Tuning from failed superconductor to failed insulator with magnetic field

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INTRODUCTION

A variety of electronic orders have been proposed and/or observed to exist within the generic phase diagram of copper oxide high-temperature superconductors (1), and most are commonly viewed as competing with the superconducting order. In the case of charge stripe order (2), however, it has been argued that electron pairing and superconducting order can actually be intertwined with the charge modulations (3). Moreover, increasingly powerful numerical calculations indicate that charge stripes are a natural consequence of doping holes into a correlated antiferromagnetic insulator (4, 5) and can exhibit superconducting correlations (6, 7). While charge stripe order is readily observed within the CuO2 planes of compounds such as La2-xBaxCuO4 (LBCO) (8), experimentally establishing a positive connection with superconductivity has been challenging. One theoretical expectation for a superconductor based on stripes is that the magnitude of the energy gap associated with pair correlations within charge stripes should be much greater than the energy associated with coherent coupling between the stripes (9), and here we test the implications of this picture. We use a transverse magnetic field first to decouple the superconducting planes in LBCO with x = 0.125 and then to destroy the superconducting order within the planes. We discover an ultraquantum metal (UQМ) phase that we argue is not easily explained by conventional fermionic quasiparticles, in sharp contrast to the high-field behavior in YBa2Cu3O6+δ, where a similar hole concentration (10). We conclude that it provides circumstantial evidence that charge stripes can exhibit robust pairing correlations.

What is already known about LBCO? The spin and charge stripe orders have been studied previously by neutron and x-ray diffraction techniques in magnetic fields perpendicular to the CuO2 planes up to approximately 10 T. For hole concentrations close to x = 0.125, the field enhances the stripe order parameters (11), while right at x = 0.125, it simply increases the charge stripe correlation length (12) [field-induced charge order has been reported in YBa2Cu3O6+δ in fields up to 28 T (13)]. While the bulk Tc is suppressed to ~5 K in zero field for x = 0.125, a substantial decrease in the ab-plane resistivity already occurs below ~40 K, with additional evidence for a transition to two-dimensional (2D) superconducting order near 16 K (14). One would expect Josephson coupling between neighboring layers to induce 3D superconducting order as soon as 2D superconductivity develops; the apparent frustration of the interlayer Josephson coupling has been explained in terms of a proposed pair-density wave (PDW) state, which involves a strong pairing amplitude on the 1D charge stripes but with opposite signs of the pair wave function on neighboring stripes (15, 16). In LBCO with x = 0.095, where the 3D superconductivity is more robust, a magnetic field perpendicular to the CuO2 planes causes a decoupling into 2D superconducting layers (17).

RESULTS

Following the first successful growth of a La2-xBaxCuO4 single crystal with x ~ 1/8 and observation of stripe order by neutron diffraction (18), considerable time was devoted to growing a series of large crystals with 0.095 ≤ x ≤ 0.155 in steps of Δx = 0.02 using infrared image furnaces and the traveling solvent floating-zone technique (19). X-ray and neutron diffraction studies have demonstrated that the structural transitions and lattice parameters are quite sensitive to x, providing valuable and reliable measures of stoichiometry (8). The crystals of x = 0.125 were originally grown for a neutron-scattering study of the spin dynamics (20) and have since been characterized by transport (14, 21) and optical conductivity (22). The crystals used in the present study, described further in Materials and Methods, were from the same growth.

A partial summary of our observations is presented in Fig. 1A as a color contour map of electrical resistivity as a function of temperature and magnetic field. The electrical resistivity is presented throughout this work in terms of the resistance per CuO2 sheet (sheet resistance), R = ρab/d, where d = 6.6 Å is the layer separation, with the magnitude in units of the quantum of resistance for electron pairs, RQ = h/(4e2) = 6.45 kilohms, where h is Planck’s constant and e is the electron charge. An increase of R through RQ is associated with the localization of electron pairs, as observed in the carrier-density–tuned superconductor-insulator transition for La2-xSrxCuO4 thin films (23). At low temperatures with increasing magnetic field H, we observe a progression from 3D superconducting order, through a reentrant 2D superconducting order, to an anomalous high-field metallic state with a sheet resistance saturated at Rs ≈ 2RQ. We denote this unanticipated state as a UQМ: It is a metal because the resistance appears not to change as the temperature is reduced toward...
Fig. 1. Phase diagram of La$_2$-Ba$_2$CuO$_4$ with $x = 0.125$ in terms of sheet resistance $R_s$ and Hall coefficient $R_H$.

