de Haas–van Alphen Effect and Fermi Surface of $YBa_2Cu_3O_{6.97}$

C. M. Fowler, B. L. Freeman, W. L. Hults, J. C. King, F. M. Mueller, and J. L. Smith
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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The component of the magnetic susceptibility periodic in $1/B$, the de Haas–van Alphen effect, has been investigated using a 100-T flux compression system. It is concluded from these measurements that $YBa_2Cu_3O_{6.97}$ exhibits a Fermi surface with at least three separate extremal frequencies for the c axis parallel to the applied field. Orbital masses, renormalization λ's, and scattering rates are estimated from an amplitude analysis.

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A direct ground-state measurement of the Fermi surface of Y-Ba-Cu-O has been called for a number of times [1–3]. In this Letter, we present measurements of the de Haas–van Alphen (dHvA) effect using 100-T fields and temperatures below 4.2 K. The basic data obtained are the oscillatory part of the YBCO magnetization as a function of applied field. As noted below, at least three well-defined frequencies have been deduced from these data. We have reported preliminary results at conferences [4], and recently there has been a report of dHvA oscillations for YBCO by Kido *et al.* using fields up to 27 T [5]. As discussed below, their oscillatory frequency of 0.54 kT is virtually identical to our lowest observed frequency.

Our experiments are carried out in large pulsed fields, and this places a number of constraints on the preparation of the samples as well as the measurement techniques. The samples are in the form of fine-grained powders embedded in a nonconducting medium so that pulsed magnetic fields can easily penetrate between and into the grains during the time of the pulse. The $YBa_2Cu_3O_{6.97}$ (YBCO) samples exhibited a T_c onset of 93 K. The technique used to prepare and characterize the YBCO samples has been discussed [6]. Superconducting powder was well stirred into epoxy and allowed to harden for 12 h in a 4.2-T field, oriented along the cylindrical axis [7]. This technique produced samples which maintained a c-axis orientation along the magnetic-field axis to better than 2° as seen by x rays and NMR. The sample density (17% by volume) is well below the percolation threshold (roughly 31%), as confirmed by scanning electron microscopy. We have considered penetration retardation by eddy current effects in some detail [8]. The thermal rise due to these effects in a typical dHvA experiment is estimated to be no bigger than 0.2 K for our YBCO powder.

The magnetization signals were detected by measuring the voltage developed across a multiturn pickup coil wound on the samples. Because these signals were much smaller than the dB/dt voltage generated by the magnetic-field pulse, a similar "empty" coil containing an inert sample was connected in opposition to the sample "full" coil. Each coil consisted of a single layer of about 100 turns of 30-μm-diam insulated copper wire. The full pure sample was machined from epoxy impregnated with YBCO powder while the empty coil was machined from straight epoxy. Both samples were accurately machined to 1 mm diameter ×4.5 mm length. Test experiments showed that a typical coil pair, with leads, was balanced to within one turn over the duration of the 100-T pulse. Still, even with only one equivalent uncompensated turn, the amplitude of the dB/dt signal from the coil pair could be substantially larger than the magnetization signals. However, the frequency spectrum of dB/dt itself has little content over 100 kHz and virtually none above 550 kHz. The output of the coil pair was, therefore, fed through a two-stage high-bandpass filter centered at 550 kHz into a 50-Ω terminated load. The use of carefully balanced coils, together with the passive filter, eliminated most signals arising from dB/dt of the field pulse. At the same time analysis of the true magnetization signals analyzed showed that they were largely unaffected by the filter.

The 100-T fields were obtained from explosive-driven flux compression generators [9]. Experimental volumes are unusually large for 100-T fields. For these experiments, the field coil is made by machining a 16-mm-diam hole in brass bar stock, 73 mm long. The magnetic fields are monitored by measuring dB/dt developed in a separate coil, of known area. Field measurements are accurate to within (1–2)%. Computer simulations of this flux compressor system show [10] that the fields produced have homogeneity of about 3 parts in 10^4, for all times, over the region occupied by the sample and empty coil volume in the central portion of the brass load block. At a nominal 100-T peak field the inhomogeneity would be about 0.03 T. Estimating the effect of such an inhomogeneity, we find that the dHvA amplitude is diminished by no more than about a factor of 2 in these experiments. Thus, our system has the high fields, low temperatures, easy flux penetration, high-field homogeneity, and oriented, high-quality samples necessary as prerequisites for dHvA measurements.

