

de Haas–van Alphen Oscillations in the Underdoped High-Temperature Superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$

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(Received 22 November 2007; published 8 May 2008)

The de Haas–van Alphen effect was observed in the underdoped cuprate $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ via a torque technique in pulsed magnetic fields up to 59 T. Above a field of ~ 30 T the magnetization exhibits clear quantum oscillations with a single frequency of 540 T and a cyclotron mass of 1.76 times the free electron mass, in excellent agreement with previously observed Shubnikov–de Haas oscillations. The oscillations obey the standard Lifshitz-Kosevich formula of Fermi-liquid theory. This thermodynamic observation of quantum oscillations confirms the existence of a well-defined, closed, and coherent, Fermi surface in the pseudogap phase of cuprates.

DOI: [10.1103/PhysRevLett.100.187005](https://doi.org/10.1103/PhysRevLett.100.187005)

PACS numbers: 74.25.Jb, 74.25.Bt, 74.25.Ha, 74.72.Bk

Progress towards a more complete understanding of the high-temperature superconductors involves a few key questions, one of which is to understand the nature of the pseudogap phase in the underdoped region of the phase diagram. Angle-resolved photoemission spectroscopy (ARPES) shows an apparent destruction of the Fermi surface (FS): below the pseudogap temperature T^* , a gap opens up along the $(\pi, 0)$ direction, producing a set of disconnected Fermi arcs [1]. Recently, quantum oscillations, a clear signature of a Fermi-liquid ground state, were observed in the Hall resistance of $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ [2]. Since quantum oscillations are a direct consequence of the quantization of closed orbits in a magnetic field, this result suggests that the FS consists of small pockets, rather than Fermi arcs. Very recently, Shubnikov–de Haas (SdH) oscillations have also been observed in the stoichiometric compound $\text{YBa}_2\text{Cu}_4\text{O}_8$ [3,4], suggesting that they are generic to the CuO_2 planes rather than some feature of the band structure specific to $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ [3,5]. The frequency of the oscillations gives the area of the FS, but neither the location in k space nor the number of pockets are known. A comparison with ARPES measurements (assuming only half of each pocket is detected) suggests a four nodal pockets scenario [6]. However, when the density of carriers is estimated from the Luttinger sum rule, an obvious violation is found in both systems studied so far. In addition, the four nodal pockets scenario presumes that these pockets are holelike, in contradiction with the negative sign of the Hall effect observed in both systems in the normal state at low temperature [7]. An alternative scenario has been proposed whereby the FS of these Y -based cuprates consists of both electron and hole pockets, probably arising from a reconstruction of the FS

[7,8]. Moreover, it was suggested that, due to their higher mobility at low temperature, only the electron pocket is detected in quantum oscillations and no other frequency has yet been observed.

Quantum oscillations are due to the oscillation of the density of states of the quasiparticles in a magnetic field. The origin of the SdH effect (detected so far in $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$), i.e., quantum oscillations of the resistance, is basically due to an increase of scattering rate which occurs when a Landau level crosses the Fermi level, inducing a maximum in the resistance. Regarding the de Haas–van Alphen (dHvA) effect, i.e., quantum oscillations of the thermodynamic magnetization, it is a direct consequence of the oscillation of the free energy in magnetic field. In particular, it is not sensitive to quantum interference effects [9], which can contaminate the electrical resistance.

In this Letter, we report the first direct observation of dHvA oscillations in $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ at very low temperatures in magnetic fields up to 59 T, using a piezoresistive cantilever. We found a dHvA frequency of $F = 540 \pm 4$ T and a cyclotron mass of $m^* = (1.76 \pm 0.07)m_0$, in excellent agreement with the values reported in the SdH measurements, i.e., $F_{\text{SdH}} = 530 \pm 20$ T and $m_{\text{SdH}}^* = (1.9 \pm 0.1)m_0$. We also present the first study of the angular dependence of the SdH frequency, confirming that the oscillations arise from a quasi-2D Fermi surface. As all aspects of the data obey the standard Lifshitz-Kosevich (LK) theory, there is no indication of any deviation from Fermi-liquid theory.