(A) Interpolated color contour plot of the sheet resistance $R_s$ as a function of temperature and magnetic field. Black vertical marks indicate measurement temperatures. The regimes of 3D and 2D superconductivity (SC) with zero electrical resistance are labeled; the UQM phase occurs at fields above the dotted line. Characteristic fields $H_{3D}$, $H_{2D}$, and $H_{UQM}$ (defined in the text) are overplotted as solid, dashed, and dotted white lines, respectively (these results are for sample A1; for an analogous plot for sample A2, see fig. S1). (B) Hall coefficient as a function of temperature, with error bars obtained by averaging over the entire field range (0 to 35 T; see fig. S2) at each temperature. $R_H$ is effectively zero below 15 K, as expected for a superconductor, and it rises to the normal-state magnitude around $\sim 40$ K. The upper dashed line indicates the magnitude of $R_H$ that would be expected in a one-band system with a nominal hole density of 0.125. Results are from sample A, as described in the text.

Before proceeding to the high-field results, we first consider the temperature dependence of $R_s$ in zero field. It is known from previous work that the anisotropy between the $c$-axis resistivity, $\rho_c$, and the in-plane resistivity, $\rho_{ab}$, is $\sim 10^4$ near 40 K and rises to $\sim 10^5$ below the onset of 2D superconducting correlations (14). This large anisotropy makes the measurement of $\rho_{ab}$ quite sensitive to sample imperfections. The inset of Fig. 2 shows the sample configuration; the crystal is directly injected into the CuO$_2$ planes, and the voltage probes contact the edges of those planes. If there is a slight misorientation of the crystal such that the $c$ axis is not precisely perpendicular to the current direction, then a small fraction of the current path will be along the $c$ axis, making the measurement sensitive to $\rho_c$. With that preface, consider the measurements of $R_s$ shown in Fig. 2 for two samples, A and B, with results from voltage contacts on both sides of A, labeled A1 and A2. The responses observed for A1 and A2 are consistent with previous work (14): a slight jump in $R_s$ and change in slope at 56 K, corresponding to a well-known structural transition and the coincident charge stripe ordering (8, 18), and a large decrease below $\sim 36$ K indicating the mean-field transition to 2D superconductivity. In contrast, sample B shows a distinct behavior, with a larger magnitude of $R_s$ at high temperature, a significant enhancement of resistance on cooling, and a peak in resistivity at 29 K. Such behavior, which resembles that of $\rho_c$ (14) and has been reproduced in other samples, indicates a contribution from $\rho_c$ consistent with a misorientation of $\sim 1.5^\circ$ (see the Supplementary Materials for further details). The differences between samples A and B, while unintended, provide valuable information regarding the field-dependent behavior.

Next, we consider the magnetic field dependence of $R_s$ measured at various fixed temperatures. Data for A1, A2, and B up to 35 T are shown in Fig. 3 (A to C). At low temperature, we find that the data collapse with a simple ad hoc scaling determined by characteristic fields $H_{3D}$ and $H_{2D}$. We define $H_{3D}$ to be the field at which we detect an initial onset of finite resistivity, indicating loss of 3D superconducting order. $H_{2D}$ corresponds to the midpoint of the reentrant 2D superconductivity, as discussed below (in practice, it was determined by the corresponding local maximum in $R_s$ for A2). Figure 3 (D to F) shows $R_s$ plotted versus $(H - H_{2D})/\Delta H$, where $\Delta H = H_{3D} - H_{2D}$. Here, we see that all three datasets are identical up to $H_{3D}$. For A1, $R_s$ then returns to zero for a finite range of field. In the same region, B shows a rise to greater than $4R_Q$, consistent with the insulating behavior in the $\rho_c$ contribution due to putative PDW order and 2D superconductivity (16, 17).

The unexpected behavior occurs for A2, where $R_s$ virtually plateaus at $R_Q$ for $H > H_{3D}$; however, it is also a consistent response for a 2D superconductor in a strong magnetic field. The field penetrates the sample as magnetic flux quanta that are screened by vortices of superconducting current. If the vortices are pinned by a combination of quenched disorder and electromagnetic interactions between layers (17, 24), then $R_s$ will be zero (as observed for A1); on the other hand, if the vortices are not pinned in one part of the sample, then dissipation will be observed (as for A2). In a model for the field-driven superconductor-insulator transition in disordered 2D superconductors, a boson-vortex duality has been proposed, which predicts that $R_s \approx R_Q$ when the vortices can flow freely, provided that $R_{4\pi} = 0$ (25, 26) as in the present case.

