SUPERCONDUCTIVITY

Uniaxial pressure control of competing orders in a high-temperature superconductor

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Cuprates exhibit antiferromagnetic, charge density wave (CDW), and high-temperature superconducting ground states that can be tuned by means of doping and external magnetic fields. However, disorder generated by these tuning methods complicates the interpretation of such experiments. Here, we report a high-resolution inelastic x-ray scattering study of the high-temperature superconductor YBa2Cu3O6.67 under uniaxial stress, and we show that a three-dimensional long-range-ordered CDW state can be induced through pressure along the a axis, in the absence of magnetic fields. A pronounced softening of an optical phonon mode is associated with the CDW transition. The amplitude of the CDW is suppressed below the superconducting transition temperature, indicating competition with superconductivity. The results provide insights into the normal-state properties of cuprates and illustrate the potential of uniaxial-pressure control of competing orders in quantum materials.

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Moderately doped high-temperature superconductors show an ubiquitous tendency toward charge order (1). Manifestations of charge ordering include striped order—an incommensurate modulation of both charge and spin that suppresses superconductivity in lanthanum-based cuprates (2, 3)—and a biaxial charge density wave (CDW) with quasi-two-dimensional (2D) short-range order in the CuO2 planes of all other cuprate families (4–12). The origin of CDW order and its relationship with superconductivity are widely debated issues. It is well-established that static CDW order and superconductivity are incompatible; however, it is not clear whether the two orders are best described as mutually incommensurate (7, 8), as different manifestations of the same pairing interaction (33), or as different aspects of a composite order parameter (14, 15). These issues have direct and important implications for the mechanism of high-temperature superconductivity (HTSC).

The YBa2Cu3O6±δ family has been particularly well studied because doping-induced structural disorder is less severe than in other families. In this compound, the competition between superconductivity and CDW order is evidenced by the depression of the superconducting transition temperature (Tc) (7, 8) and by its enhancement in magnetic fields that weaken superconductivity (8, 9). Nuclear magnetic resonance (NMR) and x-ray studies further showed that in fields larger than ~15 T, a 3D long-range ordered uniaxial CDW (16–19) is induced. This 3D order is distinct from the 2D one, although they coexist and are related to each other; for example, they have the same in-plane incommensurability. The 3D order has an identifiable thermodynamic transition (20–22), whereas the 2D CDW onsets gradually (23, 24), and it is not clear whether it constitutes an alternative ground state or is, for example, a vestige of the 3D order that is weakened by the interaction with superconductivity.

We used inelastic x-ray scattering (IXS) on a sample of YBCO6.67 (Tc = 65K, doping p = 0.12) to show that uniaxial pressure along the crystallographic a axis can induce long-range 3D CDW order, in the absence of a magnetic field.

We first specify why we chose to work with uniaxial pressures. In the doping range of interest (0.08 < p < 0.15), hydrostatic pressure yields, through self-doping and other effects, an increase of Tc (25). A recent study of the biaxial charge modulation further revealed its rapid suppression under hydrostatic pressure (26). However, it has long been established that, at least in the limit of low pressures, the effect of hydrostatic pressure on Tc reflects a net sum of larger effects of uniaxial pressures that almost cancel each other out (27, 28). In particular, close to p = ~0.12, where the charge modulation is maximized, uniaxial pressure applied along the a axis suppresses Tc (29, 30).

In this study, we pressurized the sample using a piezoelectric-based apparatus similar to that used in recent studies of ruthenium oxides (31–33) but modified to allow x-ray transmission through the apparatus and the sample. For maximum scattering intensity, the thickness of the sample should match the absorption length at the working wavelength (λ = 0.6968 Å for this experiment), which is ~40 μm. However, to withstand strong compression without buckling, the length-to-thickness ratio of the sample cannot be too large (31), and a 40-μm thickness implies a length that is inconveniently short for reliable mounting. Therefore, we prepared a thicker needle from a YBCO6.67 single crystal then used a plasma focused ion beam (PFIB) to thin down to 40-μm thickness the central portion over a length of ~200 μm (Fig. 1). The x-ray beam, with a spot size of 50 × 40 μm2, was considerably smaller than the thinned central portion of the sample, so highly uniform strain is expected in the probed volume. All the strain values in this paper were calculated by using capacitive displacement sensor built into the pressure cell, the changes in the c axis lattice parameter measured from the (0 0 6) Bragg peak and the changes in Tc (30). The highest compression we reached was εc = ~1.0%, where ac susceptibility measurements showed that Tc decreases (at a rate increasing with strain) to 48 ± 2 K (30).

