Unconventional quantum vortex matter state hosts quantum oscillations in the underdoped high-temperature cuprate superconductors

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A central question in the underdoped cuprates pertains to the nature of the pseudogap ground state. A conventional metallic ground state of the pseudogap region has been argued to host quantum oscillations upon destruction of the superconducting order parameter by modest magnetic fields. Here, we use low applied magnetic fields and millikelvin temperatures on ultrapure single crystals of underdoped YBa2Cu3O6+x to unearth an unconventional quantum vortex matter ground state characterized by vanishing electrical resistivity, magnetic hysteresis, and nonohmic electrical transport characteristics beyond the highest laboratory-accessible static fields. A model of the pseudogap ground state is now required to explain quantum oscillations that are hosted by the bulk quantum vortex matter state without experiencing sizable additional damping in the presence of a large maximum superconducting gap; possibilities include a pair density wave.

Significance

Modest magnetic fields above ~22 T in the underdoped cuprates were argued to destroy superconductivity and reveal a conventional metallic pseudogap ground state that hosts quantum oscillations. We perform high-magnetic field electrical resistivity and magnetic torque measurements using low temperatures and small applied measurement currents to reveal instead an unconventional quantum vortex matter ground state that persists beyond the highest static magnetic fields. An alternative model is required to explain the coexistence of quantum oscillations with a robust d-wave superconducting gap in the newly uncovered quantum vortex matter ground state.

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In agreement with the high resistive magnetic fields we access in this work (SI Appendix, Fig. S3).

We provide further evidence that argues against an origin of magnetic field-resilient superconductivity in the ground state of underdoped YBa$_2$Cu$_3$O$_{6+x}$ high critical temperature doping inclusions. Superconducting homogeneity is indicated by the narrow superconducting transition in the electrical resistivity as a function of temperature in high magnetic fields (Fig. 1 E and F). Further support for superconducting homogeneity is provided by the sharpness of the transition in the magnetic susceptibility ($\chi$) in very small magnetic fields (SI Appendix, Figs. S6 and S7), from which we infer a minimal volume fraction of any regions of the sample with a value of $T_c$ greater than the mean $T_c$ (defined by the step in $\chi$). The observation of low critical temperatures at high magnetic fields further argues against the inclusions of higher dopings as responsible for the persistence of superconductivity up to high magnetic fields. The systematic doping evolution of the high-field superconducting region, which reaches higher critical temperatures with increasing doping, also supports the intrinsic bulk character of the high magnetic field-resilient superconductivity (Fig. 2).

In order to discern the nature of the high-field superconducting ground state, we perform a study of the voltage-current characteristics, signatures of which are used to characterize regimes of superconducting vortex physics (9). We find a striking and systematic nonohmic voltage-current dependence at high magnetic fields and low temperatures (Fig. 3 A–E). Given that previous measurements in the vortex state were largely confined to low magnetic fields and high temperatures (9), we use a new model of unconventional quantum vortex matter developed to treat the high-magnetic field region to compare with our measurements in the high-magnetic field–low-temperature region of underdoped YBa$_2$Cu$_3$O$_{6+x}$ (10). We find that the measured nonohmic voltage–current dependence can be well captured by a model of quantum vortex matter based on self-organization of vortices in a magnetic field (10, 11). We use the term “quantum vortex matter” to describe the vortex regime in high magnetic fields and low temperatures where quantum fluctuations are expected to be relevant (10), as opposed to the more conventional vortex regime at low magnetic fields and high temperatures (9). We extract a temperature scale (10) ($T_{\text{HFF}}$) associated with the melting of quantum vortex matter into a vortex liquid (Fig. 3F) for various magnetic fields and find that the quantum vortex matter–vortex liquid phase boundary agrees well with the extracted resistive magnetic field $\mu_0H_{\text{r}}$, below which the voltage drops to a vanishingly small value for a range of temperatures (Fig. 2A). Our findings thus unearth a bulk quantum vortex matter ground state that persists up to at least 45 T and evolves to a vortex liquid with increasing temperature as shown in the phase diagrams (Fig. 2 and SI Appendix, Fig. S8; see Fig. 4E). A similar superconducting phase diagram driven by quantum fluctuations has been reported in two-dimensional materials families such as the organic superconductors (12), and may be similarly expected in the strongly...
interacting quasitwo-dimensional cuprate superconductors. Similarly, non-BCS (Bardeen–Cooper–Schrieffer)-like magnetic field-resilient superconductivity with positive curvature of the resistive magnetic field was reported in high-purity single crystals of TlBa2Cu4O8+δ (3). An interplay of superconductivity and a density wave (13) has been further proposed to yield a steep magnetic field–temperature slope of the superconducting phase boundary, as observed in our experiments.