The quantized vortices are only defined when a locally coherent supercurrent is present. A 2D superconductor can only be ordered in the limit of zero current (27); with a finite current, we weaken the superconducting correlations and introduce a finite resistance.
leads to a marked reduction in the magnitude of the $\rho_c$ contribution to $R_s$ for sample B. On the basis of the $\rho_c$ contribution to B, we estimate that $\rho_c/\rho_{ab}$ drops to $<10^5$ at $T = 3$ K (details in the Supplementary Materials), compared to $10^3$ at zero field and $T = 40$ K. We also note that, for sample B, $R_s$ depends on the measurement current for $H > H_{2D}$, as shown in fig. S5.

**DISCUSSION**

What do these results tell us about the UQM phase? The standard theoretical description of a metal is in terms of fermionic quasiparticles. To estimate the transport properties due to fermionic excitations at low temperature after suppression of superconductivity, we can extrapolate from the zero-field normal-state response previously reported for $T > 40$ K (14, 21). In particular, $\rho_c$ has a large, nonmetallic magnitude that grows with cooling and extrapolates to insulating behavior. If we assume that quasiparticles could explain our observations for A1 and A2, then we would expect to have $\rho_c/\rho_{ab} > 10^4$, inconsistent with our results for B. In-plane quasiparticles should also yield a finite value of $R_{11}$, as observed in YBa$_2$Cu$_3$O$_{6+x}$ at high field (29), which is, again, contrary to our results for LBCO.

The Hall constant is zero in the superconducting state because of the particle-hole symmetry of the Bogoliubov quasiparticles that make up the Cooper pairs. The observation that $R_{11}$ remains negligible at high field suggests that such symmetry may survive. Such behavior (zero Hall resistivity and finite longitudinal resistivity) has been observed previously in disordered 2D superconductors (30), and hence, our observations are not entirely unique. Nevertheless, the longitudinal resistivity in that case is much smaller than in the normal state, corresponding to the recently discussed “anomalous metal regime” (31). Our situation is different, with an in-plane metallic resistance much larger than in the normal state at, for example, $T = 50$ K, and hence, it requires a distinct interpretation [an alternative fermionic picture, to be discussed elsewhere, involves precisely compensating hole and electron pockets of quasiparticles; however, angle-resolved photoemission studies provide no evidence of such pockets in zero field (32, 33)].

It was noted quite some time ago that superconducting order in underdoped cuprates may be limited by phase coherence and not by electron pairing (34). In the present case, we have destroyed the superconducting phase coherence with the magnetic field, but electron pairs may survive. It has also been proposed, in a model based on coupled charge stripes, that strong electron-pair correlations might live within the charge stripes, with pair hopping between stripes providing the phase coherence (9, 35). The large magnitude of $R_s$ in the UQM state is consistent with localization of surviving electron pairs within charge stripes; nevertheless, incoherent tunneling of bosonic pairs between stripes could yield finite conductivity (failed insulator). Regarding the $c$-axis response inferred from sample B, the large magnitude of $R_s$ in the 2D regime is consistent with PDW order, which results in cancellation of interlayer pair tunneling (failed 3D superconductor) (16, 36); with the loss of that order, even limited tunneling of pairs between layers would reduce $\rho_c/\rho_{ab}$ in the UQM phase, as appears to happen. The nonlinear transport in sample B (see fig. S5) is also suggestive of pairing correlations that can be destroyed by high current density.

The experimental results leave us with the implication that the UQM phase is a Bose metal. There have been various theoretical proposals of a Bose metal state, but applications to real materials have so far been confounded by coexisting fermionic quasiparticles (31). In LBCO
at high field, quasiparticles do not appear to be relevant. While further experimental and theoretical work is needed, we believe that our circumstantial evidence supports the perspective that charge stripes in cuprates, including dynamic stripes, are good for electron pairing (5–7, 35, 37, 38), even if stripe ordering may limit the degree of superconducting phase coherence.

MATERIALS AND METHODS

Materials and sample preparation

Several samples were oriented with Laue x-ray diffraction, cut from the large crystal with a wire saw, and polished with diamond sandpaper down to 0.3-μm roughness, achieving an almost perfect rectangular shape for direct \( ab \)-plane charge transport measurements. No defects or cracks were visible on the surfaces of the samples. PELCO and Dupont 6838 silver paste and gold wires were used to contact samples in a Hall bar geometry, and samples were annealed in air flow at 450°C to minimize the contact resistance. The current contacts were made by sweeping the field, with the sweep rate varying from 1 T/min at low temperatures to 3 T/min at high temperatures.

