Signature of quantum criticality in cuprates by charge density fluctuations

The universality of the strange metal phase in many quantum materials is often attributed to the presence of a quantum critical point (QCP), a zero-temperature phase transition ruled by quantum fluctuations. In cuprates, where superconductivity hinders direct QCP observation, indirect evidence comes from the identification of fluctuations compatible with the strange metal phase. Here we show that the recently discovered charge density fluctuations (CDF) possess the right properties to be associated to a quantum phase transition. Using resonant x-ray scattering, we studied the CDF in two families of cuprate superconductors across a wide doping range (up to $p = 0.22$). At $p^* \approx 0.19$, the putative QCP, the CDF intensity peaks, and the characteristic energy $\Delta$ is minimum, marking a wedge-shaped region in the phase diagram indicative of a quantum critical behavior, albeit with anomalies. These findings strengthen the role of charge order in explaining strange metal phenomenology and provide insights into high-temperature superconductivity.

The strange metal phase in cuprate superconductors extends over a large portion of the doping-temperature phase diagram and is particularly robust at the doping level $p^*$, around which the maximum superconducting critical temperature is achieved \cite{1,2,3,4,5}. At this doping, the resistivity is linear in $T$ down to the superconducting critical temperature $T_c$, or, in the presence of strong magnetic fields, to the lowest temperatures \cite{6}. Electronic excitations lose their quasiparticle character \cite{7}, the Drude peak in the optical conductivity acquires an anomalous power-law decay in frequency \cite{8}, the magnetoresistance is linear in field \cite{9} and the spin relaxation rate is almost $T$-independent \cite{10}. The Fermi-liquid theory, although successful for strongly correlated normal metals, does not provide a proper description of this phenomenology. The unconventional properties of the strange metal are likely the result of strong electron correlation in a quasi-2D structure that leads to a complex energetic landscape where several electronic phases compete and may coexist. Indeed, a novel state has been recently proposed, with very strong long-range quantum entanglement and a thermalization time defined by the Planckian scattering time \cite{11}. Other theories suggest that quasiparticles get spoiled by strong scattering either on nearly local excitations (such as the
Metal behavior is observed only at relatively high temperatures\textsuperscript{33,34}. The experimental evidence of these two aspects has the determination of their energy following the expected doping and fluctuations at the origin of the QCP\textsuperscript{*}. For their characteristics - presence in a broad range of doping in the phase diagram, finite energy and short correlation length that implies a broad almost isotropic weight increases almost constantly with temperature, everywhere in the high temperatures, more steeply at $p = 0.19$ and $p = 0.06$, where the strange metal behavior is observed only at relatively high temperatures\textsuperscript{31,34}. High resolution spectra at the CDF critical wave vector $q_{\text{CDF}}$ provide a direct measurement of the doping dependence of the characteristic energy parameter $\Delta(T,p)$, which is minimum at $p^*$ and, at fixed $p$, grows with temperature. Moreover, the quasi-elastic spectral weight increases almost constantly with temperature, everywhere in the reciprocal space (see Supplementary Figs. 1a, b, 2a, b, and 3a). Such isotropic rise of scattered intensity was disregarded in previous studies and that the quasi-elastic signal grows linearly at $p = 0.22$, in the overdoped region, where the strange metal at high temperature is substituted by a Fermi liquid behavior at lower temperatures; $p = 0.19 > p^*$ for both YBCO and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) samples. We spanned a broad doping range, with special attention for three cases, where the absence of CDW permits a thorough study of CDF, i.e., e.g. in YBCO, from $p = 0.08$ to $p = 0.16$\textsuperscript{21}. We thus chose $p = 0.22$, in the overdoped region, where the strange metal high temperature is substituted by a Fermi liquid behavior at lower temperatures; $p = 0.19 > p^*$ for both YBCO and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212)\textsuperscript{21}, where the strange metal extends down to the superconducting critical temperature $T_c$; and $p = 0.06$, where the strange metal behavior is observed only at relatively high temperatures\textsuperscript{31,34}. For an extensive analysis of the temperature evolution of the CDF signal at $p = 0.19$, we have used medium resolution RIXS (-62 meV, see Fig. 2a) and EI-RXS data, which are quicker to measure but do not allow the direct determination of the characteristic energy $\Delta$, because the CDF contribution cannot be resolved from the elastic one (see Methods and Supplementary Fig. 7). In this case, to best single out the CDF component and minimize the contribution of the bond-stretching phonons, with an energy drop $\Delta_{\text{PH}}$ measured at a $H$ value close but higher than $H_{\text{CDW}}$, which is larger at 80 K ($\Delta_{\text{PH}}$ ~15 meV) than at 200 K. This indicates that, as opposite to the most common interpretations\textsuperscript{35,39}, the phonon softening is not associated only to long-ranged CDW but also, or mainly, to CDF.

