Thermal transport in the hidden-order state of URu$_2$Si$_2$

K. Behnia$^{1,2}$, R. Bel$^1$, Y. Kasahara$^2$, Y. Nakajima$^2$, H. Jin$^1$, H. Aubin$^1$, K. Izawa$^2$, Y. Matsuda$^{2,3}$, J. Flouquet$^4$, Y. Haga$^5$, Y. Ônuki$^5$ and P. Lejay$^6$

(1)Laboratoire de Physique Quantique(CNRS), ESPCI, 10 Rue de Vauquelin, 75231 Paris, France
(2)Institute for Solid State Physics, University of Tokyo, Kashiwanoha, Kashiwa, Chiba 277-8581 Japan
(3)Department of Physics, University of Kyoto, Kyoto 606-8502, Japan
(4)DREMC/SPSMS, Commissariat à l’Energie Atomique, F-38042 Grenoble, France
(5)Advanced Science Research Center, Japanese Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan
(6)Centre de Recherche sur les Très Basses Températures(CNRS), F-38042 Grenoble, France

(Dated: March 3, 2005)

We present a study of thermal conductivity in the normal state of the heavy-fermion superconductor URu$_2$Si$_2$. Ordering at 18K leads to a steep increase in thermal conductivity and (in contrast with all other cases of magnetic ordering in heavy-fermion compounds) to an enhancement of the Lorenz number. By linking this observation to several other previously reported features, we conclude that most of the carriers disappear in the ordered state and this leads to a drastic increase in both phononic and electronic mean-free-path.

Over the years, the phase transition which occurs at $T_0 \sim 18$ K in URu$_2$Si$_2$ has become a notorious enigma of Heavy-Fermion (HF) physics. This phase transition is associated with a large jump in heat capacity similar to the one observed in several anti-ferromagnetically ordered HF compounds. On the other hand, and in contrast with the latter, the magnetic moment in the ordered state appears to be very weak ($\sim 0.03 \mu_B$/U). Such a small magnetic moment is a feature found in many HF compounds. The puzzle of URu$_2$Si$_2$ resides in this unique combination. This is the only case of ordering by heavy electrons with large anomalies in all macroscopic properties leading to a tiny magnetic moment.

In order to resolve this apparent paradox, many models have been proposed distinct from the weak antiferromagnetism. Several exotic orders have been imagined. More recently, a feature in the $^{29}$Si NMR data has provided support for electronic phase separation in the hidden-order state. The debate has been mostly focused on the unusual thermodynamic properties of this ordering. The challenge for the theory has been to identify the degrees of freedom corresponding to the huge amount of entropy lost in the transition. Transport properties have not attracted a comparable attention. However, as indicated by the recent observation of a very large Nernst effect in the hidden-order state, they may prove to contain interesting information.

In this paper, we report on a study of thermal conductivity in URu$_2$Si$_2$ that detects a notable difference between this compound and all other HF systems which order anti-ferromagnetically. The distinct signature of this phase transition in thermal transport is a steep increase in the Lorenz number at the onset of transition. After checking the validity of the Wiedemann-Franz law in the ordered state, we will argue that results support a scenario in which most of the electronic carriers vanish and this leads to an increase in the mean-free-path of both surviving quasi-particles and heat-carrying phonons. Thus, the consequences of this phase transition on thermal transport are strikingly similar to the well-known case of the superconducting transition in the high-$T_c$ cuprates.

This observation highlights the drastic decrease in the carrier density induced by the hidden order leads in URu$_2$Si$_2$, which becomes one order of magnitude lower than in comparable magnetically-ordered HF compounds. This neglected feature provides unnoticed constraints for theoretical models.

The two single crystals of URu$_2$Si$_2$ used in this study were prepared by Czochralski method in Grenoble and in Tokai. They were designated as no. 1 (2) with a residual resistivity of $\rho_0 \sim 10.3 (19.5) \mu\Omega$ cm. One-heater-two-thermometers set-ups were used to measure both the longitudinal thermal conductivity (in both samples) and

FIG. 1: Temperature dependence of thermal conductivity of sample 1 for different magnetic fields.