The finite resistivity previously accessed above a modest magnetic field scale $\approx 22$ T in underdoped YBa2Cu3O6+x (1) can be attributed to the use in pulsed magnetic field experiments of applied measurement current densities three orders of magnitude higher than present measurements and even larger eddy current densities (Materials and Methods) at elevated temperatures (SI Appendix, Fig. S9), yielding vortex dissipation. Previous heat capacity measurements were performed at elevated temperatures and do not access low-enough temperatures to capture the unconventional quantum vortex matter regime (SI Appendix, Fig. S9) (14). Features in thermal conductivity previously interpreted as a signature of the upper critical magnetic field in YBa2Cu3O6+x (1) meanwhile differ from signatures of the upper critical magnetic field as observed in other type II superconductors (SI Appendix, Fig. S10), prompting its alternative interpretation as a density wave transition in YBa2Cu3O6+x (5).

The superconducting phase diagram in high-magnetic field–low-temperature space for the underdoped cuprates revealed by our present measurements is shown in Fig. 4E and SI Appendix, Fig. S11. We find the quantum vortex matter region characterized by vanishing electrical resistivity in the $j \to 0$ limit and nonohmic voltage–current characteristics (colored shading) to steeply rise at low temperatures, persisting beyond the highest laboratory-accessible static magnetic fields of 45 T. The newly uncovered high-magnetic field superconducting phase diagram supercedes previous proposals involving a finite electrical resistivity ground state (SI Appendix, Fig. S8 A and B). Previous proposals include a BCS-like type II superconducting phase diagram in which a Meissner superconducting state rapidly enters a conventional metallic region (1, 14) at high magnetic fields via a vortex solid (Shubnikov-phase) region (9) (SI Appendix, Figs. S8A and S12) and the alternative possibility of a vortex liquid ground state at high magnetic fields characterized by finite electrical resistivity (5, 15, 16) (SI Appendix, Fig. SSB).

We gain insight into the character of the quantum vortex matter ground state of the pseudogap by examining the quantum oscillations that are hosted in this region characterized by hysteretic magnetic torque (zero applied measurement current) evidencing vortex pinning (5, 6), vanishing electrical resistivity in the $j \to 0$ limit, and nonohmic electrical transport (Fig. 4A and B). Quantum oscillations in the electrical resistivity also appear in the quantum vortex matter region, upon the application of sufficiently elevated currents for finite resistivity to be induced from vortex dissipation (Fig. 4D). We compare quantum oscillations in the superconducting region of underdoped YBa2Cu3O6+x with those observed in other type II superconductors including NbSi2, V$_3$Si, Nb$_3$Sn, YNi$_2$B$_2$C, LuNi$_2$B$_2$C, UpdAl$_3$, URu$_2$Si$_2$, CeCoIn$_5$, CeRu$_2$, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, MgB$_2$, and others (17–19), for which theories have been developed of quantum oscillations in the mixed state (e.g., refs. 19 and 20). To estimate the extent of superconducting damping of quantum oscillations, we compare the ratio of the Landau-level spacing $\Delta_{\text{osc}}$ to the maximum superconducting gap $\Delta$ (here, $\omega_c = e\mu B/m^*$ is the cyclotron frequency and $m^*$ is the cyclotron effective mass) at magnetic fields where superconducting damping reduces the quantum oscillation amplitude (corrected for the Dingle damping factor) by a factor of two (21). In the case of underdoped YBa2Cu3O6+x, we obtain an upper bound for this ratio at the lowest magnetic field value at which quantum oscillations are observed. A low value of $\Delta_{\text{osc}}/\Delta \lesssim 0.06$ is estimated for underdoped YBa2Cu3O6+x, taking the maximum superconducting gap at zero magnetic field $\Delta \approx 30$ meV from complementary measurements (22) (consistent with the high-magnetic field resilience of superconductivity), $m^* = 1.6$ m$_0$ (m$_0$ is the free electron mass) (23), and $\mu_B H = 20$ T. This ratio in underdoped

