Letter | Editors' Suggestion

Decoupling of static and dynamic charge correlations revealed by uniaxial strain in a cuprate superconductor

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We use uniaxial strain in combination with ultra-high-resolution resonant inelastic x-ray scattering (RIXS) at the oxygen-K and copper-L_3 edges to study the excitations stemming from the charge ordering wave vector in $La_{1.875}Sr_{0.125}CuO_4$. By detwinning stripe ordering, we demonstrate that the optical phonon anomalies do not show any stripe anisotropy. The low-energy charge excitations also retain an in-plane fourfold symmetry. As such, we find that both phonon and charge excitations are decoupled entirely from the strength of static charge ordering. The almost isotropic character of charge excitations is indicative of a quantum critical behavior and remains a possible source for the strange metal properties found in the normal state of cuprate superconductors.

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Introduction. Excitation spectra entail fingerprints of their underlying ground state. In high-temperature cuprate superconductors, this has motivated numerous studies of phonon and charge excitations. In particular, excitations with momentum matching the charge-stripe ordering wave vector have been the object of intense scrutiny [1]. The problem is complicated by the presence of dynamic excitations which emerge at higher temperatures and precede the formation of static charge order [2–7]. The predominant interpretation is that they constitute quantum fluctuations of the electronic "crystal" created by charge order (CO). Such strong fluctuations could signal the proximity to a quantum critical point, such as the putative pseudogap endpoint at $p \sim 0.19$ [8,9]. It was proposed that the short-range character of such excitations could provide a quasi-isotropic scattering in momentum space, and explain the strange metal behavior in the transport properties [10–12]. Little is known yet, however, about the symmetry of these excitations. The presence of twinned domains in the tetragonal CuO_2 planes precludes investigation of their symmetry properties.

Employing an *in situ* strain device, we achieve substantial detwinning of the charge-stripe order in the prototypical cuprate $La_{2-x}Sr_xCuO_4$ with $x = 0.125$. We investigate the inelastic excitations using ultra-high-resolution resonant inelastic x-ray scattering (RIXS) at both the oxygen-K and copper-L_3 edge. No strain-induced changes in the energy and intensity of optical phonon excitations are observed, demonstrating that the anomalies in the phonon intensity and dispersion are not primarily correlated with the strength of the stripe order parameter. The simultaneous use of ultra-high-resolution at two resonant edges resolves the apparent discrepancy observed in the RIXS response at the Cu L_3 and O K edges [2,7,13] and provides a clear proof of the existence of quantum fluctuations associated with CO. Our measurements demonstrate that these excitations remain unaffected by the strain application, both around the a- and b-axis ordering wave vectors, and therefore retain a higher symmetry then the static order parameter. Moreover, we show that the anomalous softening of the phonon modes is a direct consequence of these collective modes. Our observations suggest that these fluctuations are linked to short-range electronic interactions, and the decoupling from the static charge-stripe order hints to a quantum critical behavior. Our measurements bear significant implications for the theory of charge fluctuations and the properties of strange metals.

Methods. We used single crystals of LSCO $x = 0.125$ [14]. Rectangular pieces with facets defined by the tetragonal a,

