Phonon Thermal Transport of URu_2Si_2: Broken Translational Symmetry and Strong-Coupling of the "Hidden Order" to the Lattice

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A dramatic increase in the total thermal conductivity (κ) is observed in the hidden order (HO) state of single crystal URu_2Si_2. Through measurements of the thermal Hall conductivity, we explicitly show that the electronic contribution to κ is extremely small, so that this large increase in κ is dominated by phonon conduction. An itinerant BCS or mean-field model describes this behavior well: the increase in κ is associated with the opening of a large energy gap at the Fermi surface, thereby decreasing electron-phonon scattering. Our analysis implies that the "hidden order" parameter is strongly coupled to the lattice, suggestive of a broken symmetry involving charge degrees of freedom.

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At symmetry breaking (2nd order) phase transitions, an "order parameter" must characterize the nonsymmetric, low temperature state [1]. The heavy-fermion compound URu_2Si_2 undergoes a 2nd order phase transition at $\sim$17.5 K, yet the order parameter has remained hidden for over 20 years. This is an example of one of the most fundamental problems in solid state physics: the symmetries and associated order parameters are not obvious in many interesting systems. In fact, the order parameters of antiferromagnets (AF, staggered magnetization) and superconductors (SC, condensate wave function phase) were similarly hidden for many years. New discoveries of such nontrivial hidden-order systems (e.g., 2D XY magnets exhibit order of a purely topological character) thus continually expand our understanding of symmetry in solids.

Most conceivable experimental probes have been utilized in an attempt to uncover the HO phase. Yet still, this diverse body of experimental data does not seem to convincingly converge to any existing model [2–4]. For example, some characteristics of AF ordering occur: a large peak appears in the specific heat. However, the tiny observed staggered moment is inconsistent with the entropy lost at the transition [4], instead more likely due to a small AF impurity-phase [5]. Moreover, due to strong hybridization between the conduction electrons and nearly localized U-5f^2 moments, neither a purely local nor itinerant-electron model can be applied. Indeed, there is strong evidence for a Fermi-liquid comprised of Γ_5 non-Kramers doublet degrees of freedom [6,7].

We present another route towards uncovering the HO in URu_2Si_2 via thermal conductivity (κ) measurements. $\kappa(T)$ undergoes a very large increase at the HO transition. Although this feature has been observed before [8,9], a model for $\kappa(T)$ has not been obtained in a way that elucidates the HO. First, we explicitly show that $\kappa(T)$ is dominated by phonons. We then apply a mean-field or BCS framework to describe $\kappa_{\rm ph}$ whereby electron-phonon (e-ph) scattering decreases as a gap (Δ) in the electronic density of states (DOS) opens at the HO transition. An unusually large $2\Delta(0)/k_BT_C$ ratio of $\sim$8 is needed in order to best describe the data, in agreement with point contact spectroscopy (PCS) estimates of Δ, that is indicative of strong coupling. Our analysis suggests that an itinerant density-wave model involving the charge degrees of freedom is needed in order to uncover the hidden order parameter.

κ and the thermal Hall conductivity, κ_{XY}, are measured using the steady state method with two Cernox thermometers with an error of no more than 10%. $U(Ru_{1-x}Rh_x)_2Si_2$ crystals are grown using the Czochralski method [10], with x up to 0.04 (4%).

Figure 1(a) shows $\kappa(T)$ for tetragonal $U(Rh, Ru)_2Si_2$. All samples are measured with the heat current (Q) $\parallel c$. URu_2Si_2 displays a large upturn right at the HO transition. As the Rh concentration is increased, this upturn becomes significantly reduced until entirely lost at 3%, for which AF sets in at T_N [10]. Unlike in the HO state, the AF state induced via Rh doping displays a tiny feature upon ordering. Increasing x to 4% destroys both HO and AF so that no feature is seen at all in $\kappa(T)$. In the inset of Fig. 1(b), we plot the ratio $\delta = \gamma_{\rm LT}/(\gamma_{\rm HT} - \gamma_{\rm LT})$, as a function of Rh doping. $\gamma_{\rm HT}$ and $\gamma_{\rm LT}$ are the estimates of the T-linear coefficients of the electronic specific heat, $C_E(T)$ above and below $T_{\rm HO}$, respectively, extracted from the published data [10]. δ clearly increases as a function of doping so that the evolution of the HO state with Rh substitution is clearly associated with a significant change in the DOS. The magnitude of κ is very similar at $T > T_{\rm HO}$ for each sample. This trend is unlike that of the electrical conductivity (σ), which strongly increases with Rh doping. Judging from the behavior of the nonordered 4% sample, $\kappa(T)$ sharply in-