![Figure 2](https://i.imgur.com/3e.png)
YBa$_2$Cu$_3$O$_{6+x}$ is an order of magnitude smaller than the ratio $\hbar \omega_c/\Delta \approx 0.5$ for conventional type II superconductors including NbSe$_2$, V$_2$Si, NbSn, YNi$_2$B$_2$C, LuNi$_2$B$_2$C, CeRu$_2$, and MgB$_2$ (details are in SI Appendix, Table S1). Quantum oscillations thus persist in the presence of a large maximum superconducting gap, displaying minimal superconducting damping in the case of underdoped YBa$_2$Cu$_3$O$_{6+x}$, unlike conventional type II superconductors. Similarly low ratios of $\hbar \omega_c/\Delta$ as underdoped YBa$_2$Cu$_3$O$_{6+x}$ are found in unconventional type II superconductors such as URu$_2$Si$_2$ ($\hbar \omega_c/\Delta \approx 0.08$ for $\Delta_y$) and $\approx 0.04$ for $\Delta_y$), $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_{2}$ ($\hbar \omega_c/\Delta \approx 0.05$), and UPd$_4$Al$_3$ ($\hbar \omega_c/\Delta \approx 0.006$ for $\Delta_{ab}$ and $\approx 0.3$ for $\Delta_y$) (details are in SI Appendix, Table S1).

Models of quantum oscillations in the presence of a spatially uniform superconducting gap associate a low ratio of $\hbar \omega_c/\Delta$ in unconventional type II superconductors with an anisotropic d-wave superconducting gap, compared with a higher ratio of $\hbar \omega_c/\Delta$ in the case of conventional type II superconductors characterized by an isotropic superconducting gap (24–28). Inspection of quantum oscillations in the quantum vortex matter state of underdoped YBa$_2$Cu$_3$O$_{6+x}$, however, reveals distinguishing characteristics that are challenging to reconcile with models of spatially uniform superconductivity. First, in models of spatially uniform superconductivity, whether characterized by isotropic superconducting gap over the full Fermi surface or d-wave superconducting gap over the full Fermi surface except at a gapless nodal point, the quantum oscillation amplitude is expected to exhibit a reduced temperature variation at low temperatures due to the vanishing of in-gap states (9, 19–21, 24, 29). In contrast, the quantum oscillation amplitude increases at low temperatures in underdoped YBa$_2$Cu$_3$O$_{6+x}$, consistent with the Lifshitz–Kosevich form (30) even within the quantum vortex matter state, signaling Fermi–Dirac statistics of low-energy excitations within the gap (Fig. 4C). Second, models of spatially uniform superconductivity are expected to yield increased damping both as the system transitions from the “normal” metallic regime in which the superconducting order parameter is destroyed to the vortex liquid regime in which vortices are mobile and as the system further transitions to the quantum vortex matter regime in which vortices are collectively pinned (Fig. 4E).

In our present experiments, we access quantum oscillations as the system transitions from a mobile vortex liquid to a pinned quantum vortex matter state (Fig. 4B and E). In contrast to the expectation from models of spatially uniform superconductivity, no discernible additional damping beyond that in the usual Lifshitz–Kosevich description is observed as the quantum oscillations evolve from the vortex liquid regime to the quantum vortex matter regime (Fig. 4B and C and SI Appendix, Figs. S13 and S14). These properties of the quantum oscillations we observe in the quantum vortex matter regime reveal the coexistence of finite gapless excitations with a large maximum superconducting gap.