Electrical signals were monitored by a battery of Tektronix 485 analog scopes and several Tektronix 602 digital scopes. The dM/dt signals developed across the 50-Ω load, such as those used to obtain the solid curve in Fig. 1, were recorded on a number of digital channels run at 500-MHz sampling rate for somewhat over 20 μs per channel (10 240 points per channel).

The entire dHvA system was tested in separate 4-K experiments using 1-μm-diam single-crystalline, unoriented

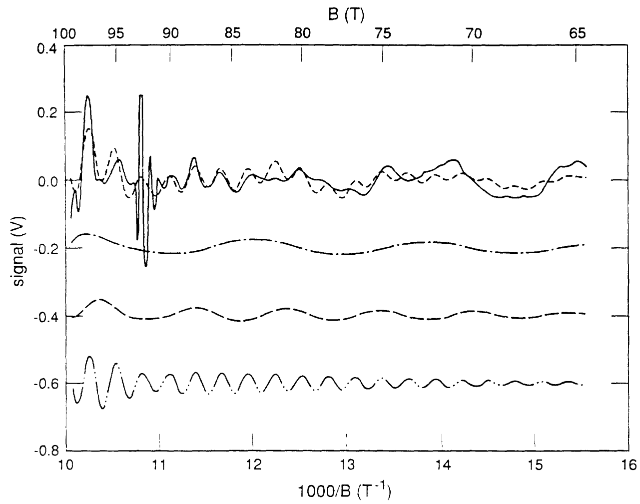

FIG. 1. Typical YBCO dM/dt dHvA pickup coil signal as a function of $1/B$. These data are for a 2.8-K experiment. As discussed in the text the dashed line has been derived from three LK frequencies. These are shown separately displaced for the 0.53-, 0.78-, and 3.51-kT orbital frequencies as the dash-dotted, long-dashed, and dash–double-dotted lines, respectively.

copper powders mixed into BN insulating 0.5-μm powder. The copper powder was about 20% by volume. These separate experiments taken in fields larger than 80 T showed a main dHvA frequency peak of 2.1 kT and amplitudes (63 mV) within 30% of Lifshitz-Kosevich (LK) [11,12] simulations as we have described in Ref. [8]. The $1/B$ Fourier transform of these data described below is presented as an inset to Fig. 2. The agreement of this frequency with the well-known copper neck frequency, 2.1 kT, lends confidence to the technique employed here. The width of the transition is larger because fewer oscillations were used.

In Fig. 1 we present typical YBCO data as the solid line for a temperature of 2.8 K plotted as a function of $1/B$, where B is the magnetic induction. An examination of the data suggests that oscillations of several spectral features exist on a scale of tens of millivolts. To probe spectral content, we have taken a Fourier transform of the pickup coil voltage shown in Fig. 1. We plot the spectral density dissipated in the 50-Ω load in Fig. 2 as the shaded area. The integral under this curve is the energy dissipated in joules. Although the spectral shapes in the YBCO and copper experiments are very different, the integrated densities are rather similar. Cooling the YBCO system makes the signal voltages larger, following typical dHvA experience. Our results have a vertical precision of about 2%. One spurious feature that appears to some degree in all of our experiments is the structure centered about 93 T in Fig. 1. Tests have shown that this structure is due to the mechanics of the flux compressor itself. The amplitude of the structure varies with the degree of coil pair imbalance, but only lasts for about 1 μs. In our Fourier transforms and wave-shape analysis, we have simply treated this structure as "data." Its delta-function-like nature does not greatly affect Fourier-transform peaks.

