Magnetic oscillations in the heavy-fermion superconductor $CeCu_2Si_2$

1990 J. Phys.: Condens. Matter 2 6859

(http://iopscience.iop.org/0953-8984/2/32/016)

J. Phys.: Condens. Matter 2 (1990) 6859–6864. Printed in the UK

LETTER TO THE EDITOR

Magnetic oscillations in the heavy-fermion superconductor $CeCu_2Si_2$

M Hunt†, P Meeson†, P-A Probst†, P Reinders†§, M Springford†, W Assmus‡ and W Sun‡

† H H Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, UK

‡ Physikalisches Institut, J W Goethe-Universitat, Robert-Mayer-Strasse 2-4, 6000 Frankfurt am Main, Federal Republic of Germany

Received 21 May 1990

Abstract. We report the first obseration of the de Haas–van Alphen effect in the heavy-fermion superconductor $CeCu_2Si_2$. Magnetic oscillations are observed above and below a magnetic phase transition which, depending on the field orientation, occurs between 2.9 and 7.2 T. Angle-resolved measurements are made in the *c–a* plane on a group of frequencies of 130–550 T which are characterised by quasiparticle masses in the range 4.5–6m_e. We find no evidence to support the view that a considerable part of the Fermi surface is composed of 'light' electrons. Further, it is concluded that the superconducting state is formed out of a fully developed coherent Fermi liquid.

It was the discovery [1] of superconductivity in $CeCu_2Si_2$ that led to the intense study of the heavy-fermion compounds. There were two reasons for this particular interest. Firstly, the specific heat, which at temperatures below ~10 K, is dominated by a term γT with $\gamma \simeq 1$ J mol^{-1} K^{-2}, suggested the existence of strongly renormalised fermion quasiparticles having a degeneracy temperature, $T_F \simeq 10$ K. As it was clear, from the magnitude of the discontinuity in the specific heat at T_c, that Cooper pairs were formed by the heavy quasiparticles, it followed that $T_c < T_F < \theta_D$ with $T_c/T_F \simeq T_F/\theta_D \simeq 0.05$. In consequence, as Steglich *et al* pointed out [1], $CeCu_2Si_2$ behaved as a 'high-temperature superconductor' and could not be described by the conventional theory of superconductivity. Secondly, the experiments in $CeCu_2Si_2$ demonstrated for the first time that Cooper pairs could form in a metal in which many-body interactions, which were probably magnetic in origin, had strongly renormalised the properties of the electrons. The subsequent discussion concerning the origin, parity and symmetry of the superconducting state in this and other heavy-fermion compounds, such as UBe_{13} and UPt_3, has been particularly stimulating [2].

In this paper we present the first measurements of the de Haas–van Alphen (DHVA) effect in $CeCu_2Si_2$. Such experiments are of value in several respects. They demonstrate rather directly the existence of a Fermi surface and hence of charged, long-lived, fermion quasiparticles. Angle resolved measurements permit a test to be made of attempts to

§ Present address: Institut für Festkörperphysik, Technische Hochshule Darmstadt, Fachbereich 5, Hochschulstrase 8, 6100 Darmstadt, Federal Republic of Germany.