### Results

**CDF at $p = 0.19$**

We have performed resonant inelastic x-ray scattering (RIXS) and energy-integrated resonant x-ray scattering (EI-RXS) at the Cu $L_2$ edge (-930 eV) at different momentum transfer components $q$ parallel to the CuO$_2$ planes along the $(H,0)$ and $(H,H)$ directions, on slightly overdoped ($p = 0.19$) samples of YBCO and Bi2212 (see Methods for details about the sample growth). Figure 1 summarizes the very-high-resolution (-38 meV) RIXS results at 80 K and 200 K, i.e., close to and well above $T_c$, respectively. To properly map the CDF signal, panels 1a and 1b represent as colormaps the difference between the spectra measured along the $(H,0)$ and $(H,H)$ directions. In this way the elastic scattering signal originated from surface defects gets mostly canceled out and the weak CDF signal emerges at small but finite energy, with broad intensity peaks at $q = q_{\text{CDW}}$ (0.30,0) for YBCO and $q = q_{\text{CDW}}$ (0.25,0) for Bi2212. Each spectrum of the $(H,0)$ series can be fitted below 150 meV with four narrow Gaussian peaks (elastic, CDF, bond-stretching phonons, phonon overtones in increasing energy loss), plus a broader feature for the spin and particle-hole excitations (see Methods, and Supplementary Fig. 5). As shown in panels 1d-i, the elastic peak intensity is temperature independent, with a monotonic decrease vs $H$ in undoped samples (see Supplementary Fig. 6a) due to the reduced amplitude of correlation functions expected at doping levels above optimum\textsuperscript{31,34}. On the contrary, the CDF intensity, at finite energy, is $T$-dependent and broad both in energy and in momentum. The FWHM of the CDF Gaussian peak, 35–60 meV, is significantly larger than the experimental energy resolution, and the energy position increases from $\Delta$ ~10 meV at 80 K to ~15 meV at 200 K. The CDF intensity plotted versus $q$ is peaked at $H = H_{\text{CDW}}$ with a FWHM ~0.15 r.l.u., corresponding to a rather short-ranged correlation length. The shape of this broad-in-$q$ peak is very similar at the two temperatures, as already observed in ref. 26, but its intensity is almost isotropically stronger at 200 K than at 80 K, for reasons discussed below. Finally, panels 1f,i show the softening of the bond-stretching phonon, with an energy drop $\Delta_{\text{PH}}$, measured at a $H$ value close below that of the CDF, which is larger at 80 K ($\Delta_{\text{PH}}$ ~15 meV) than at 200 K. This indicates that, as opposite to the most common interpretations\textsuperscript{35,39}, the phonon softening is not associated only to long-ranged CDW but also, or mainly, to CDF.

**Temperature dependence of CDF**

For an extensive analysis of the temperature evolution of the CDF signal at $p = 0.19$, we have used medium resolution RIXS (-62 meV, see Fig. 2a) and EI-RXS data, which are quicker to measure but do not allow the direct determination of the characteristic energy $\Delta$, because the CDF contribution cannot be resolved from the elastic one (see Methods and Supplementary Fig. 7). In this case, to best single out the CDF component and minimize the contribution of the bond-stretching phonons, in Fig. 2 we plot, as function of momentum, the integral of the RIXS spectra up to 35 meV energy loss (vertical band in Fig. 2a). This integral intensity is dominated by the CDF on top of other elastic and quasi-elastic components. A peak is present along the $(H,0)$ direction but not along $(H,H)$ (Fig. 2b, c, Supplementary Figs. 1a, b and 2a, b), consistently with the high resolution RIXS data and with ref. 26. Moreover, along both directions this quasi-elastic intensity increases with the temperature. It is evident that the $T$ and $q$ dependence of the CDF scattering intensity cannot be properly described by a single, temperature independent energy parameter. Therefore, we have modeled the CDF component $I_{\text{CDF}}(q,\omega)$ of the RIXS spectra according to the theory of the charge density instability in a highly-correlated Fermi liquid\textsuperscript{38}, which is particularly appropriate for the slightly overdoped samples. The theoretical x-ray scattering intensity is given by

\[ I_{\text{CDF}}(q,\omega) \propto \text{Im}[D(q,\omega)] \cdot b(\omega), \]
so that it can be independently determined from the width in measure of the phase space available for the decay of the parameter CDF spectral density decreases more rapidly. The Landau damping \( \frac{\omega}{\omega_0} \) is also linked to the CDF correlation length \( T \), i.e., the product of the dynamical density fluctuations \( \Gamma \), which is thus the energy \( \Delta \) directly measured with very high resolution RIXS at the critical wave vector. The minimum frequency \( \omega_0 \) is also linked to the CDF correlation length \( \xi \) by the relation \( \omega_0 = \gamma \omega_0 \xi^2 \), so that it can be independently determined from the width in measure of the CDF intensity peak, which is inversely proportional to \( \xi \). At a generic wave vector different from \( \xi \), the maximum of \( \text{Im}[\mathcal{I}(q_{CDF}, \omega)] \) is instead reached at \( \omega = \gamma^{-1} (\omega_0(T) + \gamma \xi^2 - \omega^2/\omega) \), an energy which is higher than \( \Delta \). In particular, when we are far away from \( q_{CDF} \) the \( T \)-independent, quadratic term becomes relevant, and this energy is maximum. We have named it \( \Omega \).

We have performed a global fit, which simultaneously considers all the YBCO data in both \((H,0)\) and \((H,H)\) directions at the \(13 \) measured temperatures, using Eqs. (1) and (2), with four critical wave-vectors in the first Brillouin zone \( q_{CDF} = (\pm q_{CDF}, 0), (0, \pm q_{CDF}) \). The experimental data and the fitting results are compared in Fig. 2b–e. The fitting leads to numerically robust estimates of \( \omega_0(T) \), \( \gamma \), and \( \Omega \) (see Methods). In particular, we find \( \omega_0 \) to increase from 5 meV at \( T_c \) to 20 meV at room temperature (see Fig. 2i), and \( \gamma \gamma \) is determined.

