Continuous Evolution of the Fermi Surface of CeRu$_2$Si$_2$
across the Metamagnetic Transition

R. Daou, C. Bergemann, and S. R. Julian

Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, UK.
Department of Physics, University of Toronto, Toronto M5S 1A7, Ontario, Canada

(Dated: Submitted to Physical Review Letters on 31 May 2005)

We present new, high resolution Hall effect and magnetoresistance measurements across the metamagnetic transition in the heavy fermion compound CeRu$_2$Si$_2$. The results force us to rethink the notion that the transition is accompanied by an abrupt f-electron localisation. Instead, we explain our data assuming a continuous change of the Fermi surface. We also point out ambiguities in the interpretation of dHvA data and give a possible solution to the problem of the “missing mass”.

PACS numbers: 71.27.+a, 72.15.Gd, 75.30.Kz, 71.18.+y

The nature and mechanism behind metamagnetic transitions (MMTs) in itinerant electron systems has recently received a surge of interest, partly due to the observation of quantum critical phenomena in metamagnetic materials such as Sr$_2$RuO$_4$. In this context, the intriguing MMT in the heavy fermion metamagnet CeRu$_2$Si$_2$ is now being revisited after two decades of intensive research. CeRu$_2$Si$_2$ possesses a massively enhanced specific heat coefficient $\gamma \approx 350 \text{mJ/moleK}^2$ and shows no superconducting or magnetic order in zero field—but an applied field of around 8 T $\cos \theta$ induces a MMT into a strongly spin-polarised state (the dependence on the angle $\theta$ between the field and the c-axis reflects the Ising character of the f-moments that underlie the Abrikosov-Suhl resonance).

The metamagnetic effect in CeRu$_2$Si$_2$ is accompanied by the general tendency of an applied magnetic field to split and suppress the Kondo resonance and regain the localised f-electron. In fact, the present view appears to be that the f-electron abruptly localises at the MMT, a notion which is largely based on interpretation of de Haas-van Alphen (dHvA) data. This is despite a number of obvious problems with this viewpoint: (a) The MMT appears to be a crossover rather than a real transition, while Luttinger’s theorem applied to f-localisation necessitates a discrete change in Fermi surface volume. (b) On the high-field side of the MMT, both the magnetisation (0.7 $\mu_B$ per Ce) and the specific heat (500 mJ/moleK$^2$) depart drastically from those of the local moment analogue CeRu$_2$Ge$_2$ (2.15 $\mu_B$ and 20 mJ/moleK$^2$). The differences lessen somewhat away from the MMT, but large discrepancies persist up to the highest fields in which CeRu$_2$Si$_2$ has been studied. (c) Analysis of the dHvA data within the 4f localisation scenario can in fact only account for 20% of the observed specific heat, i.e. there is “missing mass”. (d) Finally, the whole concept of a “large” Fermi surface that does include the f-electrons, vs. a “small” one that does not, becomes blurred for spin-polarised systems. Mathematically, the only criterion to distinguish “large” and “small” Fermi surfaces is whether the Fermi volume contains an odd or even number of electrons. If $\uparrow$- and $\downarrow$-electrons are different, one has to count each spin separately, and the Fermi volume can change by one electron at a time without invoking f-localisation. [Here the “change” in Fermi volume is largely due to accountancy changes: e.g. when hitherto disregarded bands below the Fermi level start to form small hole pockets.]

If the “large/small” localisation scenario is inappropriate for spin-polarised CeRu$_2$Si$_2$ at its MMT, the question remains as to what actually happens to the electronic structure there. The issue has been dormant for a while, but there are a number of recent, related results ascribing anomalies in the transport properties or in the magnetic response in other materials to changes or complete breakdown of their Fermi surface topology.

Here we present magnetoresistance and Hall data on a purer CeRu$_2$Si$_2$ sample, down to lower temperatures, over a wider field range than had previously been possible. Analysis of our data within an orbital magnetoconductivity model suggests that CeRu$_2$Si$_2$ continuously loses one spin-split Fermi sheet at the MMT, while the overall Fermi volume remains constant. This resolves the difficulties of the f-localisation scenario. We also reassess dHvA data and point out interpretational ambiguities, to solve the riddle of the missing mass.

