Quantum Oscillations in the Underdoped Cuprate \(\text{YBa}_2\text{Cu}_4\text{O}_8\)

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We report the observation of quantum oscillations in the underdoped cuprate superconductor \(\text{YBa}_2\text{Cu}_4\text{O}_8\) using a tunnel-diode oscillator technique in pulsed magnetic fields up to 85 T. There is a clear signal, periodic in inverse field, with frequency \(660 \pm 15\) T and possible evidence for the presence of two components of slightly different frequency. The quasi-particle mass is \(m^* = 3.0 \pm 0.3m_e\). In conjunction with the results of Doiron-Leyraud et al. for \(\text{YBa}_2\text{Cu}_3\text{O}_6.5\), the present measurements suggest that Fermi surface pockets are a general feature of underdoped copper oxide planes and provide information about the doping dependence of the Fermi surface.

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The mechanism for high-temperature superconductivity in the layered copper oxide superconductors has remained elusive for more than twenty years. At the heart of the problem is the evolution of the ground state from a Mott-Hubbard insulator to a superconductor as the number of doped holes \(p\) per planar \(\text{CuO}_2\) unit is increased. In particular, there is no agreement as to how the underdoped region should be described. The recent observation of quantum oscillations in the oxygen-ordered ortho-II phase of \(\text{YBa}_2\text{Cu}_3\text{O}_6.5\) (O-II Y123) with \(T_c = 57.5\) K, and \(p = 0.1\) \(^1\) shows that it has charged quasiparticles and a well-defined Fermi surface (FS) at low temperatures. In this Letter we report observations of quantum oscillations in the stoichiometric double-chain cuprate \(\text{YBa}_2\text{Cu}_3\text{O}_8\) (Y124) with \(T_c = 80\) K, and \(p = 0.125\) \(^2\) at fields up to 85 T, suggesting that they could be a general feature of underdoped cuprates. Our data for Y124 show that the FS pockets expand as \(p\) is increased and give a higher quasi-particle mass \(m^*\) than for O-II Y123.

The Y124 crystal was grown from flux in a ZrO\(_2\) crucible under 600 bar of O\(_2\) at 1100\(^\circ\)C. Other crystals from the same batch were of high quality with a residual Cu-O chain resistivity \(\lesssim 1\) \(\mu\)Ω cm, and a low-\(T\) thermal conductivity peak \(\kappa_b(20\) K\) = 120 \(\text{Wm}^{-1}\text{K}^{-1}\). \(^3\) Pulsed magnetic fields up to 85 T were provided by the Los Alamos 85 T multi-shot magnet \(^4\). Measurements were made using a tunnel-diode oscillator (TDO) technique \(^5\), \(^6\) in which two small counter-wound coils form the inductance of a resonant circuit. The crystal was cut into four pieces, each measuring up to \(0.35 \times 0.25 \times 0.12\) mm\(^3\), which were stacked with their \(c\)-axis directions aligned within 2\(^\circ\) of each other, and placed in one coil with the \(c\)-axis parallel to \(B\) and the axis of the coil. The resonant frequency, in our case 47 MHz, can depend on both the skin-depth (or, in the superconducting state, the penetration depth) and the differential magnetic susceptibility of the sample \(^7\). The sample and coil were immersed in \(^3\)He liquid or \(^4\)He exchange gas, temperatures \((T)\) being measured with a Cernox thermometer 5 mm away from the sample.

Fig. 1(a) shows the TDO frequency \(f\) versus \(B\) at \(T = 0.53\) K. At \(B \approx 45\) T, \(f\) falls substantially indicating an increase in the penetration of the rf field as the superconductivity is suppressed. In the expanded view of the raw data taken during the falling part of the pulse, oscillations are visible for fields \(B > 55\) T. The solid lines in Fig. 1(b) show the second derivative \(d^2f/dB^2\) of data taken at 1.6 K and reveal a clear oscillatory signal. The frequency and phase are nearly the same during the rising (36 T to 85 T in 5 ms) and falling (85 T to 36 T in 10 ms) parts of the pulse, ruling out spurious heating and electrical interference effects.

