Fermi liquid behavior of the in-plane resistivity in the pseudogap state of YBa$_2$Cu$_4$O$_8$

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Our knowledge of the ground state of underdoped hole-doped cuprates has evolved considerably over the last few years. There is now compelling evidence that, inside the pseudogap phase, charge order breaks translational symmetry leading to a reconstructed Fermi surface made of small pockets. Quantum oscillations [Doiron-Leyraud N, et al. (2007) Nature 447(7144):565–568], optical conductivity [Mirzaii S, et al. (2013) Proc Natl Acad Sci USA 110(15):5774–5778], and the validity of Wiedemann-Franz law [Grissonnache G, et al. (2016) Phys Rev B 93:064513] point to a Fermi liquid regime at low temperature in the underdoped regime. However, the observation of a quadratic temperature dependence in the electrical resistivity at low temperatures, the hallmark of a Fermi liquid regime, is still missing. Here, we report magnetoresistance measurements in the magnetic-field-induced normal state of underdoped YBa$_2$Cu$_4$O$_8$ that are consistent with a $T^2$ resistivity extending down to 1.5 K. The magnitude of the $T^2$ coefficient, however, is much smaller than expected for a single pocket of the mass and size observed in quantum oscillations, implying that the reconstructed Fermi surface must consist of at least one additional pocket.

The generic phase diagram of Fig. 1 summarizes the temperature and doping dependence of the in-plane resistivity $\rho_{ab}(T)$ of hole-doped cuprates (1). Starting from the heavily overdoped side, nonsuperconducting La$_{1.67}$Sr$_{0.33}$CuO$_4$, for example, shows a purely quadratic resistivity below ~50 K (2). Below a critical doping $p_{sc}$ where superconductivity sets in, $\rho_{ab}(T)$ exhibits superlinear behavior that can be modeled either as $\rho_{ab} \sim T + \frac{T^2}{T^*}$ or as $\rho_{ab} \sim T^*$ (1 < $n$ < 2). When a magnetic field is applied to suppress superconductivity on the overdoped side, the limiting low-$T$ behavior is found to be $T$ linear (3–5). Optimally doped cuprates are characterized by a linear resistivity for all $T > T_c$, although the slope often extrapolates to a negative intercept, suggesting that, at the lowest temperatures, $\rho_{ab}(T)$ contains a component with an exponent larger than 1 (1). In the underdoped regime, $\rho_{ab}(T)$ varies approximately linearly with temperature at high $T$, but as the temperature is lowered below the pseudogap temperature $T^*$, it deviates from linearity in a very gradual way (6). At lower temperatures, marked by the light blue area in Fig. 1, there is no compelling evidence from various experimental probes of in-plane charge order (7–11). High-field NMR (12, 13) and ultrasonic (14) measurements indicate that a phase transition occurs below $T_c$. This is also confirmed by recent high-field X-ray measurements that indicate that the charge density wave (CDW) order becomes tridimensional with a coherence length that increases with increasing magnetic field strength (15, 16). This leads to a Fermi surface (FS) reconstruction that can be reconciled with quantum oscillations (QOs) (17, 18) as well as with the sign change of the Hall (19) and Seebeck (20) coefficients. Whether the charge order is biaxial (21) or uniaxial with orthogonal domains (22) is still an open issue, but a FS reconstruction involving two perpendicular wavevectors leads to at least one electron pocket in the nodal region of the Brillouin zone (23). Depending on the initial pseudogapped FS and on the wavevectors of the charge order, FS reconstruction can also lead to additional, smaller hole pockets (24, 25). In Y123 (doping level $p = 0.11$) the QQ spectra consist of a main frequency $F_a = 540$ T and a beat pattern indicative of nearby frequencies $F_{a2} = 450$ T and $F_{a3} = 630$ T. A smaller frequency $F_b \sim 100$ T has been detected by thermometer and $c$-axis transport measurements and attributed there to an additional small hole pocket (26). The presence of the three nearby frequencies $F_a$ can be explained by a model involving a bilayer system with an electron pocket in each plane and magnetic breakdown between the two pockets (27, 28). In this scenario, the low-frequency $F_b$ could originate from quantum interference or the Stark effect (29). However, this scenario predicts the occurrence of five nearby frequencies and thus requires fine-tuning of certain microscopic parameters such as the bilayer tunneling $t_L$. Moreover, the doping dependence of the Seebeck coefficient is difficult to reconcile with a FS reconstruction scenario leading to only one electron pocket per plane.

