# Quantum Oscillations in the Underdoped Cuprate $YBa_2Cu_4O_8$

E. A. Yelland,[1,*] J. Singleton,[2] C. H. Mielke,[2] N. Harrison,[2] F. F. Balakirev,[2] B. Dabrowski,[3] and J. R. Cooper[4]

[1]*H. H. Wills Physics Laboratory, Tyndall Avenue, Bristol BS8 1TL, United Kingdom*
[2]*National High Magnetic Field Laboratory, MS-E536, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[3]*Department of Physics, Northern Illinois University, De Kalb, Illinois 60115, USA*
[4]*Cavendish Laboratory, J.J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom*

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We report the observation of quantum oscillations in the underdoped cuprate superconductor $YBa_2Cu_4O_8$ 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 ± 15 T and possible evidence for the presence of two components of slightly different frequency. The quasiparticle mass is $m^* = 3.0 \pm 0.3 m_e$. In conjunction with the results of Doiron-Leyraud *et al.* for $YBa_2Cu_3O_{6.5}$, 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 20 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 $CuO_2$ 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 $YBa_2Cu_3O_{6.5}$ (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 $YBa_2Cu_4O_8$ (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 quasiparticle 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 °C. Other crystals from the same batch were of high quality with a residual Cu-O chain resistivity $\leq 1\ \mu\Omega$ cm, and a low-$T$ thermal conductivity peak $\kappa_b(20\text{ K}) = 120\text{ W m}^{-1}\text{ K}^{-1}$ [6]. Pulsed magnetic fields up to 85 T were provided by the Los Alamos 85 T multishot magnet [7]. Measurements were made using a tunnel-diode oscillator (TDO) technique [8,9] 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 $0.35 \times 0.25 \times 0.12\text{ mm}^3$, which were stacked with their $c$-axis directions aligned within 2° 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 [10]. The sample and coil were immersed in $^3$He liquid or $^3$He exchange gas, temperatures ($T$) being measured with a Cernox thermometer 5 mm away from the sample.

Figure 1(a) shows the TDO frequency $f$ versus $B$ at $T = 0.53$ K. At $B \approx 45$ T, $f$ falls substantially indicating an

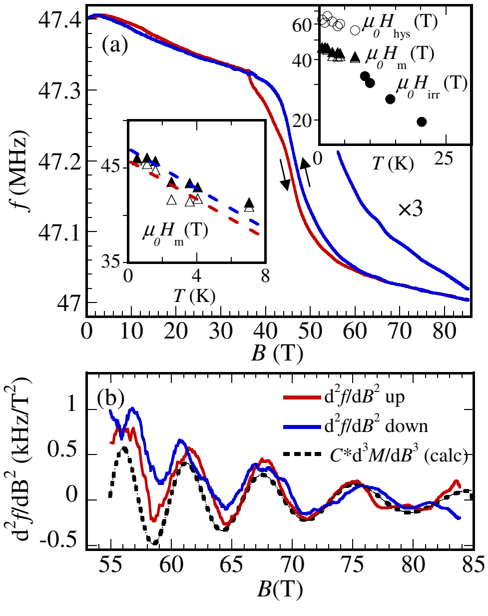

FIG. 1 (color online). (a) Resonant frequency $f$ of the TDO versus magnetic field $B$ recorded during an 85 T pulse at $T = 0.53$ K. Upper inset: $\mu_0 H_{\text{hys}}$ (∘) and $\mu_0 H_m$ (rising—△, falling—▲) from this work and $\mu_0 H_{\text{irr}}$ (•) from Ref. [16]. Lower inset shows expanded view of the $\mu_0 H_m$ data at low $T$. The dashed lines are guides to the eye. (b) Second derivative $d^2f/dB^2$ of 1.6 K data (solid lines). The dashed line is given by LK theory assuming $\Delta f \propto dM/dB$ [10], and using a suitable scale factor.

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 $M \propto B^{1/2}R_DR_T \sin(2\pi F/B + \phi)$ [11], 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 $F = (\hbar/2\pi e)A$ [11]; the scattering and temperature damping factors are, respectively, $R_D = \exp(-\pi\hbar k_F/e\ell B)$, where $k_F$ is the Fermi wave vector, $\ell$ is the mean free path and $R_T = (14.69m^*T/m_eB)/\sinh(14.69m^*T/m_eB)$. The dashed line in Fig. 1(b) shows $d^3M/dB^3$ [10] 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 $B \sim 70$ T and the factor of $1/B^6$ in the third derivative. Note, however, that the nonmonotonic $B$ dependence of the oscillation amplitude at $T = 0.53$ K and $T = 1.6$ K in Fig. 2 (but not in Fig. 1(b) due to the $1/B^6$ factor) may be signs of beating between two close frequencies.