Experiment The CeRu$_2$Si$_2$ samples were grown by F. S. Tautz using the Czochralski technique. One rectangular platelet of dimensions 0.65 x 0.5 x 0.1 mm, the short dimension along the c-axis, was soldered to at the four corners, with contact resistances estimated to be less than 0.1 mΩ at low T. We made AC four-terminal resistance measurements with sensing currents 1.6 mA (for T $\geq$ 100 mK) and 0.16 mA (T $< 100$ mK), at 77 Hz, using low-temperature transformers, on a dilution refrigerator with a base T of 6.5 mK, in magnetic fields up to 16 T.

The in-plane transverse magnetoresistivity $\rho(\parallel ab, B||c)$ was determined using the Montgomery geometry factor for rectangular sample dimensions, refined for our slightly irregularly-shaped sample through explicit numerical so-
The in-plane residual resistivity is around 6.5 mK. Our results are broadly similar to previous studies \[17\], demonstrating a much improved signal-to-noise ratio. At low-\( T \), \( \rho_{xx} \) in our measurements increases monotonically with field and lacks the unphysical plateau regions that were previously observed \[14\].

**Resistivity Power Laws** A representation of the effect of magnetic fluctuation scattering on the resistivity near a magnetic critical point can be obtained through power law analysis. The logarithmic derivative \( x = \frac{\partial \ln(\rho - \rho_0)}{\partial \ln T} \) reveals whether the system is in a Fermi liquid region with dominant electron-electron scattering \( (x = 2) \) or whether stronger fluctuation effects dominate. Such power law analysis has directly revealed the phase diagram in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) \[13\], \( \text{YbRh}_2\text{Si}_2 \) \[18\], and \( \text{La}_2\text{Sr}_{2-x}\text{RuO}_4 \) \[14\].

We have have performed additional temperature sweeps at fixed magnetic field to facilitate this kind of analysis. Fig. 2 reveals how the Fermi liquid state is confined to a low \( T \) region below the MMT and then rapidly but continuously opens out above 8 T. This is a striking qualitative difference to \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) \[13\] where, broadly speaking, the situation is the opposite, and the high-field state is the more renormalised one. At the MMT in \( \text{CeRu}_2\text{Si}_2 \), \( \rho_{xx}(T) \) actually behaves sub-linearly over a wide temperature range, with an effective power law exponent around \( x = 0.75 \). The \( A \)-coefficient in the resistivity is roughly reciprocal to the width of the blue (Fermi liquid, \( x = 2 \)) region in Fig. 2 and tracks both \( \gamma \) \[8\] and the \( 1/T_1 T \) in \( ^{99}\text{Ru} \) NMR \[19\] extremely well.

**Orbital Analysis** The most important feature in our data is the absence of a discontinuity in either the magneto- or the Hall resistivity as the MMT is crossed. If the MMT represented an abrupt 4f localisation transition, a discontinuity would almost certainly be expected since, in the simplest view, the Hall number tracks the carrier density, while the resistivity tracks the Fermi sur-
face area. Also, localization would turn CeRu$_2$Si$_2$ into an uncompensated metal, implying huge changes in the magnetoresistance which we do not observe. [In more sophisticated orbital magnetoresistance calculations, the exact shape of the expected discontinuity depends on the Fermi surface geometry, but a sudden change in one still implies an abrupt discontinuity in the other.]

It is possible, however, to interpret the low-$T$ magnetoresistance and Hall curves in Fig. 4 in terms of a less drastic Fermi surface topology transition. For this, we first need to separate the orbital contributions to the Hall effect from the anomalous ones which arise from skew electron-electron and electron-impurity scattering. For the magnetoresistance, extracting the orbital part is simple: we just need to view the lowest-$T$ data where electron-electron scattering has been cut out. It can be more elaborate to extract the orbital contribution of the Hall effect: the usual analysis $^{12}$ assumes that the $T$-dependent low-field Hall coefficient depends linearly on $\rho \chi$, so that an extrapolation to $\rho \chi = 0$ gives the orbital part. In our data, this linear relationship appears to be invalid, so we have to resort to a more qualitative discussion. The total low-$T$ Hall coefficient is around $2 \times 10^{-9}$ m$^3$/C, roughly the right magnitude expected for the orbital effect; in comparison, the anomalous contribution in other heavy fermion systems near magnetic instabilities such as YbRh$_2$Si$_2$ $^{12}$ is less than $0.1 \times 10^{-9}$ m$^3$/C even for residual resistivities 2–3 times higher than in our sample. We can therefore conclude with some confidence that the bulk of our lowest-$T$ Hall signal is orbital, not anomalous, in origin.