Given the importance of getting a reliable estimate of \( \omega_0(T) \), we have analyzed the data also in a different way. We have isolated the CDF peak close to \( q_{CDF} \) by subtracting, at each \( T \), the featureless \((H,H)\) data from \((H,0)\) ones. This subtraction allows us to remove from the quasi-elastic RIXS intensity the contribution of the elastic scattering due to surface defects, which is independent of the modulus of \( q \); at the same time, along the \((H,H)\) direction the CDF contribution is still present though rather flat, so that the shape, i.e. the FWHM, of the CDF...
peak is not altered by the subtraction procedure. We have therefore fitted the resulting \((H,0) - (H,H)\) curves to a Lorentzian peak (Fig. 2f) and observe that, with increasing \(T\), the height of the difference peak slowly decreases, i.e. the intensity increases less at \(q_{\text{CDW}}\) than far from it, and the FWHM increases (Fig. 2g, h). Both \(T\) dependences show a mild slope discontinuity around \(T_c\) (see dotted vertical lines in Fig. 2g, h) which tells us about a possible entwining between CDF and the superconducting state. Moreover, the extrapolation to zero kelvin of the high-temperature linear-in-\(T\) behavior of the FWHM provides the estimate of a finite CDF correlation length \(\xi \propto FWHM^{-1}\) for \(T \to 0\). This last occurrence sets a strong difference between CDF and CDW\(^{\infty}\) (see Supplementary Fig. 3c-e), and confirms that a standard criticality based on the divergence of a spatial correlation length cannot be contemplated for CDF.

The \(\omega_0(T)\) obtained from the FWHM are compared in Fig. 2i with those determined with the global fit on the same dataset (solid line) and with the energy \(\Delta\) extracted directly from the very-high-resolution spectra (triangles). Similar energies have been determined for Bi2212 at the same doping level (see Supplementary Fig. 2). The fact that the values of \(\Delta\) and \(\omega_0\) determined by different procedures and from different data sets, coincide within the experimental uncertainties allows us to take \(\gamma = 1\) for \(T > T_c\); although further investigations might lead to a more precise estimate of the Landau damping parameter \(\gamma\), we do not expect it to depart significantly from one, if not at low temperatures, and with superconductivity suppressed by a magnetic field, as it was also previously found by the fit of the linear-in-\(T\) resistivity of YBCO\(^{17}\).

Doping dependence of CDF

To determine the dependence of the CDF properties over a significantly broad doping range, we performed the RIXS measurements on a strongly underdoped \((p = 0.06)\) YBCO sample (see Methods). Even at such low doping level, the high resolution spectra of Fig. 3a show a CDF peak, centered at \(q = q_{\text{CDW}} - (0.35,0)\), with an energy \(\Delta(T_{\text{min}}, p = 0.06) \sim 25\) meV at 20 K (see Supplementary Fig. 6b, c). This energy value, so high already close to \(T_c\), is therefore significantly larger than that of the \(p = 0.19\) samples. Although weaker, the temperature dependence of the CDF intensity and FWHM in the \(p = 0.06\) sample is similar to that of the overdoped ones (see Fig. 3b, c and Supplementary Fig. 3a, b). We also notice that the softening of the bond-stretching phonons \((\Delta E_{\text{ph}} \sim 4\) meV at 20 K) is still present at \(p = 0.06\), though much weaker than at higher doping (see Supplementary Fig. 6c). This occurrence further supports the generality of the connection between CDF and bond-stretching phonons.
These two values significantly extend the doping range over which the $H_{\text{CDF}}$ is known for this cuprate family. The $(H,0)$--$(H,H)$ RIXS map in Fig. 3e shows that the CDF energy is larger at $p=0.22$ than at $p=0.19$, and the fit of the high resolution spectra leads to $\Delta(T_{\text{min}},p=0.22) \approx 18$ meV. Moreover, the CDF scattering intensity is smaller at $p=0.22$ than at $p=0.19$. By adding the high temperature data of three other samples (see Fig. 3f, g and Supplementary Fig. 3c–e), at very low ($p=0$) and intermediate doping ($p=0.09$ and 0.10), we can convincingly show in Fig. 3h that the CDF intensity has a maximum at $p=0.19$. This result is robust and independent of the procedures for the subtraction of the pure elastic contribution.