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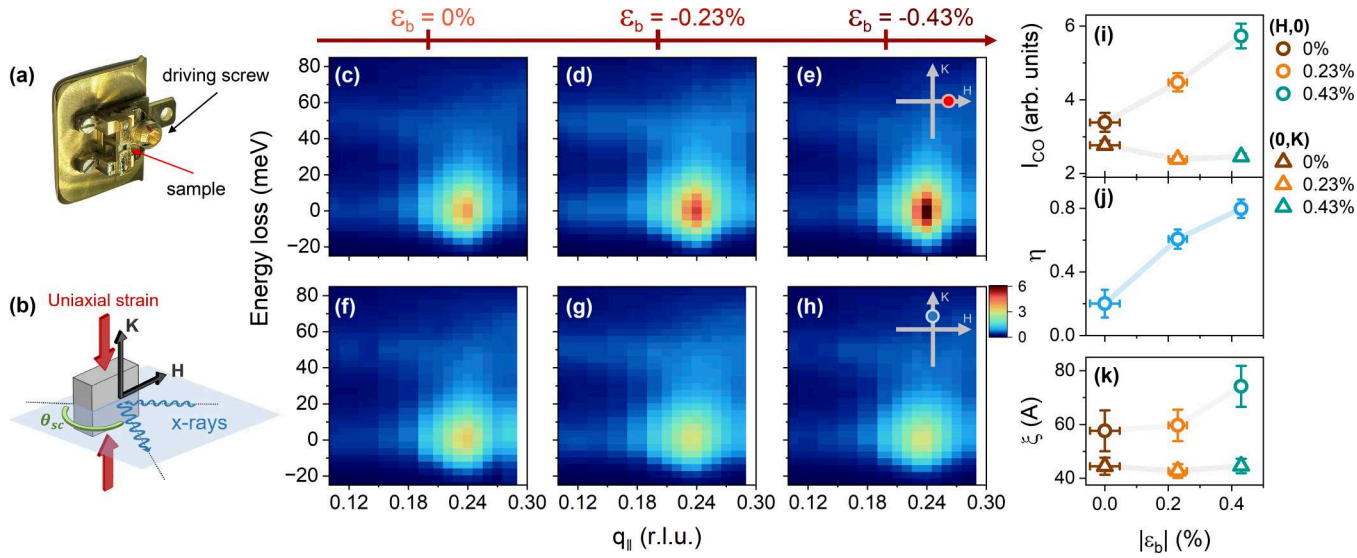

FIG. 1. RIXS spectra at oxygen K-edge under uniaxial pressure application. (a) Photograph and (b) sketch of, respectively, the *in situ* strain device and the experimental (scattering) geometry. Incident and scattered x-rays are indicated by blue wavy arrows. The reciprocal H and K directions are indicated with black arrows. The direction of application of uniaxial strain (K) is indicated with red arrows. (c)–(h) Low-energy RIXS spectra as a function of momentum along (c)–(e) the $(H, 0)$ and (f)–(h) $(0, K)$ directions. (i) Intensity of static CO along $(H, 0)$ (circles) and $(0, K)$ (triangles) directions as a function of applied strain. (j) Domain population factor η (see text for definition) as a function of strain. (k) Longitudinal correlation length as along the $(H, 0)$ and $(0, K)$ directions as function of in-plane strain ε_b.

b, and c axes were cut down to $1 \times 0.4 \times 0.7$ mm^3 in size, glued inside the strain cell, and cleaved *ex situ*. Our uniaxial pressure cell is an adaptation of the design described in Refs. [15,16]. Compressive strain is applied *ex situ* through a driving screw and gauged through a calibration curve obtained through the measurement of the 002 Bragg reflection and knowledge of the Poisson ratio [17,18];- see the Supplemental Material [19]. The RIXS scattering geometry is illustrated in Figs. 1(a) and 1(b). The sample stage can be rotated such that scans along both $q_\parallel = (H, 0)$ and $(0, K)$ directions are possible under application of b-axis strain (ε_b). The energy resolutions at the oxygen-K (copper-L_3) edge were 23 (43) meV full-width-at-half-maximum (FWHM). Wave vectors $\mathbf{q} = (q_x, q_y, q_z)$ are labeled in tetragonal reciprocal lattice units (r.l.u.) of $(2\pi/a, 2\pi/b, 2\pi/c)$, with $a = b = 3.78$ Å and $c = 13.2$ Å. Temperature was fixed to $T_c \sim 28$ K, to maximize the CO intensity [14,20].

