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Quantum Interference in the Spin-Polarized Heavy Fermion Compound CeB₆: Evidence for Topological Deformation of the Fermi Surface in Strong Magnetic Fields

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We report susceptibility experiments in magnetic fields up to 60 T and show conclusively that the de Haas–van Alphen oscillations originate from a single spin Fermi surface. New observed frequencies present in magnetic breakdown and quantum interference oscillations indicate that the Fermi-surface topology changes as a function of the applied field while maintaining a constant volume. These results are not expected from the Anderson lattice model. [S0031-9007(98)06703-9]

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Susceptibility experiments in pulsed magnetic fields have recently shown that Stark quantum interference (QI) effects can be detected in the magnetization arising from induced eddy currents in metallic LaB₆ [1]. QI is an effect unique to the magnetotransport of metals exhibiting magnetic breakdown (MB), and leads to oscillation frequencies in the magnetoresistance that are *not* present in Landau quantum oscillatory phenomena such as the de Haas–van Alphen (dHvA) effect [2–4]. In this Letter, we report the results of extending QI and MB measurements to CeB₆, which is known to have a Fermi surface (FS) similar to LaB₆ [5]. We use the results of the present measurements on CeB₆ to detect FS changes as a function of magnetic field that are not otherwise observable.

The only significant difference between the FS topologies of LaB₆ and CeB₆ is that the latter possesses $4f$ electrons that interact with the conduction electrons to result in a heavily renormalized effective mass [5–8]. The effective mass becomes radically quenched by the application of a strong magnetic field [6], going from $\sim 30m_e$ (where m_e is the free electron mass) at 5–7 T [9,10] to $\sim 3m_e$ at 50 T [11,12]. This behavior has been discussed in connection with the Anderson lattice model for the heavy fermion (HF) ground state [8,12]. A small but detectable change in the frequency α_3 , corresponding to the intersection of the large α electron ellipsoid with the XMR plane of the Brillouin zone (BZ), has also been reported [10,13], though has not been explained.

MB and QI occur within the ΓXM plane when the field is oriented along the $\langle 100 \rangle$ crystallographic axis [11,12]. Apart from the novelty of observing the BZ QI frequency with an effective mass of *zero* in a HF system, the numerous combination frequencies that are observed enable the sizes of all FS sheets and pockets to be inferred (at least for the down-spin sheet). We find that in several respects, the behavior of the dHvA oscillations is consistent with the theory for the dHvA effect in HF systems developed by Wasserman *et al.* [8]. This includes the field-induced quenching of the effective mass together with the reduc-

tion of the α_3 frequency, as well as a clear indication that only quasiparticles of a single spin contribute to the dHvA effect (as revealed by the behavior of the harmonics at high magnetic fields and the lack of spin zeros with changing mass [10]). Other aspects of quantum oscillations nevertheless reveal the limitations of this theory. In particular, the MB and QI frequencies observed in this work indicate that there is a notable deformation of the FS as a function of the magnetic field, suggesting that the interactions, which give rise to the HF ground state, are anisotropic. In spite of this change in FS topology, the overall volume of this spin-polarized sheet remains unchanged.

The single-crystal samples used for this study were grown in aluminum flux and etched down to dimensions of < 0.2 mm to minimize the effects of eddy current heating. Pulsed magnetic fields of up to 60 T were provided by the National High Magnetic Field Laboratory (NHMFL), Los Alamos, while temperatures down to 330 mK were obtained using a ³He cryostat. Additional experiments in static magnetic fields of up to 30 T using field modulation were performed below 100 mK at Louisiana State University and above 300 mK at the NHMFL, Tallahassee.

Figure 1 shows an example of a Fourier transform (FT) at 330 mK for the interval in magnetic field between 30 and 60 T. The signal on the falling side of the pulse is shown in the inset. In accordance with previous investigations [5,6,11,12], the α_3 dHvA frequency dominates the Fourier spectra. MB takes place between necks, linking the large α ellipsoids together within the ΓXM plane, and a set of eight smaller ρ ellipsoids. The simplest MB orbit $\alpha_{1,2} + 2\rho$ has been reported in both CeB₆ [11,12] and LaB₆ [1]. The most prominent QI frequencies in CeB₆ can be inferred by means of a direct comparison with similar frequencies observed in LaB₆ [1]. These are identified as $\alpha_{1,2} + \gamma$ and $2\alpha_{1,2} + \gamma + \varepsilon \equiv BZ$ in Fig. 1, where BZ is the frequency corresponding exactly to the area of the Brillouin zone. In contrast to LaB₆ [1], the $\alpha_{1,2} + \varepsilon$ QI frequency appears as only a relatively weak feature [see Fig. 3(b)].

