LETTERS

Electron pockets in the Fermi surface of hole-doped high-T_c superconductors

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High-temperature superconductivity in copper oxides occurs when the materials are chemically tuned to have a carrier concentration intermediate between their metallic state at high doping and their insulating state at zero doping. The underlying evolution of the electron system in the absence of superconductivity is still unclear, and a question of central importance is whether it involves any intermediate phase with broken symmetry[1]. The Fermi surface of the electronic states in the underdoped 'YBCO' materials $YBa_2Cu_3O_y$ and $YBa_2Cu_4O_8$ was recently shown to include small pockets[2–4], in contrast with the large cylinder that characterizes the overdoped regime[5], pointing to a topological change in the Fermi surface. Here we report the observation of a negative Hall resistance in the magnetic-field-induced normal state of $YBa_2Cu_3O_y$ and $YBa_2Cu_4O_8$, which reveals that these pockets are electron-like rather than hole-like. We propose that these electron pockets most probably arise from a reconstruction of the Fermi surface caused by the onset of a density-wave phase, as is thought to occur in the electron-doped copper oxides near the onset of antiferromagnetic order[6,7]. Comparison with materials of the La_2CuO_4 family that exhibit spin/charge density-wave order[8–11] suggests that a Fermi surface reconstruction also occurs in those materials, pointing to a generic property of high-transition-temperature (T_c) superconductors.

The Hall effect is a powerful probe of the Fermi surface of a metal because of its sensitivity to the sign of charge carriers, which distinguishes between electrons and holes. In addition, the Hall effect has been the prime transport signature of density-wave order in copper oxides such as $La_{2-y-x}Nd_ySr_xCuO_4$ (Nd-LSCO) (ref. 10) and $La_{2-x}Ba_xCuO_4$ (LBCO) (ref. 11). The Hall resistance R_{xy} of LBCO is reproduced in Fig. 1, in which it drops precipitously below a temperature T_{DW} that coincides with the well-established onset of spin/charge density-wave order in this material[8]. The drop leads to a change of sign in R_{xy}, pointing to a reconstruction of the Fermi surface from purely hole-like above T_{DW} to a combination of electron-like and hole-like sheets below T_{DW}. The fact that our high-field measurement of R_{xy} in $YBa_2Cu_3O_y$, a copper oxide material with a different structure and considerably higher purity, cation order and maximal T_c, exhibits a similar behaviour of R_{xy}, as shown in Fig. 1, raises the possibility that Fermi surface reconstruction may be a generic phenomenon in copper oxides, and hence is likely to be essential for a full understanding of high-temperature superconductors.

The Hall resistance was measured in two closely related underdoped copper oxides of the YBCO family: $YBa_2Cu_3O_y$ (Y123), with $y = 6.51$ and $y = 6.67$, and $YBa_2Cu_4O_8$ (Y124). The Y123 samples have a high degree of oxygen order, with ortho-II and ortho-VIII superstructure, respectively. The Y124 is stoichiometric, with intrinsic oxygen order. With T_c values of 57.5, 66.0 and 80 K, respectively, the three samples have a hole doping per planar copper atom of $p = 0.10$, 0.12 and 0.14, respectively, that is, they all fall in the underdoped region of the doping phase diagram ($p < 0.16$). (Sample characteristics are given in the Methods Summary.) The current was applied along the a axis of the orthorhombic structure ($J||x||a$), that is, perpendicular to the CuO chains, in magnetic fields applied normal to the CuO_2 planes ($B||z||c$). (Details of the measurements are given in the Methods Summary.) The Hall coefficient $R_H \equiv t\,R_{xy}/B$, where t is the sample thickness, is displayed as a function of magnetic field in Fig. 2 and as a function of temperature in Supplementary Fig. 1.

Our central finding is that all three materials have a negative Hall coefficient in the normal state at low temperature. This is displayed in Fig. 3, where a plot of R_H versus T at the highest field reveals a change of sign from $R_H > 0$ above T_0 to $R_H < 0$ below T_0, with $T_0 = 30$, 70 and 30 K for Y123-II, Y123-VIII and Y124, respectively, with ± 2 K uncertainty. A very similar sign change was reported in ref. 12 in Y123 samples with $T_c = 62$–64 K. Because their measurements were limited to moderate fields (below 24 T), these authors attributed the negative R_{xy} to a negative contribution to the Hall conductivity σ_{xy}