PACS numbers: 72.15.Eb, 71.27.+a, 63.20.Kr
the transverse thermal conductivity (in sample 2). Cer-
nox chips were used as thermometers in both set-ups. The ther-
molectric (Seebeck and Nernst) coefficients of sample 1 were also measured using an identical set-up and recently reported in a separate communication[14].

The thermoelectric (Seebeck and Nernst) coefficients of URuSi₂ were also measured using an identical set-up in absence of lat-

tice conductivity and inelastic scattering of electrons, to the Wiedemann-Franz (WF) law, in absence of lattice conductivity and inelastic scattering of electrons, this number becomes equal to the Sommerfeld value, \( L_0 = \frac{\pi^2}{3} \left( \frac{\hbar k_B}{e} \right)^2 = 24.4 mW/(K^2m) \). Fig. 2 displays the temper-

ture dependence of the normalized Lorenz number, \( L/T \), in the two samples of URu₂Si₂ used in this study. As seen in the figure, in both cases ordering leads to a sudden increase in \( L/T \). In other words, thermal conduc-
tion, even after normalization to the charge transport, still displays an enhancement. This is in sharp contrast with the other compounds mentioned above. In those cases, ordering leads to a decrease in \( L/T \), the enhance-
ment in thermal conduction is not large enough to match the increase in the charge transport channel[17, 18, 19].

In order to explore the possible origin of this singular behavior of URu₂Si₂, let us begin by separating the effect of the phase transition on different types of heat carriers. As seen in Fig. 2, the large magnitude of \( L/T \) (≈ 18) at the onset of transition indicates that the contribution of the quasi-particles to heat transport constitutes a tiny fraction of the total thermal conductivity. The situation is similar in UPd₂Al₃ where \( \frac{L(T = T_N)}{L_0} \sim 11[14] \). However, this is not the case of PrB₆, NdB₆, GdB₆ or CeRhIn₅. In the latter systems, with quasi-particles carrying most or all of heat, the observed decrease in \( L/T \) is undoubtedly due to a change in the inelastic scattering of electrons. Above \( T_N \), spin fluctuations scatter conduction electrons and their sudden freezing by the onset of ordering leads to a steep increase in conductivity[18, 19]. Now, inelastic scattering is more efficient in impeding the transport of heat than charge, since those scattering events which imply little change in the momentum of the scattered quasi-particle leave a much stronger signature in thermal resistance. In this context, a sudden drop in \( \frac{L}{L_0} \) with or-
dering is a signature of more frequent small wave-vector scattering events in the ordered state. In other words, the presence of magnetic fluctuations above \( T_N \) tends to amplify the relative weight of large-q scattering and to rectify the excess in thermal resistivity produced by inelastic e-e scattering. Interestingly, this picture seems relevant even for UPd₂Al₃. In spite of the much smaller relative weight of quasi-particles in heat transport (which account for less than ten percent of the total), the transition is accompanied with a reduction of \( \frac{L}{L_0} [17] \). Therefore, one is brought to explore the possible reasons which makes the case of URu₂Si₂ so different. Why does the occurrence of the hidden order lead to an excessive enhancement of thermal conductivity?

One hypothetic possibility is the existence of an exotic heat transport introduced by the hidden order. In or-
der to check this, we have measured the Righi-Leduc (or the thermal Hall) effect in the ordered state of URu₂Si₂. This effect, which refers to the emergence of a transverse thermal gradient in response to a longitudinal heat current (and in presence of a perpendicular magnetic field) is associated with a finite value of the off-diagonal thermal conductivity tensor \( \kappa_{xy} \). It has been employed success-
fully to separate the electronic and lattice components of heat conduction in the superconducting state of YBa₂Cu₃O₇−δ[20]. Among heat carriers, only those which are skew-scattered in presence of a magnetic field are expected to contribute to \( \kappa_{xy} \). A verification of the
The solid line represents the field-dependence of $\kappa_{xy}$ in the same sample multiplied by the Sommerfeld value.