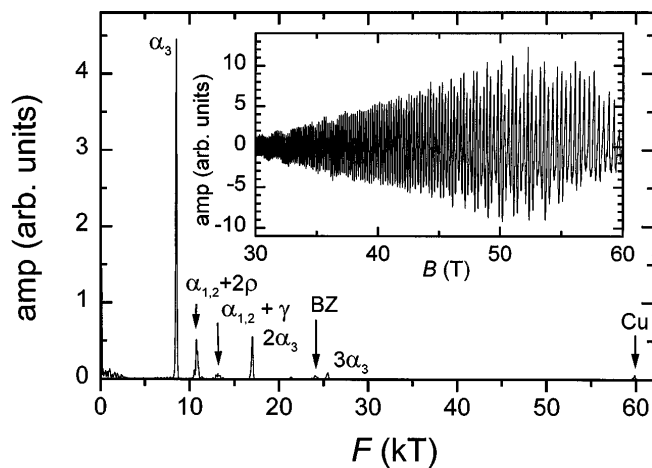


FIG. 1. A FT of the oscillations measured in the susceptibility of CeB₆. An additional peak at ~ 60 kT originates from the polycrystalline Cu comprising the detection coils. The data are shown in the inset.

Figure 2 shows fits below 2.1 K to the temperature dependences of the amplitudes of the α_3 frequency and the $\alpha_{1,2} + 2\rho$ MB frequency at 40 T. The fitted effective masses $m_{\alpha_3}^*$ and $m_{\alpha_{1,2}+\rho}^*$ for these frequencies are in close agreement with previous high field estimates [11,12]. Above the λ point of superfluid ⁴He, however, all of the quantum oscillations become attenuated due to eddy current heating effects. Exactly the same effect was observed in previous pulsed magnetic field experiments on CeB₆ [11,12], and can be attributed to the formation of a gas bubble in the center of the field which acts to impede the cooling of the sample. The extent to which the α_3 frequency is attenuated above the λ point corresponds to a sample temperature of ~ 5 – 6 K on extrapolating the fit in Fig. 2. The amplitude of the α_3 frequency thus provides a rough thermometer by which an additional high temperature point for the BZ QI frequency can be obtained. It is apparent in Fig. 2, that the amplitude of the

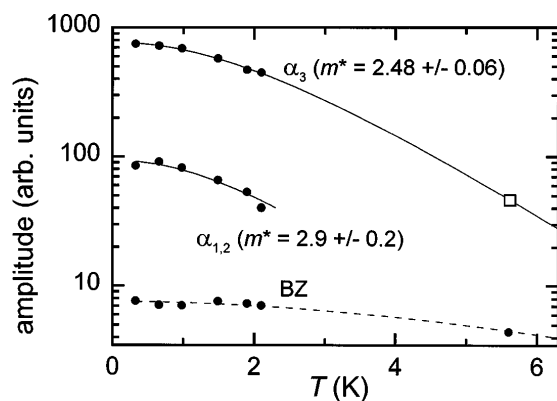


FIG. 2. Plots of the temperature dependences of the FT amplitudes of the α_3 , $\alpha_{1,2} + 2\rho$ and BZ frequencies including fits of the dHvA amplitudes to the Lifshitz-Kosevich (LK) theory (solid lines). For the α_3 frequency, the fit has been extrapolated to higher temperatures (open square). For the BZ frequency, the dashed line is a guide for the eye.

BZ frequency already starts to become attenuated at this temperature; i.e., a much lower temperature than in LaB₆ [1]. While in LaB₆ the QI oscillations were damped only at very high temperatures (~ 70 K) by electron-phonon scattering [1]; in the case of CeB₆, it clearly must be electron-electron interactions that are responsible. Strong electron-electron interactions contribute a T^2 term to the resistivity [14], although this is partially “quenched” in strong magnetic fields.