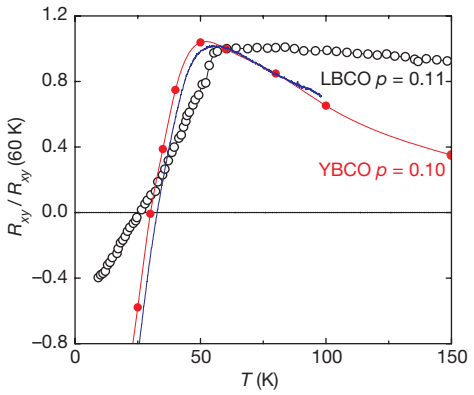

Figure 1 | Hall resistance of LBCO and YBCO. Hall resistance R_{xy} versus T, normalized at 60 K, for LBCO at $p = 0.11$ ($x = 0.11$; black circles from ref. 11) and YBCO at $p = 0.10$. Our data on YBCO were obtained on two different Y123-II samples (with $y = 6.51$), one measured in a continuous temperature sweep at a constant field of 45 T (at the NHMFL; blue curve) and the other measured via field sweeps up to 61 T (at the LNCMP; red circles, taken at 55 T).

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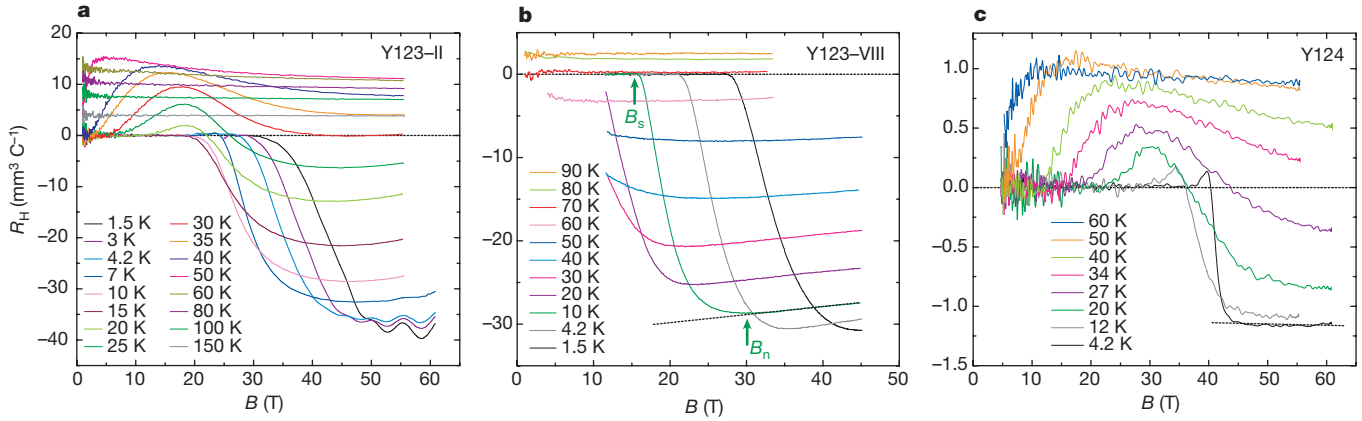

Figure 2 | Hall coefficient versus magnetic field. Hall coefficient $R_H = tR_{xy}/B$ as a function of magnetic field B at the indicated temperatures. **a**, Y123-II ($p = 0.10$); **b**, Y123-VIII ($p = 0.12$); **c**, Y124 ($p = 0.14$). The arrows in **b** indicate the fields B_s and B_n described in the text and defined in the Supplementary Information. The 4.2-K isotherm of Y124 illustrates nicely the basic components of R_H: the flat negative part at high field (above B_n) is the normal-state value, whereas the positive overshoot just above B_s is due to a vortex flux-flow contribution.

coming from vortices (flux flow). By going to much higher fields, we can now rule out this interpretation, as discussed in detail in the Supplementary Information, in which the negative R_H is shown to be unambiguously a property of the normal state, the consequence of a drop in $R_H(T)$ that starts below a field-independent temperature T_{max}. The value of T_{max} at the three doping levels studied here is 50, 105 and 60 K, for Y123-II, Y123-VIII and Y124, respectively, with ± 5 K uncertainty (see arrows in Fig. 3).

Three groups have previously detected this drop in low-field measurements of underdoped Y123, with $B < 15$ T, on crystals with $T_c(0) = 60$–70 K (refs 13–15). Because these earlier studies were limited to high temperatures ($T > T_c(0)$), they failed to reveal that the drop is just the start of a large swing to negative values. By measuring R_{xx} and R_{xy} along both a and b axes, it was shown[15] that the drop in $R_H(T)$ is a property of the planes, not the chains. From the perfect linearity of R_{xy} versus B it was also concluded that the drop is not due to flux flow[15].