We used a detwinned single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ flux grown in a nonreactive BaZrO_3 crucible. The dopant oxygen atoms were ordered into an ortho-II superstructure of

alternating full and empty CuO chains, yielding a superconducting transition temperature $T_c = 57.5$ K [10]. The sample used for the torque experiment is a small part ($140 \times 140 \times 40 \mu\text{m}^3$) of the sample studied in Ref. [2]. The Hall effect of the original piece was measured at different orientations of the magnetic field with respect to the CuO₂ planes, in order to study the angular dependence of the SdH frequency. Torque measurements were performed with a commercial piezoresistive microcantilever [11] down to 0.4 K at the LNCMP in Toulouse, using pulsed magnetic fields up to 59 T. The sample was glued with Araldite epoxy to the cantilever. A one-axis rotating sample holder allowed the angle ($\theta \sim 5^\circ$) to be varied between the normal to the CuO₂ planes and the magnetic field at ambient temperature. The cantilever was set inside a vacuum tight resin capsule filled at room temperature with ⁴He gas to ensure thermalization of the sample. This capsule sits in the ³He/⁴He mixture of the dilution fridge. The temperature gradient between the sample located at the end of the cantilever and the thermometer located in the mixing chamber was estimated by measuring the critical field of a known compound under the same experimental conditions. The temperature gradient is about 0.2 ± 0.1 K at 0.4 K and negligible above 1 K. This uncertainty does not affect the value of the physical parameters extracted from the data. The variation of the piezoresistance of the cantilever is measured with a Wheatstone bridge with an ac excitation at a frequency of 63 kHz (see lower inset of Fig. 1).

Raw data of torque $\tau = |\vec{M} \times \vec{B}|$ versus magnetic field are shown in Fig. 1 for the up and down field sweeps between $T = 4.1$ K and $T = 0.4$ K. As observed in the mixed state of other type-II superconductors at low field

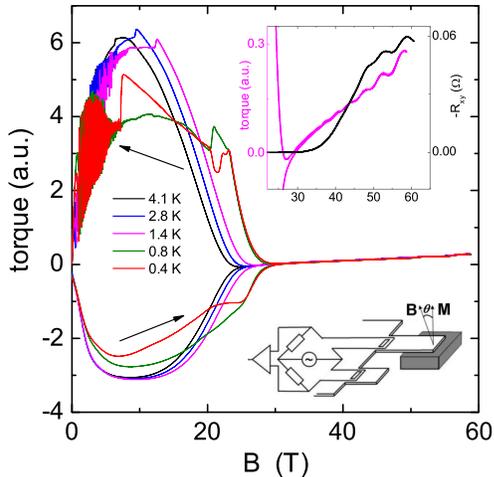


FIG. 1 (color online). Torque versus magnetic field at different temperatures. Both up (50 ms) and down (250 ms) field sweeps are shown, as indicated by the arrows. Lower inset: Schematic of the cantilever. Upper inset: Magnetic field dependence of the torque ($\theta \simeq 5^\circ$) at $T = 1.4$ K and of the Hall resistance ($\theta = 0^\circ$) measured in the same sample at $T = 1.5$ K.

[12], there is a strong hysteresis effect, which reflects the penetration (expulsion) of magnetic field into (out of) the sample when the magnetic field increases (decreases). In this regime, “flux jumps” are clearly observed and are associated with a thermal instability corresponding to a reorganization of the magnetic flux as it enters (exits) the sample [12,13]. The hysteresis disappears above B_{irr} and the torque becomes almost linear with magnetic field. B_{irr} marks the field where the electrical resistance starts to rise and corresponds to the vortex liquid phase. The upper inset of Fig. 1 shows the torque and the (normalized) Hall resistance versus magnetic field measured in the same sample at $T \simeq 1.5$ K. Above a field of ~ 30 T, clear dHvA oscillations are observed [14]. We have checked that the amplitude of oscillations is identical for the up and down field sweeps, which shows that no detectable self-heating occurs in the sample during the measurement.

In the following, we will discuss only the oscillatory part of torque, obtained by subtracting a monotonic background from the raw data. The amplitude of the first harmonic of the dHvA oscillations is interpreted using the LK theory for a two-dimensional Fermi liquid:

$$\tau_{\text{osc}} = BA \tan \theta \sin \left[2\pi \left(\frac{F}{B} - \gamma \right) \right], \quad (1)$$

where F is the oscillation frequency and γ is a phase factor. We neglect any contribution from magnetic breakdown, spin damping, or any additional damping coming from the effect of superconductivity on the dHvA amplitude. The amplitude of the fundamental frequency is given by $A \propto R_T R_D$, where R_T and R_D are the thermal ($R_T = \alpha T m^* / B \sinh[\alpha T m^* / B]$) and Dingle ($R_D = \exp[-\alpha T_D m^* / B]$) damping factors, respectively, where $\alpha = 2\pi^2 k_B m_0 / e \hbar$ ($\simeq 14.69$ T/K) and T_D is the Dingle temperature [14].

