Anomalous isotopic effect near the charge-ordering quantum criticality

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Within the Hubbard-Holstein model, we evaluate the various crossover lines marking the opening of pseudogaps in the cuprates, which, in our scenario, are ruled by the proximity to a charge-ordering quantum criticality (stripe formation). We provide also an analysis of their isotopic dependencies, as produced by critical fluctuations. We find no isotopic shift of the temperature \( T^0 \) marked as a reduction of the quasiparticle density of states in various experiments, and a substantial positive shift of the pseudogap-formation temperature \( T^* \). We infer that the superconducting critical temperature \( T_c \) has almost no shift in the optimally- and overdoped regimes while it has a small negative isotopic shift in the underdoped, which increases upon underdoping. We account also for the possible dynamical nature of the charge-ordering transition, and explain in this way the spread of the values of \( T^* \) and of its isotopic shift, obtained with experimental probes having different characteristic timescales.

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There are several experimental evidences \([1]\) that the peculiar properties of the cuprates, both in the normal and in the superconducting phase are controlled by a Quantum Critical Point (QCP), located near the optimal doping \( \delta = \delta_{opt} \). In this framework the phase diagram of the cuprates is naturally partitioned into a (nearly) ordered, a quantum critical, and a quantum disordered region corresponding to the under-, optimally, and overdoped regions respectively. The ordered region occurs below a second-order transition line \( T_\delta(\delta) \), which depends on the nature of the underlying ordering and, upon increasing \( \delta \), ends at \( T = 0 \) in a QCP (at \( \delta = \delta_c \gtrsim \delta_{opt} \)). The above correspondence between the theoretical and experimental partitioning of the phase diagram, leads to a close connection \([1]\) between the hypothetical \( T_\delta(\delta) \) and the crossover line \( T^*(\delta) \), below which a pseudogap behavior is observed in NQR, NMR relaxation-rate, XANES, and ARPES measurements \([2,3]\) for a recent overview on La\(_{2-x}\)Sr\(_x\)CuO\(_4\) (LSCO). In the proximity of quantum criticality, critical fluctuations mediate a singular interaction among the quasiparticles which can account for both the violation of the Fermi-liquid behavior observed in the normal phase of the cuprates, and the strong pairing mechanism leading to high-\( T_c \) superconductivity \([4,5,6]\). Various realizations of this scenario have been proposed, associated with different quantum criticalities, e.g., antiferromagnetic \([6]\), excitonic \([5]\), change in the symmetry of the superconducting order parameter \([7,8]\), or incommensurate charge-density wave \([9]\).

In the QCP framework, any mechanism shifting the position of the QCP is mirrored by corresponding shifts in \( T_\delta(\delta) \) and in the superconducting critical line \( T_c(\delta) \). In particular, the observation of isotopic effects (IE's) on \( T_c \) \([9,10]\) and on \( T^* \) \([11,12]\) suggests that a lattice mechanism underlies the instability marked by the QCP. We here consider the single-band Hubbard-Holstein model as a minimal model to describe the strongly correlated electrons coupled to the lattice, giving rise to a QCP for the onset of a phonon-induced incommensurate charge ordering (CO) \([12,13]\). This introduces density inhomogeneities on a semimicroscopic scale, and corresponds to the onset of stripes \([14]\), coming from the high-doping regime. Thus, in our approach, the critical line \( T_c(\delta) \) corresponds to the line for CO, \( T_{CO}(\delta) \), which in real materials can be masked by pair formation and lattice effects.

In this letter, we first determine the mean-field (m-f) critical line \( T_{CO}^0(\delta) \), which we identify with the weak-pseudogap crossover line \( T^0(\delta) \gg T^*(\delta) \) observed in Knight-shift, transport, and static susceptibility measurements \([2]\), as the incipient depression of the single-particle density of states (DOS). Indeed, for \( T < T_{CO}^0 \) one expects the CO fluctuations to become substantial, leading to a reduction of the quasiparticle DOS, which accounts for the weak-pseudogap behavior. Then, we investigate the effect of fluctuations near the CO QCP in order to determine: a) the fluctuation-corrected critical line \( T_{CO}(\delta) \), which we relate to the pseudogap crossover line \( T^*(\delta) \); b) the IE on \( T_{CO}(\delta) \), from which we shall also infer the effect on \( T_c(\delta) \). In particular we shall describe the highly non-trivial effect of quantum criticality in determining IE's on \( T^* \) and \( T_c \), which can be strong and weak, respectively, and opposite in sign in the underdoped cuprates. We shall also show why both \( T^* \) and its isotopic shift are observed to be larger in experiments with shorter characteristic timescales. The near absence of IE on \( T_c \) near and above \( \delta_{opt} \) will also be naturally accounted for.