The observed coexistence can potentially be explained by nonuniform models of superconductivity that are spatially modulated at a finite wave vector, such as the pair density wave (PDW) recently reported in experiments such as scanning–tunneling microscopy (31–44). Unlike models of spatially uniform superconductivity that are characterized by nodal points (9, 19–21, 24, 29), models of finite wave vector superconductivity result in “nesting” over only a portion of the Fermi surface and consequently yield partially gapped Fermi surface and lines of gapless excitations (31). PDW models predict a nodal–antinodal dichotomy in which the antinodes are gapped by a large maximum superconducting gap, while gapless “Fermi arcs” occur near the nodes (31, 32, 34, 37). The reconstruction of the gapless nodal Fermi arcs in PDW models yields a sharply defined nodal Fermi pocket, providing a possible explanation for our observation of quantum oscillations hosted in a quantum vortex matter ground state, which are largely undamped by the large maximum superconducting gap. Alternatively, a nodal Fermi pocket has been modeled to arise from Fermi surface reconstruction by biaxial charge density wave order (45). Our observations potentially point to quantum oscillations in the quantum vortex matter regime due to the interplay of superconductivity and biaxial charge density wave order.

Any model of quantum oscillations in the unconventional quantum vortex matter ground state of the pseudogap region must also explain features such as the isolated nodal Fermi pocket found by complementary observations of forward–backward quantum oscillations (21), low measured value of linear specific heat capacity in high magnetic fields (46), the high magnetic field saturation of quantities such as the specific heat capacity (1), and the spin susceptibility from the Knight shift (47). An open question pertains to the extent to which vortex physics persists over the broader doping, temperature, and magnetic field range of the pseudogap region of the underdoped cuprate phase diagram (31, 34, 48).
Fig. 4. Quantum oscillations coexisting with the vortex matter phase and the magnetic field–temperature phase diagram for YBa$_2$Cu$_3$O$_{6.4}$. (A) Quantum oscillations coexist with hysteresis in magnetic torque (zero applied measurement current, $\theta = 9^\circ$) from vortex pinning extending up to the irreversibility field $\mu_0H_N$, beyond 45 T, coincident with the vanishing electrical resistivity region that also extends beyond 45 T at $T = 0.04$ K (SI Appendix, Fig. S3). (B) Quantum oscillations in the quantum vortex matter (QVM) regime at the lowest temperature $T = 0.04$ K are seen to be of similar size to quantum oscillations in the finite electrical resistivity vortex liquid (VL) region at elevated temperature $T = 1.0$ K (SI Appendix, Fig. S5). (C) Lifshitz–Kosevich (LK) temperature dependence of the quantum oscillation amplitude at the lowest measured temperatures. Inset shows that the growth of quantum oscillation amplitude continues to the lowest measured temperatures, as brought out by a low-temperature expansion (Materials and Methods). $A(T)$ is the quantum oscillation amplitude at temperature $T$, $A_0$ is the amplitude at the lowest measured temperature, and $X = 2\pi^2k_B^2T/m^*e\mu_0H$ is the temperature damping coefficient in the LK formula (50). (D) Shubnikov–de Haas oscillations after background subtraction in the QVM regime upon applying elevated current densities to induce vortex dissipation. Here, $\rho_{xx} = \rho_{xx}(B) / B$ is an effective resistivity in the nonohmic regime. (E) Superconducting phase diagram in which high magnetic field-resilient QVM ground state is revealed in the present measurements, melting to a VL with elevated temperature.

Materials and Methods

Sample Preparation for Transport Measurements. The electrical transport is measured on pristine detwinned oxygen-ordered single crystals of YBa$_2$Cu$_3$O$_{6.4}$ grown by the flux technique. Samples with typical dimensions of (0.8 to 1.5) $\times$ (0.5 to 1.0) $\times$ (0.03 to 0.08) mm were selected for the electrical transport measurements. Gold pads of standard six-contact geometry were deposited onto the top surface with 160-nm thickness and to the sides with 80-nm thickness using thermal evaporation methods. Top and side views of a typical transport sample are shown in SI Appendix, Fig. S1. Samples with gold pads were annealed at temperatures above 500 °C with flow of high-purity oxygen ($\geq$99.999%) to set the oxygen content $x$ and meanwhile, allow the gold pads to diffuse into the bulk of the crystal. All measurements in this work were performed with current flowing along the $a$ axis, with crystals detwinned under uniaxial stress of 100 MPa at 250 °C. Cu–O chain superstructures were formed in samples under vacuum pressures of below $3 \times 10^{-2}$ mbar. Samples with current contact resistances of $\approx 0.5 \Omega$, made using gold wires attached with DuPont 4929WN, were used for high-field measurements. SI Appendix, Fig. S2 shows the superconducting transitions in the susceptibility for the measured samples, with transition widths similar to previous reports. Hole doping $p$ is inferred from the critical temperature $T_c (49)$, defined as the midpoint of the superconducting transition.