It is clear from an examination of Fig. 2 that the several separate features seen in the shaded spectral density may be harmonically related. To check this possibility, we have fitted the data of Fig. 1 with the dHvA ampli-

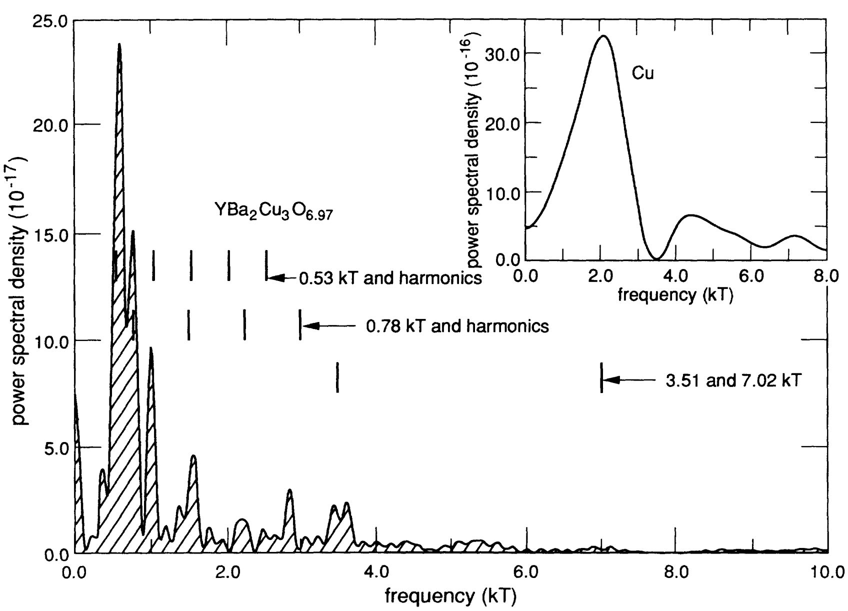

FIG. 2. The $1/B$ Fourier transform of the pickup coil signal. Peaks in the power spectral density as a function of $1/B$ probe possible cyclotron extremal cross-sectional frequencies of the dHvA effect. As discussed in the text and in Table I, extremals of 0.53, 0.78, and 3.51 kT have been extracted from a LK amplitude analysis. Inset: Data on unoriented 1-μm copper powder taken in fields above 80 T. The neck frequency of Cu is 2.1 kT.

tude of a LK form [11]. The spectral densities of several different YBCO dHvA measurements, as well as the one shown in Fig. 2, show that there were three independent base frequencies, 0.53, 0.78, and about 3.51 kT, together with several harmonics. We have used these base frequencies and have varied three other LK parameters for each frequency: the orbital mass m^*, the Dingle temperature or scattering rate, T_D, and the phase ϕ. Only a single amplitude constant was permitted for all frequencies since the same coil calibration applies to all of the frequencies and since the amplitudes of the harmonics are determined from that of each fundamental. The LK amplitude was constructed for a given choice of parameters. The dM/dt was calculated numerically. The amplitudes for the separate frequencies were added and rms differences compared with the dHvA signals. All parameters other than the frequencies were arbitrarily set initially to 1, 5, or 10 in separate minimization runs. The rms fitting error was minimized by making tiny ($\delta=0.005$) steps in each of the parameters for each of the three frequencies. We have not tried to fit other possible frequencies such as the small feature at 1.8 kT in Fig. 2 because we do not think such fitting would be statistically significant. If arbitrary frequencies were used, the fitting procedure sometimes made two frequencies identical. We conclude that three independent frequencies and their LK harmonics are well supported by these data.

After only a few cycles, independent of initial starting values, the three frequency parameters arrived at converged values. This was true even if we tried separate one-, two-, or three-frequency fits. One should note that in this wave-shape amplitude method, all of the dHvA harmonics are being simultaneously fitted. If we used values from Fig. 2, the minimization smoothly and systematically approached the same converged values. The reader will notice that the procedure we have employed here is an extremal in the deferred limit concept, slowly approaching the global minimum in the parameter space. We have found that this procedure of taking a multiplicity of very tiny parameter steps in a perhaps severely nonlinear variational problem is superior to other and more complicated minimization techniques. In Fig. 1 we have presented separately the amplitude for each of the three frequencies as well as a comparison with their sum. Note that each separate LK amplitude for each of the three frequencies includes the sum of many LK harmonics. The amplitude of each higher harmonic is exponentially smaller. These ratios of harmonic amplitudes chiefly determine m^* and T_D for a fixed temperature. The parameters derived from different YBCO dHvA experiments and temperatures were similar. After cycling, the "warmest" 4-K data had about 3 times the rms fitting error of the 2.8-K data and a corresponding worse signal-to-noise ratio. Nevertheless the same three independent

TABLE I. YBCO experimental and theoretical Fermi-surface areas and masses (LANL is this work; ANL-NW and NRL-WM are theoretical band-structure results from Refs. [13] and [14], respectively). The cyclotron frequencies ω_c are for a 100-T field. The scattering times are τ. The λ is the renormalization of the cyclotron mass. The different values from Ref. [13] reflect small changes between 1987 and 1991.