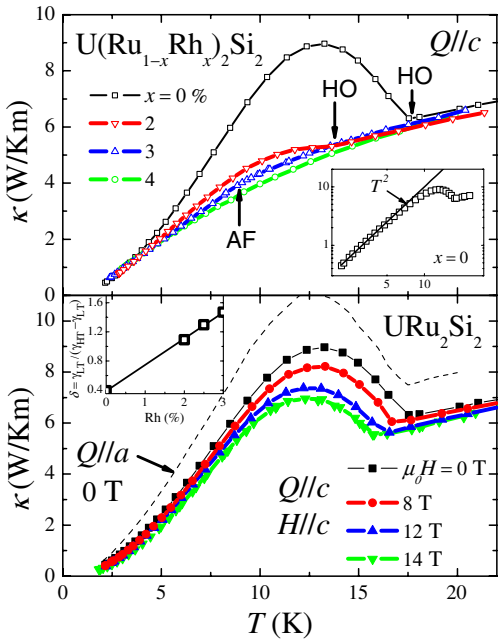

FIG. 1 (color online). (a) $k(T)$ for $U(Ru_{1-x}, Rh_x)_2Si_2$. A dramatic increase in κ is observed at the hidden order transition, sharply suppressed with Rh doping. AF order is realized at $x = 3\%$, with only a slight feature in κ. No ordering was observed at $x = 4\%$. Inset: The pure sample displays a low-T T^2 power law, indicating phonon conduction in the presence of e-ph scattering. (b) $\kappa(T)$ of pure URu_2Si_2 with heat (Q) applied along the a axis (dashed line) and the c axis (closed squares). The H dependence of the c axis $\kappa(T)$ is shown (closed symbols) up to 14 T, with $H \parallel c$. Inset: $\delta = \gamma_{LT}/(\gamma_{HT} - g_{LT})$ as a function of Rh doping (%). γ_{LT} and γ_{HT} were extracted from the published $C(T)$ data.

creases at the HO transition in URu_2Si_2. Figure 1(b) shows $\kappa(T)$ for $Q \parallel H \parallel c$ and $Q \parallel a$ in zero field for URu_2Si_2. Clearly, $\kappa_a > \kappa_c$, in contrast to σ for which $\sigma_c > \sigma_a$ [11]. Interestingly, as H is applied, the peak in $\kappa(T)$ is reduced in a more gradual way than the effects of doping. T_{HO} is also gradually reduced with $H \parallel c$ in accordance with the H vs T phase diagram [12].

Here, we argue here that the observed increase in κ is dominated by phonons rather than electrons [13], before discussing any implications for the HO. As suggested by the data in Fig. 1, the κ anisotropy and systematic change with Rh doping is opposite to that of σ. In addition, κ obeys a T^2 power law behavior at low T [inset Fig. 1(a)], which is consistent with dominant phonon conduction in the presence of e-ph scattering [14].

The thermal Hall (Righi-Leduc) conductivity (κ_{XY}) is the best way of ascertaining κ_E. The Weidemann-Franz law (WFL), $\kappa_E = L_0\sigma T$, is unreliable since the results of Fig. 1 imply that κ_{ph} is dominant. Assuming that e-ph scattering dominates, the WFL fails because of the preponderance of small angle scattering events ($q_{ph} \ll k_F$) at low, yet intermediate T, which disproportionately affect the electronic heat (as opposed to charge) current [14]. κ_{XY} provides a direct measurement of κ_E since electrons moving perpendicular to H will be deflected in a transverse direction, unlike phonons [15,16].

