Anomalous de Haas–van Alphen Oscillations in $CeCoIn_5$

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We present de Haas–van Alphen oscillation measurements showing a strong spin dependence of the quasiparticle mass enhancement in the heavy fermion superconductor $CeCoIn_5$ at high magnetic fields. There is evidence that the Fermi-liquid temperature dependence of the oscillations, embodied in the Lifshitz-Kosevich equation, is breaking down on the most strongly renormalized Fermi surface sheets.

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The de Haas–van Alphen (dHvA) effect has been an invaluable tool in the study of strongly correlated electron systems, providing not only information about Fermi surface geometry, but also, via the temperature dependence of the quantum oscillatory magnetisation, *Fermi surface specific* values of the highly renormalized quasiparticle masses. In particular, dHvA measurements have identified quasiparticles with masses of up to 100 times the bare electron mass, and give striking confirmation that the entropy of the f-electron spins in the Kondo lattice is, at low temperatures, encoded in a huge quasiparticle density of states, as is suggested by the enhanced linear coefficient of the specific heat, $\gamma = C(T)/T$ [1,2], where T is the temperature.

In many of the strongly correlated electron systems to which the dHvA technique has been applied [3], good agreement is found between γ and the measured quasiparticle masses. There are notable exceptions, however, [2,4,5] where the measured quasiparticle masses add up to significantly less than the measured linear specific heat coefficient. Exotic theories of heavy fermion behavior invoking neutral fermionic quasiparticles have been proposed to explain the "missing" mass in these systems, [6–8], as has the possibility of a spin-split Fermi surface with an undetected heavy spin component [9]. The latter possibility received support from measurements by Harrison *et al.* above the metamagnetic transition in CeB_6 [10], which suggested that quasiparticles of only a single spin orientation were contributing to the dHvA signal.

Signatures of spin-dependent masses have also been observed in $CeRu_2Si_2$ [4] and $CePd_2Si_2$ [5]. In these cases the dHvA signal contained contributions from quasiparticles of both spin orientations, and in $CePd_2Si_2$ the spin-split signal for one of the quasiparticle orbits was sufficiently well-resolved that both masses could be extracted, the masses differing by roughly a factor of 2.

In this Letter, we report high resolution dHvA results that show previously undetected spin dependence of the quasiparticle mass in $CeCoIn_5$, the first time this property has been observed in the absence of metamagnetism. We give values for masses of each spin orientation and their evolution with magnetic field.

We further report a possible low-temperature breakdown of the Fermi-liquid, *Lifshitz-Kosevich* (LK) theory of dHvA magnetisation, which affects quasiparticles on all observed Fermi surface orbits as the field is reduced towards a proposed quantum critical point at 5.1 T [11–13].

Non-Fermi-liquid (NFL) behavior associated with quantum critical points has been investigated in many materials using a number of transport and thermodynamic probes, but, to date, has not been directly observed in the dHvA effect. Such an observation would therefore be of great significance, as dHvA is a true $T \rightarrow 0$ probe, with signals becoming stronger in the low millikelvin temperature range. This can be seen from the LK expression [14–16], which describes the dHvA effect and incorporates the temperature dependence of the oscillations in the single factor

$$R_T = \frac{\chi}{\sinh(\chi)} \quad \text{where } \chi = 2\pi^2 p k_B T/\hbar\omega_c, \qquad (1)$$

where the cyclotron frequency is related to the effective quasiparticle mass m^* by $\omega_c = eB/m^*$, and p is the dHvA harmonic number.

Previous dHvA studies of $CeCoIn_5$ [17,18] have revealed all the Fermi surface sheets predicted by band-structure calculations (in which the f electrons were considered to be hybridized into the conduction electron sea). The thermodynamics is dominated by two large, quasi-2D sheets, α and β. We present here a study of the quasiparticle masses on these α and β sheets using the field modulation method in fields of up to 18 T and at temperatures down to 6 mK.