Results. Oxygen-K edge RIXS maps along the two high-symmetry directions are shown for three pressure points in Figs. 1(c)–1(h). Elastic scattering from CO is observed at $q_{co} = (\delta, 0)$ and $(0, \delta)$ with $\delta \sim 0.235$ r.l.u.—consistent with previous reports [21,22]. To quantify the CO intensity versus applied strain, we model the RIXS spectra with an elastic peak, three inelastic excitations, and a smooth function for the electron-hole continuum [Fig. 2(a)]; see the Supplemental Material. The intensity of CO is quantified by the area of the elastic line as a function of in-plane momentum. Its analysis as a function of strain is shown in Figs. 1(i) and 1(j). Without strain application, CO is nearly equally strong along the a and b axis. The minor discrepancy may originate from a small residual compressive strain induced by thermal contraction of the device. With application of b-axis strain, the a-axis charge order intensity I_H is enhanced roughly linearly in the investigated range of strain values [Fig. 1(i)]. Simultaneously, b-axis CO intensity I_K decreases, in agreement with Ref. [17]. The correlation length, extracted as $\xi = \frac{1}{\text{HWHM}}$ [23] (with HWHM being the half-width at half-maximum of the CO peak), slightly increases with strain along the a axis, from (9 ± 1) to (12 ± 1) unit cells. No significant changes are observed along the b axis. These correlation lengths are consistent with the existing literature [20,24]. The domain population factor $\eta = 2(I_H - I_K)/(I_H + I_K)$, plotted in Fig. 1(j), increases by more than a factor of four in the applied strain interval. We note that the magnitude of the reduction of CO along the b axis is less than the enhancement along the perpendicular direction, a behavior consistent with previous measurements on $YBa_2Cu_3O_{6+x}$ [25,26].

The high-energy resolution RIXS spectra allows us to scrutinize the charge excitations visible in the 0–100 meV energy range. A typical RIXS spectrum near is shown in Fig. 2(a). The low-energy inelastic region, visible in Figs. 2(b)–2(d), contains three excitation branches—with energy scales of about 20, 50, and 70 meV. By comparison with inelastic neutron and x-ray scattering experiments [27–29], the two highest-energy modes are identified with the oxygen bond-buckling (BB) and bond-stretching (BS) modes. The nature of the low-energy (LE) excitation (∼20 meV) will be discussed later. From the fitting model (see also the Supplemental Material [19]) the dispersion relations and excitation amplitudes are extracted. The LE excitation has a pronounced intensity maximum centered approximately at the CO wave vector. The momentum width corresponds to a short correlation length of $\xi \sim 6a$. Its energy shows a minimum close to q_{co} and a dispersion of ∼5 meV [Fig. 2(b)]. The energy of the BS phonon far from the CO wave vector has a characteristic $\cos(\pi k_{x,y})$ behavior [black dotted line in 2(e)], and shows a gradual decrease toward the zone boundary [27]. This is usually attributed to