We now have to explain the kinks in both $\rho_{xx}$ and $\rho_{xy}$. Such kinks can be the signature of weaker electronic topological (sometimes called Lifshitz, or 2$^+$) transitions that are associated with the Fermi level moving through a van Hove singularity in the density of states of a 3D metal. $^{22}$ From a Fermi surface point of view, these van Hove singularities correspond to the band edges of the strongly renormalised f-band, i.e. to the point where the heavy $\psi$-surface disappears. In other words, the Zeeman spin-splitting of the heavy $\psi$-surface leads to progressive shrinking of the majority spin $\uparrow$-sheet (recall that $\psi$ is a hole sheet). If we look at the relevant energy scales, the $\psi$-surface has a Fermi temperature of order the Kondo temperature, $T_K \approx 20$ K. With a Wilson ratio of $\approx 2$ typical for Kondo systems, the MMT field of 8 T indeed appears sufficient to drive the whole of the $\uparrow$-band below the Fermi energy so that the $\uparrow$-volume takes up the whole BZ and its surface shrinks to a point. Above the MMT, the heavy $\uparrow$-electron is then no longer itinerant.

While CeRu$_2$Si$_2$ is of course a multi-band metal, the essence of the above ideas is captured by a spherical, spin-split Fermi surface in a simple parabolic band. To quantify the spin-splitting, $k_F^2_{\uparrow} - k_F^2_{\downarrow}$—essentially the Zeeman splitting—is taken proportional to $B$ while the total Fermi volume is held constant. The conductiv-

![FIG. 3: Magneto- and Hall resistivity of a model system consisting of a spin-split, hole-like spherical Fermi surface where one of the spin sheets shrinks to a point at $B_{\text{MMT}}$. The pictures show the field evolution of the Fermi surface.](image-url)

ity contributions $\sigma_{xx} \propto k_f^2/(1 + \cot^2 \phi_H)$ and $\sigma_{xy} \propto k_f^2 \cot \phi_H/(1 + \cot^2 \phi_H)$ of both sheets can be added, and the resulting resistivities are shown in Fig. 3. $\phi_H$ is the Hall angle: $\cot \phi_H = eB_{\text{loc}}/h k_F$, and the mean free path $l_{\text{loc}}$ forms the only free parameter of the model.

The comparison with the actual data in Fig. 4 shows very good qualitative agreement: the main features in the low-$T$ data are remarkably well reproduced, such as the direction and relative size of the kinks at $B_{\text{MMT}}$, and the formation of the high-field plateau in the magnetoresistance. A more quantitative comparison is made difficult by the presence of the other bands, and would require much more sophisticated calculations.

One peculiarity of our data is that no further Fermi surface evolution seems to take place beyond the MMT. A previous scenario $^{21}$ suggested that the $\uparrow$-level should go through the f-level hybridization gap and create a small but expanding electron sheet on the high-field side of the MMT. Additional calculations that we have done show that this new electron sheet should have a dramatic influence on the Hall effect, a feature that is entirely absent in the data. Our experiment therefore forces us to conclude that the $\uparrow$-level remains within the hybridization gap for fields up to at least 16 T.

The presence of a hybridization gap appears to conflict with the observation of enhanced specific heat $^{8}$ and differential susceptibility at the MMT which indicates a peak, not a gap, in the density of states. However, it has been suggested that fluctuation contributions can further enhance the specific heat near Fermi surface topology transitions $^{22}$, producing an inverse square root divergence at the MMT. The observed specific heat data $^{8}$ does in fact resemble such a fluctuation peak superposed on a band edge. Also, the lattice softening at the
transition already points towards significant effects of coupled volume and magnetisation fluctuations on the thermodynamic properties.

**D Huff A Fermi Surface Data** At first glance, existing and our own dHvA data seem at odds with the continuous Fermi surface evolution discussed above, since a frequency of 28 kT is observed which matches a large hole orbit ( ω ) in band calculations that assume complete f-localisation. This constitutes the main evidence for the existing, abrupt “large/small” transition scenario.

However, ω is in fact one of two orbits with roughly the same dHvA frequency for on-axis fields, the other being a hole orbit ( μ ) in the extended sheet of the itinerant band structure which is applicable at the very least to the low-field state of CeRu₂Si₂. This accidental degeneracy of dHvA frequencies has been somewhat overlooked, even though the μ-orbit is actually seen, unambiguously and below the MMT, when CeRu₂Si₂ is pressurised. Above the MMT, the 28 kT branch disappears for θ > 10°, which is indeed expected for μ from the quantitative band structure—in contrast, ω should be visible much more clearly away from the c-axis, as it is e.g. in CeRu₂Ge₂, due to its smaller off-axis cross-section and more favourable curvature. Also, if the 28 kT frequency is reassigned to the μ-orbit, its moderate mass enhancement is exactly what is expected from comparison with other orbits like κ/δ on the same sheet (m*/m ≈ 10).