With the exception of the α_3 frequency [Fig. 3(a)], which occurs within a different plane of the BZ, a closer inspection of the FT reveals that there are numerous satellite peaks associated with the other frequencies. In Fig. 3(b), for example, further MB frequencies of the form $\alpha_{1,2} + (2 + n)\rho$ can be resolved, corresponding to additional excursions of the quasiparticles around the ρ ellipsoids. This series of frequencies can be used to estimate the size of the ρ ellipsoids in CeB₆ at high magnetic fields. The equivalent spectral features in LaB₆ could not be resolved owing to the small size (~ 5 T compared to ~ 124 T in CeB₆) of the ρ ellipsoids in that material. An additional weak feature ($\alpha_{1,2}$) in Fig. 3(b) suggests that MB can also take place without the ρ ellipsoids being involved, although this is obviously less favorable. Satellite peaks corresponding to $\alpha_{1,2} + \gamma \pm n\epsilon \pm n\rho$ and $BZ \pm n\rho$ are also observable for the QI frequencies in Figs. 3(c) and 3(d), respectively. In these cases, either of the branches of the QI trajectory can take additional excursions around the ρ ellipsoids to result in both negative and positive contributions to the enclosed area.

Upon compiling the results of FT's over different intervals of magnetic field, in Fig. 4 it becomes clear that nearly *all* of the frequencies (open symbols) have changed considerably from their values at low magnetic fields [5,12,13]. The field dependences become more

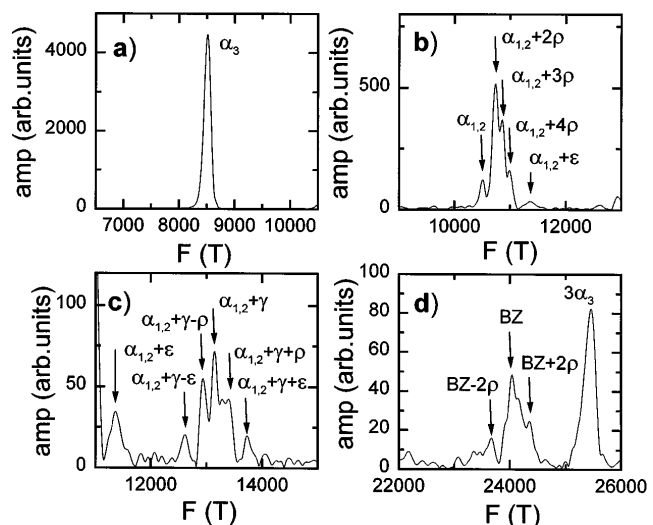


FIG. 3. A closer look at the principal Fourier peaks revealing the absence of satellite peaks in the vicinity of the α_3 frequency (a), but the presence of many satellite peaks around the $\alpha_{1,2} + 2\rho$ (b), $\alpha_{1,2} + \gamma$ (c), and BZ (d) frequencies.

acute (full symbols) when we correct for the background magnetization [15], which saturates at $\sim 1\mu_B$ per formula unit (μ_B being the Bohr magneton). Fewer field points are shown for the $\alpha_{1,2}$, γ , ε , and ρ FS cross sections, since the satellite peaks in Figs. 3(b)–3(d) cannot be resolved over too short an interval of magnetic field. Whereas the area of the α_3 FS cross section falls with increasing magnetic field in Fig. 4, that of the $\alpha_{1,2}$ cross section increases; i.e., the large electron ellipsoid α changes in shape as a function of magnetic field. The change of its ellipticity r from ~ 1.146 at 10 T to ~ 1.234 at 40 T also accounts for the respective increase and decrease of the areas of the γ and ε FS cross sections, as inferred from the analysis of satellite FT peaks in Figs. 3(b)–3(d). The ρ pocket, on the other hand, appears not to change at all.

Previous studies [12] have shown that the quenching of the effective mass $m_{\alpha_3}^*$ associated with the α_3 frequency appears to be adequately explained by the theory of Wasserman *et al.* for the dHvA effect in HF systems [8]. Of the N $4f$ electron channels (where N is thought to be 2 in CeB₆ [12]), only the $m = -J$ channel contributes significantly to the amplitude of the quantum oscillations. The quenching of the effective mass is then thought to result from the lifting of the hybridized many-body band by the Zeeman energy $-mg\mu_B B$ (where g is the electron g factor), so that its effective mass

$$m^*/m_B = 1 + 2Dn_f k_B T_A / N(k_B T_A - mg\mu_B B)^2 \quad (1)$$

becomes field dependent. Here, D is the bandwidth of the unperturbed conduction electron band of mass m_B , n_f is the mean occupancy of the $4f$ levels, and T_A is the Kondo temperature. While this mean field theory was originally intended for the limit $|mg\mu_B B| \ll k_B T_A$, fits to Eq. (1) have been performed in the opposite limit [8,12]; i.e., $T_A/g \sim 4$ K according to Harrison *et al.* [12].