Wiedemann-Franz correlation between $\kappa_{xy}$ and $\sigma_{xy}$ has been reported for copper[21].

As seen in Fig.3, at T=5.5K, that is well below $T_0$, the magnitude of $\kappa_{xy}/T$ is comparable with $L_0\sigma_{xy}$. In other words, in the field range extending from 0 to 9T, the ratio $(\kappa_{xy}/(\sigma_{xy}T L_0))$ remains between 0.6 and 1.2. The apparent non-linear field-dependence of $\kappa_{xy}$ is presumably due to the temperature instability during field sweeps which can lead to an uncertainty of 30 percent on $\Delta_T$ (which is of the order of a few mK). Moreover, the magnetoresistance of the regulating Cernox thermometer was not corrected. Using the technical literature data[22], we estimate that at 5K and 9T, it can lead to an overestimation of $\kappa_{xy}$ by 4 percent. If the hidden-order state was host to any unconventional type of heat carriers exposed to skew scattering, then one would expect a $(\kappa_{xy}/(\sigma_{xy}T))$ significantly larger than $L_0$. Even with the level of experimental uncertainty achieved here, it appears safe to conclude that this is not the case. If there is any heat transport by magnetic excitations, it cannot be distinguished from lattice heat transport, at least at this stage. Therefore, in the following discussion, any contribution to heat transport by bosonic excitations would be addressed as part of the conventional phonon heat conductivity.

If heat conduction in URu$_2$Si$_2$ is the sum of electronic ($\kappa_e$) and lattice ($\kappa_{ph}$) components as usual, then let us separate them in order to see what sets the phase transition occurring at 18K apart. Assuming $\kappa_e = L_0\sigma T$, i.e. supposing the validity of the WF law for the electronic contribution to thermal conductivity, one obtains $\kappa_e(T)$ and $\kappa_{ph}(T)$ as displayed in Fig. 4. The striking feature of the figure is the enhancement of $\kappa_{ph}$ below $T_0$. Note that if the $\Delta_T$ drops for the electronic component, as observed in other systems, then the extracted enhancement in the lattice contribution would become even stronger. We will argue below that this feature, exclusive to URu$_2$Si$_2$, is a consequence of the vanishing of most of the itinerant electrons and a concomitant decrease in the electronic scattering of phonons.

The opening of an energy gap upon ordering in URu$_2$Si$_2$ was detected as early as the discovery of this phase transition. An activated behavior is clearly resolved in the temperature dependence of both resistivity and the specific heat[1, 2, 3]. The magnitude of the energy gap extracted from these measurements (50-110K) is somewhat larger than the gap observed in spin excitation spectrum (1.8 meV ~ 21 K)[3, 10]. The quantification of the fraction of the Fermi Surface destroyed by the opening of this gap, however, is less straightforward. By monitoring the change in the magnitude of the linear electronic specific heat ($\gamma = C_{el}/T$), Fisher and co-workers estimated that the fraction of the Fermi Surface removed is 31 percent[22], not very different from earlier estimations employing the same method[3]. Now, the change in $\gamma$ induced by AF ordering in UPd$_2$Al$_3$ (from 210 mJ/K$^2$ above $T_N$ to 150 below) implies the removal of a comparable fraction of the Fermi Surface[24]. However, the consequences of ordering for thermal conductivity in the two systems are visibly different.

If the transition affects both the effective mass and the density of carriers, then the change in specific heat does not simply reflect the fraction of Fermi Surface lost. There are two distinct experimental observations indicating that the change in $\gamma$ underestimates the fraction of the Fermi Surface lost in the transition in URu$_2$Si$_2$: i) The five-fold jump in the Hall coefficient $R_H$ induced by the transition[14, 22], which (taken at its face value) reflects a large decrease in carrier density; ii) The three-fold increase in the linear term of the thermopower, $S/T$, which points to an enhancement of the entropy per carrier in the ordered state[14]. In such a case, the change in entropy per volume monitored by $\gamma$ is much smaller than the change in carrier density. Neither of these occur in the case of UPd$_2$Al$_3$. 