Figure 2 displays the temperature dependence of the amplitude of the oscillations (“mass plot”) for different field windows. The usual behavior expected for thermal damping of the dHvA oscillations is observed. Solid lines are fits of Eq. (1), from which we deduce an effective mass $m^* = (1.76 \pm 0.07)m_0$, in agreement with the value obtained from Shubnikov–de Haas measurements [2]. No significant variation of the effective mass with magnetic field is observed, which confirms that the LK model is adequate to describe the data. Using $v_F = \hbar k_F / m^*$ and the Onsager relation (assuming a circular orbit) $\pi k_F^2 = 2\pi^2 F / \Phi_0$, the average Fermi velocity is 8.4×10^4 m s⁻¹. This value is in fact very similar to that found by ARPES at the antinode in both La_{2-x}Sr_xCuO₄ [15] and Bi₂Sr₂CuO_{6+δ} [16]. This may indicate that the pockets detected by dHvA are not located at the same position in k space as the Fermi arcs seen by ARPES [1].

Figure 3(a) displays the oscillatory torque versus $1/B$ between $T = 5.2$ K and $T = 0.4$ K. At the lowest temperatures, 8 periods can be resolved, corresponding to the

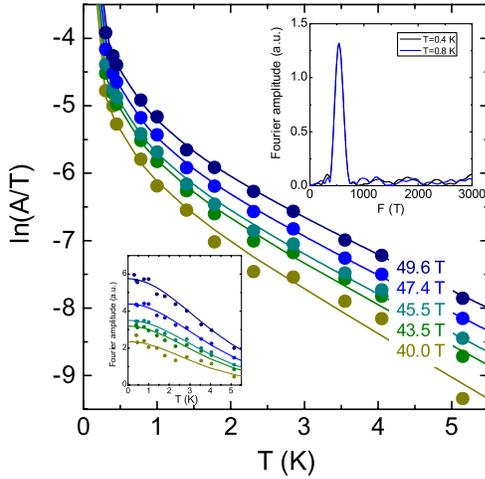


FIG. 2 (color online). Main panel: Temperature dependence of the oscillation amplitude A (where A is determined from discrete Fourier analysis with a Blackmann window) plotted as $\ln(A/T)$ versus T for various mean magnetic field values. Solid lines are fits to Eq. (1) with $m^* = 1.76m_0$. Lower inset: Same data plotted as A versus temperature. Upper inset: Fourier transform of the oscillatory part of the torque at $T = 0.4$ K and $T = 0.8$ K between 35 and 58 T, revealing a single frequency $F = 540$ T.

Landau levels $n = 9$ to $n = 16$. Oscillations can be observed down to a field of ~ 30 T, in particular, in the resistive superconducting transition (see upper panel of Fig. 1). Figure 3(b) displays the corresponding field dependence of the amplitude of the dHvA oscillations divided by the R_T factor (“Dingle plot”). Within our

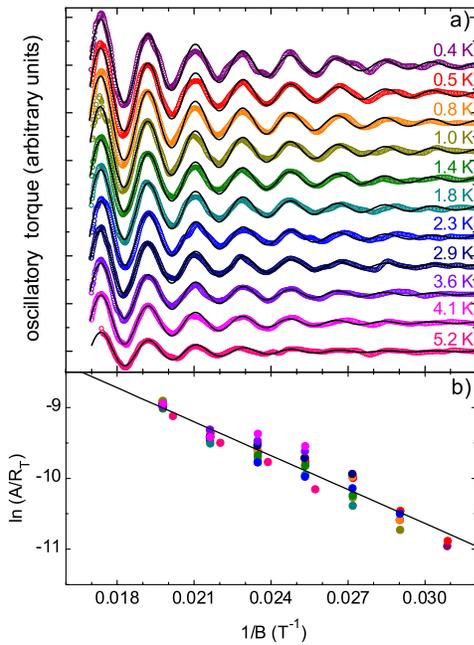


FIG. 3 (color online). (a) Field dependence of the oscillatory part of the torque for the various temperatures explored and (b) corresponding Dingle plots. Solid lines are best fits to Eq. (1) with $m^* = 1.76m_0$ and $T_D = 6.2$ K.