The results for the $p=0.06$, 0.19 and 0.22 samples suggest that not only the intensity but also the energy associated to the CDF depends on the doping level, calling for a study at intermediate values of charge fluctuations in YBCO. a High resolution RIXS map along the $(H,0)$ direction taken at $T=20$ K for a strongly underdoped ($p=0.06$) YBCO. The map is presented after subtracting the fit of the pure elastic peak from the raw spectra. b, c Height and FWHM as a function of the temperature of the Lorentzian profiles used to fit the CDF peak. Here, the peak has been obtained from the difference $(H,0)$--$(H,H)$ of the intensity of the medium resolution RIXS spectra, integrated within the energy range shown in Fig. 2a. The error bars represent the 95% confidence interval of the Lorentzian fit. The solid line is a linear fit of the data. d The $H_{\text{CDF}}$ of the CDF peaks in the YBCO and Ca-YBCO samples we have measured is in agreement with the $H_{\text{CDF}}$ doping dependence of CDW previously found in YBCO (see squares, taken from ref. 31). The same linear trend is indeed also followed by the samples at $p=0.06$ and $p=0.22$, where no signature of charge order was previously detected. e High resolution RIXS map of the difference $(H,0)$--$(H,H)$ taken at $T=60$ K for an overdoped ($p=0.22$) Ca-YBCO sample. f The integrated intensity at $T=290$ K is presented as a function of the momenta along the $(H,0)$ direction for YBCO samples at different level of doping $p$. Curves are presented after a vertical translation, so to align the integrated intensities at the lowest and highest momentum values. g The CDF peak, determined by the difference $(H,0)$--$(H,H)$, is shown at $T=290$ K for strongly underdoped ($p=0.06$) YBCO, for a slightly overdoped ($p=0.185$) YBCO, and for an overdoped ($p=0.22$) Ca-YBCO thin film. For clarity, the weak, high temperature CDF peaks are here presented after smoothing. h The height of the CDF peak, normalized to the maximum value measured at $p=0.19$, is plotted vs doping for the YBCO and Ca-YBCO. The violet region is a guide to the eye.
of $p$. However, in those cases the task is more difficult because the CDW peak at zero energy cannot be resolved from the CDF signal at very low energy, being the respective critical wave-vectors almost coincident (see Fig. 3d). Therefore we have studied the CDF far from $q_{\text{CDW}}$, where the CDW contribute negligibly to the scattering intensity, i.e., along the $(H,H)$ direction and on the tails of the $(H,0)$ scan. What we can determine in this way is $\Omega$, the bosonic characteristic energy of the CDF previously mentioned. We expect $\Omega$ to be related to, and larger than, the energy $\Delta$ at $q_{\text{CDW}}$. For convenience, we assume that $\Omega$ is constant with respect to temperature, since the $T$-dependent $\delta_0$ term is little relevant in the expression of $\Omega$ for each doping $p$. Consequently, the energy-integral of the quasi-elastic intensity far from $q_{\text{CDW}}$ can be simply attributed to a bosonic distribution function of a single characteristic energy $\Omega$ at all temperatures. Even before fitting the experimental data we observe that the $T$ dependence of the intensity is very similar at all $q$ positions along the $(H,H)$ and that it strongly depends on the doping level (see Fig. 4a, b). We fitted those curves with a simple function $A + 4 \beta_0 \left[1 + 2 \left(\frac{\omega}{\Omega}\right)^2 - 1\right]^{-1}$ where $\beta_0$ is the CDF intensity at zero temperature and $A$ accounts for the non-CDF scattering contributions (see Methods and Supplementary Fig. 4). We can thus be confident in using the same method at intermediate doping. Interestingly, $\Omega$ is 15–20 meV larger for $p = 0.06$ than for $p = 0.19$, which is close to what we have also observed at $q_{\text{CDW}}$ for $\Delta$ (see Fig. 4c).

The values of $\Delta$, converted into kelvin, are shown in the phase diagram of Fig. 4d for a set of YBCO, NBCO and Bi2212 samples, including those previously used in ref. 26. Below and above $p^*$ these points line up with the border of the strange metal phase as determined by transport\cite{tomioka2009, mcintyre2016, mcintyre2017} (shaded regions in Fig. 4d) and define a characteristic wedge with a minimum at $p^*$.

**Discussion**

Thanks to an innovative analysis of RIXS data, applied to a large set of measurements (6 doping levels, 3 families of samples, wide temperature range, high and low resolution in energy), we provide here a consistent assessment of the doping and temperature dependences of the CDF intensity and energy in superconducting cuprates. We find that the CDF scattering intensity is strongest in proximity of $p \approx 0.19$ and at low $T$, while it fades both when increasing the temperature and when moving the doping away from $p^*$. Moreover, the energy $\Delta(T_{\text{min}}, p)$, i.e. the minimum of the parabolic relation for the CDF dispersion in the $q$-space, is lowest at $p \approx 0.19$, while it increases with temperature at all dopings. The results are summarized in Fig. 4e, where we depict the CDF dispersion relation using the propagator of

![Fig. 4](https://doi.org/10.1038/s41467-023-42961-5)
Eq. (2) calculated with the parameters experimentally determined. We find a good indication that the CDF can be the fluctuations associated to a quantum critical point at $p \approx 0.19$. In fact, the CDF is enhanced exactly at the putative QCP. Moreover, the CDF energy $\Delta$ coincides, within the experimental errors, to the temperature where the strange metal ends, i.e., where the linear-in-$T$ resistivity is lost. These correspondences point towards a strong role of charge excitations in driving the phase transition, giving rise to the strange metal. However, since recent inelastic neutron scattering and nuclear magnetic resonance results support a quantum criticality at $p^*$ in $La_2-x\text{Sr}_x\text{CuO}_4$ related to spin excitations$^{13,44}$, at our present knowledge, we cannot exclude a subdominant spin nature of the quantum fluctuations we have single out with RIXS. Finally, it is worth reminding that the CDF are strongest at a doping level, slightly above the optimal doping, $p^* \approx 0.19$. The superfluid density$^{34}$, the superconducting critical current density$^{45,46}$ and the upper critical field$^{47}$ are the highest.

In a scenario of quantum criticality, one should expect the correlation length of the CDF to diverge at $p^*$ when approaching zero temperature. This is not happening in our data for $p \approx 0.19$, also because the superconducting order clearly influence the behavior of CDF below $T_c$. We can postulate that, under high magnetic field suppressing superconductivity, the CDF might be drastically different and $\xi$ diverges. However, this scenario is in contradiction with the observation that the linear resistivity of the strange metal extends down to very low $T$ in the absence of superconductivity. In fact, as we attribute the linear resistivity to the quasi-isotropic scattering of carriers by CDF, a CDF peak becoming very narrow at low $T$ would not guarantee the needed isotropic scattering mechanism. To resolve this contradiction, we can think either of a frustrated criticality at $p \approx 0.19$, with $\Delta$ remaining finite as $T \to 0$, or of $\Delta \to 0$ due to the divergence of the damping factor $\gamma$, with $\omega$ staying finite. In the latter case, small domains hosting CDF with finite correlation length $\xi$ would undergo a critical slowing down resulting in an almost persistent glassy state. In other words, the tendency towards a long-range order at the QCP would be frozen in an incoherent ensemble of CDF droplets. This picture would be consistent with the existence of an anomalous QCP.