Piece	Area (kT)	Masses (m_e)	Dingle (K)	Shape	ω_c (THz)	τ (ps)	λ
Pill (small cylinder at $k_z=0$)							
LANL	0.53 ± 0.02	7.0 ± 2.5	1.7 ± 0.6	Min.	2.5	0.7	
ANL-NW	0.40	−2.1					2.3
	1.24	−3.2					1.2
NRL-WM	0.17	−1.6					3.4
Pill (small cylinder at $k_z=\pi/c$)							
LANL	0.78 ± 0.02	7.2 ± 2.5	2.1 ± 0.7	Max.	2.4	0.6	
ANL-NW	0.42	−2.3					2.1
	1.25	−3.8					0.9
NRL-WM	0.22	−1.7					3.2
Neck (1D Cu chain in $k_z=\pi/c$)							
LANL	3.51 ± 0.10	7.4 ± 2.6	3.4 ± 1.2	Min.	2.4	0.4	
NRL-NW	3.70	1.45					4.1
Barrel (1) (Cu plane)							
LANL	Unobserved						
ANL-NW	12.05	−1.5					
	12.70	−1.3					
NRL-WM	12.41	−1.4					
Barrel (2) (Cu plane)							
LANL	Unobserved						
ANL-NW	12.68	−1.6					
	13.84	−1.9					
NRL-WM	12.57	−1.5					

frequencies emerged. The signal-to-noise ratio was about 10:1 for the low-temperature data.

A number of theoretical models which allow for a Fermi surface in high-temperature superconductors (HTSC) have been proposed and reviewed [1]. Here we consider the relationship of band models and local-density-functional theory to these data. To a great degree all HTSC band calculations are in excellent agreement on a scale of a few mRy. In Table I, we present our results and the results of Fermi-surface predictions by Yu *et al.* [13] and by Pickett, Cohen, and Krakauer [14].

If we compare our results with the theoretical band-structure cross sections, a considerable agreement emerges. Since the open small sheet cylinders are oriented along the c or z axis, the crystallographic group allows at least two extremal cross sections per sheet. (One is the Γ-X-S-Y plane and one is in the T-R-U-Z plane [13,14].) We compare in Table I the ratio of experimental to theoretical masses in order to extract renormalization λ's from the relation $|m^*|=(1+\lambda)|m_{\mathrm{band}}|$. As is well known, dHvA masses are harder to extract than extremal cross sections and hence have less accuracy. The electronic cyclotron masses found are strongly renormalized, but not to an extraordinary degree. The renormalization λ's are in the range of 1-4. Although some of these λ's are comparable with those of $A15$ materials, the larger λ's are consistent with the higher superconducting critical temperature of YBCO using Nambu-Eliashberg theory [15]. We have not yet observed the larger barrel orbits predicted by the band calculations of both theoretical groups. We plan to use a 200-T system to search for these higher cyclotron frequencies.

The scattering times τ are derived from the Dingle temperatures T_D. In making this comparison we have implicitly assumed that all of T_D is due to impurities or dislocations. Some of T_D may be due to field inhomogeneity and phase smearing effects. Nevertheless the evidence is that electrons will be able to complete one orbit before scattering ($\omega_c\tau\approx 1$). This internally consistent independent evidence demonstrates that a dHvA effect should be visible. Note that the scattering time of the neck is about twice as large as that of the pill and that this orbit derived from the 1D copper chain structures is more sensitive to the formation of twins, which is consistent with the higher T_D in Table I. We plan to make a similar analysis on the barrel sheets when measured to make intercomparisons.

We conclude that YBCO has a Fermi surface consistent with local-density-approximation predictions that the renormalization λ's are consistent with Nambu-Eliashberg predictions of T_c, and that the internal evidence of the scattering rates and cyclotron frequencies allow the dHvA orbits to be observed.

Many colleagues have contributed greatly to the success of these measurements over the last three years. This work was performed under the auspices of the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences. This work was performed using the Pulsed Magnetic Field Facility of the National High Magnetic Field Laboratory at Los Alamos National Laboratory.

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