Thermal Transport in the Hidden-Order State of URu_2Si_2

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We present a study of thermal conductivity in the normal state of the heavy-fermion superconductor URu_2Si_2. Ordering at 18 K 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 the phononic and electronic mean free path.

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Over the years, the phase transition that occurs at $T_0 \sim 18$ K in URu_2Si_2 has become a notorious enigma of heavy-fermion (HF) physics. This phase transition is associated with a large jump in heat capacity [1–3] similar to the one observed in several antiferromagnetically 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 ($\sim 0.03\mu_B/U$) [4]. Such a small magnetic moment is a feature found in many HF compounds. The puzzle of URu_2Si_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 [5–11]. It is widely suspected that there is a hidden-order parameter [12] distinct from the weak antiferromagnetism. Several exotic orders have been imagined [5,6,9,10]. More recently, a feature in the ^{29}Si NMR data has provided support for electronic phase separation in the hidden-order state [13]. 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 comparable attention. However, as indicated by the recent observation of a very large Nernst effect in the hidden-order state [14], they may prove to contain interesting information.

In this Letter, we report on a study of thermal conductivity in URu_2Si_2 that detects a notable difference between this compound and all other HF systems which order antiferromagnetically. 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 argue that the 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 quasiparticles 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_2Si_2, which becomes 1 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_2Si_2 used in this study were prepared by the 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 setups were used to measure both the longitudinal thermal conductivity (in both samples) and the transverse thermal conductivity (in sample 2). Cernox

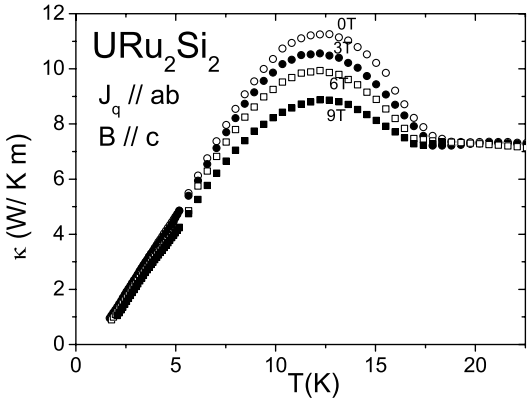

FIG. 1. Temperature dependence of thermal conductivity of sample 1 for different magnetic fields.

chips were used as thermometers in both setups. The thermoelectric (Seebeck and Nernst) coefficients of sample 1 were also measured using an identical setup and recently were reported in a separate communication [14].

Figure 1 displays the thermal conductivity, $\kappa(T)$, of URu_2Si_2 as a function of temperature for different magnetic fields. The data, measured on sample 1, are similar to the results obtained for sample 2. As seen in the figure, the onset of the transition at $T_0 \sim 18$ K is accompanied with an enhancement of thermal conductivity leading to the appearance of a visible maximum of thermal conductivity in the ordered state. As seen in the figure, this upturn in $\kappa(T)$ is reduced by the application of a magnetic field. Since the magnetic field is known both to gradually destroy the ordered moment and to reduce T_0 [15,16], the latter observation is not surprising.

In this regard, the case of URu_2Si_2 appears identical to other compounds studied in the vicinity of a magnetic order. This is the case of UPd_2Al_3 (which orders antiferromagnetically at $T_N \sim 14$ K) [17], $CeRhIn_5$ ($T_N = 3.8$ K) [18], as well as rare-earth compounds of the generic formula RB_6 (with $R = \mathrm{Pr}, \mathrm{Nd}, \mathrm{Gd}$ and $4 < T_N < 16$ K) [19]. In all these cases, heat transport in the ordered state improves due to the sudden freezing of a major scattering mechanism of the heat carriers.

However, URu_2Si_2 presents a unique feature which becomes visible by comparing the conduction of heat, κ, and charge, σ, and contrasting the change in each induced by the onset of ordering. One convenient method for such a comparison is to focus on the temperature dependence of the Lorenz number, a ratio of thermal to charge conductivities: $L = \frac{\kappa}{\sigma T}$. According to the Wiedemann-Franz (WF) law, in the absence of lattice conductivity and inelastic scattering of electrons, this number becomes equal to the Sommerfeld value, $L_0 = \frac{\pi^2}{3}(\frac{k_B}{e})^2 = 24.4\ \mathrm{nW}/(\mathrm{K}^2\,\mathrm{m})$. Figure 2 displays the temperature dependence of the normalized Lorenz number, $\frac{L}{L_0}$, in the two samples of URu_2Si_2 used in this study. As seen in the figure, in both cases ordering leads to a sudden *increase* in $\frac{L}{L_0}$. In other words, thermal conduction, 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 $\frac{L}{L_0}$: the enhancement in thermal conduction is not large enough to match the increase in the charge transport canal [17–19].