0953-8984/90/326859 + 06 \$03.50

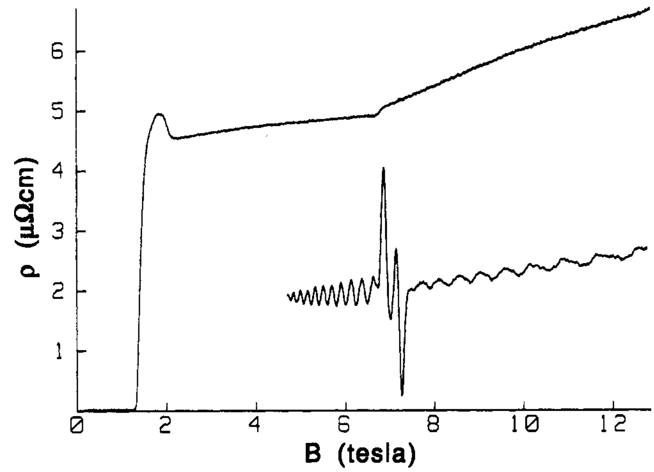

Figure 1. Magnetoresistance and DHVA effect in $CeCu_2Si_2$ measured at 20 mK. For the magnetoresistance the current is directed along the *c*-axis and is transverse to the field which is directed along the *a*-axis. The approximately linear magnetoresistance above $H_{c2}(T)$ is extrapolated to $H = 0$ in order to determine the zero-field resistivity at temperature T. Shown also are DHVA effect (magnetisation) oscillations, both above and below the phase transition region, measured at the same temperature and crystal orientation.

calculate the electronic structure of this highly correlated system and, in the present case, of the suggestion that both 'light' (essentially unrenormalised) and heavy carriers coexist [3, 4]. Finally, quasiparticle masses and their anisotropies may also be measured. Of the four known heavy-fermion superconductors, UPt_3, UBe_{13}, URu_2Si_2 and $CeCu_2Si_2$, only the first has, to our knowledge, been investigated in any detail using the DHVA effect, by Taillefer *et al* [5]. That work yielded the surprising result that the measured Fermi surface was rather well reproduced by a conventional LDA electronic structure calculation [6], even though the quasiparticle masses were found to be substantially and approximately uniformly enhanced (by a factor of between 12 and 25) over their respective band values. In contrast, DHVA measurements in $CeCu_6$, for which the mass enhancement is ~200, do not appear to agree with LDA calculations [7].

The present experiments were performed on a single crystal which had been grown using the cold boat technique described elsewhere [8]. No presence of impurities was detected in the as-grown single crystal, either by x-ray diffraction or by electron probe micro-analysis. From resistivity measurements the superconducting critical temperature was found to be $T_c = 0.72 \pm 0.02$ K, in line with previous measurements and the known sensitivity of T_c to the exact stoichiometry [4]. To characterise the samples further, the transverse magnetoresistance was measured at temperatures in the range 20 to 720 mK, a typical experiment being shown in figure 1. The pronounced 'bump' above $H_{c2}(T)$, whose origin is unclear, is followed by two regions of positive, approximately linear magnetoresistance with a phase transition region separating them. As we shall discuss below, two closely spaced transitions in this field region are clearly revealed by the magnetic measurements. By extrapolation of the linear magnetoresistance above $H_{c2}(T)$ to zero field, the resistivity is found to be well represented by, $\rho = \rho_0 + AT^2$, with coefficients, $\rho_0 = 4.5 \pm 0.5\ \mu\Omega$ cm and $A = 2.5 \pm 0.2\ \mu\Omega$ cm K^{-2}. The T^2-dependence is obeyed to better than 1% over this temperature range, the error in A arising from

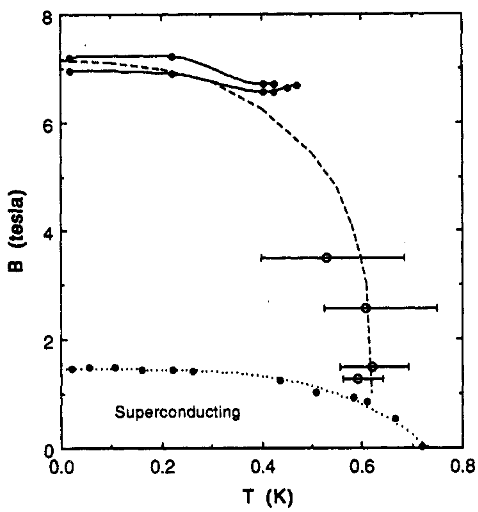

Figure 2. Phase diagram for $CeCu_2Si_2$ constructed in the B–T plane ($\boldsymbol{B}$ parallel to $\boldsymbol{a}$) from the measurements illustrated in figure 1 (full circles). The open circles are inferred from NMR measurements in polycrystalline material by Nakamura *et al* [9]. The broken curve is inferred by Steglich [12] from the earlier magnetoresistance measurements of Rauchschwalbe [4].