More generally, the observation of QOs is a classic signature of Landau quasiparticles. In underdoped Y123, the temperature dependence of the amplitude of the oscillations follows Fermi–Dirac statistics up to 18 K, as in the Landau–Fermi liquid theory (30). This conclusion is supported by other observations, such as the validity of the Wiedemann–Franz law (31) in underdoped Y123.

Significance

High-temperature superconductivity evolves out of a metallic state that undergoes profound changes as a function of carrier concentration, changes that are often obscured by the high upper critical fields. In the more disordered cuprate families, field suppression of superconductivity has uncovered an underlying ground state that exhibits unusual localization behavior. Here, we reveal that, in stoichiometric YBa$_2$Cu$_4$O$_8$, the field-induced ground state is both metallic and Fermi liquid-like. The manuscript also demonstrates the potential for using the absolute magnitude of the electrical resistivity to constrain the Fermi surface topology of correlated metals and, in the case of YBa$_2$Cu$_4$O$_8$, reveals that the current picture of the reconstructed Fermi surface in underdoped cuprates as a single, isotropic electron-like pocket may be incomplete.

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and the quadratic frequency and temperature dependence of the quasiparticle lifetime $\tau(\omega, T)$ measured by optical spectroscopy (32) in underdoped HgBa$_2$CuO$_{4+\delta}$ (Hg1201). An important outstanding question is whether the in-plane resistivity of underdoped cuprates also exhibits the behavior of a canonical Landau–Fermi liquid, namely, a quadratic temperature dependence at low $T$. In underdoped cuprates, several studies have shown $\rho_{ab} \sim T^2$, but always at elevated temperatures (indeed, most are above $T_c$) and only over a limited temperature range that never exceeds a factor of 2.5 (6, 33–36) (see Table S1 for a detailed list of studies). At low temperatures, either $\rho_{ab}(T)$ starts to become nonmetallic (36), suggesting that the $T^2$ behavior observed at intermediate temperatures could just be a crossover regime, or a quadratic behavior has previously been hinted at (19), rather than shown explicitly. Here, we present high-field in-plane magnetoresistance measurements in underdoped YBa$_2$Cu$_4$O$_8$ (Y124) that are consistent with the form $\rho_{ab}(T) = \rho_0(T) + AT^2$ from $T = T_c$ down to temperatures as low as 1.5 K, that is, over almost two decades in temperature. In addition, we investigate the magnitude of the resultant $A$ coefficient and compare it with some of the prevalent FS reconstruction scenarios. In conclusion, we find that the magnitude of $A$ is difficult to reconcile with the existence of a single electron pocket per plane, with an isotropic mass.

Results

We have measured the $a$-axis magnetoresistance (i.e., perpendicular to the conducting CuO chains) of two underdoped Y124 ($T_c = 80$ K) single crystals up to 60 T at various fixed temperatures down to 1.5 K. From thermal conductivity measurements at high fields, the upper critical field $H_{c2}$ of Y124 has been estimated to be $\sim 45$ T (37). Raw data for both samples are shown in Fig. 2. Above $T_c$, that is, in the absence of superconductivity, the transverse magnetoresistance can be accounted for, over the entire field range measured, by a two-carrier model (35) using the following formula:

$$\rho(H) = \rho(0) + \frac{\alpha H^2}{1 + \beta H^2}$$

where $\rho(0)$ is the zero-field resistivity and $\alpha$ and $\beta$ are free parameters that depend on the conductivity and the Hall coefficient of the electron and hole carriers (35) (see Fig. S1 for a comparison of the two-band and single-band, quadratic forms for the magnetoresistance). To obtain reliable values of $\rho(H \to 0, T) = \rho(0)$ and corresponding error bars for each field sweep, the data were fitted...
to Eq. 1 in varying field ranges using the procedure described in detail in Figs. S2–S5 and Tables S2 and S3. Precisely the same form is used to fit the high-field data at all temperatures studied below $T_c$. This procedure has been found to yield reliable $\rho(0)$ values in both cuprate (5) and pnictide (38) superconductors. Extrapolation of the high-field data to the zero-field axis $\rho(0)$, as shown by dashed lines in Fig. 2, allows one then to follow the evolution of $\rho(0)(T)$ down to low temperatures. The extrapolated $\rho(0)$ values are plotted versus temperature in Fig. 3 (symbols) for both crystals, along with the zero-field temperature dependence of the resistivity (solid line). The dashed lines in Fig. 3 correspond to fits of the $\rho(0,$ $T$) data to the form $\rho(0,T) = \rho_0 + AT^2$. A fit of the data to the form $\rho(0,T) = \rho_0 + AT^2$ yields $n = 1.9 \pm 0.2$ is shown in the Fig. S6. The Insets of Fig. 3 are corresponding plots of $\rho(0,T)$ versus $T^2$ to highlight the approximately quadratic form of $\rho(0,T)$. From the dashed line fits, we obtain $\rho_0 = 7.5 \pm 1.0$ and $10.0 \pm 1$ and $8.5 \pm 0.5 \mu \Omega$-cm-K$^{-2}$ for samples 1 and 2, respectively.

Discussion

The first key result of this study is our observation, within experimental resolution, of a quadratic temperature dependence of the in-plane resistivity in Y124 down to low temperatures, which indicates that the low-lying (near-nodal) electronic states inside the pseudogap phase of underdoped cuprates bear all of the hallmarks of Landau quasiparticles. Intriguingly, the magnitude of the $T^2$ term, $A = 9.5 \pm 1.5 \mu \Omega$-cm-K$^{-2}$, is similar to that measured at high temperature, for example, above 50 K in underdoped Y123 at $\rho = 0.11$ ($A \sim 6.6 \mu \Omega$-cm-K$^{-2}$) (33) as well as in single-layer Hg1201 above 80 K where $A$ varies between $\sim 10$ and 15 $\mu \Omega$-cm-K$^{-2}$ for 0.055 $\leq \rho \leq 0.11$ (34). Note that, in Y124, such comparison cannot be made because the temperature dependence of the resistivity is not quadratic above $T_c$. In Y123, it is known that an incipient CDW is formed below $T^*$, which onset at about $T_{CDW} \sim 130–150$ K in the doping range at $p = 0.11-0.14$ (39, 40). At a lower temperature $T_{SR} \sim 50$ K, high-field NMR (12) and ultrasound (14) measurements for $p = 0.11$ have revealed a phase transition, below which long-range static charge order appears and FS reconstruction is believed to take place. Taking into account that the $\rho(0)$ values shown in Fig. 3 are extrapolated from this high-field phase, there would appear to be no significant change in the $A$ coefficient between the low-$T$ regime where long-range CDW sets in and the high-$T$ regime where only incipient CDW order is detected. This is reminiscent of the situation in NbSe$_2$ where there is negligible change of the resistivity at the CDW transition $T_{CDW} = 33$ K (41). This behavior can be understood if no substantial change of the FS occurs at the CDW transition (for a discussion, see ref. 42). To make an analogy with Y124, we first acknowledge that the effect of the pseudogap is to suppress quasiparticles near the Brillouin zone boundaries. A FS reconstruction due to charge order with dominant wavevectors ($Q_x$, 0) and (0, $Q_y$) will create a small electron-like pocket composed of the residual “nodal” density of states, in contrast to the large hole-like FS characterizing the overdoped state. This FS reconstruction can be seen as a folding of the FS, and in terms of transport properties, the same nodal states will be involved in scattering processes below $T^*$ and below $T_{SR}$ when the FS is reconstructed. Thus, the similar value of $A$ at high and low temperatures can be reconciled. Note that, in canonical 1D CDW systems such as NbSe$_2$ (43) and organics metals (44), there is a marked change of slope of the resistivity below the charge ordering temperature due to nesting of part of the original FS.