Evidence for Bulk Superconductivity. An important question concerning the observation of superconductivity that persists up to high magnetic fields is whether the superconductivity is of bulk character. Fig. 1 $I$ and $J$ presents measurements of the electrical resistivity vs. temperature for $p = 0.108$, 0.123. The narrow absolute width of the superconducting transition in electrical resistivity at high magnetic fields indicates no significant increase in inhomogeneity of the superconducting state in strong magnetic fields when $T_c$ is suppressed. Furthermore, an inclusion of a small superconducting volume fraction of a higher doping, were it to exist, would manifest in magnetic susceptibility measurements, which is not observed, as shown in SI Appendix, Fig. S3.

The observation of significant vortex pinning in the magnetic torque (i.e., hysteresis in Fig. 4A and SI Appendix, Fig. S4C) accompanying vanishing electrical resistivity in the $\theta \rightarrow 0$ limit below the superconducting transition indicates a bulk superconducting state. No such sharp transition or hysteretic signature in the bulk magnetic torque is expected to occur for surface or filamentary superconducting states (8), which furthermore, typically exhibit small finite electrical resistivity rather than the vanishing electrical resistivity that is observed. The above findings and the systematic nature and sample independence of our results point to the intrinsic, bulk character of the magnetic field-resilient superconducting state in the pseudogap ground state characterized by low critical temperature and low critical current that we find to be persistent beyond the highest-accessible static magnetic fields of 45 T.

Low-Temperature Growth of Quantum Oscillation Amplitude Compared with Lifshitz–Kosevich Expansion. The Fermi–Dirac distribution yields a temperature-dependent quantum oscillation amplitude in the Lifshitz–Kosevich form (50). This low-temperature growth of quantum oscillation amplitude is given by

$$R_T = \frac{X}{\sinh X},$$

with $X = 2\pi^2k_B^2T/m^*e\mu_0H$, where $k_B$ is Boltzmann’s constant, $T$ is temperature, $m^*$ is the quasiparticle effective mass, $e$ is the electron charge, and $\hbar$ is the reduced Planck constant (50). For small $T$, a series expansion of the temperature dependence term yields

$$R_T \approx 1 - \frac{X^4}{6} + O(X^6).$$

For small $T$, therefore, the quantum oscillation amplitude linearly increases with decreasing $X^2$ approaching the $T \rightarrow 0$ limit. The low-temperature growth in quantum oscillation amplitude is captured by the relative change of quantum oscillation amplitude at a finite temperature $A(T)$ with respect to the amplitude at the lowest measured temperature $A_0$, given by

$$\frac{1 - A(T)}{A_0} = \frac{A_0 - A(T)}{A_0} = \frac{X^4}{6}.$$

A plot of $(A_0 - A(T))/A_0$ against $X^2$ would therefore yield a straight line with a gradient equal to 1/6 at low temperatures for low-energy excitations within the gap. In contrast, in the absence of low-energy excitations,
gapped quantum oscillation models would yield a much reduced change in amplitude as a function of $X^2$ at low temperatures well below the gap temperature scale (24). Fig. 4 C, inset shows the growth in quantum oscillation amplitude plotted against $X^2$ with a quasiparticle effective mass $m^* / m = 1.676$ (51). The rapid low-temperature growth of the quantum oscillation amplitude yields a linear slope of 0.20 (2) at low temperatures, in notable contrast to the expectation of little to no growth in the case of gapped quantum oscillations in the low-temperature limit. A full temperature dependence of the quantum oscillation amplitude up to a temperature of 18 K is shown in SI Appendix, Fig. S4.

Data Availability. All data are included in the manuscript and/or SI Appendix.

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