Broken rotational symmetry on the Fermi surface of a high-T_c superconductor

B. J. Ramshaw1,6, N. Harrison1, S. E. Sebastian2, S. Ghannadzadeh3,7, K. A. Modic1,8, D. A. Bonn4, W. N. Hardy4, Ruixing Liang4 and P. A. Goddard5

Broken fourfold rotational (C4) symmetry is observed in the experimental properties of several classes of unconventional superconductors. It has been proposed that this symmetry breaking is important for superconducting pairing in these materials, but in the high-T_c cuprates this broken symmetry has never been observed on the Fermi surface. Here we report a pronounced anisotropy in the angle dependence of the interlayer magnetoresistance of the underdoped high transition temperature (high-T_c) superconductor YBa2Cu3O6.58, directly revealing broken C4 symmetry on the Fermi surface. Moreover, we demonstrate that this Fermi surface has C2 symmetry of the type produced by a uniaxial or anisotropic density-wave phase. This establishes the central role of C4 symmetry breaking in the Fermi surface reconstruction of YBa2Cu3O6+δ and suggests a striking degree of universality among unconventional superconductors.

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INTRODUCTION

Broken C4 symmetry is observed in a number of experiments on unconventional superconductors, including transport, nuclear magnetic resonance (NMR), neutron scattering, X-ray scattering and scanning tunneling microscopy. In the iron-based superconductors broken C4 symmetry is observed directly on the Fermi surface, and has been taken as an indication that this broken symmetry drives the high T_c. The question of whether the same type of symmetry breaking is relevant in the copper-oxide high-T_c superconductors has been left open because it has never been observed on the Fermi surface. Without a link to the Fermi surface, it is hard to make a compelling argument that the experimental observations of broken C4 symmetry are relevant to the phenomenon of high-T_c.

The Fermi surface of a metal is constrained by the symmetry of its electronic environment. A precise determination of Fermi surface geometry, therefore, provides information about symmetry-breaking states of matter that feed back into the electronic structure. In the underdoped high-T_c cuprates it is now well established that a charge-density wave (CDW) competes with superconductivity, but a direct experimental connection is still missing between the CDW and the Fermi surface. Quantum oscillations (QOs) are used to determine the Fermi surface geometry of the high-T_c cuprates. To map the Fermi surface geometry of YBa2Cu3O6+δ we performed interlayer (ρzz) magnetoresistance measurements in a fixed magnetic field of 45 T even in the absence of interlayer coherence.

RESULTS

Since the 1930s it has been known that a change in resistance with an applied magnetic field—magnetoresistance—provides geometric information about a metallic Fermi surface, and early magnetoresistance experiments were instrumental in developing the modern quantum theory of metals. Real metals have non-spherical Fermi surfaces, thus magnetoresistance can be a strong function of the angle between the magnetic field and the crystal axes, giving rise to AMR. The signatures of AMR are particularly strong for quasi-2D Fermi surfaces, and thus this technique is well suited for determining the Fermi surface geometry of the high-T_c cuprates. To map the Fermi surface geometry of YBa2Cu3O6.58 we performed interlayer (ρzz) magnetoresistance measurements in a fixed magnetic field of 45 T even in the absence of interlayer coherence. The field-angle dependence of ρzz was obtained by rotating the sample in situ, sweeping the polar angle θ between the magnetic field and the crystalline c-axis for several values of the azimuthal angle ϕ (see the inset of Fig. 1a for angle definitions). Our primary observation is twofold anisotropy of the AMR as a function of ϕ—immediately apparent in Fig. 1—with the AMR increasing much more rapidly on rotating the field.
The angle-dependent magnetoresistance of YBa$_2$Cu$_3$O$_{6.58}$. 