The standard Lifshitz-Kosevich (LK) form for the oscillatory magnetization is \(M \propto B^{1/2}R_D R_T \sin(2\pi F/B + \phi)\) \(^8\), where \(\phi\) is a phase, and in conventional metals the oscillation frequency \(F\) is related to a zero-field extremal FS cross-section \(A\) by the Onsager relation \(F = (h/2\pi e)A\); the scattering and temperature damping factors are respectively \(R_D = \exp(-\pi R_T e F e/\ell B)\) where \(k_F\) is the Fermi wave vector, \(\ell\) is the mean free path and \(R_T = (14.69 m^T/m_e B)/\sinh(14.69 m^T/m_e B)\). The dashed line in Fig. 1(b) shows \(d^2M/dB^2\) \(^7\) calculated from the LK formula with \(F = 660\) T, \(\phi = \pi/2\), \(m^* = 3.0m_e\), \(\ell = 400\) Å and a suitable scale factor. Note that this estimate of \(\ell\) assumes pure de Haas-van Alphen oscillations; any Shubnikov-de Haas component would imply a higher value. The model describes the data well, the decrease in amplitude with \(B\) arising from the weak \(B\) dependence of \(R_D\) at \(B \approx 70\) T and the factor of \(1/B^6\) in the third derivative. Note however that the non-monotonic \(B\)-dependence of the oscillation amplitude at \(T = 0.53\) K and \(T = 1.6\) K in Fig. 2 (but not in Fig. 1b due to the \(1/B^6\) factor) may be signs of beating between two close frequencies.

Fig. 2 shows \(\Delta f\), the TDO frequency minus a smooth monotonic background \(^9\), versus \(1/B\), at various temperatures. The oscillations are periodic in \(1/B\) as expected from quantized cyclotron orbits of Fermi-liquid-like quasiparticles. They are also damped rapidly at
higher $T$, consistent with thermal smearing of the FS. The agreement in frequency and phase with Fig. 1(b) shows that the oscillations are not an artefact of background subtraction.

In Ref. 3 it was pointed out that no hole pockets are present in a band calculation for O-II Y123 10. However small hole pockets of mainly chain character can be formed by allowing small shifts of the Fermi level $\Delta E_F \approx 25 \text{meV}$. Our observation of quantum oscillations in Y124, for which calculations find no small pockets near $E_F$ 11, suggests that the FS pockets are likely to be a general feature of the copper oxide planes of underdoped cuprates.

The insets to Fig. 1 show the $T$-dependence of $\mu_0 H_m$, the field of the well-defined peak in $|df/dB|$ and $\mu_0 H_{\text{hys}}$ where the hysteresis between the rising- and falling-field curves ceases to be detectable. $H_m$ is where vortex pinning becomes weak enough for the rf field to penetrate further than the London penetration depth (but still less than the normal state skin depth). In cuprate superconductors, vortex pinning becomes very small above an irreversibility field $H_{\text{irr}}$, which is usually much less than the estimated upper critical field $H_c2$, although these two fields may converge as $T \to 0$. Our values of $H_m$ are similar to $H_{\text{irr}}$ determined previously using torque magne-

The frequency determined from LK fits to the data in Fig. 2 and the peak positions in the Fast Fourier transform (FFT) spectra shown later in Fig. 3(a) both give $F = 660 \pm 15 \text{T}$. This corresponds to a FS pocket of only 2.4% of the Brillouin zone (BZ) area $\Delta_{BZ}$ ($\frac{\pi}{\sqrt{3}} \Delta_{BZ} = 27.9 \text{kT}$ for Y124 16). If we ascribe the oscillations to four hole-pockets as suggested for O-II Y123 3, the hole density $p_{QO} = 0.195 \pm 0.005$ compared to $p = 0.125 \pm 0.005$ estimated from the $a$-axis thermopower 17 18. For O-II Y123, the corresponding values of $p_{QO} = 0.152 \pm 0.006$ and $p = 0.1 \pm 0.01$ also differ by a factor 1.5. If antiferromagnetism 19 20 or other order doubles the unit cell, there would be only four half-pockets in the reduced BZ and $p_{QO}$ would be a factor 2 smaller. The same reduction in $p_{QO}$ is given by earlier calculations using the $t$-$J$ model 21. In both cases there is a discrepancy between $p$ and $p_{QO}$ but this is not an issue if both electron and hole pockets are present 22.

FIG. 2: Changes in resonant frequency $\Delta f$ of the tunnel-diode oscillator circuit versus $1/B$ recorded during 85 T pulses at various temperatures. A smooth monotonic background has been subtracted 3. The dotted lines are equally spaced in $1/B$. The oscillatory signal is periodic in $1/B$ with frequency $F = 660 \pm 15 \text{T}$.
m suppressed. The limited data raise the possibility that potential. It is an approximate value since there is no FS coupling BCS superconductor it is equal to 1. γ obtained from ARPES spectra of the parent Mott insulator for YBa$_2$Cu$_3$O$_{6+x}$ pseudogap energy scale measurements on many hole-doped cuprates suggest that the "special point" where heat capacity and other measurements on many hole-doped cuprates suggest that the pseudogap energy scale $E_G$ goes to zero. Fig. 4b shows the $p$-dependence of $E_G$ and the specific heat jump at $T_c$, for YBa$_2$Cu$_3$O$_{6+x}$ [24]. The latter is usually $\sim \gamma T_c$ where $\gamma$ is the Sommerfeld coefficient, for example for a weak coupling BCS superconductor it is equal to $1.43\gamma T_c$.