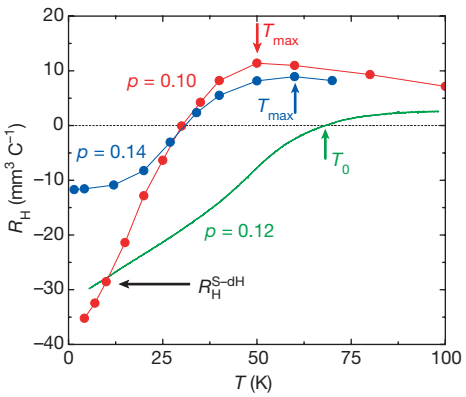

Figure 3 | Normal-state Hall coefficient versus temperature. Hall coefficient R_H versus T for Y123-II, Y123-VIII and Y124 (data multiplied by ten), at $B = 55$, 45 and 55 T, respectively. T_0 is the temperature at which R_H changes sign, equal to 30, 70 and 30 $\pm$ 2 K, respectively. T_{max} is the temperature at which R_H is maximum, equal to 50, 105 and 60 $\pm$ 5 K, respectively. The black arrow indicates the value of the Hall coefficient expected for a single electron Fermi pocket of the size imposed by Shubnikov–de Haas oscillations of frequency F, namely $R_H^{S-dH} = -V_{cell}/en_{S-dH}$, where $n_{S-dH} = F/\Phi_0 = 0.038$ electrons per unit cell. (The data for Y124 is multiplied by a factor of ten to put it on a scale comparable to Y123-II and Y123-VIII. The order-of-magnitude reduction of the measured Hall voltage comes in large part from the short-circuiting effect of the CuO chains along the b axis, which in this stoichiometric material, unlike in Y123-II and Y123-VIII, remain highly conductive down to low temperature; see text.)

The most natural explanation for the negative R_H is the presence of an electron pocket in the Fermi surface. (In principle, it could also come from a hole pocket with portions of negative curvature[16].) In a scenario in which the Fermi surface contains both electron and hole pockets, the sign of R_H depends on the relative magnitude of the respective densities n_e and n_h, and mobilities[17] μ_e and μ_h. ($\mu \equiv e\tau/m^*$, where e is the electron charge, τ^{-1} is the scattering rate and m^* is the effective mass.) Given that these materials are hole-doped, we expect $n_h > n_e$. The fact that $R_H < 0$ at low T therefore implies that $\mu_e > \mu_h$ at low T. Given strong inelastic scattering, this inequality can then easily invert at high T, offering a straightforward mechanism for the sign change in R_H. This happens in simple metals like Al and In (ref. 17) and is typical of compensated metals (with $n_e = n_h$). In high-purity samples of $NbSe_2$, a quasi-two-dimensional metal that undergoes a charge density-wave transition at $T_{CDW} \approx 30$ K, $R_H(T)$ drops from its positive and flat behaviour above T_{CDW} eventually to become negative below $T_0 \approx 25$ K (ref. 18), as reproduced in Supplementary Fig. 5. In impure samples, however, $R_H(T)$ remains positive at all T values (ref. 18; Supplementary Fig. 5), showing that the electron/hole balance can depend sensitively on impurity/disorder scattering.

A scenario of electron and hole pockets for YBCO resolves a puzzle in relation to the Shubnikov–de Haas oscillations observed in Y123-II (ref. 2). The puzzle is the apparent violation of the Luttinger sum rule, which states that the total carrier density n must be equal to the total area of the two-dimensional Fermi surface. From the oscillation frequency $F = 530$ T, one gets a carrier density $n_{S-dH} = 0.038$ carriers per planar Cu atom per pocket via $F = n_{S-dH}\Phi_0$, where $\Phi_0 = 2.07 \times 10^{-15}$ T m^2 is the flux quantum. Assuming that the pocket is a hole pocket (of arbitrary curvature) and there is nothing else in the Fermi surface, and assuming also that n must be equal to the density of doped holes ($n = p = 0.10$), the Luttinger sum rule is clearly violated, whether the relevant Brillouin zone includes one or two (or any number) of these pockets[19] (whether $n = n_{S-dH} = 0.038$ or $n = 2n_{S-dH} = 0.076$). If, on the other hand, the Fermi surface contains other sheets (not seen in the Shubnikov–de Haas oscillations) besides the observed pockets, then the sum rule can easily be satisfied.