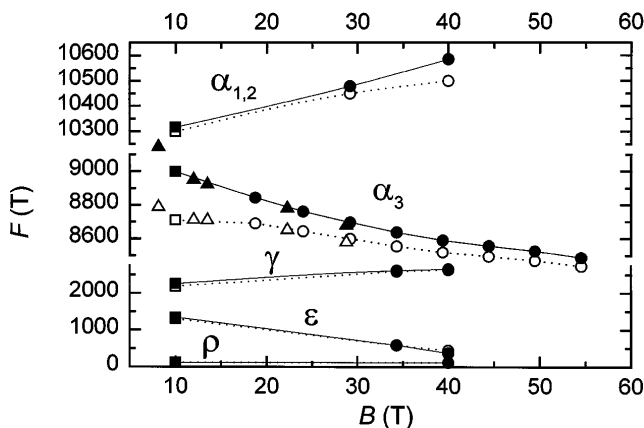


FIG. 4. Plots of the field dependences of the various FS cross-section areas in CeB₆ before (open symbols) and after (solid symbols) correction for the background magnetization given in Ref. [15]. Circles, triangles, and squares correspond to pulsed field measurements, static field measurements and additional data points taken from Refs. [5,12,13], respectively.

So far there has been no direct evidence which shows that the dHvA effect originates from a single spin FS sheet. This is nevertheless required in order to verify the applicability of the Anderson lattice model in the high magnetic field limit. For a single spin FS sheet, we should expect the Fourier amplitudes of the harmonics to decrease in an approximately exponential fashion with increasing harmonic index p . This is due to the fact that p enters into the arguments of the thermal and Dingle scattering damping factors, which are approximately exponential in form [16,17]. According to the LK theory for a FS consisting of two nondegenerate spins, each harmonic should be damped by an additional spin factor $R_S = \cos(\pi p g m^*/2m_e)$, which varies between -1 and $+1$ depending on p . In this case, the Fourier amplitudes should not decrease exponentially. Note that it is this factor that would lead to spin zeros in the ($p = 1$) fundamental frequency with m^* varying by a factor of 5 with applied field. Magnetic field modulation measurements of the α_3 frequency between 7 and 30 T show a continuous increase in amplitude with no spin zeros [10]. Figure 5 shows a FT for the high magnetic field interval $50 < B < 60$ T, clearly indicating that the harmonics of both the α_3 and $\alpha_{1,2} + 2\rho$ frequencies decay in an almost ideal exponential manner, therefore providing irrevocable proof that the dHvA oscillations of the α ellipsoids originate from a spin-polarized FS.

Assuming the limit $|mg\mu_B B| \ll k_B T_A$, Wasserman *et al.* found that the real component of the self-energy causes only an additional phase shift of the oscillations within the thermodynamic potential [8]. Given that fits to Eq. (1) have been made for $|mg\mu_B B| \geq k_B T_A$ [8,9,12], we can no longer justify this approximation. Upon including the *full* real component of the self-energy into the thermodynamic potential, some manipulation yields that there is a shift in frequency

$$\Delta F = 2Dn_f k_B T_A m_B / N \hbar e (k_B T_A - mg\mu_B B). \quad (2)$$

Inserting the fitted values of $2Dn_f / N k_B T_A \sim 260$ and $mg\mu_B / k_B T_A \sim -0.16 \text{ T}^{-1}$ of Harrison *et al.* [12] into Eq. (1) and assuming $m_B \sim 0.6m_e$ [8] and $T_A \sim 4$ K,