The scenario discussed above for Y124 is also consistent with the observation of an anisotropic scattering rate $\Gamma$ in cuprates. In overdoped Tl$_2$Ba$_2$CuO$_{6+\delta}$ (Tl2201) (45), for example, it has been shown that the scattering rate is composed of two distinct terms, a $T^2$ term that is almost isotropic within the basal plane and a $T$-linear scattering rate that is strongly anisotropic, vanishing along the zone diagonals and exhibiting a maximum near the Brillouin zone boundary, where the pseudogap is maximal. Inside the pseudogap regime, therefore, one expects the $T$-linear scattering rate to become much diminished, leaving the $T^2$ scattering term as the dominant contribution to $\rho_A(T)$.

In correlated metals, both the $T^2$ resistivity and the $T$-linear specific heat are the consequence of the Pauli exclusion principle. Thus, the strength of the $T^2$ term in $\rho(T)$ is empirically related to the square of the electronic specific heat coefficient $\gamma_0$ via the Kadowaki–Woods ratio (KWR) $A/\gamma_0^2$ (46). An explicit expression

$$\rho \sim T^2$$
for the KWR has been derived for correlated metals taking into account unit cell volume, dimensionality, and carrier density (47). In a single-band quasi-2D metal, the $A$ coefficient reads as follows:

$$A_{\text{KWR}} = \left( \frac{8\pi e^2 c k_F^2}{\hbar^3} \right) \frac{m^*}{k_F^2},$$  \[2\]

where $a$ and $c$ are the lattice parameters, and $m^*$ and $k_F$ are the (isotropic) effective mass and Fermi wavevector, respectively. A similar expression has also been obtained recently using the Kubo formalism (48). As shown in Supporting Information, comparison of $A_{\text{KWR}}$ with experimentally determined values for both single-band and multiband correlated oxides shows good agreement for $A_{\text{KWR}}$ values spanning over three orders of magnitude.

Electron–electron collisions involve two quasiparticles that reside within a width of order $k_BT$ near the Fermi energy, providing the factor $T^3$. However, the total electron momentum is conserved in normal electron–electron scattering for the simple metals with a nearly free-electron–like FS. Additional mechanisms, such as Umklapp or interband scattering, are thus needed to understand the dissipation (see refs. 49–51 for a discussion). With regards to the hole-doped cuprates, it is debatable whether the KWR should hold at all within the pseudogap phase. However, given the increasing amount of data pointing to a rather conventional state at sufficiently low temperature, we believe that such analysis and comparison are appropriate, and in the following, we consider briefly a number of these possible mechanisms (47–51) in turn.

**Umklapp Scattering.** Assuming a single electron pocket per CuO$_2$ plane, Umklapp collisions can lead to dissipation for electron–electron scattering because the condition on the reconstructed FS, $k_p > G/4$, is fulfilled in the reconstructed Brillouin zone ($G$ is a reciprocal lattice vector). In Y124, the QO frequency linked to the electron pocket ($F_e = 660 \pm 30$ T) converts into $k_F = 1.42 \pm 0.03$ nm$^{-1}$ (52, 53), whereas the most recent QO measurements have indicated that $m^* = 1.9 \pm 0.1 m_e$ (54, 55). Eq. 2 thus gives an estimate of the $A$ coefficient for the electron pocket, $A_e = 86 \pm 20$ nΩ cm K$^{-2}$, that is, $A_{\text{KWR}} = 43 \pm 10$ nΩ cm K$^{-2}$, taking into account the two CuO$_2$ planes. Significantly, this is almost five times larger than the experimental value. Assuming that the KWR ratio holds in underdoped cuprates, it implies that a reconstructed FS containing only one electron pocket (with an isotropic $m^*$) cannot account fully for the magnitude of the $T^2$ resistivity term in Y124. Similar conclusions are also drawn from comparison of the Fermi parameters reported for underdoped Y123 (18) and the measured $A$ coefficient (33) (albeit at elevated temperatures).