Therefore, while the MMT certainly induces slight changes in the observed (back-projected) dHvA frequencies on the minor sheets, the existing dHvA data can be brought in line with the continuous Fermi surface transition scenario. In this, the heavy ↓-orbit remains unobserved—but this is not too worrisome since (as ψ ) it is quite elusive even below the MMT unless the field is exactly in the plane. This orbit carries most of the density of states and accounts for the “missing mass” that presented such a puzzle in the f-localisation scenario.

The fluctuation scenario invoked in the previous section helps to explain some of the remaining puzzles in the dHvA data. At the MMT, the dHvA signal is strongly damped even at the lowest temperatures, but this damping is not accompanied by a peak in the low-T resistivity (see Fig. 1). This points to the importance of low-κ fluctuations which destroy quantum coherence but do not affect transport properties. The anomalous damping, and enhanced temperature sensitivity of the dHvA effect, is seen on all sheets of the Fermi surface, not just on the heavy orbit, suggesting that the specific heat divergence is not purely a quasi-1D band edge effect, but again that fluctuations play the main role.

In conclusion, our new interpretation of the MMT as a continuous, Fermi surface volume conserving, topology driven transition deals with all the issues (a)–(d) mentioned in the introduction. Our data forcefully agree with the notion that the MMT is actually a crossover and that CeRu₂Si₂ remains a Fermi liquid at all fields, so a formulation in terms of continuous Fermi surface transitions appears as the natural and indeed the only viable viewpoint. Our analysis suggests that the ↑-surface shrinks to a point at the MMT—essentially recasting previous ideas in a continuous fashion—and it precludes any further Fermi surface topology changes up to 16 T.

We benefited from discussions with J. Flouquet, P. Gegenwart, K. Ishida, D. E. Khmelnitskii, G. G. Lonzarich, A. P. Mackenzie, and D. van der Marel. This work was funded by the U.K. EPSRC. C.B. acknowledges the support of the Royal Society.

---

[1] R. S. Perry et al., Phys. Rev. Lett. 86, 2661 (2001).
[2] S. A. Grigera et al., Science 294, 329 (2001).
[3] J. Flouquet et al., Physica B 319, 251 (2002).
[4] M. J. Besnus et al., Solid State Commun. 55, 779 (1985);
  J. D. Thompson et al., ibid. 56, 169 (1985).
[5] P. Haen et al., J. Low Temp. Phys. 67, 391 (1987).
[6] T. A. Costi, Phys. Rev. Lett. 85, 1504 (2000).
[7] H. Aoki et al., Phys. Rev. Lett. 71, 2110 (1993).
[8] H. P. van der Meulen et al., Phys. Rev. B 44, 814 (1991).
[9] C. A. King and G. G. Lonzarich, Physica B 271, 161 (1991).
[10] A. Bohm et al., J. Magn. Magn. Mat. 76, 150 (1988).
[11] F. S. Tautz et al., Physica B 206, 29 (1995).
[12] S. Paschen et al., Nature 432, 881 (2004).
[13] M. Lee et al., Phys. Rev. Lett. 92, 187201 (2004).
[14] N. Kikugawa et al., Phys. Rev. B 70, 134520 (2004).
[15] S. Kambe et al., Solid State Commun. 96, 175 (1995).
[16] F. Weickert et al., Physica B (in press).
[17] S. Kambe et al., J. Low Temp. Phys. 102, 477 (1996).
[18] J. Custers et al., Nature 424, 504 (2003).
[19] K. Ishida et al., Phys. Rev. B 57, 11054 (1998).
[20] A. Fert and P. M. Levy, Phys. Rev. B 36, 1907 (1987).
[21] D. M. Edwards and A. C. M. Green, Z. Phys. B 103, 243 (1997).
[22] For a review, see Y. M. Blanter et al., Phys. Rep. 245, 159 (1994).
[23] M. Takashita et al., J. Phys. Soc. Jpn. 65, 515 (1996).
[24] H. Yamagami and A. Hasegawa, J. Phys. Soc. Jpn. 61, 2388 (1992); ibid. 62, 592 (1993).
[25] H. Aoki et al., J. Phys. Soc. Jpn. 70, 774 (2001).