In order to explore the possible origin of this singular behavior of URu_2Si_2, 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 $\frac{L}{L_0}$ ($\sim$18) at the onset of the transition indicates that the contribution of the quasiparticles to heat transport constitutes a tiny fraction of the total thermal conductivity. The situation is similar in UPd_2Al_3 where $\frac{L(T=T_N)}{L_0} \sim 11$ [17]. However, this is not the case of PrB_6, NdB_6, GdB_6, or $CeRhIn_5$. In the latter systems, with quasiparticles carrying most or all of the heat, the observed decrease in $\frac{L}{L_0}$ 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 that imply little change in the momentum of the scattered quasiparticle leave a much stronger signature in thermal resistance. In this context, a sudden drop in $\frac{L}{L_0}$ with ordering 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 UPd_2Al_3. In spite of the much smaller relative weight of quasiparticles in heat transport (which account for less than 10% 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 that make the case of URu_2Si_2 so different. Why does the occurrence of the hidden order lead to an excessive enhancement of thermal conductivity?

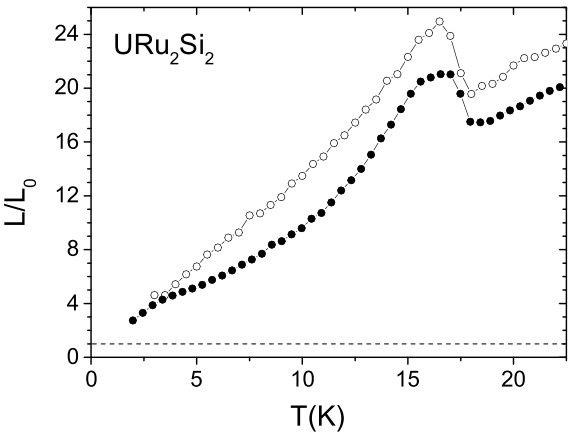

FIG. 2. The zero-field Lorenz number normalized to the Sommerfeld value as a function of temperature for sample 1 (solid circles) and sample 2 (open circles).

One hypothetic possibility is the existence of an exotic heat transport introduced by the hidden order. In order to check this, we have measured the Righi-Leduc (or the thermal Hall) effect in the ordered state of URu_2Si_2. This effect, which refers to the emergence of a transverse thermal gradient in response to a longitudinal heat current (and in the presence of a perpendicular magnetic field), is associated with a finite value of the off-diagonal thermal conductivity tensor κ_{xy}. It has been employed successfully to separate the electronic and lattice components of heat conduction in the superconducting state of $YBa_2Cu_3O_{7-\delta}$ [20]. Among heat carriers, only those which are skew scattered in the presence of a magnetic field are expected to contribute to κ_{xy}. A verification of the Wiedemann-

Franz correlation between κ_{xy} and σ_{xy} has been reported for copper [21].

As seen in Fig. 3, at $T = 5.5$ K that is well below T_0, the magnitude of κ_{xy}/T is comparable with $L_0\sigma_{xy}$. In other words, in the field range extending from 0 to 9 T, the ratio $\kappa_{xy}/(\sigma_{xy}TL_0)$ remains between 0.6 and 1.2. The apparent nonlinear field dependence of κ_{xy} is presumably due to the temperature instability during field sweeps, which can lead to an uncertainty of 30% on $\Delta_y T$ (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 5 K and 9 T, it can lead to an overestimation of κ_{xy} by 4%. 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_2Si_2 is the sum of electronic (κ_e) and lattice ($\kappa_{\rm ph}$) components as usual, then let us separate them in order to see what sets the phase transition occurring at 18 K apart. Assuming $\kappa_e = L\sigma T$, i.e., supposing the validity of the WF law for the electronic contribution to thermal conductivity, one obtains $\kappa_e(T)$ and $\kappa_{\rm ph}(T)$ as displayed in Fig. 4. The striking feature of the figure is the enhancement of $\kappa_{\rm ph}$ below T_0. Note that if $\frac{L}{L_0}$ drops for the electronic component, as observed in other systems, then the extracted enhancement in the lattice contribution would become even stronger. We argue below that this feature, exclusive to URu_2Si_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_2Si_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 [1–3]. The magnitude of the energy gap extracted from these measurements (50–110 K) is somewhat larger than the gap observed in the spin excitation spectrum (1.8 meV $\sim$ 21 K) [4,16]. 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 ($\gamma = C_{\rm el}/T$), Fisher and co-workers estimated that the fraction of the Fermi surface removed is 31% [23], not very different from earlier estimations employing the same method [3]. Now, the change in γ induced by antiferromagnetic (AFM) ordering in UPd_2Al_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 the Fermi surface lost. There are two distinct experimental observations indicating that the change in γ underestimates the fraction of the Fermi surface lost in the transition in URu_2Si_2: (i) The fivefold jump in the Hall coefficient $R_{\rm H}$ induced by the transition [14,25], which (taken at its face value) reflects a large decrease in carrier density; (ii) the threefold 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 γ is much smaller than the change in carrier density. Neither of these occur in the case of UPd_2Al_3.