The fact that R_H is negative at low T implies that the Shubnikov–de Haas frequency that was seen in Y123-II (ref. 2) must come from the high-mobility electron pocket, because the amplitude of Shubnikov–de Haas oscillations depends exponentially on mobility, as $\exp(-\pi/\mu B)$. The hole-like portions of the Fermi surface are either open or have a lower mobility at $T \to 0$. The largest value of R_H that a single electron pocket of density $n_{S-dH} = 0.038$ electrons per unit cell can produce is $R_H^{S-dH} = -V_{cell}/en_{S-dH} = -29$ mm^3 C^{-1}. Within the uncertainty in the geometric factor, this is the magnitude of R_H

measured in Y123-II and Y123-VIII at low temperature (see Fig. 3). (A similar estimate cannot be made for Y124, because in this particular case, unlike in Y123-II and Y123-VIII with their imperfect CuO chains that localize charge carriers[2], the chains remain metallic down to low temperature, and thus partially short-circuit the Hall voltage, causing a reduction in Hall resistance by as much as a factor of ten or so (see ref. 15).) The picture that emerges is in sharp contrast with the single Fermi 'arc' at $(\pi/2, \pi/2)$ seen in angle-resolved photoemission spectroscopy (ARPES) studies on other copper oxides in zero field (see discussion in refs 1 and 2). A possible explanation for the discrepancy is that ARPES detects only the hole pocket (one side of it) and the Shubnikov–de Haas oscillations only the electron pocket (thanks to its high mobility in the elastic scattering regime). This would suggest a Fermi surface similar to that proposed for electron-doped copper oxides (see below), with a hole pocket at $(\pi/2, \pi/2)$ and an electron pocket at $(\pi, 0)$.

One might ask why a negative R_H has not been seen in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ (BSLCO) and $La_{2-x}Sr_xCuO_4$ (LSCO), the other hole-doped copper oxides to have been measured up to 50 T (refs 20 and 21). Indeed, although R_H can drop by nearly a factor of three between 150 and 1.5 K in BSLCO, in a manner not unlike what is seen here in YBCO, it never becomes negative. A possible explanation is that the negative R_H in the YBCO copper oxides is associated with the presence of CuO chains. In Y123-II, however, electrical anisotropy in the *a*–*b* plane is unity below 100 K (ref. 2), implying that the chain subsystem is non-conducting at low temperatures. In Y124, the double-chain unit remains metallic down to low T and will therefore have an associated Hall coefficient. However, in isostructural, non-superconducting Pr124, in which only the chains are conducting, R_H is found to be positive at low T (ref. 22). A more likely alternative is that the much stronger disorder scattering characteristic of BSLCO and LSCO compared to Y123 or Y124 suppresses μ_e more severely than μ_h. In such a case, the electron pocket, although present, manifests itself only in the temperature dependence of R_H, not its sign. As mentioned above, this is what happens in $NbSe_2$: adding impurities eliminates the sign change in R_H (see ref. 18 and Supplementary Fig. 5).

Because the band structures of Y123 (see ref. 2 and references therein) and Y124 (see ref. 4 and references therein) calculated within the local density approximation do not support electron pockets, the fundamental question is: how do electron pockets come to exist? The combination of a small Fermi surface volume from Shubnikov–de Haas oscillations and a negative R_H pointing to electron pockets, in both Y123 and Y124, argues strongly for a reconstruction of the local density approximation Fermi surface. The standard mechanism for such reconstruction is the onset of a density-wave instability, as encountered in numerous materials. Examples in quasi-two-dimensional materials include $NbSe_2$, already mentioned, and the transition-metal oxide $Ca_3Ru_2O_7$ (ref. 23).

In copper oxides, at least three mechanisms can be invoked for a reconstruction of the Fermi surface that would result in electron and hole pockets. The first scenario is an antiferromagnetic phase with a (π, π) ordering wavevector, which causes the large hole-like Fermi surface to reconstruct into a small hole pocket at $(\pi/2, \pi/2)$ and a small electron pocket at $(\pi, 0)$. This model was used to explain the sign change in the low-temperature R_H measured in the electron-doped copper oxide $Pr_{2-x}Ce_xCuO_{4-\delta}$ (PCCO) (ref. 7), on crossing a critical concentration x_c close to where long-range antiferromagnetic order ends. In PCCO, R_H is positive at all temperatures at high doping ($x > 0.19$) and negative at low doping ($x < 0.15$), but it changes sign at intermediate dopings, from positive below $T_0 =$ 30–40 K to negative above[6]. The latter behaviour is similar (but opposite in sign) to that of YBCO in the range $0.10 < p < 0.14$, suggesting that the transport properties of both materials should be interpreted in terms of electrons and holes with different, T-dependent mobilities. In the case of hole-doped copper oxides, however, it is less likely that antiferromagnetic order is the relevant mechanism, because long-range antiferromagnetic order is thought to be confined to lower doping ($p < 0.05$). Nevertheless, the possibility should be investigated, particularly as a large magnetic field may push the phase transition to higher doping[19].