The raw resistance (a) as a function of $\theta$ at 45 T and 15 K, for several values of $\phi$ spanning between the $\hat{a}$-axis ($\phi = 0^\circ$) and $\hat{b}$-axis ($\phi = 90^\circ$). The rapid drop in resistivity beyond $\theta = 60^\circ$ is due to the onset of superconductivity when insufficient magnetic field is parallel to the $\hat{c}$-axis. The inset defines the field angles $\phi$ and $\theta$ with respect to the crystallographic $\hat{a}$, $\hat{b}$, and $\hat{c}$ axes. This data was taken at a temperature of 15 K to increase the polar angular range over which the normal resistive state is accessed and to thermally suppress QOs. Broken $C_4$ symmetry is clearly shown in a polar plot of the data (b), where the radius is the polar angle $\theta$, and the amplitude and color correspond to the magnitude of the resistance.

**Fig. 2** Quasiparticle trajectory and magnetoresistance for different interlayer tunneling symmetries. Schematic Fermi surface with a cyclotron orbit. Gray arrows indicate the Fermi velocity along a cyclotron orbit (black line). Dispersion along the $k_z$ direction modulates the $\hat{z}$ component of the velocity and changes the sign of $v_z$ around the cyclotron orbit. Panel a shows the three dominant $\hat{c}$-axis dispersion shapes allowed by symmetry in YBa$_2$Cu$_3$O$_{6.58}$ (top panels) and their subsequent contributions to the AMR (bottom panels). Full rotational symmetry is preserved for the $\cos k_z$ dispersion, while $\sin \phi \cos k_z$ has $C_2$ symmetry. $\sin 2\phi \cos k_z$ has $C_4$ symmetry but produces $C_4$ symmetric AMR once all cyclotron orbits are accounted for. For clarity we have presented these Fermi surfaces with isotropic $k_z$ to emphasize the symmetry of the warping: in Fig. 4a these warplings will be superimposed on the actual diamond-like $k_z$ that we obtain from modeling the AMR.
existence of such a symmetry-breaking Fermi surface in YBa$_2$Cu$_3$O$_{6.58}$ is if the Fermi surface reconstruction itself breaks C$_4$ symmetry—the unreconstructed Fermi surface of YBa$_2$Cu$_3$O$_{6.58}$ is only slightly distorted from C$_4$ symmetry by the weakly orthorhombic crystal structure. While the crystal structure of YBa$_2$Cu$_3$O$_{6.58}$ inherently breaks C$_4$ symmetry, we argue that the small symmetry-breaking strain field arising from the orthorhombicity serves to preferentially align an underlying electronic instability. Our simulations suggest that anisotropy in the scattering rate; anisotropy in the interlayer hopping that is still finite in all directions; or an in-plane anisotropy in the Fermi wavevector (i.e., a Fermi surface elongated along one in-plane direction), cannot produce magnetoresistance with the angular structure we observe (see Supplementary Information for details).

Another prominent feature in the data is the suppression of AMR along the $\phi = 45^\circ$ direction, particularly above $\phi = 50^\circ$. In addition to the interlayer velocity, AMR is responsive to the in-plane geometry of the Fermi surface. For a cylindrical surface with simple cos $k_z$ warping and an isotropic Fermi radius $k_F$ (see Fig. 2b), the AMR evolves with field angle $\theta$ as $\rho_{zz}(\theta) \propto 1/(J_0(k_Z \tan \theta))^2$, where $c$ is the $c$-axis lattice constant (see footnote for more complicated warping geometries the actual form of $\rho_{zz}(\theta)$ is different, but it is still the product $k Zc$ that sets the angular scale over which the maxima in $\rho_{zz}$ appear). The AMR shows maxima wherever $k Zc \tan (\theta) \equiv 0$ (see Fig. 2b). A critical feature of this form of $\rho_{zz}$ is that the product $k Zc$ sets the in-plane scale in $\theta$ over which these maxima in $\rho_{zz}$ appear: a smaller $k Z$ pushes the resistance maxima out to higher angles.