Below 100 mK, the temperature was measured by a RuO_2 thermometer calibrated against nuclear orientation and cerous magnesium nitrate (CMN) thermometers. Error bars calculated from these calibrations are shown in all plots. Heat sinking of the sample was via annealed silver wires, which have very high thermal conductivity and provide a "thermal short" to the mixing chamber of our dilution refrigerator, where the thermometers were located. Continued evolution of the signal down to 6 mK assured us that thermal contact was maintained. Above 100 mK, a calibrated Ge thermometer was used. Rigorous testing at different modulation fields showed that no eddy current heating occurred at the frequencies used during these experiments.

High-purity crystals of $CeCoIn_5$ were grown by the flux technique, with starting materials in the atomic ratio Ce:Co:In = 1:1:20. The alumina crucible was encapsulated in an evacuated quartz ampoule, heated to 1050 °C and cooled rapidly to 750 °C. The temperature was then reduced to 450 °C at 0.5 °C/h, and single crystals were obtained by spinning off excess In in a centrifuge.

Figure 1 shows the temperature dependence of oscillations from the α_3 orbit at 13–15 T. With the field parallel to the c axis ($\theta = 0$) there are slight but significant departures from the conventional LK formula [Fig. 1(a)]. The origin of these departures is clarified on rotating the field to a position 10° off axis, where a drastic failure of the LK formula is observed; this shows that conventional LK analysis is not appropriate for this system [Fig. 1(b)].

We believe that the correct explanation for this apparent failure is a spin dependence of the mass enhancements on the spin-split Fermi surface [4,5], as follows. A magnetic field **B** lifts the spin degeneracy of each quasiparticle band, splitting the Fermi surface into majority-spin and minority-spin surfaces (assuming a pseudospin of degeneracy 2). The overall dHvA signal is a superposition of the signals due to these spin-up (majority) and spin-down (minority) surfaces:

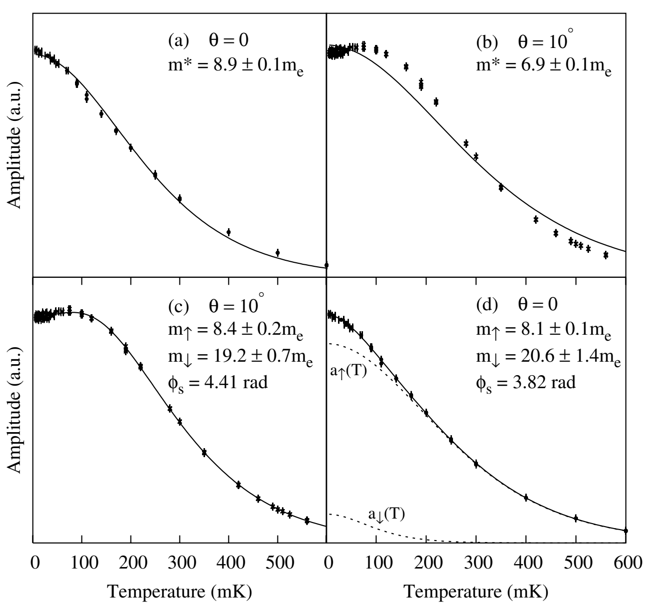

FIG. 1. Temperature dependence of dHvA amplitudes on the α_3 orbit with $\mathbf{B} \parallel \mathbf{c}$ [(a) and (d)], and **B** at 10° to **c** [(b) and (c)]. The solid lines in (a) and (b) are least-squares fits of the conventional LK formula (1), those in (c) and (d) are fits of the spin-dependent LK expression (5). 10° off axis, the two components interfere destructively, suppressing the amplitude at low temperature, while on axis they interfere constructively, giving the appearance of a nonsaturating signal.