The second scenario is a theoretical phase with *d*-density-wave order, which would also cause a (π, π) folding of the Fermi surface and thus could produce electron pockets near $(\pi, 0)$ (ref. 24). The third scenario is a density-wave phase akin to that encountered experimentally in LBCO and Nd-doped LSCO. In these systems, a Fermi surface reconstruction, signalled by a precipitous drop in R_H (Fig. 1), coincides with the density-wave transition, observed directly via neutron diffraction. Reference 25 has recently shown that within mean-field theory a '1/8-stripe' spin/charge density-wave order of this kind does reconstruct the Fermi surface of a generic hole-doped copper oxide in a way that tends to produce an electron pocket.

Although the similarities between the La_2CuO_4 and YBCO families are highly suggestive, there are some important differences. First, whereas there is unambiguous direct evidence for static spin/charge density-wave order in LBCO and Nd-LSCO from neutron diffraction[8,9], there is no such evidence so far in YBCO. This could be because the putative density-wave phase in YBCO involves fluctuating rather than static order[26], or short-range rather than long-range order[27]. Second, the anomalies in the transport properties associated with density-wave order are sharp in LBCO (ref. 11) and Nd-doped LSCO (ref. 10), whereas in YBCO the temperature dependence of R_H is smooth. Note, however, that anomalies in the former materials seem to be sharp only when the low-temperature structural transition coincides with the density-wave transition. When structural and density-wave transitions do not coincide, as in Eu-doped LSCO (ref. 28), the anomalies also seem to be smooth.

In summary, the normal state of underdoped YBCO is characterized by a negative Hall coefficient, revealing the presence of an electron pocket in the Fermi surface, the mobility of which in these clean copper oxides is high enough to outweigh the contribution from other, hole-like parts of the Fermi surface. This implies that the Shubnikov–de Haas oscillations observed recently in the same materials must come from those electron pockets. It also suggests that the generally positive Hall coefficient seen in other hole-doped copper oxides results from electron mobilities that are too low because of stronger disorder scattering. Because electron pockets are not supported by the band structure of YBCO, we conclude that they must come from a reconstruction of the Fermi surface, which occurs at a critical doping above $p = 0.14$. In the absence of any direct evidence so far for long-range density-wave order in YBCO, our findings call for theoretical investigations of more unconventional scenarios.

METHODS SUMMARY

Samples. The Y123 samples are fully detwinned crystals of $YBa_2Cu_3O_y$ grown in non-reactive $BaZrO_3$ crucibles from high-purity starting materials (see ref. 2 and references therein). For Y123-II (or Y123-VIII), the oxygen content was set at $y = 6.51$ (or 6.67) and the dopant oxygen atoms were made to order into an ortho-II (or ortho-VIII) superstructure, yielding a superconducting transition temperature $T_c = 57.5$ K (or 66.0 K). The samples are uncut, unpolished thin platelets, the transport properties of which are measured via gold evaporated contacts (resistance $<1\,\Omega$), in a six-contact geometry. Typical sample dimensions are 20–50 × 500–800 × 500–1,000 μm^3 (thickness × width × length). The $YBa_2Cu_4O_8$ crystals were grown by a flux method in Y_2O_3 crucibles and an Ar/O_2 mixture at 2,000 bar, with a partial oxygen pressure of 400 bar (ref. 29).

Estimates of hole doping. The hole doping p in Y123 is determined from a relationship between T_c and the c axis lattice constant[30]. For our Y123-II (or Y123-VIII) samples, the measured T_c implies $p = 0.099$ (or 0.120). We assume the doping in Y124 is the same as in a sample of Y123 with the same T_c of 80 K, namely $p = 0.137 \approx 0.14$.

Resistance measurements. Longitudinal (R_{xx}) and transverse (R_{xy}) resistances are obtained from the voltage drop measured diagonally on either side of the sample width, for a field parallel (up) and anti-parallel (down) to the c axis: $R_{xx} \equiv (V_{up} + V_{down})/2I_x$ and $R_{xy} \equiv (V_{up} - V_{down})/2I_x$. Measurements on Y123-II and Y124 were performed at the LNCMP in Toulouse, in a pulsed resistive

magnet up to 61 T. Measurements on Y123-II and Y123-VIII were performed at the NHMFL in Tallahassee, in a steady hybrid magnet up to 45 T.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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