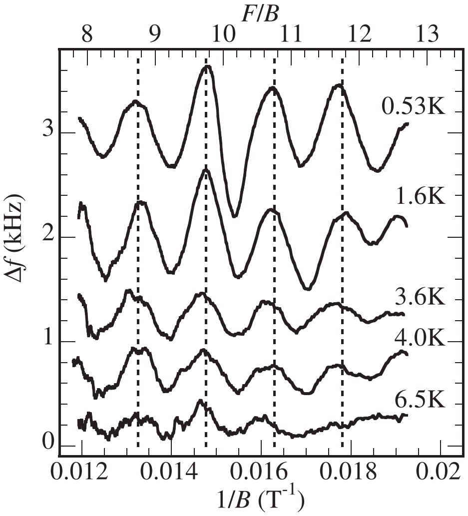

FIG. 2. Changes in resonant frequency $\Delta f$ of the tunnel-diode oscillator circuit versus $1/B$ recorded during 85 T pulses at various temperatures. A smooth monotonic background has been subtracted [13]. The dotted lines are equally spaced in $1/B$. The oscillatory signal is periodic in $1/B$ with frequency $F = 660 \pm 15$ T.

Figure 2 shows $\Delta f$, the TDO frequency minus a smooth monotonic background [13], 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. [1] it was pointed out that no hole pockets are present in a band calculation for O-II Y123 [14]. However, small hole pockets of mainly chain character can be formed by allowing small shifts of the Fermi level $\Delta E_F \approx 25$ meV [15]. Our observation of quantum oscillations in Y124, for which calculations find no small pockets near $E_F$ [15], 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 $\mu_0H_m$, the field of the well-defined peak in $|df/dB|$ and $\mu_0H_{\text{hys}}$, 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 magnetometry [16] on another crystal from the same batch (upper inset) and very recently from the resistivity of other Y124 crystals [12]. Somewhat unexpectedly we find that for the present crystal $\mu_0H_{\text{hys}}$ is 20 T larger than $\mu_0H_m$. The sudden onset of hysteresis at $B = 36$ T occurs when the insert magnet is energized. This and experiments in a faster-sweeping 65 T magnet show that the hysteresis increases with $|dB/dt|$. The lower inset shows our values of $\mu_0H_m(T)$ on a larger scale [17].

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 $F = 660 \pm 15$ T. This corresponds to a FS pocket of only 2.4% of the Brillouin zone (BZ) area $A_{\text{BZ}}$ ($\frac{\hbar}{2\pi e}A_{\text{BZ}} = 27.9$ kT for Y124 [18]). If we ascribe the oscillations to four hole-pockets as suggested for O-II Y123 [1], the hole density $p_{\text{QO}} = 0.195 \pm 0.005$ compared to $p = 0.125 \pm 0.005$ estimated from the $a$-axis thermopower [3,4]. For O-II Y123, the corresponding values of $p_{\text{QO}} = 0.152 \pm 0.006$ and $p = 0.1$ [1] 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 $p_{\text{QO}}$ would be a factor 2 smaller. The same reduction in $p_{\text{QO}}$ is given by earlier calculations using the $t$-$J$ model [21]. In both cases there is a discrepancy between $p$ and $p_{\text{QO}}$ but this not an issue if both electron and hole pockets are present [22].

