and M. R. Norman, Phys. Rev. B 55, 14554 (1997), and references therein.

The presence of two collective modes has also been inferred from optical and experiments in BSCCO systems in Refs. [13, 20].

[1] P. W. Anderson, Science 316, 1705 (2007).
[2] T. A. Maier, D. Poilblanc, and D. J. Scalapino, Phys. Rev. Lett. 100, 237001 (2008).
[3] W. Hanke, M. L. Kiesel, M. Aichhorn, S. Brehm, and E. Arrigoni, Eur. Phys. J. Special Topics 188, 15 (2010).
[4] Ar. Abanov, A. Chubukov, and J. Schmalian, Adv. Phys. 52, 119 (2003), and references therein.
[5] C. M. Varma, Phys. Rev. Lett. 75, 898 (1995); Phys. Rev. B 55, 14554 (1997), and references therein.
[6] L. Benfatto, S. Caprara, and C. Di Castro, Eur. Phys. J. B 17, 95 (2000).
[7] W. Metzner, D. Rohe, and S. A. Kivelson, Adv. Cond. Mat. Phys. Volume 2010, Article ID 807412, 188, Phys. Rev. B 55, 12443 (1996).
[8] S. A. Kivelson, I. P. Bindloss, E. Fradkin, V. Oganesyan, J. M. Tranquada, and C. Howald, Rev. Mod. Phys. 75, 1201 (2003) and references therein.
[9] C. Castellani, C. Di Castro, and M. Grilli, Phys. Rev. Lett. 75, 4650 (1995).
[10] For a review, see, e.g., D. R. Garcia and A. Lanzara, Adv. Cond. Mat. Phys. Volume 2010, Article ID 807412, 188, Phys. Rev. B 55, 12443 (1996).
[11] T. A. Maier, D. Poilblanc, and D. J. Scalapino, Phys. Rev. Lett. 75, 4650 (1995).
[12] W. Hanke, M. L. Kiesel, M. Aichhorn, S. Brehm, and E. Arrigoni, Eur. Phys. J. Special Topics 188, 15 (2010).
[13] W. Hanke and B. Keimer, Nat. Phys. 87-144.
[14] V. B. Zabolotnyy, J. Fink, B. Bchner, D. J. Scalapino, I. M. Vishik, W. S. Lee, F. Schmitt, S. Johnston, B. J. Sternlieb, J. D. Axe, Y. Nakamura, and S. Uchida, Nature (London) 375, 561 (1995).
[15] P. Abbamonte, A. Rusyd, S. Smadici, G. D. Gu, G. A. Sawatzky, and D. L. Feng, Nat. Phys. 1, 155 (2005).
[16] K. Yamada, C. H. Lee, K. Kurahashi, J. Wada, S. Waki moto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R. J. Birgeneau, M. Greven, M. A. Kastner, and Y. J. Kim, Phys. Rev. B 57, 6165 (1998).
[17] An explanation of the waterfalls observed in BSCCO in terms of an electronic (spin) generated self-energy was proposed by Susmita Basak, Tanmoy Das, Hsin Lin, J. Nieminen, M. Lindroos, R. S. Markiewicz, and A. Bansil, Phys. Rev. B 80, 214520 (2009). In this case, however, only one (spin) mode was considered and the momenta where the waterfalls occur were not reported.
[18] A. V. Chubukov, D. K. Morr, and O. A. Kivelson, Phil. Mag. B 74, 563 (1996); A. V. Chubukov and D. K. Morr, Phys. Rep. 288, 347 (1998).
[19] S. Caprara, M. Sulipizi, A. Bianconi, C. Di Castro, and M. Grilli, Phys. Rev. B 59, 14980 (1999).