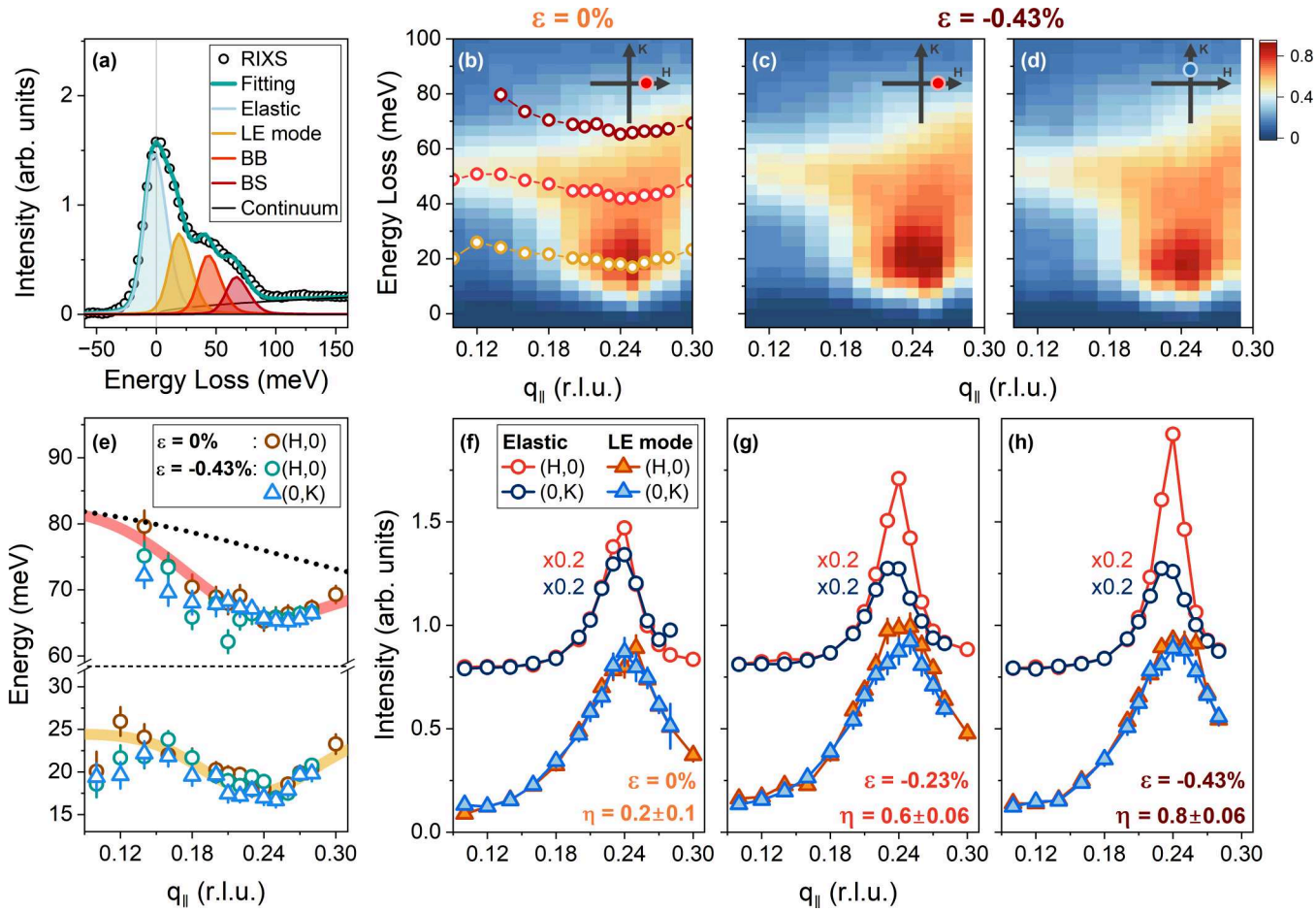

FIG. 2. Oxygen-K edge resonant x-ray scattering. (a) Example of a fitted spectrum at Q=(0.26,0). (b)–(d) RIXS spectra as a function of momentum $\mathbf{q}_\parallel$ after subtraction of the elastic signal. Applied strain values and directions are indicated above the panels and in the insets, respectively. (e) Energy of the bond-stretching mode and of the low-energy mode as a function of momentum for two different values of applied strain, for both H (circles) and K (triangles) directions. The black dotted line is a cosine function, while the red and yellow solid lines are guides to the eye. (f)–(h) Intensity of elastic peak (empty circles) and low-energy mode (filled triangles) for both $(H, 0)$ (red and orange symbols) and $(0, K)$ (blue and light-blue symbols) directions, for increasing strain application.

the screening provided by holes on extended orbitals [30]. Close to q_{co}, it shows a marked softening of ∼10 meV [Figs. 2(b) and 2(e)], in agreement with neutron scattering measurements [31–33]. The dispersion relations, as well as the intensities versus momentum, are—within our instrumental sensitivity, which we can estimate as 5 meV, see the Supplemental Material [19]—independent of the applied uniaxial pressure. In particular, as shown in Fig. 2(e), the anomalous softening of the BS phonons is independent of strain and is equal along the $(H, 0)$ and $(0, K)$ directions. The intensity of the LE mode also shows the same behavior; see Figs. 2(f)–2(h). These observations are simultaneous with the uniaxial-strain-induced detwinning of the static CO.

These experimental observations are confirmed and reproduced at the copper-L edge (Fig. 3). With the assumption that the measured excitations have the same energy at the two edges, we are able to give a unified description of the RIXS response. The domain population ratio η is consistent at the two edges [Fig. 3(a)]. The q dependence of the intensity observed on the LE mode is identical to that extracted at the oxygen-K edge, as highlighted in panel (d) of Fig. 3. The intensity of the bond-stretching mode is, likewise, identical along the a and b axis along the entire direction, and independent of strain within our experimental sensitivity [Fig. 3(c)]. The intensity of a phonon is, in RIXS, correlated with the electron-phonon coupling (EPC) [34–36]. In systems where the photoexcited electrons are strongly localized, like at the copper-L_3 edge in cuprates, the RIXS phonon cross section is first order proportional to the EPC [37]. Even in systems with mobile electrons, like at the oxygen-K edge, a positive correlation still persists [36,38]. This implies that the EPC is identical along the a and b axis, despite a twofold increase in the CO intensity. These observations reinforce the results at the oxygen-K edge and prove that the same excitations are observed on both resonances. Their response to uniaxial strain, consistent at both absorption edges, is schematically visualized in Fig. 4.