Our observations stimulate important questions about the possible interconnection between charge ordering, lattice dynamics, strange metal and superconductivity in cuprates. While the presence of CDF leads to a softening of the high-energy bond-stretching phonons, as we have observed in Fig. 1f, i, it is very plausible a direct coupling of the CDF with the acoustic phonons, whose intensity is observed in the medium-resolution RIXS spectra, measured at $20$ and $290$ K i) at nine different temperatures for the $p = 0$ and $0.06$ samples; ii) at four different temperatures for the $p = 0.09$ sample. Relaxing the energy resolution up to 62 meV has allowed us to perform a thorough temperature dependence. The full agreement between the results coming from high and medium resolution RIXS spectra, we have presented in the manuscript, demonstrates that the pollution coming from the bond stretching phonons to the CDF dependencies are negligible.

The incident X-rays have been chosen so that their energy is tuned to the maximum of the Cu $L_2$ absorption peak at around 931 eV, and their polarization is linear, perpendicular to the scattering plane (o-polarization). They impinge on the sample surface, normal to the YBCO c-axis, and are scattered by an angle $2\theta$. The momentum transfers are given in units of the reciprocal lattice vectors $\vec{q} = 2\pi/a, b = 2\pi/b, c = 2\pi/c$. We have worked at $20 - 150^\circ$ in order to get $|\vec{q}| = 0.91\text{Å}^{-1}$, which allowed us to cover the whole first Brillouin zone along the $(H,0,0)$ direction ($0.5\text{Å}^{-1} < 0.81\text{Å}^{-1}$). We have changed the in-plane wave vector component $q_y$, fixing $2\theta$ and rotating the samples around $\theta$ only (charge order in cuprates is indeed only weakly $\theta$-dependent).

The RIXS spectra in the manuscript have been first corrected for self-absorption$^{48,49}$ and then normalized to the integral of the interorbital $dd$ excitations, in the range $[-3\text{eV}, -1\text{eV}]$. The self-absorption correction compensates for the possible reabsorption of scattered photons along their path out of the sample from the scattering point. The probability of self-absorption depends on the absorption coefficients of the incident and scattered photons (which depend on their energy and polarization) and on the scattering geometry. It also depends on the total thickness of the sample. We have implemented the exact form of the correction that takes into account all the parameters: $\theta$ and $\chi$, angles between the normal to the sample surface and the incident and emitted photon propagation direction; $\Delta_{1,2}$, total and resonant part of absorption coefficient for the incident/scattered photons, with $\Delta_{2} = \Delta_{1} + \Delta_{0}$ and $\Delta_{0}$ non-resonant pre-edge absorption coefficient not contributing to the RIXS signal; $d$, sample thickness. The quality of the correction depends on the knowledge of the absorption coefficients, which critically depend on the photon energy and polarization in the proximity of the absorption resonance.
Conversely, the angular dependence is univocally known. The absorption coefficients were evaluated from the XAS spectra measured on every sample during each RIXS experiment. Their relative values are sufficient for the correction in case of thick samples (as single crystals, whose thickness can be approximated to infinite), whereas the absolute value of $\alpha_{\text{d}}$ is needed for thin films, but it can be estimated from tabulated data of $\alpha_0$.

For clarity, we report here the self-absorption correction formulas. Calling $\eta_i(h_{\nu_2})$ the ideal RIXS spectrum measured with incident photon energy $h_{\nu_1}$ and polarization $\varepsilon_i$, the self-absorption modifies the spectral intensity point by point leading to:

$$I_i(h_{\nu_2}) = \frac{\eta_i}{\alpha_{\text{d}}} C_i(h_{\nu_2}, \varepsilon_2) D_i(h_{\nu_2}, \varepsilon_2) \eta_i(h_{\nu_2})$$

with

$$C_i(\theta, \chi, \frac{\alpha_{\text{d}}}{\alpha_{\text{d}}} (h_{\nu_2}, \varepsilon_2)) = \left(1 + \frac{\alpha_{\text{d}}}{\alpha_{\text{d}}} (h_{\nu_2}, \varepsilon_2) \sin \theta \right)^{-1}$$

$$D_i(\theta, \chi, \frac{\alpha_{t}}{\alpha_{\text{d}}}, \alpha_{\text{d}}, d) = 1 - \exp \left[ -C_i \cdot \frac{d}{\sin \theta} \right]$$

We notice that for thick samples, $d \to \infty$ and $d = 1$. Therefore, the spectrum $\eta_i(h_{\nu_2})$ that ideally would be measured in the absence of self-absorption from a thick sample is derived from the measured one $I_i(h_{\nu_2})$ as

$$\eta_i(h_{\nu_2}) = \frac{\alpha_i}{\alpha_{\text{d}}} C_i D_i I_i(h_{\nu_2})$$

Thanks to this procedure we can safely compare the shape and intensity of spectra measured at different $\theta$ and $\chi$ angles (i.e., at different $q$ points) for a given sample and excitation energy (fixed absorption coefficients, also when the temperature is varied). Spectra of different samples can also be compared, although with some extra precautions due to the uncertainties related to the $\alpha$ coefficients that are not identical in different samples.

We also emphasize here that the self-absorption correction we have applied for both thin films and single crystals does not modify the FWHM and the $H_{\text{CDF}}$ of the CDF peak (See Supplementary Fig. 8).