**Multiband Scenario.** In a second scenario initially proposed by Baber (49), the momentum transfer between two distinct reservoirs can also lead to dissipative scattering. In underdoped Y123, a small QO frequency ($F_e \sim 95$ T) was discovered and attributed to a hole pocket based on the doping dependence of the Seebeck coefficient (26). A FS comprising at most one electron and two hole pockets with the measured areas and masses is consistent with a scenario based on the FS reconstruction induced by the CDW order observed in Y123 (24) (Fig. 4). It is also compatible, within error bars, with the value of the electronic specific heat coefficient (33) for both the electron and hole pockets, respectively. $A_{\text{tot}}$ is the expected $A$ coefficient for a parallel resistor model taking into account one electron and two hole pockets and the two CuO$_2$ planes.

$$A_{\text{tot}} = \left[ 2 \left( \frac{1}{A_e} + \frac{2}{A_h} \right) \right]^{-1} = 14.7 \text{nΩ cm K}^{-2}$$

$A_e = 56 \pm 9 \text{nΩ cm K}^{-2}$ and $A_h = 89 \pm 42 \text{nΩ cm K}^{-2}$ (note that the large error here is due to the relative error in the effective mass). Finally, applying the parallel-resistor formula and taking into account the bilayer nature of Y123, the $A$ coefficient is estimated to be $A_{\text{KWR}} \sim 12 \pm 6$ nΩ cm K$^{-2}$, in reasonable agreement with the value measured at high temperature (4 $\sim 6.6$ nΩ cm K$^{-2}$) (33).

CDW order has not yet been directly observed by X-ray or by NMR in Y124, but the similar QO frequency (presumably to arise from the electron pocket) and the sign change in the Hall coefficient observed in both families (58) point to a very similar FS reconstruction. Assuming the presence of additional (as yet-undetected) hole pockets in the reconstructed FS of Y124 and given that $A = 9.5 \pm 1.5$ nΩ cm K$^{-2}$, we can estimate using Eq. 2 the effective magnitude of the $A$ coefficient associated with an individual hole pocket to be $A_h = 49 \pm 10$ nΩ cm K$^{-2}$. Given the strong sensitivity of $A_{\text{KWR}}$ to the absolute values of $m^*$ and $k_F$, this estimate is considered to be in good agreement with the value of $A_h$ deduced for Y123. This is also in agreement with the two-band description of transport data in Y124 (35) and the doping dependence of the Seebeck (20) and Hall (19) coefficients in YBCO. The magnitude of the $T^2$ term in $\rho_{xx}(T)$ can thus be considered as yet further evidence that the reconstructed FS of underdoped Y123 and Y124 contains not only the well-established electron pocket, but also at least one additional hole-like pocket. This is in agreement with the FS reconstruction scenario proposed within the biaxial CDW model (24, 25), assuming that the initial FS is the pseudogapped FS (e.g., without states at the antinode) and not the band structure-derived FS.
The multiband scenario has a number of other implications. First, in the Hg1201 family, QOs have been measured at a doping level $p = 0.09$ with a frequency $F = 840 \pm 30$ T and an effective mass $m^* = 2.45 \pm 0.15 m_0$ (59). Accordingly, Eq. 2 gives $A_{KWR} = 73 \pm 10 \hbar^2c^2m^{-2}$ (34). Above $T_c$, the measured $A$ coefficient is $A \sim 10 \hbar^2c^2m^{-2}$ (34). This large discrepancy between the estimated and measured values of $A$ again indicates that the reconstructed FS of Hg1211 may also contain an additional pocket or pockets that have not yet been observed in QO experiments (60). Second, in Y123, QOs have been observed over a wide range of doping yet $F$ is found to increase only by $\sim 20\%$ between $p = 0.09$ and $p = 0.152$ (61). These measurements also reveal a strong enhancement of the quasiparticle effective mass as optimal doping is approached and suggest a quantum critical point at a hole doping of $p_{opt} \sim 0.17$. If the electron pocket was indeed the only pocket that persists in the reconstructed phase, this marked enhancement in $m^*$ should lead to a corresponding enhancement of the $A$ coefficient on both sides of $p_{opt}$, as has been observed, for example, in the transport properties of the isovoltly substituted nitride family BaFe$_2$(Si$_x$P$_{1-x}$)$_2$ (38). In cuprates, however, the situation is far from clear. Certainly, there is no sign of a divergence of the $A$ coefficient near $p \sim 0.18$ from existing high-temperature measurements (34). Moreover, measurements of the low-temperature in-plane resistivity of several overdoped LSCO samples in high magnetic field have revealed a time- and temperature-independent “anomalous” or “extended” criticality regime around $p \sim 0.19$ where the coefficient of the $T$-linear term is maximal yet there is no sign of divergence or an enhancement in $A$ (from the overdoped side) (5). Within this multiband scenario, it is possible that the marked enhancement in $m^*$ of the electron pocket, and thus in $A_e$, is offset by changes in $A_h$ (assuming that the hole pockets are always present).