Discussion. Acquisition of high-resolution RIXS spectra allowed us to extract three low-energy excitations. The BB mode, at 50 meV, does not show anomalies in the proximity of q_{co} (see Figs. S4 and S5 in the Supplemental Material), consistent with previous results. We therefore concentrate our discussion on the other two modes. A fundamental question revolves around the relationship between the energy

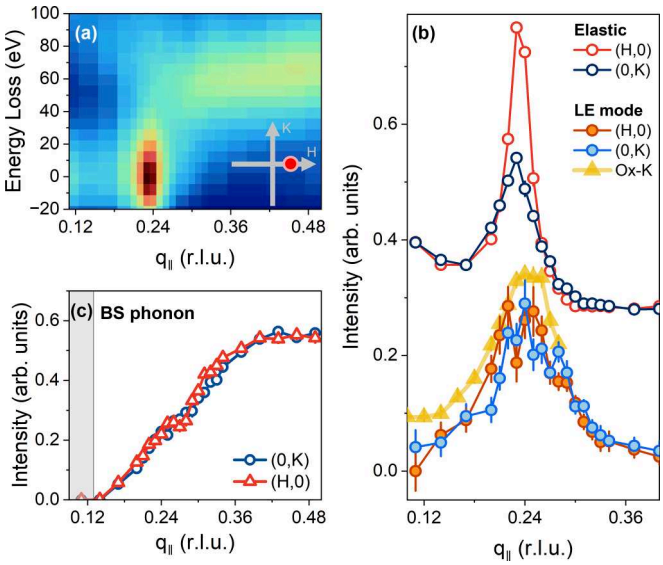

FIG. 3. Copper-L edge resonant x-ray scattering. (a) RIXS maps at Cu L_3 edge along the $(H, 0)$ direction at the maximum applied strain (-0.43%). (b) Comparison of the amplitude of CO fluctuations at the oxygen (yellow triangles) and copper edges (orange and light blue circles). Oxygen data has been scaled and shifted by 0.04 in the vertical direction. (c) Intensity of the bond-stretching phonon along the $(H, 0)$ and $(0, K)$ directions at the maximum applied strain.

of the BS mode and charge order. The softening of the dispersion from the $\cos(\pi k_{x,y})$ behavior expected from LDA calculations [39] [dotted line in Fig. 2(e)], as well as anomalies in the linewidth and intensity, have been reported in many cuprate families by several spectroscopic techniques [2,6,13,40,41]. This self-energy effect could stem from an

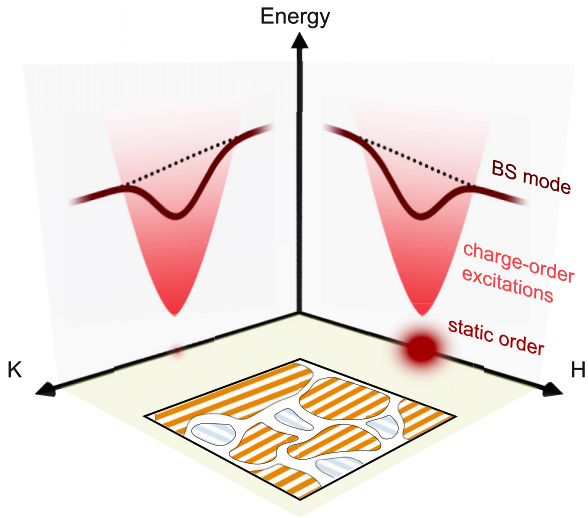

FIG. 4. Sketch summarizing our experimental results. The intensity of static CO shows a substantial detwinning upon strain application. The energy and intensity of charge fluctuations, as well as the softening of the BS mode, retain tetragonal symmetry. The square at the bottom sketches the situation in real space, with an imbalance of CO domains.