For Y124, every two-dimensional (2D) FS sheet in the BZ will give a contribution to $\gamma$ of $1.46m^*/m_e$ mJ mol$^{-1}$K$^{-2}$. This is independent of the number of carriers in the sheet and arises because in 2D both $\gamma$ and $m^*$ are proportional to the energy derivative of the FS area [20] multiplied by the same enhancement factor. Our value $m^* = 3.0 \pm 0.3m_e$ thus implies a contribution $\gamma = 4.4 \pm 0.4$ mJ mol$^{-1}$K$^{-2}$ for every 2D FS pocket of the observed frequency present in the BZ. An upper limit obtained from specific heat measurements of polycrystalline Y124 [24] is $\gamma = 9$ mJ mol$^{-1}$K$^{-2}$. This is a "normal state" value at $T = 0$K and zero field, obtained by applying an entropy conserving construction to $\gamma(T)$ from $T > T_c$ to $T \ll T_c$, and is consistent with the measured jump of $\gamma = 15$ mJ mol$^{-1}$K$^{-2}$ at $T_c$. If an estimated chain contribution of $3.5 \pm 0.5$ mJ mol$^{-1}$K$^{-2}$ is subtracted, this leaves a plane contribution $\gamma_{\text{plane}} = 5.5 \pm 0.5$ mJ mol$^{-1}$K$^{-2}$. Hence comparison of heat capacity data with our results casts doubt on the original model [1] involving four hole pockets near the $(\pm \pi/2, \pm \pi/2)$ points where photoemission (ARPES) experiments on underdoped crystals give evidence for Fermi arcs [27].

Four half-pockets of holes in a reduced BZ still give an electronic heat capacity that is a factor $\sim 8.8/5.5 = 1.6 \pm 0.2$ larger than the above estimate of $\gamma_{\text{plane}}$. Recent Hall effect measurements [22] suggest that the quantum oscillations may be due to a single electron pocket in the reduced BZ, centered at $(\pi,0)$. This would be consistent with $\gamma_{\text{plane}}$ but implies that the proposed hole pockets [22] only make a very small contribution to the heat capacity. In contrast to heavy fermion compounds, where the large heat capacity often suggested that quantum oscillations from the heavy electrons were not being detected in some of the early experiments, in the present case it is the small heat capacity that provides significant constraints to theoretical models for the FS pockets.
The Fermi energy ($E_F$) can be calculated if we assume that the FS sheets responsible for the oscillations are nearly 2D, that is, open in the $c$-axis direction. For a parabolic energy dispersion, we find $E_F = 295\text{K}$ for Y124 and 375 K for O-II Y123. Intriguingly these are of the same order as the pseudogap energies $Y_{124}$ and 375 K for O-II Y123. Note that these values of $E_F$ are consistent with the values of $p$ quoted earlier.

If the pockets of carriers are still present at lower fields and higher $T$, these low values of $E_F$ would lead to $T$-dependent diamagnetism, which although small, would be much more anisotropic than the spin susceptibility. This provides another means of testing theoretical models and making comparisons with ARPES data. Anomalous $T$-dependent magnetic anisotropy has been detected in the normal state of various cuprate superconductors and the similarity with Landau-Peierls diamagnetism in the organic conductor HMTSF-TCNQ has been noted.

In summary, we have observed quantum oscillations in the 80 K cuprate superconductor Y124 that have a larger orbit area than in O-II Y123, with $T_c$ of 57 K, and a considerably larger effective mass. Comparison with heat capacity data places strong constraints on the number of pockets present in the BZ, and supports models with a reduced BZ and small FS.

After completing the present measurements, we became aware of Hall resistivity results for YBa$_2$Cu$_4$O$_8$ giving values of $F$ and $m^*$ that agree with ours. JRC and EAY thank A. Carrington, S.M. Hayden, N.E. Hussey, J.W. Loram and J.L. Tallon for helpful discussions and collaboration and the EPSRC (U.K.) for financial support. This work is supported by DoE grants LDRD-DR-20070085 and BES Fieldwork grant, “Science in 100 T”. Work at NHMFL is performed under the auspices of the National Science Foundation, DoE and the State of Florida.

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