$$\tilde{M}(T) = a_\uparrow(T)\sin\left(2\pi p \frac{F_\uparrow(B)}{B} + \phi_o\right) + a_\downarrow(T)\sin\left(2\pi p \frac{F_\downarrow(B)}{B} + \phi_o\right) \quad (2)$$

where $a_\uparrow$ and $a_\downarrow$ are now the amplitudes of the spin-up and spin-down oscillations, respectively, and contain R_T. The Onsager formula, $F = \hbar A/2\pi e$, tells us that as the band energies shift with field, $F_\uparrow$ and $F_\downarrow$ vary correspondingly. However, the frequencies yielded by a Fourier decomposition of the signal are not related to the true field-dependent Fermi surface areas. Taking

$$F_\sigma = F_o + B\left(\frac{\partial F_\sigma}{\partial B}\right)_o + O_\sigma(B^2) + \dots \quad (3)$$

we obtain

$$\tilde{M}(T) = \sum_\sigma a_\sigma(T)\sin\left(\frac{2\pi p(F_o + O(B^2) + \dots)}{B} + \phi_o + 2\pi p\left(\frac{\partial F_\sigma}{\partial B_o}\right)\right), \quad (4)$$

where $\sigma = \uparrow\downarrow$. We see that the observed frequency, $F_o + O(B^2) + \dots$, will only be field dependent if the Fermi surface areas are nonlinearly dependent on field. If such higher orders are negligible, however, only a single peak is observed, with frequency $2\pi F_o$ and amplitude

$$[(a_\uparrow(T) + a_\downarrow(T))^2\cos^2\phi_s + (a_\uparrow(T) - a_\downarrow(T))^2\sin^2\phi_s]^{1/2}, \quad (5)$$

where

$$\phi_s \equiv \pi p\left(\frac{\partial F_\uparrow}{\partial B}\right)_o - \pi p\left(\frac{\partial F_\downarrow}{\partial B}\right)_o; \qquad a_\sigma \propto \frac{1}{pm_\sigma}R_{T_\sigma}. \quad (6)$$

This expression gives the dHvA oscillation amplitude predicted by "spin-dependent" LK theory.

Applying expression (5) to the off-axis data in Fig. 1 yields an excellent fit [Fig. 1(c)]. The fit to the on-axis data is similarly improved [Fig. 1(d)], but in this case the two spin components *add*, rather than *subtract* as in the off-axis data, and the shape of the curve does not differ quite so dramatically. The phase ϕ_s at which the two spin components combine is a free parameter in the fit and is given in the figure. Also shown in the figure are the mass values obtained from each fit; it can be seen from 1(c) and 1(d) that the analysis in terms of spin-dependent masses yields the same values of m_σ for both the on-axis and off-axis data, as should be expected. Similar results were obtained for the α_1 orbit.

Analysis of higher harmonic signals should yield a mass enhancement that is effectively p times that obtained from the fundamental. Strong second harmonic signals in our data allowed us to perform such an analysis, and we indeed found this to be very accurately the case.

Although we have assigned labels $\uparrow\downarrow$, it is important to point out that it is unclear from our data which spin component is the heavier one. In the conventional slave-boson picture of the heavy fermion state, the bands are asymmetric, so it appears that quasiparticles of one spin orientation should become lighter and the other heavier as the bands polarize. At low fields, it is the majority Fermi surface that becomes heavier, but at high fields, following a metamagnetic-like transition, the minority-spin quasiparticles are expected to be more strongly renormalized, as they have a higher probability of undergoing spin-flip scattering [19,20]. In this picture, which may be relevant to the systems in which spin-dependent masses have previously been observed, the Fermi surface shrinks discontinuously at the metamagnetic transition. In $CeCoIn_5$, however, the situation may be slightly different, as metamagnetism is not apparent, and the system retains a large Fermi surface at high fields.