FIG. 3: The field dependence of $\kappa_{xy}$ at T=5.5K in sample 2. The solid line represents the field-dependence of $\sigma_{xy}$ in the same sample multiplied by the Sommerfeld value.

FIG. 4: Lattice ($\kappa_{ph}$) and electronic ($\kappa_e$) components of thermal conductivity in sample 1, assuming $\kappa_e = L_0\sigma T$ in the whole temperature range.
Moreover, by comparing the physical properties of URu$_2$Si$_2$ and UPd$_2$Al$_3$ at low temperatures, one finds three independent lines of evidence suggesting that the carrier density in the former is one order of magnitude smaller than the latter. a) The Hall coefficient in the zero-temperature limit, remarkably large in URu$_2$Si$_2$ ($R_H \sim 10^{-8} \text{m}^3/\text{C}$ corresponding to 0.05 carriers per U in a simple one-band picture [14, 22]), exceeds by a factor of twenty the same quantity in UPd$_2$Al$_3$ [26]. Note that neither multi-band effects (which would eventually reduce the total $R_H$) or skew scattering (estimated in the zero-temperature limit using the Pauli susceptibility [27]) can explain the magnitude of $R_H$ in URu$_2$Si$_2$. b) In dHvA studies, a Dingle temperature of similar magnitude ($T_D \sim 0.2K$) was obtained, in spite of the fact that the residual resistivity of the URu$_2$Si$_2$ sample (with $\rho_0 \sim 9.5\mu\Omega \text{cm}$) studied was much higher than the UPd$_2$Al$_3$ one ($\rho_0 \sim 1.4\mu\Omega \text{cm}$). In other words, the same carrier mean-free-path corresponds to an electric conductivity which is almost one order of magnitude lower in URu$_2$Si$_2$ [20]. c) The superconducting penetration depth, $\lambda$, is almost 2.5 times larger in URu$_2$Si$_2$ than in UPd$_2$Al$_3$ [29]. Since $\frac{1}{\lambda} \propto \frac{n^*}{m^*}$, this implies that the ratio of the superfluid density, $n^*$, to the effective mass, $m^*$, is more than six times larger in the former compound.

If ordering in URu$_2$Si$_2$ leads to the removal of ninetenth of the Fermi surface as suggested by the above-mentioned data (and imaginable in a Spin Density Wave scenario), its intriguing signature on thermal transport will find a natural explanation. Lattice thermal conductivity is known to increase abruptly in many Charge Density Wave transitions because of the vanishing of electronic scatterers [31, 32]. Sizable increase in phonon thermal conductivity due to the opening of a superconducting gap [33] is not unusual either. The case of URu$_2$Si$_2$ is more intriguing as it leads to an increase in both thermal and electric conductivities in spite of the loss of a huge fraction of charged carriers. In other words, the partial destruction of the Fermi surface leads to an increase in the scattering time of both phonons and electrons [34]. Thermal transport in this context presents a curious similarity with the more familiar case of cuprates. In YBa$_2$Cu$_{3}O_{7-\delta}$, the opening of the d-wave superconducting gap leads to an enhancement of both phononic and electronic components of thermal conductivity [21].

The diluted carrier concentration in URu$_2$Si$_2$ may prove to be an important piece of the puzzle. Until now, the debate has focused on the small magnetic moment of 0.03 $\mu_B$/U without considering the density of itinerant electrons per uranium which is also unusually small. An intimate connection between these two properties remains an open question. They may be two distinct consequences of the Fermi surface nesting at $T_0$. Further exploration of transport properties under pressure, where the hidden order is replaced by a large moment AF state is clearly desirable. Theoretically, thermal transport by nodal quasi-particles in an unconventional density wave state appears as an interesting subject to explore.