--- The model and the mean-field instability --- Our two-dimensional (2D) Hamiltonian is

\[
H = -t \sum_{\langle i,j \rangle, \sigma} (c_{i\sigma}^\dagger c_{j\sigma} + H.c.) - t' \sum_{\langle i,j \rangle, \sigma} (c_{i\sigma}^\dagger c_{j\sigma} + H.c.) - \mu_0 \sum_i n_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} \sum_{q \neq 0} V_c(q) \rho_{q\bar{q}-\bar{q}}
\]
where \(c_\sigma^{(t)}\) are the fermion operators, \(a_\sigma^{(t)}\) are the phonon operators, \(\langle i, j \rangle\) and \(\langle (i, j) \rangle\) indicate nearest- and next-nearest-neighbor sites, coupled by the hopping parameters \(t\) and \(t'\) respectively. The lattice spacing has been set to unity. The chemical potential \(\mu_0\) is coupled to the local electron density \(n_{i\sigma}\). The on-site Hubbard repulsion, and \(\rho_q = \sum_{k, \sigma} c_{k, \sigma}^\dagger c_{k, \sigma}\). \(\tilde{V}_C(q) \approx V_C/|q|\) at small \(|q|\) is the Coulomb interaction between electrons in a 2D plane embedded in the three-dimensional (3D) space \(\omega_\ell\). \(\omega_\ell\) is the phonon frequency and \(g\) is the Holstein electron-phonon coupling. In the limit \(U \to \infty\), this model was solved with a standard slave-boson technique, at leading order within a large-N expansion in Refs. [3,4]. The most relevant result was that a CO instability with a finite wavevector \(q_c\), incommensurate with the underlying lattice, was found at \(T = 0\) when \(\tilde{\delta}\) is reduced below a critical value \(\tilde{\delta}_{c}^{(0)}\). Besides a small, weakly momentum-dependent, residual repulsion, nearby this instability the critical charge fluctuations mediate a singular scattering

\[
\Gamma(q, \omega_n) \approx -\frac{V}{\xi_0^{-2} + |q - q_c|^2 + \gamma|\omega_n|},
\]

of strength \(V\), between the quasiparticles. Here \(\omega_n\) is a bosonic Matsubara frequency, \(\gamma \sim t^{-1}\) is a characteristic timescale, and \(\xi_0^{-2}\) is the m-f inverse square correlation length, which measures the distance from criticality. Eq. [2] displays the behavior of a Gaussian QCP with a dynamical critical index \(z = 2\) [2].

Quite remarkably, the complicated formal structure of the quasiparticle scattering, mediated by slave bosons, phonons and by the Coulomb interaction, is well represented near criticality by a RPA resummation \(\Gamma(q, \omega_n) \approx V_{eff}(q)/[1 + V_{eff}(q)\Pi(q, \omega_n)]\) of an effective static interaction \(V_{eff}(q) = U(q) + V_C(q) - \lambda t\) [1,9]. Here \(\Pi\) is the fermionic polarization bubble in a 2D lattice, \(\lambda \equiv 2g^2/t\omega_\ell\) is the dimensionless electron-phonon coupling, and \(U(q) \simeq A + B|q|^2\) is the residual short-range repulsion between quasiparticles. For the correspondence of \(V_{eff}(q)\) with the parameters of Eq. [1] see Ref. [10].