Considering next the heavier quasiparticles on the β sheet, Fig. 2(a) shows that conventional LK analysis also fails to account for these data. As with the α data in Fig. 1, the expression (5) gives an excellent fit [Fig. 2(b) solid line], but the heavier-spin component is now extremely large. The dashed curve shown in Fig. 2(b) represents the conventional LK expression fitted only to the data *above 50 mK*. We note the high quality of this fit down to 20 mK, and the effective mass of $48.8 \pm 0.6m_e$, which corresponds precisely to the value of $49m_e$ quoted by Settai *et al.* [17], who did not measure below 20 mK.

In further analysis of these results, we estimated the renormalized linear specific heat coefficient γ. Using the WIEN2K package [21], γ_{band} for each band, and m_b for each observed orbit were calculated; the *average* enhancement for each band $<m_{\text{exp}}/m_b>$ was then obtained, and γ_{band} was enhanced by this factor [1]. These calculations suggested that the light ($m_\uparrow$) masses alone can account for only $66 \pm 15\%$ of the experimental value of γ determined by Kim *et al.* [22]. Our results thus give further weight to the suggestion that a heavier-spin component accounts for the missing mass in some heavy fermion systems.

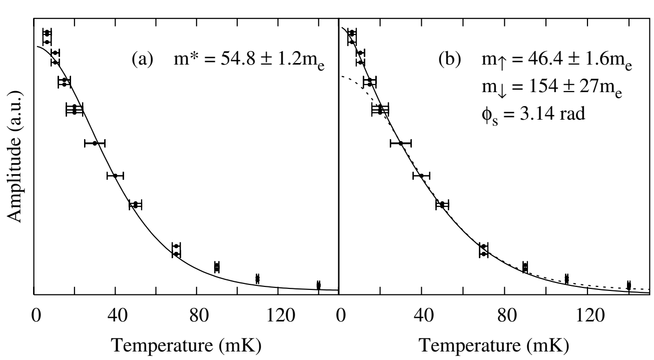

FIG. 2. Temperature dependence of the β_2 signal amplitude between 13 and 15 T, with $\mathbf{B} \parallel \mathbf{c}$. Again, we find that the conventional LK formula fails to fit the data (a). The spin-dependent amplitude expression (5) fits well [(b), solid line], but yields a very high mass spin component. The dotted line in (b) represents a fit of the conventional LK formula to the data above 50 mK; this fit yields $m^* = 48.8 \pm 0.6m_e$.

As mentioned above, $CeCoIn_5$ is believed to have a magnetic-field-tuned quantum critical point at 5.1 T. Refs. [11–13] suggest that at high fields, above 8 T, the system is a Fermi liquid. As the field is reduced, NFL behavior is seen above a temperature T^* which appears to vanish as the quantum critical point is approached.

The signal from the β orbits could not be tracked to below 10 T, but high quality data was obtained for the α sheet down to 5.25 T. Table I gives the mass enhancements on the α sheet extracted from studies on and off axis from 6–7 T. The amplitude data for the α sheet in this field range and below were similar to those from the β orbits at high field: LK analysis with spin-dependent masses was required for a good fit to all the data; a conventional LK curve gives an excellent fit for $T > 30$ mK, but the amplitude does not saturate below this temperature. It was, however, observed that there is no agreement between the on-axis and off-axis mass values below $\sim$7 T, in marked contrast to the excellent agreement observed at high field. The phase of $\phi_s = 3.14$ [23] is particularly significant, as it implies *fully constructive interference* of the two component spin-signals, one of which must derive from very high mass quasiparticles, *independent of* the magnetic field orientation. This behavior is hard to reconcile with our understanding of a conventional quasi-2D metal. In fact, under some conditions of measurement, the behavior is even more extreme, with the upward curvature at low temperature becoming so steep that a good description of the data cannot be achieved for any values of ϕ_s and m_σ (Fig. 3 inset).

We believe that these results near the quantum critical point may indicate non-Fermi-liquid behavior of quasiparticles in $CeCoIn_5$, surviving at temperatures as low as 6 mK, leading to a breakdown of LK theory.