experimental resolution, it is not possible to resolve an extra attenuation corresponding to the effect of superconductivity on the dHvA amplitude [17,18]. By neglecting any additional damping factor coming from the mixed state, the solid line in Fig. 3(b) yields an upper limit of $T_D = 6.2 \pm 1.2$ K [using $m^* = (1.76 \pm 0.07)m_0$], which converts to $\omega_c\tau = 0.7 \pm 0.2$ at $B = 35$ T. Assuming a cylindrical FS, we can deduce a mean free path $l = 16$ nm. Black solid lines in Fig. 3(a) are fits to Eq. (1) obtained by setting $m^* = 1.76m_0$ and $T_D = 6.2$ K for all temperatures. The deduced oscillation frequency and phase factor are $F = 540 \pm 4$ T and $\gamma = 0.15 \pm 0.05$, respectively. This fitting procedure confirms that the LK formula, which describes the dHvA oscillations for a 2D Fermi liquid, is appropriate. This conclusion is at odds with recent theories that invoke mechanisms other than the Landau quantization of quasiparticles [19,20].

Figure 4(a) shows the oscillatory Hall resistance versus magnetic field measured in the same sample at $T = 3$ K for different angles θ . Several measurements have been done at different temperatures for each orientation, from which we have deduced an oscillation frequency F plotted in Fig. 4(b). Within the error bars, F increases as $1/\cos(\theta)$ as expected for a quasi-2D part of the FS [14]. It simply reflects the increase of the area of the cyclotron orbit when the magnetic field is tilted from the normal of the CuO_2 plane.

dHvA measurements have been widely used to study the FS of metals [14]. As a thermodynamic measurement, it firmly confirms the existence of well-defined quasiparticles at the FS with a substantial mean free path. Quantum oscillations are detected with torque starting at a field corresponding to the onset of the electrical resistance, but there is no clear evidence in the amplitude of the oscillations of a transition from the mixed state to the normal state [21]. It may be due to the fact that, in order to resolve an additional damping, the dHvA signal has to be measured in a field range much larger than in Fig. 3(b). dHvA oscillations have been observed in the mixed state of many superconductors [17,18,22–25], and in all cases the oscil-

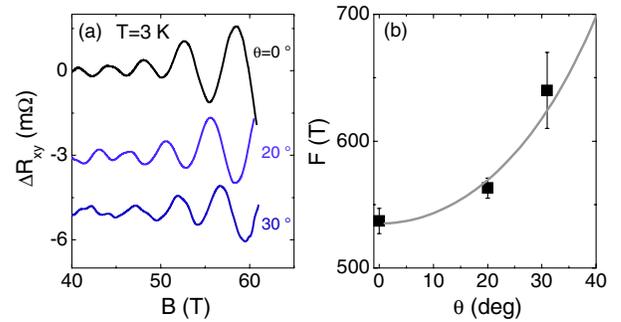


FIG. 4 (color online). (a) Oscillatory part of the Hall resistance versus magnetic field at different angles at $T = 3$ K. (b) Angular dependence of the deduced Shubnikov–de Haas frequency. Solid line is a fit to $F(\theta = 0)/\cos(\theta)$.

lations have the same frequency as in the normal state. In particular, dHvA and SdH effects have been observed in the organic superconductor κ -(BEDT-TTF)₂Cu(NCS)₂ [26] which shares many characteristics with high T_c superconductors such as low dimensionality and short coherence length, but which has a much lower H_{c2} [27–29]. The striking similarity between the data in κ salt (see Fig. 2 of Ref. [26]) and that in the upper inset of Fig. 1 suggests that the normal state could be reached around our maximum field in YBa₂Cu₃O_{6.5}. This value is lower than the one given by the extrapolation of the Nernst data to higher fields [30], which suggests that H_{c2} could be as large as 150 T (corresponding to the field scale associated with the short coherence length in cuprates). However, one should keep in mind that in such a layered superconductor there is no true phase transition between the vortex liquid and the normal state, but a crossover with an extended range of fluctuating superconductivity [31]. In this crossover regime, one can expect the electrical resistance to be representative of the normal state.