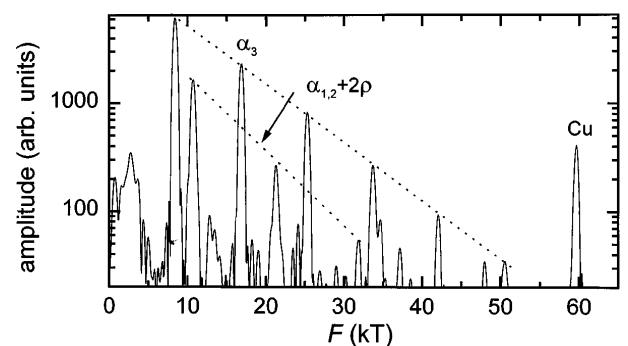


FIG. 5. A logarithmically plotted FT for the field interval $40 < B < 60$ T, showing that the Fourier harmonics of both the α_3 and $\alpha_{1,2} + 2\rho$ frequencies fall in a near-perfect exponential manner, as indicated by the dotted line.

we find that according to this theory, ΔF changes from ~ 200 T at fields of 10 T to ~ 60 T at fields of 40 T. This change of frequency by ~ 140 T on increasing the magnetic field from 10 to 40 T is of similar order to that found experimentally for the α_3 frequency in Fig. 4. From the evidence based solely on the α_3 frequency, it might be concluded that the theory for the dHvA effect in HF systems provides a reasonably accurate description of CeB₆ at high magnetic fields (i.e., $|mg\mu_B B| > k_B T_A$), in which the dHvA effect originates predominantly from the $m = -J$ channel.

The limitations of this model become apparent when we consider the remainder of the FS. While the fall of the α_3 cross-section area appears to fit the model, the increase of the $\alpha_{1,2}$ area does not; i.e., if the α_3 and $\alpha_{1,2}$ cross sections correspond to the same $m = -J$ FS sheet, then according to Eq. (2), ΔF for both of them must change in the same direction. The deformation of the FS topology suggests that the hybridization potential giving rise to the many-body band is anisotropic and that the degree of anisotropy varies as a function of the applied magnetic field. The mean field theory, on the other hand, treats only the simple case of an isotropic spherical FS. In spite of the change in ellipticity r by $\sim 8\%$, the total volume of the $m = -J$ sheet, evaluated using the ellipsoidal approximation $n_{-J} = \frac{4}{3}\pi r a_0^3$ (where a_0 is the minor axis), remains constant to within 0.2%.

While the constant volume of the $m = -J$ FS sheet appears to be consistent with Luttinger's theorem [18], this volume accounts for only ~ 0.58 electrons per formula unit. Models describing the application of the Anderson lattice model to high magnetic fields expect all spins to contribute to the FS [8,19,20], although the different spin components are not expected to occupy the same volume in k space. Measurements of the electronic component γ of the specific heat in high magnetic fields provide an alternative means of determining the mass enhancement, which includes both the $m = -J$ and $m = +J$ contributions. Such measurements have indicated that there exists a sizable discrepancy between the thermal and quasiparticle effective masses [21]. Recent investigations of the dHvA effect in high magnetic fields, in which the dHvA signal was thought to originate from two spin channels, attempted to account for this discrepancy by notionally introducing a spatial separation $\Delta k \sim 1.25 \times 10^9 \text{ m}^{-1}$ between the α ellipsoids [12]. A large component of electron density was then proposed to exist in the region of the necks. The more extensive information on the FS topology obtained in this work suggests instead that the α ellipsoids overlap; i.e., $\Delta k \sim -0.7 \times 10^9 \text{ m}^{-1}$ at high fields. This together with the fact that observed dHvA oscillations correspond to only a single spin FS sheet suggests that the discrepancy between γ and m^* is even larger than originally thought. According to the theoretical models [8,20], this discrepancy could be accounted for by the existence of a heavier $m = +J$ FS sheet [20]. From the ratio between γ and m^* , we can infer that this sheet can

be no more than ~ 2 to 3 times heavier than that of the $m = -J$ sheet, in which case it should still contribute clearly to the dHvA signal. The absence of a dHvA signal for this sheet appears to suggest that the $m = +J$ quasiparticles are incoherent. Such a scenario has recently been discussed by Edwards and Green [20] in an attempt to explain a similar absence of one spin component of the dHvA oscillations in CeRu₂Si₂ above the metamagnetic phase transition. The incoherence of these quasiparticles was attributed to the strong oscillatory contribution to the self-energy which introduces additional phase smearing effects.

To summarize, while dHvA experiments show that the quantum oscillations are characteristic of a HF band consisting of only a single spin, the additional information provided by MB and QI reveals that the FS topology deforms as a function of the applied magnetic field. This together with the absence of a $m = +J$ contribution to the dHvA signal highlights some of the limitations of the present models for the dHvA effect in HF systems.

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