κ_E is estimated in Fig. 2. Figure 2(b) displays the raw ΔT_{Hall} signal, measured isothermally as a function of H for $T = 2$, 5, 10, and 20 K [17]. The expression $\kappa_{XY} = (\nabla_{Hall}T/\nabla_a T)\kappa_c$ [16] is then utilized. $\nabla_a T$ is the longitudinal gradient produced by a $Q(\parallel a)$ identical to that which produces $\nabla_{Hall}T$. κ_a ($\nabla_a T$) is independent of $H \parallel a$. κ_c is assumed not to change with $H \parallel a$ similar to what is observed in M and ρ. κ_{XY} is shown in Fig. 2(c).

The *longitudinal* κ_E can be derived from the $\kappa_{XY}(H)$ plot using the more robust assumption: $\kappa_{XY}/\sigma_{XY} = \kappa_{XX}/\sigma_{XX} = \kappa_E/\sigma_c$ [16]. The quantity $\sigma_{XY}/\sigma_c = \rho_{XY}/\rho_a = \tan\Theta_H(T)$ is then measured [Fig. 2(b), inset] to calculate $\kappa_E(H)$ at each T, which is extrapolated at low H to 0 T to determine κ_E [Fig. 2(a)]. At 2 and 5 K, the error bars are approximately the size of the symbols. Because of errors in geometry, the error in the magnitude of κ_E may be up to 50%.

κ_E makes up a small proportion of the total κ, shown more clearly in the inset of Fig. 2(a), along with κ_E expected from the WFL. The WFL clearly overstates the

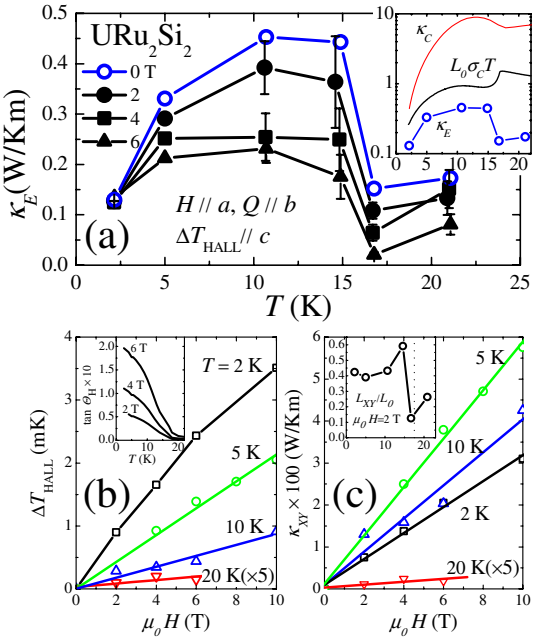

FIG. 2 (color online). (a) The electronic contribution to $\kappa(T)$ calculated from the thermal Hall effect in pure URu_2Si_2 measured in H up to 6 T (closed symbols). The zero field $\kappa_E(T)$ was extrapolated from the high-field data (open circles). At low T, the error bars are about the size of the symbols. Inset: Comparison of the total $\kappa_C(T)$, the electronic contribution calculated using the Weidemann-Franz law ($L_0\sigma_C T$), and the zero field data in the main panel (κ_E). (b) Raw thermal Hall signal as a function of H at fixed T. Inset: Electrical Hall angle as a function of T at various fixed H. (c) κ_{XY} as a function of H at various fixed T. Inset: Hall Lorenz ratio, L_{XY}, as a function of T and normalized to L_0, For clarity, only selected data sets are shown for panels (b) and (c). Lines are guides for the eye.

magnitude given by our estimate of κ_E, but approaches it at low T as expected. The WF T dependence near T_{HO} is also incorrect based on our results. κ_E itself appears to also undergo an increase of a much smaller magnitude than κ_{ph}, consistent with the claim that the electronic mean free path increases at the transition [18]. Note that the magnitude and T dependence of $L_{XY} = \kappa_{XY}/T\sigma_{XY}$ at 2 T (and by implication, L) are very different from previous reports [8,9]. We make the general observation that indiscriminate use of the Lorenz ratio $L = \kappa/\sigma T$ using the raw $\kappa = \kappa_{ph} + \kappa_E$ data should be thoughtfully reexamined.