The instability condition, which occurs for reasonable values \(\lambda \sim 1\), is \(1 + V_{eff}(q_c)\Pi(q_c, \omega = 0) = 0\). At \(T = 0\) this determines \(q_c\) and the position of the m-f QCP \(\delta_{c}^{(0)}\). For realistic parameters we find \(q_c \approx (\pm 1, 0)\) or \((0, \pm 1)\), and \(\delta_{c}^{(0)} \approx 0.2\) [1,3]. By expanding \(1 + V_{eff}\Pi\) near the instability at \(T = 0\), we find \(\xi_0^{-2} \propto \delta - \delta_{c}^{(0)}\), which gives the Gaussian index \(\nu = 1/2\) for \(\xi_0\). The m-f critical line \(T_{CO}(\delta)(\text{the solid-line curve in Fig. 1})\), which starts from the m-f QCP at \(\delta_{c}^{(0)}\) is obtained by considering the T dependence of the bare polarization bubble, which reduces to the simple Fermi-liquid form, \(\propto T^2\), at low \(T\). We identify this m-f transition with the experimental crossover line \(T^{0}\), extrapolating to \(T = 0\) at a doping, which we identify with our \(\delta_{c}^{(0)}\). From the data reported in Ref. [3], for LSCO we estimate \(\delta_{c}^{(0)} \approx 0.22\). Then we evaluate \(T_{CO}(\delta)\) by taking standard quasiparticle (i.e., dressed by the slave bosons) band parameters \((t_{qp} = 0.2\ eV, t'_{qp} = -0.05\ eV, \omega_0 = 0.07\ eV, \text{leading to} \ A = 0.2\ eV\ and \ B = 0.17\ eV\ \text{in the residual repulsion} \ U)\). \(V_C = 0.22\ eV\ and \ g = 0.21\ eV\ are adjusted to match the experimental extrapolation of \(T^{0}\) with the \(T = 0\) m-f instability (i.e., \(\delta_{c}^{(0)}\)). We point out that the agreement between \(T_{CO}(\delta)\) and \(T^{0}(\delta)\) at finite \(T\) is obtained without any further adjustment of the parameters. Similar parameters are taken for Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8\) (Bi2212) to fit the data in Refs. [17,18], with \(V_C = 0.22\ eV\ and \ g = 0.23\ eV\).

FIG. 1. The phase diagram of the cuprates according to the CO-QCP scenario for LSCO (a) and Bi2212 (b). The solid line is the m-f critical line ending at \(T = 0\) in the m-f QCP at \(\delta_{c}^{(0)}\). The lowest dashed line in panel (a) marks the 3D critical line in the presence of fluctuations, ending in the QCP at \(\delta_e\). We took \(\Omega_\perp = 1\ meV\ (\text{see text})\). The dot-dashed line in panels (a,b) indicates the “dynamical instability” condition (see text) for \(\omega_{probe} = 1\ meV\). The intermediate dashed line in panel (a) represents the “dynamical instability” condition for \(\omega_{probe} = 1\ mu\ eV\). The experimental points for \(T^0(\pm)\) and for \(T^*\) measured with fast (×) and slow (□) probes for LSCO are from Ref. [2], those for Bi2212 are from Refs. [17,18]. The experimental critical temperatures \(T_c\) are also shown (△).

— The phase diagram beyond mean-field — The fluctuations shift the m-f QCP and critical line to their actual position [1,2]. Specifically the fluctuations which mediate the effective critical interaction, Eq. [2], can be included in the polarization bubble via the diagrams of Fig. 2a, leading to corrections beyond RPA. From the explicit evaluation of these diagrams we find the (self-consistent) correction to the mass term \(m \equiv \gamma^{-1} \xi^{-2}\) of the fluctuation propagator, Eq. [2],
where $D = (\Omega_\alpha + |\omega_n|)^{-1}$ is the charge-fluctuation critical propagator, $u \sim V^2/(\gamma^2 T^3)$ is the coupling resulting from the two 4-leg vertices represented in Fig. 2b. The critical modes have a 2D dispersion $\Omega = \gamma^{-1} |q - q_c|^2 + m$, up to an ultraviolet bandwidth cutoff $\Omega_{max} \sim \gamma^{-1}$, resulting from the underlying lattice. Since we deal with a phonon-driven CO instability, as it also follows from the detailed dynamical analysis of Ref. [3], $\omega$ appears as the ultraviolet frequency cut-off. By introducing the DOS $N(\Omega) = \sum_q \delta(\Omega - \Omega_q)$ and the spectral-density representation of $D$, we rewrite Eq. (3) in the form

$$m = m_0 + 12u \sum_{|\omega_n| < \omega_0} \sum_q D(q, \omega_n; m),$$

(3)

where $D = (\Omega_\alpha + |\omega_n|)^{-1}$ is the charge-fluctuation critical propagator, $u \sim V^2/(\gamma^2 T^3)$ is the coupling resulting from the two 4-leg vertices represented in Fig. 2b. The critical modes have a 2D dispersion $\Omega = \gamma^{-1} |q - q_c|^2 + m$, up to an ultraviolet bandwidth cutoff $\Omega_{max} \sim \gamma^{-1}$, resulting from the underlying lattice. Since we deal with a phonon-driven CO instability, as it also follows from the detailed dynamical analysis of Ref. [3], $\omega$ appears as the ultraviolet frequency cut-off. By introducing the DOS $N(\Omega) = \sum_q \delta(\Omega - \Omega_q)$ and the spectral-density representation of $D$, we rewrite Eq. (3) in the form