In summary, our study of heat transport in URu$_2$Si$_2$ detected a drastic enhancement in lattice thermal conductivity consequent to the loss of a large fraction of the Fermi surface. Both the electronic and phonon lifetime are enhanced in the ordered state which appears to be associated with a remarkably low level of carrier concentration.

K. B. acknowledges the hospitality of the University of Tokyo, where this research was partially carried out.

[1] T. T. M. Palstra et al., Phys. Rev. Lett. 55, 2727 (1985)
[2] W. Schlabitz et al., Z. Phys. B 62, 171 (1986)
[3] M. B. Maple et al., Phys. Rev. Lett. 56, 185 (1986)
[4] C. Broholm et al., Phys. Rev. Lett. 58, 1467 (1987); Phys. Rev. B 43, 12809 (1991)
[5] V. Barzykin and L. P. Gor’kov, Phys. Rev. Lett. 70, 2479 (1993)
[6] P. Santini and G. Amoretti, Phys. Rev. Lett. 73, 1027 (1994)
[7] A. E. Sikkema et al., Phys. Rev. B 54, 9322 (1996)
[8] Y. Okuno and K. Miyake, J. Phys. Soc. Jpn. 67, 2469 (1998)
[9] H. Ikeda and Y. Ohashi, Phys. Rev. Lett. 81, 3723 (1998)
[10] P. Chandra et al., Nature, 417, 831 (2002)
[11] V. P. Mineev and M. E. Zhitomirsky, cond-mat/0412055, unpublished.
[12] N. Shah et al., Phys. Rev. B 61, 564 (2000)
[13] K. Matsuda et al., Phys. Rev. Lett. 87, 87203 (2001); J. Phys.:Condens. Matter 15, 2363 (2003)
[14] R. Bel et al., Phys. Rev. B 70, 220501(R) (2004)
[15] S. A. M. Mentink et al., Phys. Rev. B 53, R6014 (1996)
[16] F. Bourdarot et al., Phys. Rev. Lett. 90, 067203 (2003)
[17] M. Hiroi et al., J. Phys. Soc. Japan 66, 1595 (1997)
[18] M. Sera et al., Phys. Rev. B 54, R5207 (1996)
[19] J. Paglione et al., cond-mat/0404266 (2004)
[20] K. Krishana, J. M. Harris, and N. P. Ong, Phys. Rev. Lett. 75, 3529 (1995)
[21] Y. Zhang et al., Phys. Rev. Lett. 84, 2219 (2000)
[22] G. Heine and W. Lang, Cryogenics 38, 377 (1998)
[23] R. A. Fisher et al., Physica B 163, 419 (1990)
[24] C. Geibel et al., Z. Phys. B 84, 1 (1991)
[25] J. Schoenes et al., Phys. Rev. B 35, 5375 (1987); A. L. Dawson et al., J. Phys.:Condens. Matter 1, 6817 (1989)
[26] M. Huth et al., Phys. Rev. B 50, 1309 (1994)
[27] H. Ohkuni et al., J. Phys. Soc. Japan 66, 945 (1997)
[28] Y. Haga et al., J. Phys. Soc. Japan 68, 342 (1999)
[29] In very clean URu$_2$Si$_2$ samples with $\rho_0 \sim 1.27\mu\Omega \text{cm}$, the Dingle temperature drops to 35mK. (See Ohkuni et al., Phil. Mag. B 79, 1045 (1999))
[30] A. Amato, Rev. Mod. Phys. 69, 1119 (1997)
[31] M. D. Núñez-Regueiro, J. M. Lopez-Castillo and C. Ayache, Phys. Rev. Lett. 55, 1931 (1985)
[32] E. B. Lopes et al., J. Phys.: Condens. Matter, 4 L357 (1992)
[33] S. Belin, K. Behnia and A. Deluzet, Phys. Rev. Lett. 81, 4728 (1998)
[34] The increase in the electronic mean-free-path induced by the transition is also reflected in the 30-fold enhancement of the Hall angle below $T_0$. [12]