### EI-RIXS measurements

Energy Integrated Resonant X-ray scattering (EI-RIXS) measurements have been performed at the UE46-PGM beamline of BESSY II at Helmholtz Zentrum Berlin on the slightly overdoped YBCO ($\rho = 0.185$) and Bi2212 ($\rho = 0.19$) samples. The geometry was identical to that of the RIXS experiments. The samples have been mounted on an ultra-high vacuum diffractometer, in contact with the cold finger of a liquid-Helium-flow cryostat. The incident photons are linearly polarized in the direction perpendicular to the scattering plane (0-polarization). The scattered photons have been detected using a standard photodiode without discrimination of either polarization and energy, implying that the measured intensities represent an integration over all elastic and inelastic scattering processes. Momentum-space scans have been measured at several temperatures in the range between 10 and 300 K.

### Alignment and fit of the high resolution RIXS spectra

For each sample, we have first determined the zero energy loss line by measuring a resonant spectrum close to $\Gamma=(0,0)$: here, the low energy region is indeed dominated by the purely elastic intensity of the specular, which prevails over any other inelastic contribution, coming from charge order or phonons. The upper error on the position of the zero can be estimated in the order of 2 meV, as we have shown in detail in the supplementary material of ref. 26. All the spectra measured at higher $q$ values have been aligned among each other, and to the reference spectrum at $\Gamma$, by aligning the maxima of the first derivatives of the quasi-elastic regions.

To extract quantitative information from the spectra, we have fitted those along the $(H,O)$ direction considering the quasi-elastic region as the sum of: i) an energy resolution limited, purely elastic peak (green Gaussian in Fig. 1c); ii) a low energy peak linked to CDF (red Gaussian in Fig. 1c), whose FWHM is a free fit parameter and whose position is first left free to change, then fixed to its value close to $q_c$, which is more stable (being its intensity overshadowed by that of the specular peak at low $q$ values and by that of the breathing phonons at high $q$ values); iii) a Gaussian associated to the bond-stretching phonons (orange curve in Fig. 1c), whose position and FWHM are free fit parameters; iv) a Gaussian associated to the phonon overtones, with variable FWHM and position set to twice the position of the bond-stretching phonon peak; v) an antisymmetrized Lorentzian function, associated to the paramagnons, as shown in ref. 64.

A similar fit has been done on the spectra measured along the $(H,0)$ direction: for each sample i) the intensity of the purely elastic peak is at any $q$ very close to that along the $(H,O)$ direction (which justifies the difference spectra plotted in Fig. 1a, b) (see Supplementary Fig. 5d); ii) the position of the CDF Gaussian is much higher than that extracted along the $(H,O)$ direction (see Supplementary Fig. 5f), in agreement with theoretical previsions (see Eqs. (1) and (2)), while the intensity of the peak is almost $q$-independent; iii) the bond-stretching phonons do not soften, differently from what observed along the $(H,O)$ direction (see Supplementary Fig. 5h).

Here, it is worth mentioning that the positions in energy of the CDF, $\Delta$, and of the bond-stretching phonons are affected by an uncertainty of about 4 meV. As previously discussed in ref. 26, this is well below the energy resolution of our instrument. Indeed these two peak positions are determined by the difference between the centroid of the pure elastic peak and the centroid either of the CDF peak at $q = q_{\text{CDF}}$ or of the bond-stretching phonon peak. This estimation is much more accurate than the instrumental bandwidth: the position of the centroid of a data distribution is well defined and very stable, even when the noise level is relatively high, once the line-shape of the data distribution is well-known.

Our high-resolution spectra could in principle be fitted considering only two peaks in the quasi-elastic region, i.e., the pure elastic peak and the bond-stretching phonon one. However, we have also considered a third peak between these two, which we have assigned to charge density fluctuations. This is mainly for two reasons. First, the fit improves significantly, since we directly account for the presence of the strong intensity at $q_{\text{CDF}}$ and finite energy, which is evident from the maps in Figs. 1 and 3. Secondly, it is rather common for Cu L$_3$ RIXS spectra of cuprates, measured with good resolution (35–40 meV BW) to make a three-peak fitting for the undoped compounds: one for the elastic component at zero energy, one for the bond-stretching phonons above 50 meV, and one of intermediate energy for other low-energy phonons modes, such as the bond-buckling. However, in doped samples, charge order excitations become relevant, and their spectral intensity dominates the region between the elastic and the bond-stretching phonon peak. A three-peak fit (elastic, CDF and bond-stretching) can be performed here, without including an additional, fourth component connected to the bond-buckling phonons. Indeed, their intensity at the Cu L$_3$ edge is much smaller than the other low-energy phonons, as the acoustic and the bond-stretching. We note that, on the contrary, for O K edge RIXS the buckling mode is more intense and cannot be neglected. Moreover, in the range of $q$ where the CDF is the strongest and its energy $\Delta$ is determined, i.e. at $q$ close to $q_{\text{CDF}} = 0.25-0.35$ r.l.u., their intensity, being proportional to $\cos^2(qR)$, is even weaker. Therefore, we decided not to include the buckling...
phonon contribution in our fitting, in order to restrict the number of free parameters and obtain more robust results. For completeness, we have anyway made a test with four peaks, adding a fourth Gaussian component, centered at the energy where buckling phonons have been previously measured in YBCO and Bi2212 by other techniques. We found that the intensity of this additional phonon mode is low enough to leave unaltered the position of the CDF contribution, i.e., the CDF energy $\Delta$ (see Supplementary Fig. 9). Of course, a softening of the bond-buckling phonon branch due CDF is expected, as it has been already measured in cuprates at the O $K$ edge. However, the intensity we measure at the Cu $L_{2,3}$ edge is too low to study this phonon anomaly and to attribute to this phonon anomaly any change in the CDF parameters as extracted by the three-peak fit.