Finally, the frequency $F = 540$ T converts to a carrier density 0.038 carrier per planar Cu atom. Independently of whether there are 2 or 4 pockets, it gives a number of carriers which is not in agreement with the doping level (10%). However, a scenario based on a (π, π) reconstruction of the FS can explain both the negative Hall effect (electron pocket) in the normal state [7] and the apparent violation of the Luttinger sum rule. It assumes that the frequency observed with SdH and dHvA effects corresponds to an electron pocket, whose mobility is much higher than that of a larger hole pocket not seen in the measurements. To validate this scenario, however, the detection of another frequency corresponding to a total hole density of 0.138 hole per planar Cu atom is still required.

In summary, we have observed the first direct evidence of dHvA oscillations in YBa₂Cu₃O_{6.5}. The frequency and the effective mass are in excellent agreement with previous SdH measurements in the same compound. This observation confirms the existence of a coherent closed Fermi surface at low temperature in the underdoped side of the phase diagram of cuprates and suggests that the ground state obeys Fermi-liquid theory.

We thank K. Behnia, N. Hussey, and G. Rikken for useful discussions. We acknowledge support from EuroMagNET under EC Contract No. 506239, the Canadian Institute for Advanced Research, the ANR project ICENET, and funding from NSERC, FQRNT,

EPSRC, and the Canada Research Chairs Program.

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- [1] M. R. Norman *et al.*, Nature (London) **392**, 157 (1998).
- [2] N. Doiron-Leyraud *et al.*, Nature (London) **447**, 565 (2007).
- [3] A. Bangura *et al.*, Phys. Rev. Lett. **100**, 047004 (2008).
- [4] E. Yelland *et al.*, Phys. Rev. Lett. **100**, 047003 (2008).
- [5] A. Carrington and E. A. Yelland, Phys. Rev. B **76**, 140508 (2007).
- [6] S. R. Julian and M. R. Norman, Nature (London) **447**, 537 (2007).
- [7] D. LeBoeuf *et al.*, Nature (London) **450**, 533 (2007).
- [8] S. Chakravarty, Science **319**, 735 (2008).
- [9] R. W. Stark and C. B. Friedberg, Phys. Rev. Lett. **26**, 556 (1971).
- [10] R. Liang *et al.*, Physica (Amsterdam) **336C**, 57 (2000).
- [11] E. Ohmichi and T. Osada, Rev. Sci. Instrum. **73**, 3022 (2002).
- [12] M. M. Mola *et al.*, Phys. Rev. Lett. **86**, 2130 (2001).
- [13] L. Legrand *et al.*, Physica (Amsterdam) **211C**, 239 (1993).
- [14] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, England, 1984).
- [15] T. Yoshida *et al.*, J. Phys. Condens. Matter **19**, 125209 (2007).
- [16] A. Kaminski *et al.*, Phys. Rev. B **71**, 014517 (2005).
- [17] T. J. B. M. Janssen *et al.*, Phys. Rev. B **57**, 11 698 (1998), and references therein.
- [18] J. D. Fletcher *et al.*, Phys. Rev. B **70**, 144501 (2004), and references therein.
- [19] A. S. Alexandrov, J. Phys. Condens. Matter **20**, 192202 (2008).
- [20] A. Melikyan and O. Vafek, arXiv:0711.0776.
- [21] K. Yasui and T. Kita, Phys. Rev. B **66**, 184516 (2002), and references therein.
- [22] J. E. Grabner and M. Robbins, Phys. Rev. Lett. **36**, 422 (1976).
- [23] F. Mueller *et al.*, Phys. Rev. Lett. **68**, 3928 (1992).
- [24] C. Bergemann *et al.*, Physica (Amsterdam) **230–232B**, 348 (1997).
- [25] N. J. Clayton *et al.*, Phys. Rev. B **65**, 064515 (2002).
- [26] T. Sasaki *et al.*, Phys. Rev. B **67**, 144521 (2003).
- [27] S. Belin *et al.*, J. Supercond. **12**, 497 (1999).
- [28] T. Sasaki *et al.*, Phys. Rev. B **57**, 10 889 (1998).
- [29] R. Lortz *et al.*, Phys. Rev. Lett. **99**, 187002 (2007).
- [30] Y. Wang *et al.*, Science **299**, 86 (2003).
- [31] F. Rullier-Albenque *et al.*, Phys. Rev. Lett. **99**, 027003 (2007).