Charge transport measurements

A standard four-probe configuration was used for charge transport measurements. Zero-field resistivity was measured using a helium cryostat, Signal Recovery 7265 lock-in amplifiers, Keithley 6220 current sources, and Keithley 2182A nanovolt meters.

The measurements as a function of magnetic field were performed with the 35-T–resistive magnet and a \(^3\)He cryostat at the DC Field Facility, National High Magnetic Field Laboratory. A straight probe was used to measure two samples at the same time. Signal Recovery 7265 and Stanford Research 865A lock-in amplifiers, Keithley 2182A nanovolt meters, and Lake Shore 372 AC resistance bridges were used to measure magnetoresistivity and Hall coefficient. Magnetoresistivity measurements were done with low-frequency ac current at 31.6 \( \mu \)A for 0.35 K \( \leq T \leq 5 \) K and with currents of 31.6, 100, and 316 \( \mu \)A for 5 K \( \leq T \leq 50 \) K. Distinct frequencies were used (13 Hz for sample A and 16 Hz for sample B) to avoid cross-talk. Hall measurements were done with currents of 31.6 \( \mu \)A for 5 K \( \leq T \leq 25 \) K and 100 \( \mu \)A for 25 K \( \geq T \) to minimize heating effects at low temperatures and the noise level at high temperatures. All measurements were performed by fixing the temperature and sweeping the field, with the sweep rate varying from 1 T/min at low temperatures to 3 T/min at high temperatures.

ac \( dV/dI \) measurements of nonlinear resistivity were performed with Signal Recovery 7265 and Stanford Research 865A lock-in amplifiers, Keithley 2182A nanovolt meters, and DL Instruments 1211...
current preamplifiers, with an instrumentation setup similar to that used in a previous work (28). ac voltage across the sample was measured by applying a dc bias (10 to 200 μA; provided by Keithly 6221 current sources) and a small ac excitation (~10 μA) at 13 and 16 Hz. Joule heating was negligible at high temperatures (Fig. 4) and relatively small compared to nonlinear resistivity signals at low temperatures. dV/dl measurements were performed at 0.4 K ≤ T ≤ 5 K.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/6/eaav7686/DC1.

Estimation of c-axis misorientation for sample B. Fig. S1. Temperature-magnetic field phase diagram for A2. Fig. S2. Field dependence of Hall voltage at various temperatures. Fig. S3. Sheet resistance at constant magnetic field. Fig. S4. ac nonlinear resistivity at H_{B,H}. Various temperatures. Fig. S5. Additional results for sample B. Fig. S6. Comparison between A1 and previous results for a similar sample.

REFERENCES AND NOTES

1. B. Reimer, S. A. Kivelson, M. R. Norman, S. Uchida, J. Zaanen, From quantum matter to high-temperature superconductivity in copper oxides. Nature 518, 179–186 (2015).

2. J. Orenstein, A. J.Millis, Advances in the physics of high-temperature superconductivity. Science 288, 468–474 (2000).

3. E. Fradkin, S. A. Kivelson, J. M. Tranquada, Colloquium: Theory of intertwined orders in high temperature superconductors. Rev. Mod. Phys. 79, 457–482 (2007).

4. B-X. Zheng, C-M. Chung, P. Corboz, G. Ehlers, M.-P. Qin, R. M. Noack, H. Shi, S. R. White, S. Zhang, G.-K.-L. Chan, Stripe order in the underdoped region of the two-dimensional Hubbard model. Science 358, 1155–1160 (2017).

5. E. W. Huang, C. B. Mendel, L. Su, S. Johnston, H.-C. Jiang, B. Moritz, T. P. Devereaux, Numerical evidence of fluctuating stripes in the normal state of high-Tc cuprate superconductors. Science 358, 1161–1164 (2017).

6. S. R. White, D. J. Scalapino, Pairing on striped J–F–lattices. Phys. Rev. B 79, 220504 (2009).

7. P. Corboz, T. M. Rice, M. Troyer, Competing states in the J-T model: Uniform d-wave state versus stripe state. Phys. Rev. Lett. 113, 046402 (2014).

8. M. Hütter, M. v. Zimmermann, G. D. Gu, Z. J. Xu, J. S. Wen, G. Y. Xu, H. J. Kang, A. Zhleudev, J. M. Tranquada, Stripe order in superconducting La2−xSrxCuO4 at 0.05 ≤ x ≤ 0.155. Phys. Rev. B 83, 104506 (2011).

9. E. W. Carlson, D. Orgad, S. A. Kivelson, V. J. Emery, Dimensional crossover in quasi-one-dimensional and high-Tc superconductors. Phys. Rev. B 62, 3422–3437 (2000).