Global fit of the medium energy resolution RIXS spectra

Medium energy resolution RIXS spectra have been integrated over the [$-100, 35$] meV energy range to extract quantitative information on the intensity evolution of the CDF peak with momentum $q$ and temperature $T$, irrespective of the details of its spectral line shape, as shown in Fig. 2a. The $(q, T)$ curves were fit simultaneously to extract robust estimates of the parameters appearing in Eqs. (1) and (2), as shown for slightly overdoped YBCO (see Fig. 2b) and Bi2212 (see Supplementary Fig. 2a, b).

Bose fit of the medium energy resolution RIXS spectra

The Bose function $A + I_0 \left[ 1 + 2 \left( e^{\frac{\Delta q_{\text{CDF}}}{k_B T}} - 1 \right) \right]^{-1}$ used in the main text depends, among the other, on the values of the variables $A$ and $I_0$. Their sum represents the total quasi-elastic intensity in the zero temperature limit $I_0$ is the CDF intensity, which far from $q_{\text{CDF}}$ we consider $q$-independent, while $A$ accounts for the non-CDF contributions, which is strongly $q$-dependent, and minimum at high $q$ values (consequently to the $q$-dependence of the specular elastic peak, see Supplementary Fig. 6a). For each sample, we have determined the $A$ and $I_0$ values from the high-resolution spectra, given the relative weight of the purely elastic Gaussian and of the CDF Gaussian in the [$-100, 35$] meV energy range. The doping dependence of the $\Omega$ values, i.e. CDF energies along the $(HH)$ direction estimated from the fit, is however very robust, and the solidity of the $\Omega$ values goes beyond any possible uncertainties in the correct determination of $A$ and $I_0$.

In Fig. 4a, b the difference between the data taken on samples at various dopings is very clear. In particular, the slopes of the quasi-linear trend of the integrated intensity versus temperature are very different (see also Supplementary Fig. 4a). In a Bose scenario, this means that the energies $\Omega$ are different. More precisely, we can estimate the ratio between the $\Omega$ values at different dopings, which is a good indicator of the situation. This approach is based on the fact that the Bose function can be put in a universal form by plotting it in terms of the reduced temperature $T/\Omega$. Thus, different curves will collapse onto the universal function if they are plotted as a function of $T/\Omega$. Because the scaling factors are different in the two cases, the definition of reduced temperature, thence the energy $\Omega$, is different. This is very clear from Supplementary Fig. 4b, where we have plotted as a function of $T/\Omega$ the two curves of the strongly underdoped and of slightly overdoped samples at $q = 0.44$ r.l.u. Here, the ratio is 1.5, which is in agreement with the $\Omega_{3.45}/\Omega_{3.19}$ ratio (47/31), resulting from the Bose fit presented in the main text. This ratio of the two scaling factors does not depend on the choice of one of the two values of $\Omega$, and is $T$-independent because of the universality of the Bose plot.

Focusing instead on the absolute values of $\Omega$, the values determined from the fit are in fairly good agreement with the values we have directly measured in the high-resolution spectra. Indeed, the position of the CDF Gaussian along the $(HH)$ direction is, depending on the $q$ value, in the 35–38 meV energy range (see Supplementary Fig. 5f): this value is only ~15% higher than that we have found using the Bose fit.

Data availability

All data shown in the main text and in the supplementary information are available at the Zenodo repository, under accession code https://doi.org/10.5281/zenodo.8430044.