The data on the 2D biaxial CDW are shown in Fig. 2. The scattering intensity of the biaxial CDW peaks at the reduced momentum transfer qCDW = (h, k, l) = (0, 0.31, 0.5), in reciprocal lattice units (r.l.u.) of the orthorhombic crystal structure (7, 8). By contrast, the 3D CDW peaks at qCDW = (h, l, l) = (0, 0.31, 0.65) and qCDW = (0, 0.31, l), around which measurements were carried out and where the structure factor of the CDWs is maximum (34, 35). The color maps in Fig. 2, A and B, show the raw IXS intensity at εc = 0% (Fig. 2A) and at ~1.0% (Fig. 2B). Both data sets were measured below Tc (30). Two characteristic features of the CDW are clearly visible: the quasi-elastic “central” peak at qCDW and the superconductivity-induced Kohn anomaly in the phonon spectra. In unstressed conditions, the latter consists of a ~15% softening of the low-lying acoustical phonon (at ~8 meV) at qCDW (35). Comparing Fig. 2, A and B, it can be seen that the quasi-elastic peak is enhanced by the
applied pressure. In Fig. 2C, it is seen that this enhancement occurs smoothly. At the highest strain, the integrated intensity of the peak is close to two times larger than that of the unstrained sample (Fig. 2D). Its half-width-at-half-maximum (HWHM) σ, which is inversely proportional to the modulation correlation length \( \xi = \frac{1}{2\pi \sigma} \), decreases modestly under pressure (Fig. 2D). We did not resolve any shift of \( Q_{2D} \) with \( \epsilon_{x x} \). Last, a comparison of Fig. 2, A and B, reveals an increase of the low-energy spectral weight under pressure. Its phenomenology and relation to the Kohn anomaly will be discussed below.

We observed a much more notable response to uniaxial pressure in the scattering pattern at \( Q_{3D} \). We first looked at the strain dependence of the elastic peak intensity along the \( Q = (0, 0.315, L) \) line at \( T = 50 \text{ K} \) (Fig. 3A). At \( \epsilon_{x x} \approx -0.8\% \), a small, narrow peak appears at \( Q_{3D} \). When the compression is further increased, to \( \epsilon_{x x} \approx -1.0\% \), this peak becomes much more intense. It appears on top of the broad profile centered around \( I = 0.5 \) (\( L = 6.5 \)) that arises from the 2D CDW. The profile of the 3D peak along \( K \) is shown on Fig. 3B. A weak 3D peak is visible at compressions as low as \( \epsilon_{x x} \approx -0.5\% \); however, the increase in intensity from 0.8 to 1.0% compression dwarfs the evolution at lower compressions.

Fig. 1. Strain device. (A) Photograph of the piezoelectric device. (B) Sectional cut of the device and picture of the PFIB-thinned sample used for this experiment. (C) Unit cell of YBa$_2$Cu$_3$O$_{6+x}$ (here with \( x = 1 \)). Strain is applied perpendicular to the CuO chains.

Fig. 2. Strain dependence of the IXS spectra around \( Q_{2D} \). (A) IXS intensity versus total momentum transfer for the unstrained sample at \( T < T_c \).

The square root of the intensity has been plotted to enhance the contrast between the phonon and the elastic line. The calculated dispersion of the low-lying acoustical mode is plotted as a dashed line. The solid line is a guide to the eye to the observed dispersion of this mode (fitted values are represented by the dots). (B) Same as in (A), but for \( \epsilon_{x x} \approx -1.0\% \). The stars indicate the energy of a soft optical phonon. (C) Strain dependence of the quasi-elastic line intensity across \( Q_{2D} \) along the \( (0, K, 6.5) \) direction. (D) Strain dependence of the HWHM and of the integrated intensity (normalized to the unstrained value) of the quasi-elastic line at \( Q_{2D} \) integrated along \( K \) and normalized to the unstrained value.
The evolution of the integrated intensity, and the HWHM along $K$, are shown in Fig. 3C. At $\varepsilon_{xx} \sim -1.0\%$, the HWHM is $\sigma_{xx} \sim 0.002$ r.l.u. It is resolution-limited along $L$ ($\sigma_{rr} \sim 0.02$ r.l.u.). These correspond to respective (lower bounds for the) correlation lengths of $\xi_b = \frac{1}{\pi \nu_b} \sim 80 \sim 310$ Å and $\xi_c = \frac{1}{\pi \nu_c} \sim 8c \sim 94$ Å (the limitations of the scattering geometry did not permit investigation of $\xi_c$). These correlation lengths are larger than the values reported under a field of 26 T. The correlation lengths of the 2D CDW at 1.0% compression are $\xi^{2D} \sim 36b \sim 65$ Å and $\xi^{3D} \sim c \sim 12$ Å. To estimate the correlation volume $\Xi = \xi_b \times \xi_c \times \xi_{xx}$, we estimate $\xi_{xx} \sim \xi_b$ for both the 2D and 3D CDWs. For the 3D order, we find $\Xi \sim 51,000$ unit cells under uniaxial pressure for $T - T_c$ exceeding by more than two orders of magnitude that of the 2D CDW at ambient conditions ($\Xi \sim 250$ unit cells).

The temperature evolution of the 3D CDW at $\varepsilon_{xx} \sim -1.0\%$ is shown in Fig. 3, D to F. The peak is very strong at 60 K and weaker but still visible at 70 K. That is higher than the onset temperature of the 3D order observed under high field and higher than $T_c$ of unstressed sample. On the low-temperature side, strong competition with superconductivity is apparent. At $T = 41$ K (below $T_c$), the peak at $Q_{3D}$ has already lost $\sim$90% of the integrated intensity recorded at 50 K and can hardly be distinguished from the background of the 2D order at lower temperatures. This is a much more rapid suppression than that seen for the 2D CDW ($T_c$).