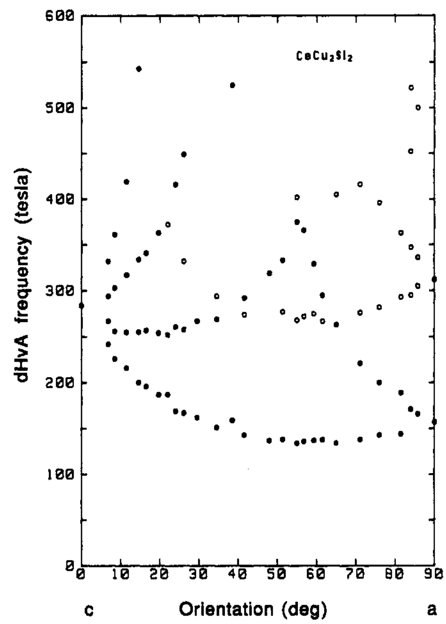

Figure 3. Angle-resolved measurements of the DHVA effect in $CeCu_2Si_2$ in the c–a plane. The experimental points indicated as open circles are considered to be harmonics.

uncertainties associated with sample geometry. The magnitude of A found here differs substantially from the values of ~10 and 11.7 $\mu\Omega$ cm K^{-2} reported by Rauchschwalbe [4] and Ōnuki *et al* [9] respectively. We note however that the transport and thermodynamic properties of $CeCu_2Si_2$ are very sensitive to the metallurgy and are capable of wide variations [10] which are not always correlated with T_c. The residual resistivity ρ_0 is similar to the lowest reported by Rauchschwalbe [4] for single crystals of this material but is still some five times larger than the value that we were able to obtain in our original DHVA experiments [11] on $CeCu_6$.

An example of the DHVA effect oscillations in $CeCu_2Si_2$ at 20 mK is shown as an inset in figure 1 where magnetic oscillations are seen both above and below the phase transition region. The magnetisation, which is measured by a second-derivative technique, reveals the presence of a double transition which, at this orientation (a-axis), occurs at 7.0 T and 7.2 T, with only the first of these being revealed in the magnetoresistance experiments. From the location of the magnetic anomalies we have then constructed a partial phase diagram in the B–T plane for a particular orientation of $\boldsymbol{B}$, as shown in figure 2. Shown also in figure 2 by the broken line is the single-phase boundary deduced by Steglich [12] from the earlier magnetoresistance measurements of Rauchschwalbe [4]. Heavy fermion compounds show a clear tendency towards antiferromagnetic order and the presence of an SDW-type structure in superconducting $CeCu_2Si_2$ has been inferred from NMR measurements by Nakamura *et al* [13], whose results are also included in figure

Table 1. Measured quasiparticle masses in $CeCu_2Si_2$ using the DHVA effect. Both frequencies and masses are averaged over the field range indicated.

Orientation	Field (T)	Frequency (T)	m^*/m_e
a-axis	4.8–5.8	171 ± 2	4.62 ± 0.07
	7.5–11.5	162 ± 2	5.15 ± 0.10
c-axis	9.5–11.5	295 ± 5	5.81 ∓ 10.05

2. Our observation of the DHVA effect in this region of the phase diagram would suggest that the magnetic order is of long range. Aside from the superconducting transition, we could find no evidence in the present measurements of magnetic or resistive indications of phase boundaries at temperatures above 0.47 K. We believe the double transition illustrated in figure 1 to be intrinsic property of the single crystal, as the angle-resolved DHVA measurements, to which we now turn, have the appropriate symmetry, so we may exclude the presence of say a bi-crystal.