References

1. Keimer, B., Kivelson, S. A., Norman, M. R., Uchida, S. & Zaanan, J. From quantum matter to high-temperature superconductivity in copper oxides. Nature 518, 179–186 (2015).
2. Zaanan, J. Why the temperature is high. Nature 430, 512–513 (2004).
3. Legros, A. et al. Universal $T$-linear resistivity and Planckian dissipation in overdoped cuprates. Nat. Phys. 15, 142–147 (2019).
4. Varma, C. M. Linear in temperature resistivity and associated mysteries including high temperature superconductivity. Rev. Mod. Phys. 92, 031001 (2020).
5. Castellani, C., Di Castro, C. & Grilli, M. Non-Fermi-liquid behavior and d-wave superconductivity near the charge-density-wave quantum critical point. Z. Phys. B 103, 137 (1996).
6. Mitrano, M. et al. Anomalous density fluctuation in a strange metal. Proc. Natl Acad. Sci. USA 115, 5392 (2018).
7. Van der Marel, D. et al. Quantum critical behaviour in a high-Tc superconductor. Nature 425, 271–274 (2003).
8. Giraldo-Gallo, P. et al. Scale-invariant magnetoresistance in a cuprate superconductor. Science 361, 479–481 (2018).
9. Berthier, C. et al. NMR investigation of low energy excitations in high $T_c$ superconductors. Phys. Scr. T49A, 131–136 (1993).
10. Zaanan, J. Planckian dissipation, minimal viscosity and the transport in cuprate strange metals. SciPost Phys. 6, 061 (2019).
11. Patel, A. A., McGreevy, J., Arovas, D. P. & Sachdev, S. Magneto-transport in a model of a disordered strange metal. Phys. Rev. X 8, 021049 (2018).
12. Dumitrescu, P. T., Wentzell, N., Georges, A. & Parcollet, O. Planckian metal at a doping-induced quantum critical point. Phys. Rev. B 105, L180404 (2022).
13. Seibold, G. et al. Strange metal behaviour from charge density fluctuations in cuprates. Commun. Phys. 4, 7 (2021).
14. Wù, W., Wang, X. & Tremblay, A.-M. Non-Fermi liquid phase and linear-in-temperature scattering rate in overdoped two-dimensional Hubbard model. Proc. Natl Acad. Sci. USA 119, e211581119 (2022).
15. Varma, C. M. Non-Fermi-liquid states and pairing instability of a general model of copper oxide metals. Phys. Rev. B 55, 14554 (1997).
16. Delacrétaz, L., Goutéraux, B., Hartnoll, S. & Karlsson, A. Bad metals from fluctuating density waves. SciPost Phys. 3, 25 (2017).
17. Sachdev, S. Where is the quantum critical point in the cuprate superconductors? Phys. Status Solidi (b) 247, 537 (2010).
18. Chen, S.-D. et al. Incoherent strange metal sharply bounded by a critical doping in Bi2212. Science 366, 1099–1102 (2019).
19. Sachdev, S. Quantum Phase Transitions 2nd edn (Cambridge Univ. Press, 2011).
20. Andergassen, S., Caprara, S., Di Castro, C. & Grilli, M. Anomalous isotopic effect near the charge-ordering quantum criticality. Phys. Rev. Lett. 87, 056401 (2001).
21. Abanov, A., Chubukov, A. V. & Schmalian, J. Quantum-critical theory of the spin fermion model and its application to cuprates: Normal state analysis. Adv. Phys. 52, 119–218 (2003).
22. Huang, E. W. et al. Fluctuating intertwined stripes in the strange metal regime of the Hubbard model. Phys. Rev. B 107, 085126 (2023).
23. Li, Y. et al. Hidden magnetic excitation in the pseudogap phase of a high- $T_c$ superconductor. Nature 468, 283–285 (2010).
24. Wahiberg, E. et al. Restored strange metal phase through suppression of charge density waves in underdoped YBa$_2$Cu$_3$O$_{6.6}$-Science 373, 6562 (2021).
69. Graf, J. et al. Bond stretching phonon softening and kinks in the angle-resolved photoemission spectra of optimally doped Bi$_2$Sr$_{1.6}$La$_{0.4}$Cu$_2$O$_{8+\delta}$ superconductors. Phys. Rev. Lett. 100, 227002 (2008).

70. Pintschovius, L. et al. Oxygen phonon branches in YBa$_2$Cu$_3$O$_7$. Phys. Rev. B 69, 214506 (2004).

Acknowledgements

The authors acknowledge Zhi-Xun Shen, Tom Devereaux, Wei-Sheng Lee, Jan Zaanen, Ulf Gran and Alexander Krikun for insightful discussions, Roberto Fumagalli for helping with EI-RXS measurements, and Alexei Kalaboukhov for the support with the PLD used for the deposition of the Ca-YBCO thin films. This work was performed in part at Myfab Chalmers. The RIXS experimental data were partly collected at the beam line ID32 of the European Synchrotron (ESRF) in Grenoble (France) using the ERIXS spectrometer designed jointly by the ESRF and Politecnico di Milano. We acknowledge Diamond Light Source for providing the beam time at the I21-RIXS beamline under proposal MM23880. We thankfully acknowledge HZB for the allocation of synchrotron radiation beam time, at the beamline UE46-PGM1 of BESSY II where the EI-RXS measurements have been carried out. This work is supported by the project PRIN2017 “Quantum-2D” ID 2017Z6TS58 of the Ministry for University and Research (MIUR) of Italy (L.M., M.M.S., S.C., M.G., G.G.), by the Swedish Research Council (VR) under the projects 2018-04658, 2022-04334 (F.L.), 2020-04945 (R.A.) and 2020-05184 (T.B.), by the European Union’s Horizon 2020 research and innovation programme (grant agreement No 730872). S.C. and M. G. also acknowledge financial support from the University of Rome Sapienza, through the projects Ateneo 2020 (Grant No. RM120172ABCC7CC7), Ateneo 2021 (Grant No. RM12117A4A7FD11B), Ateneo 202 (Grant No. RM12218162CF9D05). M.G. acknowledges financial support from PNRR MUR project PE0000023-NQSTI.

Author contributions

R.A., L.B. and G.G. conceived and designed the experiments with suggestions from F.L., C.D.C. and M.G. L.M., R.A., M.M.S., G.G., L.B., N.B.B., A.N., K.J.Z., M.G.-F., Q.L. and Y.Y.P. performed the RIXS measurements. E.S., R.A., M.M.S., L.M. and G.G. performed the EI-RXS measurements. R.A., T.B. and F.L. grew and characterized the YBCO films. Q.G. and X.Z. grew and characterized the Bi2212 single crystal. R.A., L.B., M.M.S., P.C. and G.G. analyzed the RIXS experimental data. R.A., L.B., G.G, M.M.S., S.C., C.D.C., M.G., F.L., T.B. and L.M. discussed and interpreted the results. R.A. and G.G. wrote the manuscript with the input from M.G., C.D.C., F.L., L.B., M.M.S., S.C. and contributions from all authors.

Funding

Open access funding provided by Chalmers University of Technology.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41467-023-42961-5.

Correspondence and requests for materials should be addressed to Riccardo Arpaia or Giacomo Ghiringhelli.

Peer review information Nature Communications thanks Saleh Hasan Naqib, and the other, anonymous, reviewers for their contribution to the peer review of this work. A peer review file is available.

Reprints and permissions information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2023