$$m = m_0 + \frac{24u}{\pi} \int_{m}^{\Omega_{max}} d\Omega N(\Omega) \int_0^\infty dz \frac{z}{z^2 + \Omega^2} \left[ b(z) + \frac{1}{2} - \frac{1}{\pi} \arctan \frac{z}{\omega_0} \right],$$

(4)

where $b(z) = \text{exp}(\frac{z}{T}) - 1^{-1}$ is the Bose function. At $T = 0$, Eq. (4) with $m = 0$ leads to a finite shift of the 2D QCP $\delta_0 - \delta_c \propto \omega_0$ (see Fig. 1). At finite $T$, as a consequence of a constant DOS for the 2D modes, the integral in Eq. (4) is logarithmically divergent for $m \to 0$, leading to a vanishing of the renormalized $T_{CO}$ (Mermin-Wagner theorem). This divergence is removed by considering the more realistic anisotropic 3D character of the critical fluctuations, introducing a small energy scale $\Omega_\perp$, below which the mode DOS is no longer constant, and displays a 3D square-root behavior $N(\Omega < \Omega_\perp) \sim \sqrt{\Omega}$ at criticality. This is enough to make the integral in Eq. (4) convergent and allows to determine the critical line (Eq. (4) with $m = 0$) in the anisotropic 3D case as reported in Fig. 1a (lowest dashed line). The inclusion of fluctuations brings the critical line from temperatures of the order of typical electronic energies ($T_{CO} \sim \gamma$) down to much lower temperatures $T_{CO}$ of the order of the observed $T^*$'s. Indeed, within the CO-QCP scenario, the pseudogap arises at $T < T^*$ because the quasiparticles feel an increasingly strong attractive interaction by approaching the critical line $T_{CO}(\delta)$. In the particle-hole channel, this interaction can produce a gap due to the incipient CO. At the same time in the particle-particle channel, the strong attraction can lead to pair formation even in the absence of phase coherence. Therefore $T^*(\delta)$ closely tracks the underlying transition line $T_{CO}(\delta)$.

On the other hand, the spread in the measured values of $T^*$ depending on the experimental probe (see, e.g., Ref. [2]), indicates that the CO instability may be “dynamical”, $\Omega_\perp$ being smaller than (or comparable to) typical frequencies of the experimental probes, $\omega_{\text{probe}}$. In this case the self-consistency condition (4) includes $\omega_{\text{probe}}$ as the infrared cut-off in the integral over $\Omega$, $m$ being replaced by $\omega_{\text{probe}}$. This determines the doping and temperature dependence of the dynamical instability lines.

Two examples are reported in Fig. 1 for $\omega_{\text{probe}} = 1$ meV (dot-dashed line if Fig. 1a,b), as in typical neutron scattering experiments, and $\omega_{\text{probe}} = 1$ $\mu$eV (second dashed line from bottom in Fig. 1a), as in static experiments (NQR, NMR). The corresponding experimental data for $T^*$ in LSCO are also reported for comparison. We determine the coupling $V$ between the charge fluctuations and the quasiparticles, which is the only parameter for which an a priori estimate is difficult, by imposing that the fluctuation-corrected QCP is located at the $T = 0$ extrapolation of the $T^*(\delta)$ curves. We used $\gamma = 0.7$ eV$^{-1}$ and $\gamma = 0.4$ eV$^{-1}$ for LSCO and Bi2212 respectively, and $V = 0.54$ eV for both. It is worth noticing that, similarly to the case of $T_{CO}^{(0)}(\delta)$, the agreement between the calculated $T_{CO}(\delta)$ and the experimental points $T^*(\delta)$ is obtained without further adjustable parameters. We also notice that, contrary to the shift of $\delta_c$ at $T = 0$, the slope of the curve $T_{CO}(\delta)$ is weakly dependent on $\omega_0$.