10. S. E. Sebastian, C. Proust, Quantum oscillations in hole-doped cuprates. Annu. Rev. Condens. Matter Phys. 6, 411–430 (2015).

11. M. Hütter, M. v. Zimmermann, Z. J. Xu, J. S. Wen, G. D. Gu, J. Xu, Tranquada, Enhanced charge stripe order of superconducting La2−xSrxCuO4 in a magnetic field. Phys. Rev. B 87, 014501 (2013).

12. J. Kim, A. Kagedan, G. D. Gu, C. S. Nelson, Y.-J. Kim, Magnetic field dependence of charge stripe order in La2−xSrxCuO4 (x = 0). Phys. Rev. B 77, 180513 (2008).

13. S. Gerber, H. Jang, H. Nojiri, S. Matsuzawa, H. Yasumura, D. A. Bonn, W. N. Hardy, H. J. Kang, A. Zhleudev, J. M. Tranquada, Stripe order in superconducting La2−xSrxCuO4 at the pair quantum resistance. Nature 472, 458–460 (2011).

14. K. S. Raman, V. Oganesyan, S. L. Sondhi, Biot-Savart correlations in layered superconductors. Phys. Rev. B 79, 174528 (2009).

15. M. P. A. Fisher, Quantum phase transitions in disordered two-dimensional superconductors. Phys. Rev. Lett. 65, 923–926 (1990).

16. N. P. Breznay, M. A. Steiner, S. A. Kivelson, A. Kapitulnik, Self-duality and a Hall-insulator phase near the superconductor-to-insulator transition in indium-oxide films. Proc. Natl. Acad. Sci. U.S.A. 113, 280–285 (2016).

17. D. S. Fisher, M. P. A. Fisher, D. A. Huse, Thermal fluctuations, quenched disorder, phase transitions, and transport in type-II superconductors. Phys. Rev. B 84, 134501 (2011).

18. Z. Shi, P. G. Baity, T. Sasagawa, D. Popović, Unveiling the phase diagram of a striped cuprate at high magnetic fields: Hidden order of Cooper pairs. arXiv:1801.06903 [cond-mat.supr-con] (21 January 2018).

19. D. LeBeuf, N. Doiron-Leyraud, J. Levalois, R. Daou, J-B. Bonnemaison, N. E. Hussey, L. Balicas, B. J. Ramshaw, R. Liang, D. A. Bonn, W. N. Hardy, S. Adachi, C. Proust, L. Tailllefer, Electron pockets in the fermi surface of hole-doped high-Tc superconductors. Nature 450, 533–536 (2007).

20. N. P. Breznay, A. Kapitulnik, Particle-hole symmetry reveals failed superconductivity in the metallic phase of two-dimensional superconducting films. Sci. Adv. 3, e1700612 (2017).

21. A. Kapitulnik, S. A. Kivelson, B. Svinkin, Colloquium: Anomalous metals: Failed superconductors. Rev. Mod. Phys. 91, 011102 (2019).

22. T. Valla, A. V. Fedorov, L. Lee, J. C. Davis, G. D. Gu. The ground state of the pseudogap in cuprate superconductors. Science 314, 1914–1916 (2006).

23. R. H. C. K. Tanaka, S.-K. Mo, T. Sasagawa, M. Fujita, T. Adachi, N. Mannella, K. Yamada, Y. Koike, Z. Hussain, Z.-X. Shen, Energy gaps in the failed high-Tc superconductor La2−xSrxCuO4. Nat. Phys. 5, 119–123 (2009).

24. V. J. Emery, S. A. Kivelson, Importance of phase fluctuations in superconductors with superfluid density. Nature 374, 434–437 (1995).

25. V. J. Emery, S. A. Kivelson, O. Zachar, Spin-gap proximity effect mechanism of high-temperature superconductivity. Phys. Rev. B 56, 6120–6147 (1997).

26. S. Rajakumar, J. Okamoto, M. Matthey, F. Vechnev, T. Thampy, G. D. Gu, A. Cavalleri, Probing optically silent superfluid stripes in cuprates. Science 359, 575–579 (2018).

27. J. Zaanen, O. Y. Osman, H. V. Kruis, Z. Nussinov, J. Tworzydło, The geometric order of stripes and Luttinger liquids. Philos. Mag. B 81, 1485–1531 (2001).

28. S. T. Carr, A. M. Tsvelik, Superconductivity and charge-density waves in a quasi-one-dimensional spin-gap system. Phys. Rev. B 65, 195121 (2002).

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