Fig. 3 Two possible anisotropic CDW reconstruction schematics for YBa$_2$Cu$_3$O$_{6.58}$. The unreconstructed cuprate Fermi surface is a large hole-like cylinder. The bilayer copper oxide planes of YBa$_2$Cu$_3$O$_{6.58}$ give rise to bonding and anti-bonding bands, whose interlayer velocities have opposite signs (one quarter of the Fermi surface has been cut away for clarity). The right hand panels are interlayer velocities have opposite signs (one quarter of the Fermi surface section, and an in-plane diamond shape—strikingly similar to the Fermi surface reconstruction predicted to occur via CDW. We can now combine our qualitative Fermi surface information—interlayer tunneling with C$_2$ symmetry on at least one Fermi surface section, and an in-plane diamond shape—and quantitatively model our data by numerically solving the Boltzmann transport equation (see Supplementary Information for details). To avoid over-parametrization of the data we model only the three $F = 530 \text{T}$ “breakdown” surfaces (plus symmetry-related copies), which are known to dominate the $c$-axis conductivity. We fix the cross-sectional area of the orbits to the value of $A_0 = 2\pi \hbar / 530 \approx 5.1 \text{nm}^2$ obtained from QO measurements on YBa$_2$Cu$_3$O$_{6.58}$ (ref. 44), and allow a single value of $t$ to vary as a free parameter for all three surfaces. We find that the AMR is best modeled by a sum of 41% of a surface with sin $\phi$ cos $k_z$, 27% of a surface with cos $k_z$, and 32% of a surface with sin $2\phi$ cos $k_z$. These proportions are reasonably in line with what is expected from the number of possible breakdown orbits and their magnetic breakdown probabilities. In Fig. 4 we show that the most important features of the AMR which we first identified on qualitative grounds—C$_4$ symmetry, negative AMR near $\phi = 90^\circ$, and suppression along the $\phi = 45^\circ$ direction at high $\theta$—are captured by this model. It is important to note that
resistivity is not a linear function of the interlayer hopping parameters: combining all three symmetry warpings onto a single section of Fermi surface cannot reproduce the data. The consistency between our Fermi surface model and other experiments can now be checked. The quasiparticle lifetime we extract from the simulations is $\tau = 0.24 \pm 0.05 \text{ ps}$ — in agreement with the 0.27 ps reported from QOs. Zero-field resistivity measurements at this doping find an in-plane resistive anisotropy that is collapsing towards one at low temperatures: this behavior is attributed to the conductivity of the 1D copper-oxide chains freezing out at low temperature. Our model, which breaks $C_4$ symmetry only in the $c$-axis dispersion of the Fermi surface, naturally preserves the near-isotropy of the $ab$-plane conductivity. If the 1D copper oxide chain layer were clean enough to produce AMR—and there are evidence from spectroscopy to believe that this is not the case—then the resultant in-plane resistivity would be highly anisotropic: such in-plane anisotropy is not observed experimentally. AMR from an open Fermi surface produced by the chain layer would also not produce the observed upturn in the AMR we observe above $\theta = 40^\circ$ when $\phi = 90^\circ$(see Supplementary Information for details). AMR measurements can detect sections of Fermi surface that are invisible to QOs, such as open sheets and surfaces with higher scattering rates (shorter $\tau$). AMR, therefore, has the potential to observe sections of Fermi surface in YBa$_2$Cu$_4$O$_{6.58}$ formed by CDW reconstruction that have remained unobserved by QOs. Our simulation reveals, however, that the AMR can be fully accounted for by the same bilayer-split electron pocket that appears in QO experiments, with no additional sheets or pockets. This is consistent with previous suggestions that there is only a single Fermi pocket in the Brillouin zone of underdoped YBa$_2$Cu$_4$O$_6$ (refs 50, 51). We note that the small hole pocket reported at this doping has a cross-section ($k_F$) that is too small to contribute any significant AMR below $\theta=75^\circ$, thus we do not include the possibility of this surface in our model.