Moreover, by comparing the physical properties of URu_2Si_2 and UPd_2Al_3 at low temperatures, one finds three independent lines of evidence suggesting that the carrier density in the former is 1 order of magnitude smaller than the latter. (a) The Hall coefficient in the zero-temperature

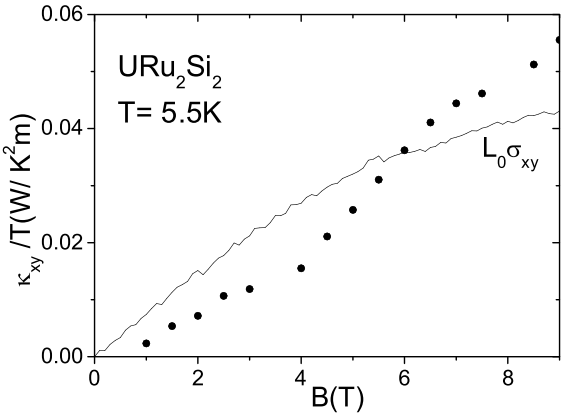

FIG. 3. The field dependence of $\frac{\kappa_{xy}}{T}$ at $T = 5.5$ K in sample 2. The solid line represents the field dependence of σ_{xy} in the same sample multiplied by the Sommerfeld value.

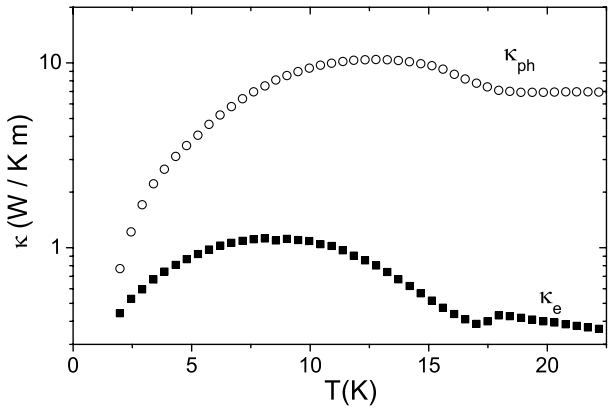

FIG. 4. Lattice ($\kappa_{\rm ph}$) and electronic (κ_e) components of thermal conductivity in sample 1, assuming $\kappa_e = L_0\sigma T$ in the whole temperature range.

limit, remarkably large in URu_2Si_2 ($R_H \sim 10^{-8}$ m^3/C corresponding to 0.05 carriers per U in a simple one-band picture [14,25]), exceeds by a factor of 20 the same quantity in UPd_2Al_3 [26]. Note that neither multiband effects (which would eventually reduce the total R_H) or skew scattering (estimated in the zero-temperature limit using the Pauli susceptibility [25]) can explain the magnitude of R_H in URu_2Si_2. (b) In de Haas–van Alphen studies, a Dingle temperature of similar magnitude ($T_D \sim 0.2$ K) was obtained, in spite of the fact that the residual resistivity of the URu_2Si_2 sample (with $\rho_0 \sim 9.5\ \mu\Omega$ cm) studied was much higher than the UPd_2Al_3 one ($\rho_0 \sim 1.4\ \mu\Omega$ cm) [27,28]. In other words, the same carrier mean free path corresponds to an electric conductivity which is almost 1 order of magnitude lower in URu_2Si_2 [29]. (c) The superconducting penetration depth λ is almost 2.5 times larger in URu_2Si_2 than in UPd_2Al_3 [30]. Since $\frac{1}{\lambda^2} \propto \frac{n_s}{m^*}$, this implies that the ratio of the superfluid density, n_s, to the effective mass, m^*, is more than 6 times larger in the former compound.

If ordering in URu_2Si_2 leads to the removal of nine-tenths 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]. A sizable increase in phonon thermal conductivity due to the opening of a superconducting gap [33] is not unusual either. The case of URu_2Si_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 $YBa_2Cu_3O_{7-\delta}$, the opening of the d-wave superconducting gap leads to an enhancement of both phononic and electronic components of thermal conductivity [20].

The diluted carrier concentration in URu_2Si_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 AFM state is clearly desirable. Theoretically, thermal transport by nodal quasiparticles in an unconventional density wave state [9] appears to be an interesting subject to explore.

In summary, our study of heat transport in URu_2Si_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 lifetimes are enhanced in the ordered state which appears to be associated with a remarkably low level of carrier concentration.

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