To gain further insight on the relationship between the 2D and 3D orders, we investigated the pressure and temperature dependence of the phonon modes in the inelastic part of the spectra. In Fig. 4A, we show the phonon spectra in the absence of applied pressure at $T = 50$ K, along the $(0, 0, 1)$ direction. Well away from $Q_{3D}$, three peaks are visible. These are well reproduced in ab initio lattice dynamics calculations (30) and correspond respectively to an acoustic phonon mode of the $\Delta'$, irreducible representation (36) at $\sim$8 meV, two optical modes ($\Delta'$ and $\Delta''$) at $\sim$11 meV (which are not distinguishable in the measurement), and another $\Delta'$ optical mode at $\sim$15 meV. It can be seen that in the absence of strain, the acoustic-mode softening associated with the 2D CDW (34), and shown at $Q_{2D}$ in Fig. 2A, extends along $L$ and is visible at $Q_{3D}$.

In Fig. 4B, we show the same spectra but with $\varepsilon_{xx} \sim -10\%$ and $T = 41$ K (to stay below $T_c$). Away from $Q_{3D}$, the spectra are essentially unchanged. Near $Q_{3D}$, on the other hand, a very strong phonon softening is observed, albeit not of the acoustic mode, which now disperses exactly as predicted by the ab initio calculations as $K$ is swept through $Q_{3D}$. In other words, the Kohn anomaly seen in Fig. 4A, at $T = 50$ K and $\varepsilon_{xx} \sim 0\%$, is no longer present under $\varepsilon_{xx} \sim -1.0\%$. This suppression of the acoustical Kohn anomaly can also be seen in Fig. 4C, where we show the $L$-dependence of the phonon spectra from $Q_{3D}$ to $Q_{3D}$ for both $\varepsilon_{xx} \sim 0\%$ and $\varepsilon_{xx} \sim -1.0\%$. Without pressure, the acoustic phonon is soft along the entire $L$-line, which is in agreement with the data in Fig. 4A. At $\varepsilon_{xx} \sim -1.0\%$, we can follow the hardening of the acoustical mode—the disappearance of the Kohn anomaly—as we traverse from $Q_{2D}$ to $Q_{3D}$. The mode that softens approaching $Q_{3D}$ is a distinct feature, which we therefore identify as one of the optical modes. The temperature dependence of the phonon modes at $Q_{3D}$ is shown in Fig. 4D and E. At 70 K, the optical phonon is already very soft, which indicates that, unlike the acoustical Kohn anomaly, the optical mode softening...
Further work is required to determine which of these optical modes is driven soft under uniaxial pressure, to determine whether the softening is complete and to understand the mechanism yielding the disappearance of the acoustical phonon Kohn anomaly at $Q_{3D}$. The absence of phonon softening in single-electron calculations indicates that electronic correlations need to be included in any theoretical treatment of the phonon softening and CDW formation (45).

From a theoretical point of view, unidirectional CDW are unstable against disorder, and a vestigial nematic state is expected instead. It has previously been discussed how short-range biaxial modulation of the charge density might emerge from an intrinsic unidirectional CDWs instability in the presence of quenched disorder (23, 46, 47), which locally reorients small uniaxial domains. Inhomogeneous distribution of the disorder strength has been invoked (18) to explain why the 2D order appears to strengthen under large magnetic fields even as unidirectional 3D order appears. Although one might expect the 3D order to grow at the expense of the 2D one, in this model this process occurs on top of a general strengthening of CDW order as superconductivity is suppressed by the magnetic field, and inhomogeneity in the strength of disorder allows spatially separated, coexisting domains of 2D and 3D order.

Similarly, our data indicate that strain-tuning efficiently strengthens the CDW and supports the formation of the 3D order, likely primarily in those regions where the pinning strength is the weakest. It will be interesting to see in the future whether the strength of the 2D order does eventually decrease as strain is further increased. More generally, further theoretical work is required to understand the strain-induced strengthening of the CDW, which cannot be solely attributed to the competition with superconductivity because the 3D CDW peak can already be induced above the nominal $T_c$ of the sample.

Uniaxial stress will allow the relationship between the superconductivity and CDW to be investigated with high precision in future experiments. A magnetic field suppresses type II superconductivity inhomogeneously because of the presence of vortices, resulting in broad transitions. By contrast, the homogeneous tuning provided by stress could, for example, allow use of thermodynamic probes to determine whether CDW and superconductivity can coexist microscopically. Our piezoelectric-based apparatus constitutes a versatile tool that can be implemented in a large variety of experimental setups—in particular, at synchrotron facilities—thus opening
perspectives for the study of correlated-electron materials.

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Competing interests: C.W.H. has 31% ownership of Razorbill Instruments, which has commercialized apparatus on which that used in this work are based.

Data and materials availability: Raw data and simulation codes used for all figures in this paper and the supplementary materials are available at (48).

SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
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