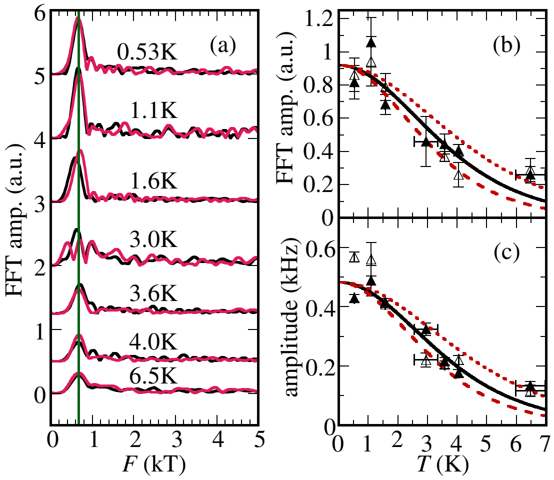

FIG. 3 (color online). (a) Fast Fourier transforms in $1/B$ of $\Delta f(B)$ over the range $60 < B < 85$ T. The red (black) lines show data for the rising (falling) part of the pulse. A single peak is present in the FFTs with a frequency $F = 660 \pm 15$ T. An extra, less-reproducible, peak near 200 T has been removed from some of the FFTs by subtracting a slowly varying background. (b) FFT amplitude versus $T$. Open (closed) symbols show rising (falling) field data. The solid line shows the LK damping factor $R_T$ with best-fit value $m^* = 3.0 \pm 0.3m_e$. (c) amplitude of oscillatory function of the form $\sin(2\pi F/B + \phi)$ with $F = 660$ T fitted to $\Delta f(B)$ in the range $67 < B < 77$ T. The best-fit $R_T$ curve, shown by a solid line, has $m^* = 3.1 \pm 0.3m_e$. Dashed lines show the LK formula for $m^* = 2.5m_e$ and $m^* = 3.5m_e$.

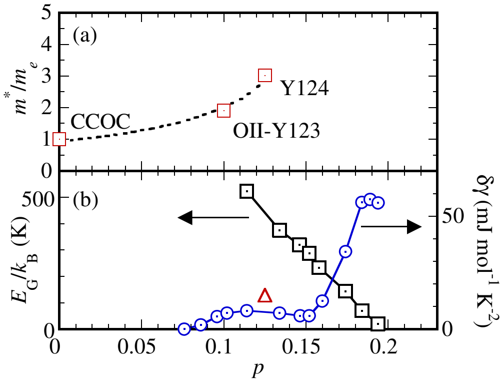

FIG. 4 (color online). (a) $m^*/m_e$ values (⊡) for Y124 (this work), O-II Y123 [1] and $Ca_2CuO_2Cl_2$ (CCOC), the latter from the dispersion of Cu-O orbital states well below the chemical potential measured by ARPES [23]. CCOC is a parent Mott insulator with $p = 0$ and no FS. The dashed line is a guide to the eye. (b) Heat capacity anomaly $\delta\gamma$ at $T_c$ for various $YBa_2Cu_3O_{6+x}$ samples (⊙) and the pseudogap energy $E_G$ (⊡) extracted from the same heat capacity data using a triangular gap model [24]. $\delta\gamma$ at $T_c$ is also shown (△) for Y124 [26].

FFTs are shown in Fig. 3(a) for all temperatures measured, for the rising and falling parts of the pulse. The amplitudes of the peak at 660 T were fitted to $R_T$ giving $m^* = 3.0 \pm 0.3m_e$ as shown in Fig. 3(b). Figure 3(c) shows the results of a separate LK analysis of the amplitudes of a sin curve fitted to the data between 67 and 77 T, giving $m^* = 3.10 \pm 0.3m_e$. Our best value is $m^* = 3.0 \pm 0.3m_e$.

Figure 4(a) shows the overall variation of $m^*/m_e$ with $p$ that is obtained by combining the present result with that of Ref. [1]. The value $m^*/m_e$ for $p = 0$ was obtained from ARPES spectra of the parent Mott insulator $Ca_2CuO_2Cl_2$ [23] for states well below the chemical potential. It is an approximate value since there is no FS and the usual Fermi liquid mass enhancement effects are suppressed. The limited data raise the possibility that $m^*/m_e$ could become very large as $p$ approaches 0.19, 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. Figure 4(b) shows the $p$ dependence of $E_G$ and the specific heat jump at $T_c$, for $YBa_2Cu_3O_{6+x}$ [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 [25] multiplied by the same enhancement factor due to electron-phonon and electron-electron interactions. 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 [26] 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 $\delta\gamma = 15$ 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 $\gamma_{\mathrm{plane}} = 5.5 \pm 0.5$ 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 $\gamma_{\mathrm{plane}}$. 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 $\gamma_{\mathrm{plane}}$ 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 $E_F = 295$ K for Y124 and 375 K for O-II Y123. Intriguingly these are of the same order as the pseudogap energies $E_G$ obtained from heat capacity and magnetic susceptibility [24,26] which are $E_G = 570 \pm 30$ K for O-II Y123 and $E_G = 360 \pm 25$ K for Y124. Note that these values of $E_G$ 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 [28].

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 $YBa_2Cu_4O_8$ [12] giving values of $F$ and $m^*$ that agree with ours.

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[*] Present address: School of Physics & Astronomy, University of St. Andrews, KY16 9SS, United Kingdom.

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