Some scattering process clearly changes at T_{HO}. There is separate evidence [19,20] that a gap opens over parts of the Fermi surface (FS) at the T_{HO}. This naturally leads to a decrease in e-ph scattering (and increase of κ_{ph}) that can be modeled using the BCS formalism (BRT), previously studied in superconductors [21]. First, we fit $\kappa(T)$ [22] of the 4% Rh doped sample, for which no transition is observed. This sample serves as a "background" that yields an independent way to extract the magnitudes of the various scattering rates for URu_2Si_2. The modification to e-ph scattering via the BRT formalism is then applied to describe URu_2Si_2. The fit, along with the experimental data points are shown in Fig. 3(a) for the 4% sample.

For $T < T_{HO}$, Δ becomes nonzero, effectively removing carriers and inhibiting e-ph scattering. The BRT e-ph scattering rate is given by: $\tau_O^{-1} = g(\hbar\omega/k_BT, \Delta/k_BT) \times \tau_N^{-1}$; the scattering rate for phonons in the ordered state (τ_O^{-1}) is taken to be equal to the normal state value (τ_N^{-1}), renormalized by a function g. τ_N^{-1} has the form $A\omega$, where ω is the phonon frequency and A is proportional to the electron density. The function g accounts for the fact that phonons with energy $\hbar\omega < 2\Delta$ are no longer scattered by electrons and causing an increase in $\kappa(T)$ [23]. This increase in $\kappa(T)$ then tracks $\Delta(T)$. Since only a portion of the FS is gapped below T_{HO}, the total e-ph scattering rate was partitioned using a parallel conduction model such that: $\tau_{e\text{-ph}}^{-1} = A_1 g\omega + A_2\omega$. The parameter $\delta = A_2/A_1$ was then adjusted to fit the experimental $\kappa(T)$, using a PCS estimate for $\Delta(T)$ (displayed as $2\Delta/k_BT_{HO}$, Fig. 3(a) inset) [19]. Note that δ is the same parameter extracted from $C_E(T)$ shown in the inset of Fig. 1(b) since A_1 and A_2 are proportional to the gapped and ungapped DOS, respectively. Quantum oscillation measurements indicate that below T_{HO}, the FS consists of equal numbers of disconnected electron and hole pockets comprising no more than 4% of the Brillouin zone [24]. Therefore, it is reasonable that an isotropic gap opens in some of the pockets while leaving the others unaffected through the HO transition. Disconnected sections of the FS may be treated in parallel to compute κ.

The results of the BRT/PCS model comparison are shown in Fig. 3(a). The increase at T_{HO} is clearly present in the calculation, the overall magnitude coming close to the data for $\delta = 0.4$. This value for δ is remarkably close to that independently extracted from $C_E(T)$ [Fig. 1(b) inset]. We have also applied this same model to $C_E(T)$ [23]