**Alternative Scenarios.** Finally, we consider an alternative scenario for the pseudogap in which only Fermi arcs exist at low field and ask what is the magnitude of the $A$ coefficient expected in such a scenario. This is relatively easy to do, at least approximately. For $Y124$, $p = 0.14$. Thus, the full FS is expected to occupy 57% of the Brillouin zone. If we assume that the quasiparticles on the full FS have a comparable mass to those found in overdoped cuprates, that is, $m^* \sim 5m_0$, one obtains $A_{KWR} = 2.5 \hbar^2c^2m^{-2}$ for the full unreconstructed FS. At low $T$, the “normal-state” specific heat coefficient $\gamma_0$ in Y124 is estimated using the entropy conservation construction to be approximately one-third of its value at high temperature, that is, above $T^* = 520$. For a Fermi arc that is one-third the length of the full quadrant, $A_{KWR}$ is correspondingly tripled (again assuming an isotropic $m^*$). Thus, the magnitude of $A$ with such a scenario is similar to the coefficient found at low temperatures and from high-field studies. It is important to recognize, however, that a Fermi arc has only hole-like curvature, and thus can in no way account for the two-carrier form of the magnetoresistance in Y124, nor for the negative sign of the Hall coefficient at low temperatures and high fields.

Until now, all estimates and comparisons have been made under the assumption that the effective mass does not vary around the FS. In an alternative scenario (62), the effective mass of the diamond-shaped electron pocket is taken to be anisotropic. The effective mass deduced from QOs is large because it is dominated by the corner of the pocket that corresponds to the hot spot of the CDW. By contrast, transport is dominated by those regions of the FS with the highest Fermi velocity, that is, the light quasiparticles in the near-nodal state. This scenario can explain the factor of five discrepancy between the expected value of the KWR ratio and the experimental one (assuming a single electron pocket), but the anisotropy needs to be abnormally strong and would need to increase with doping to explain the behavior of the effective mass deduced from QOs (61). Similar considerations would also apply if the scattering rate $\Gamma$, rather than the effective mass, were strongly anisotropic (63).

The picture of overdoped cuprates now emerging from X-ray spectroscopy is of a lengthening of the correlation length $\xi$ associated with the charge ordering with increased field strength (15) in agreement with NMR and thermodynamic measurements. At what value of $\xi$, relative to the mean free path of the remnant quasiparticles, does FS reconstruction appear, or is manifest in the magnetotransport properties, is a crucial open question. At high temperatures, where the mean free path is short, the quasiparticle response will be susceptible even to short-range charge order. At low temperatures, however, the situation is less clear. The findings reported in this manuscript point to a strong influence of the incipient charge order on the transport properties over a wide region of the temperature–doping–magnetic-field phase diagram, and calls for a systematic study of the evolution of the low-$T$ in-plane resistivity of overdoped cuprates inside the pseudogap regime.

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