interaction between the phonon and the static order having C_2 symmetry, i.e., a Kohn anomaly picture [42]. In this scenario, bond-stretching phonons should split into two branches, one propagating parallel to the stripe modulation (showing the anomalous softening) and one moving perpendicular to them [42]. Without stripe detwinning ($\eta \to 0$), this is, however, difficult to verify experimentally. By uniaxial pressure application, we reach an almost perfect detwinning ($\eta \to 1$). We do not, however, detect any change in the intensity and dispersion of phonons as a function of applied strain, despite a twofold change in the intensity of static charge-order, see Fig. 2. The softening persists unaltered along both in-plane directions. In a Kohn-type anomaly scenario [43,44], the phonon softening is proportional to the EPC $g(q)$ and the susceptibility: $\delta\omega(q)^2 \propto g(q)^2\chi(q, 0)$ [45]. The application of b-axis strain increases the volume of domains ordering along the $(H, 0)$ direction, and enhances (decreases) $\chi(q, 0)$ ($\chi(0, q)$), as also testified by the change in correlation length [Fig. 1(k)]. Since our Cu L_3 measurements reveal that $|g|$ remains constant along the two Cu-O bond directions, we can conclude that a Kohn-type anomaly is not consistent with our measurements. If the breathing branch were split into two branches propagating parallel and perpendicular to the stripes, the strain should modulate the intensities of the two in opposite ways along the $(H, 0)$ and $(0, K)$ directions, effectively changing the center of mass of the distribution. Within our sensitivity ($\sim$5 meV), we cannot detect such a shift.

The root cause of the softening of the BS mode must therefore be searched elsewhere. Previous works suggested that dynamical fluctuations of finite energy could induce a renormalization of phonon energy [32,33]. Hints of the existence of such excitations were provided only very recently in RIXS experiments on different cuprate materials. A loss of spectral weight in the inelastic range below $\sim$40 meV has been reported in the low-energy spectra upon heating [3,7,8,13,49], a behavior incompatible with a pure phonon mode. It is similar when going to the strongly underdoped and overdoped regime [8,13].

RIXS literature on copper-L [2,6] and oxygen-K [7,13] edges suggested, however, divergent interpretations. These anomalies have been assigned to either strong softening of bond-stretching modes [2,6] or to the direct presence, in the experimental spectra, of a low-energy electronic excitation [7,13]. Both scenarios are based on the presence of a continuum of quantum fluctuations associated with charge ordering [2,3,7,50], associated with a quantum critical point in the doping-temperature phase diagram of cuprates [11,12]. The static order then develops out of this sea of excitations once their characteristic timescale becomes long enough.

Within a Landau free energy paradigm [51,52], the charge excitations inherit the stripe symmetry, below a characteristic energy scale ω_c linked to their effective mass. This characteristic energy ω_c in turn scales with an energetic ground-state distance to a quantum critical point. That is, $\omega_c \to 0$ for $x \to x_c$ where x_c is the critical doping for stripe order. Excitations above the characteristic scale ω_c are expected to display critical behavior: the emergent scale invariance makes them the same on both sides of the quantum critical point [53]. We therefore assign the LE mode with the abovementioned critical excitations, in agreement with recent suggestions

[6–8]. The observed isotropy of the quantum fluctuations suggests that the critical energy ω_c lies well below our current experimental resolution (∼20 meV). As such, our experiments suggest proximity to a critical point and the critical nature of the observed fluctuations. This indirect demonstration is important, as direct scaling behavior is typically smeared by doping-induced disorder. Future improvements in RIXS instrumentation may allow improved resolution that would allow access to the noncritical excitations of the stripe-ordered state.

In conclusion, our results demonstrate an electronic nature of charge order beyond electron-phonon coupling. Given that both the phonon anomalies and the sea of dynamical fluctuations both are insensitive to strain, but lies in the same energy range and react similarly to temperature, we postulate that the former finds its root cause in the latter. The symmetry of these excitations is decoupled from the static stripe order, suggesting closeness to a quantum critical point.

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Data availability. The data that support the findings of this article are openly available [57], embargo periods may apply.

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