![FIG. 2. (a) The two (vertex and selfenergy) corrections to the fermionic bubbles (solid line) due to critical charge fluctuations (wavy line). (b) Effective 4-leg vertices for critical charge fluctuation field (wavy line) resulting from the integration over the fermion loops (solid line).](image-url)

— Novel isotopic effects — The m-f weak-pseudogap crossover temperature $T_{CO}^{(0)} \sim T^0$ is determined by $\lambda$ only. Therefore, it does not depend on $\omega_0$ and is not expected to display any isotopic dependence. On the other hand, quantities determined by the fluctuations crucially involve $\omega_0$. New physical effects can then arise in the isotopic substitutions. Since $\omega_0$ decreases by increasing the ionic mass, the corrections to $\delta_c^{(0)}$ and to $T_{CO}^{(0)}(\delta)$ become smaller and the fluctuation-corrected quantities $\delta_c$ and $T_{CO}$ shift to higher doping, closer to the m-f values, as shown in Fig. 3, where we report the line $T_{CO}(\delta)$, calculated via Eq. (4), with parameters to fit the $T^*$ data of LSCO (see above), together with its isotopic shift calculated for $^{16}O \rightarrow ^{18}O$ substitution (i.e., for a five percent reduction of $\omega_0$). Correspondingly we expect that the portion of the curve $T_c(\delta)$ on the left of $T^0(\delta)$ is rigidly translated along the horizontal axis. The rational behind this translation is that the whole physics of these materials is essentially determined by the proximity to the critical line $T_{CO}(\delta)$ and to the related QCP. On the other hand, on the right of the m-f critical line $T_{CO}^{(0)}(\delta) \sim T^0(\delta)$ the fluctuations are small and the physical processes at $T > T^0(\delta)$ are captured by the m-f description, where
λ (not \(\omega_0\)) is relevant. As a consequence \(T^0(\delta)\) and the portion of \(T_c(\delta)\) near and above \(T^0(\delta)\) are not expected to be shifted by IE’s.

In the underdoped region, there are two evident consequences of the isotopic shift, which becomes more substantial: The shift upon reducing \(\omega_0\) is negative (as usual) in \(T_c\), but, contrary to standard theories based on CO pseudogap \([21]\), it is positive in \(T_{CO} \sim T^*\). Moreover, when the slope of \(T_{CO}(\delta)\) is large, a rather small isotopic shift in \(\delta_c\) can result in a substantial shift in \(T_{CO} \sim T^*\). The steeper \(T^*\), the larger is the IE. On the other hand, since the curve \(T_c(\delta)\) is rather flat, particularly in the optimal and moderately under-doped regimes, the expected IE on \(T_c\) in these compounds is small in agreement with long-standing experiments \([7,8]\). This large difference in the IE for \(T_c\) and \(T^*\), \((\Delta T_c/\Delta M)/(\Delta T^*/\Delta M) \ll 1\), is indeed experimentally observed in HoBa\(_2\)Cu\(_4\)O\(_8\) (HBCO-124), and reported in Ref. \([11]\), where it is also noticed that there is a “striking similarity between isotopic substitution and underdoping with respect to both \(T_c\) and \(T^*\”)”.

Although we are not aware of any systematic analysis of the doping dependencies of \(T_c, T^*\) and their isotopic shifts in HBCO-124, this observation finds its natural interpretation within our QCP scenario, where the isotopic substitution produces a shift of the QCP and is therefore nearly equivalent to underdoping.

The slope of \(T_{CO}(\delta)\) increases by increasing \(\omega_{\text{probe}}\), while the QCP is unshifted. Therefore, the IE on \(T_{CO} \sim T^*\) is enhanced, and we have the general trend that faster probes should detect a larger IE on \(T^*\), since the effect of fluctuation diminishes. Although we cannot account for the near-absent or negative IE on \(T^*\) within the almost static probes in YBa\(_2\)Cu\(_4\)O\(_8\) \([16]\), this general trend is in qualitative agreement with a much stronger effect observed in the isostructural HBCO-124 with fast neutron scattering \([1]\). Indeed, this fast-probe experiment should be represented by the curve \(T_{CO}(\delta)\) correspond-
[20] A. J. Millis, Phys. Rev. B 48, 7183 (1993).
[21] I. Eremin, et al., Phys. Rev. B 56, 11 305 (1997).
[22] A. Lanzara, et al., J. Phys.: Condens. Matter 11, L541 (1999).
[23] J. Hofer, et al., Phys. Rev. Lett. 84, 4192 (2000).
[24] T. Schneider and H. Keller, [cond-mat/0011381].