It is important to note that our model relies on the validity of the single-particle picture and Boltzmann transport equations. While these assumptions may be valid for the small Fermi pocket we observe—and the observation of the same pocket via QOs...
strongly suggests this to be true — there is much experimental evidence that large portions of the Fermi surface exhibit decidedly non-quasiparticle behavior. This includes the linear-in-temperature zero-field resistivity above the pseudogap temperature. One possibility is that high magnetic fields suppress the pseudogap and restore the entire Fermi surface to a Fermi-liquid state — the possible Fermi surface reconstruction presented in Fig. 3d, e would require such a scenario for the pocket near the anti-nodal region to be ungauged. This possibility suffers from the fact that no other pieces of Fermi surface besides the small electron pocket are observed in high magnetic fields. Despite the apparent ability of single-particle Boltzman equations to model the high-field magneto-transport, it remains an important open question as to how to connect the zero-field metallic state to that observed in high magnetic fields.

**DISCUSSION**

In order to obtain a Fermi surface with $C_2$ symmetry from the $C_4$ symmetric unreconstructed Fermi surface, the mechanism of Fermi surface reconstruction must itself break $C_4$ symmetry. We suggest that the diamond-like shape we measure with AMR disordered compared to more traditional CDW metals, its surface, and suggests that the physics underlying both the nematic state is directly observable on the Fermi surface reconstruction, as has been proposed theoretically. This is similar to the iron pnictide superconductors, non-evidence that large portions of the Fermi surface exhibit decidedly asymmetric in the $b$-axis components of the wavevectors persists to at least 45 T, and that these CDWs are responsible for the Fermi surface reconstruction, as has been proposed theoretically. A second route to a diamond-like pocket with $C_2$ symmetry in the $\hat{c}$-axis tunneling requires a strong nematic distortion of the Fermi surface (see Fig. 3d, e). In high magnetic fields this surface is then reconstructed by a unidirectional CDW. This scenario only requires the single CDW wavevector that acquires three-dimensional coherence in high fields.

Independent of the specific model, our analysis shows that Fermi surface reconstruction in $YBa_2Cu_3O_{6.58}$ is anisotropic, and that the entire AMR signature can be accounted for by the same Fermi surface that is responsible for QOs and a negative Hall coefficient in high magnetic fields. We have also established that although the CDW in the cuprates is relatively weak and disordered compared to more traditional CDW metals, its symmetry and wavevector directly determine Fermi surface properties. This is similar to the iron pnictide superconductors, where the nematic state is directly observable on the Fermi surface, and suggests that the physics underlying both the superconductivity and the quantum criticality in these two classes of materials share a similar origin. Our Fermi surface model predicts that for tetragonal $HgBa_2Cu_3O_6$ where both short-range CDW order and a small Fermi surface have been observed, sufficient uniaxial strain should favour anisotropic CDW formation and give rise to anisotropic AMR similar to what we have observed in $YBa_2Cu_3O_{6.58}$.

**METHODS**

Detwinned single crystals of $YBa_2Cu_3O_{6.58}$ were grown and prepared as described in Liang et al. C-axis contacts were prepared as described in Ramshaw et al. Resistivity measurements were conducted in the 45 Tesla hybrid magnet at the National High Magnetic Field Lab in Tallahassee using a 2-axis rotator. The resistance was measured with a 4-point contact geometry using an SRS 830 lockin amplifier; the sample was driven with $f = 500 \mu A$ at a frequency of $f = 37.7$ Hz.

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**AUTHOR CONTRIBUTIONS**

S.G., P.A.G., N.H., B.J.R, and S.E.S. performed the 45 Tesla AMR measurements at the NHMFL in Tallahassee, Florida. B.J.R and K.A.M. performed additional sample characterization at the NHMFL in Los Alamos, New Mexico. D.A.B, W.N.H, R.L., and B.J.R. grew and prepared the $YBa_2Cu_3O_{6.58}$ samples. N.H. and B.J.R. performed the data analysis and wrote the manuscript with input from all co-authors.

**COMPETING INTERESTS**

The authors declare no competing interests.

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