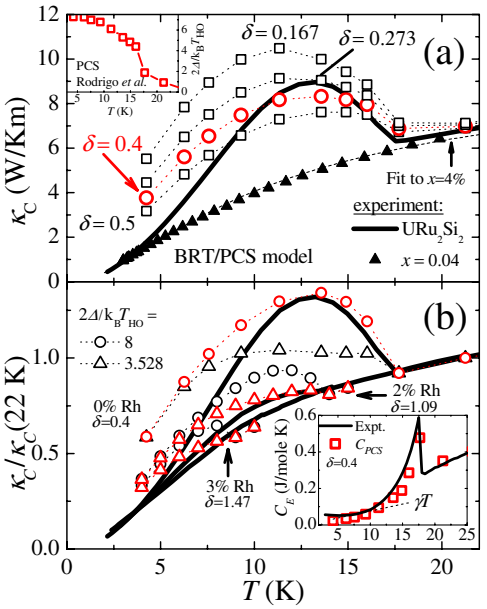

FIG. 3 (color online). (a) The BRT model (open symbols, described in text) describes the observed increase in $\kappa(T)$ for URu_2Si_2 (solid line) very well. A Debye-Calloway approximation (dashed line) was fit to the experimental $\kappa(T)$ for the x = 4% sample (open up-triangles). The energy gap, $2\Delta(T)/k_BT_{HO}$ determined by Rodrigo *et al.* (inset) was directly inserted into the BRT model. The parameter δ (defined in Fig. 1) was added in varying amounts (described in text) in parallel to the BRT model, in order to simulate a partially gapped Fermi surface. (b) An energy gap model using the $\Delta(T)$ data of Rodrigo *et al.* also describes the heat capacity measurements rather well, shown in the lower right inset, for which a value of $\delta = 0.5$ (open squares) best accounts for the ungapped amount of DOS. A mean-field solution for $\Delta(T)$ was inserted into the BRT equation for the Rh doped samples, using a δ value determined from the inset of Fig. 1(b). The ratio $2\Delta(0)/k_BT_C$ was progressively increased in order to match the experimental results. The red symbols indicate the best fit to the data for both panels.

and accounting for a partial gapping of the FS, yielding rather good agreement. The consistency of this model among different measurements validates our analysis. The value of δ obtained from $C(T)$ may be used to study $\kappa(T)$ for the Rh doped samples.

We further explore this model by inserting the BCS formula for $\Delta(T)$ into the BRT scattering rate. This allows for a more accurate estimate of $\Delta(T)$ near T_{HO}, and provides a means of analyzing the Rh-substituted samples, but for which $\Delta(T)$ is unknown. As shown in Fig. 3(b), the use of a BCS $\Delta(T)$ comes closer to the observed κ for the pure sample as the value of $2\Delta(0)/k_BT_C$ is progressively increased from the "weak-coupling" limit (3.528 for an isotropic gap) to a "strong-coupling" limit (e.g., 8). The value $2\Delta(0)/k_BT_C \sim 8$, in good agreement with the PCS data, is considerably larger than typical strong-coupled superconductors (up to ~5 in K_3C_{60} [23]), and is beyond the scope of the Migdal-Eliashberg theory [25]. Interestingly, as x is increased to 2% the behavior of κ becomes

closer to that expected for weak coupling (adjusted to account for changes in δ). This behavior is also evident for 3% Rh. As these Rh doped phases involve the same $5f$ electrons with only marginal relative changes in bandwidth and electron-electron interactions judging from high-field electrical transport [26], the strong-coupling value of $2\Delta(0)/k_BT_C$ observed for pure URu_2Si_2 can not only be due to strong electron correlation effects. Instead, note that typical charge density wave (CDW) systems show $2\Delta(0)/k_BT_C \sim 6$–24 [21,27]. Such large values are usually interpreted as a consequence of the enormous elastic energy costs associated with a coupling between the CDW and the lattice. We suggest that a similarly strong coupling to the lattice in the HO state is present.

The fact that this mean-field (BCS) model for $\kappa_{\rm ph}$ reproduces the general form of the experimental results is an important clue to the mechanism for the HO transition. This model is essentially of an itinerant nature, implying instabilities in the FS that open a gap. For example, both the SC and CDW ground states can be described using the BCS formalism [21]. Indeed, a $\kappa_{\rm phonon}$ increase at SC/CDW/SDW transitions has been observed to occur [14,28] in a similar way as observed here. A ''density-wave'' phase would also be very sensitive to the effects of doping, both in the reduction of T_C, and possibly a change in the coupling strength, since the transition would be dependent on the subtle topology of the FS. The large value of $2\Delta(0)/k_BT_C(\sim 8)$ that is needed to describe pure URu_2Si_2 suggests that the hidden order parameter may be strongly coupled to the lattice, while the AF phase is not. This claim is consistent with anomalies detected in the elastic constants at $T_{\rm HO}$; in particular, the observed softening of a shear mode implies a lattice instability [29]. If the HO is associated with the modulation of charge degrees of freedom (e.g., U electrical quadrupolar moments implicit to Γ_5 doublets [6,7]) then this would naturally lead to a strong coupling to the lattice via electrostatic forces, e.g., within a quadrupolar density-wave scenario. Note that exotic d-wave order parameter models [2,3] should not have such an electrostatic coupling because the associated charge distributions possess no net electrical moment.

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