In an attempt to explore alternative explanations for the anomalous temperature dependence of our dHvA data, we have considered a modified form of LK theory due to Wasserman and Springford [24], which can crudely be thought of as allowing for temperature dependence of the effective mass. Using a (NFL) self-energy whose real part

TABLE I. Spin-dependent mass enhancements and phase extracted from fits of expression (5) to dHvA amplitude data on the α Fermi surface sheet between 6 and 7 T.

	$m_\uparrow$	$m_\downarrow$	ϕ_s
$\theta = 0$			
α_1	21.2 ± 0.2	94 ± 7	3.14
α_2	24.2 ± 0.4	94 ± 8	3.14
α_3	14.5 ± 0.6	30 ± 8	2.42
$\theta = 10°$			
α_1	21.3 ± 2.4	39 ± 5	3.14
α_2	24.7 ± 3.5	50 ± 13	3.14
α_3	17.6 ± 0.8	40 ± 4	3.14

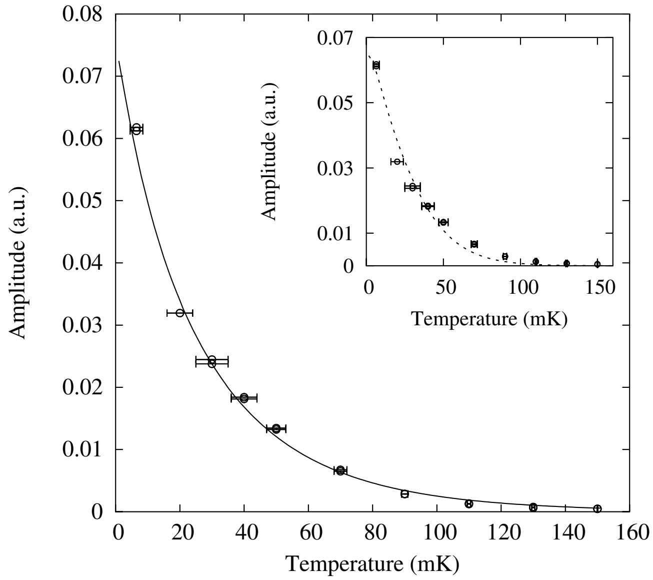

FIG. 3. Temperature dependence of the α_2 signal at 6–7 T. The data show a rather extreme rise below 20 mK, and the spin-dependent LK formula does not give a good fit (inset). However, a NFL form of the LK expression, appropriate to an antiferromagnetic quantum critical point, fits well (main plot). As discussed in the text, this result is suggestive only.

varies as $\sqrt{\omega}$, which is appropriate to a 2D antiferromagnetic quantum critical point, this expression gives a reasonably good fit to the data in Fig. 3 (main plot). We perform this analysis for illustrative purposes: to determine the particular frequency dependence of the self-energy, and to rule out a possible explanation in terms of spin-dependent LK theory with, for example, reduced scattering of a very heavy mass relative to a much lighter one, would require further careful measurements extending to still lower temperatures, an effort for which the present results provide strong motivation.

In conclusion, we have observed spin-dependent mass enhancements at high field in $CeCoIn_5$. These appear without the nonlinear splitting of the Fermi surface that would be expected if metamagnetism were influencing the system, as it is in other materials which exhibit this behavior. At low fields: masses from on-axis and off-axis measurements, which had agreed at high field, no longer agree close to the quantum critical point; the phase $\phi_s = 3.14$ yielded by spin-dependent mass analysis implies fully constructive interference of spin-up and spin-down signals independent of field orientation; and neither conventional nor spin-dependent LK analyses give a good fit to data from some orbits, as illustrated in Fig. 3. We believe these to be signatures of a departure from the Fermi-liquid LK temperature dependence of the dHvA oscillations, possibly due to quantum critical fluctuations.

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