Shown in figure 3 are the angle-resolved measurements of the DHVA effect for field directions in the *c–a* plane; (the crystal structure of $CeCu_2Si_2$ is the body-centred tetragonal $ThCr_2Si_2$ structure). A group of frequencies in the range 130–550 T has been observed and from the temperature dependence of the amplitude of the quantum oscillations we have determined the quasiparticle masses. In the *a*-direction this was possible for fields both above and below the transition region whereas in the *c*-direction the transition region had fallen to a field value below which magnetic oscillations could no longer be detected with sufficient accuracy. As shown in table 1, in the *a*-direction, we observe a small change of both F and m^* in passing through the phase transition region. Given that F represents only a small fraction ($\simeq 5\%$) of the Brillouin zone cross section, and in consequence is expected to be sensitive to any reconstruction of the Fermi surface, we interpret this measurement as indicating that the Fermi surface is modified to only a minor extent. The mass change cannot be compared directly with the measurements of Andraka *et al* [14] who found that the specific heat coefficient (C/T) of superconducting samples of $CeCu_2Si_2$ was changed by $<5\%$ in a field of 23 T, as their measurements were made at the rather higher temperature of 2.4 K. Visible also in figure 1 is a change in the amplitude and harmonic content of the quantum oscillations on either side of the transition. This cannot be accounted for solely in terms of a change in quasiparticle scattering but is consistent with a change in the Zeeman (spin-splitting) factor in the amplitude. The small change in m^* may contribute to this as may also a change in the effective g-factor.

The Fermi surface results presented here are we believe as yet insufficient to make a useful comparison with the self-consistent LMTO band structure calculation of Jarlborg *et al* [15]. However a general comment may be made with regard to the calculation of Sticht *et al* [3], who have proposed a quasiparticle band structure for $CeCu_2Si_2$ calculated on the basis of a Kondo lattice *ansatz* for the Ce 4f state and using the LDA potential parameters for all the remaining electrons. They predict a Fermi surface consisting of large sheets of essentially unrenormalised 'light' electrons coexisting with sheets of strongly renormalised heavy quasiparticles. A similar conclusion has been drawn by Steglich *et al* [16] from an analysis of specific heat and thermal conductivity experiments and also by Rauchschwalbe [4] from a consideration of the superconducting properties. However, in the present experiments, we stress that the measured masses of 4.5–6m_e, although relatively small in absolute terms in the context of heavy-fermion compounds, are very similar to our earlier measurements in $CeCu_6$ for a comparably small Fermi surface feature, and are representative we believe of *strongly* renormalised quasiparticles; in $CeCu_6$ by a factor of order 200 at zero field [7]. Were 'light' electrons to exist in $CeCu_2Si_2$, their signature in the present experiments, *particularly that deriving from the predicted larger sheets of Fermi surface*, would be expected to considerably exceed in amplitude that due to the feature that has been observed. We are forced to conclude therefore that we can find no evidence in these experiments for the presence of large sheets of 'light' carriers.

In summary, we believe that the present investigation of magnetic quantum oscillations in $CeCu_2Si_2$ has established the following points.

(i) Measurements in the normal phase have confirmed the presence of charged, long-lived, heavy-fermionic excitations. We conclude from this and from the observation of an accurately T^2-dependent resistivity in the same samples, that the superconducting state is formed out of a fully developed, coherent, Fermi liquid.

(ii) Whilst the present angle-resolved measurements yield an, as yet, incomplete description of the Fermi surface, they provide a reference against which we can find no evidence for the existence of large sheets of Fermi surface with essentially 'light' masses. This is clearly in contrast to the renormalised band picture of Sticht *et al* [3]. However, a firm conclusion to this effect must await more complete measurements. We note in contrast that, from DHVA measurements in the isostructural heavy-fermion $CeRu_2Si_2$, Lonzarich [17] has concluded that an appreciable variation in mass renormalisation occurs. $CeRu_2Si_2$ is characterised by an appreciably smaller linear specific heat coefficient, $\gamma = 350$ mJ mol^{-1} K^{-2}, and is not superconducting down to $T \simeq 20$ mK.

(iii) Measured quasiparticle masses of 4.5–6m_e for Fermi surface features having $F \sim 130$–550 T, are closely comparable to measurements [11] in $CeCu_6$. Given a value of $\gamma \simeq 1$ J mol^{-1} K^{-2}, we conclude that much of the (heavy-quasiparticle) Fermi surface is undetected in the present experiments. We attribute this in part to the presence of sample imperfections, as indicated by the relatively large value of residual resistivity of $\rho_0 = 4.5$ $\mu\Omega$ cm, and undoubtedly in part to the even higher quasiparticle masses characterising the larger sheets of Fermi surface.

(iv) Part of the complex phase diagram in the B–T plane has been mapped out and will be discussed by us elsewhere [18]. At some orientations, DHVA oscillations are observed to survive a phase transition with a small change in both frequency and quasiparticle mass.

The financial support of this work by the SERC is gratefully acknowledged, and one of us, P-AP, acknowledges partial support from Swiss National Funds.

References

[1] Steglich F, Aarts J, Bredl C D, Lieke W, Meschede D, Franz W and Schaffer H 1979 *Phys. Rev. Lett.* **43** 1892

[2] For recent discussions see, for example,
Wolfle P 1988 *J. Magn. Magn. Mater.* **76 + 77** 492
Joynt R 1988 *Supercond. Sci. Technol.* **1** 210
Hess D W, Tokuyasu T A and Sauls J A 1989 *J. Phys.: Condens. Matter* **1** 8135
Hasselbach K, Taillefer L and Flouquet J 1989 *Phys. Rev. Lett.* **63** 93

[3] Sticht J, d'Ambrumenil N and Kubler J 1986 *Z. Phys.* B **65** 149

[4] Rauchschwalbe U 1987 *Physica* B **147** 1
[5] Taillefer L and Lonzarich G G 1988 *Phys. Rev. Lett.* **60** 1570
[6] Norman M R, Albers R C, Boring A M and Christensen N E 1988 *Solid State Commun.* **68** 245
[7] Chapman S, Hunt M, Meeson P, Reinders P H P and Springford M 1990 *J. Phys.: Condens. Matter* submitted
[8] Sun W, Brand M, Bruls G and Assmus W 1990 *Z. Phys.* at press
[9] Ōnuki Y, Hirai T, Kumazawa T, Komatsubara T and Oda Y 1987 *J. Phys. Soc. Japan* **56** 1454
[10] See, for example.
Steglich F, Aarts J, Bredl C D, Lieke W, Meschede D, Franz W and Schafer H 1980 *J. Magn. Magn. Mater.* **15–18** 889
[11] Reinders P H P, Springford M, Coleridge P T, Boulet R and Ravot D 1986 *Phys. Rev. Lett.* **57** 1631; 1989 *J. Magn. Magn. Mater.* **63+64** 297
[12] Steglich F 1989 *J. Phys. Chem. Solids* **50** 225
[13] Nakamura H, Kitaoka Y, Yamada H and Asayama K 1988 *J. Magn. Magn. Mater.* **76+77** 517
[14] Andraka B, Fraunberger G, Kim J S, Quitmann C and Stewart G R 1989 *Phys. Rev.* B **39** 6420
[15] Jarlborg T, Braun H F and Peter M 1983 *Z. Phys.* B **52** 295
[16] Steglich F, Ahlheim U, Franse J J M, Crewe N, Rainer D and Rauchschwalbe U 1985 *J. Magn. Magn. Mater.* **52** 54
[17] Lonzarich G G 1988 *J. Magn. Magn. Mater.* **76+77** 1
[18] Hunt M, Meeson P, Probst P-A, Reinders P, Springford M, Assmus W and Sun W 1990 *Proc. 25th Yamada Conf. on Magnetic Phase Transitions (Osaka, 1990)* [*J.Magn. Magn. Mater.* at press]; 1990 *Proc. 19th Int. Conf. on Low Temperature Physics (LT19) (Brighton, UK, 1990)* [Physica B at press]