Hidden Order Transition in URu$_2$Si$_2$ and the Emergence of a Coherent Kondo Lattice

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Using a large-N approach, we demonstrate that the differential conductance and quasi-particle interference pattern measured in recent scanning tunneling spectroscopy experiments (A.R. Schmidt et al. Nature 465, 570 (2010); P. Aynajian et al., PNAS 107, 10383 (2010)) in URu$_2$Si$_2$ are consistent with the emergence of a coherent Kondo lattice below its hidden order transition (HOT). Its formation is driven by a significant increase in the quasi-particle lifetime, which could arise from the emergence of a yet unknown order parameter at the HOT.

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Heavy-fermion materials exhibit a plethora of exciting phenomena [1] which are believed to arise from the competition between Kondo screening [2] and antiferromagnetic ordering [3]. One of the most puzzling phenomena arises in the heavy-fermion compound URu$_2$Si$_2$ which exhibits an onset of Kondo screening around $T_c \approx 55K$ [4, 5], and undergoes a second order phase transition at $T_0 = 17.5K$ [4, 5] into a state with a still unknown (hidden) order parameter. Currently, an intense debate focuses on the nature of this hidden order transition (HOT) and its microscopic origin [6, 7]. Important new insight into this question has recently been provided by ground-breaking scanning tunneling spectroscopy (STS) experiments [8, 9]. Above the HOT, the differential conductance, $dI/dV$, exhibits a characteristic Fano lineshape. In contrast, below $T_0$, a soft gap opens up in $dI/dV$ [8, 9] and a quasi-particle interference (QPI) analysis reveals a band structure similar to that expected in the (heavy Fermi liquid) phase of a screened Kondo lattice [8]. Whether the observed changes in $dI/dV$ below the HOT are consistent with the observed QPI pattern and with the emergence of a coherent Kondo lattice, is an important question whose answer will provide crucial insight into the nature of the hidden order transition.

In this Letter, we address this question and demonstrate that the experimentally observed $dI/dV$ [8, 9] and QPI pattern [8] below the HOT are consistent with the emergence of a coherent Kondo lattice (CKL) and its electronic band structure. In particular, $dI/dV$ exhibits characteristic signatures of the Kondo lattice band structure [9], such as an asymmetric gap, and a peak inside the gap which arises from the van Hove singularity of the heavy $f$-electron band. In addition, the temperature evolution of $dI/dV$ [8, 9] suggests that the formation of the CKL below the HOT is primarily driven by a significant increase in the coherence, i.e., lifetime, of the heavy quasi-particles. Since the creation of a CKL is not expected to be the primary source of the observed second order phase transition at $T_0$, we suggest that the increased quasiparticle coherence is a result of the yet unknown order parameter that emerges at the HOT.

The starting point for our study is the Kondo-Heisenberg Hamiltonian [11, 12]

\[ H = \sum_{k,\sigma} \varepsilon_k c_{k,\sigma}^\dagger c_{k,\sigma} + J \sum_{r,\alpha,\beta} S_{r}^K \cdot c_{r,\alpha}^\dagger \sigma_{\alpha,\beta} c_{r,\beta} + \sum_{r, r', f} I_{r, r'} S_{r}^K \cdot S_{r'}^F. \]

We use a hole-like two-dimensional (2D) conduction ($c$-electron) band dispersion $\varepsilon_k = 2t (\cos k_x + \cos k_y) - \mu$ with nearest-neighbor hopping $t$ and chemical potential $\mu$ to reproduce the 2D QPI dispersion [8]. $c_{k,\alpha}^\dagger (c_{k,\alpha})$ creates (annihilates) a $c$-electron with spin $\alpha$ and momentum $k$. $J > 0$ is the Kondo coupling, $S_{r}^K$ is the $S = 1/2$ spin operator of a magnetic atom at site $r$ and $\sigma$ are the Pauli matrices. $I_{r, r'}$ is the antiferromagnetic coupling between magnetic atoms. In the large-$N$ approach [14, 15], one represents $S_{r}^K$ by pseudo-fermion operators, $f_\alpha^r$, and decouples the Hamiltonian via the mean fields [16, 18]

\[ s(r) = \frac{J}{2} \sum_\alpha \langle f_{r,\alpha}^r f_{r,\alpha}^\dagger \rangle; \quad \chi(r, r') = \frac{I_{r, r'}}{2} \sum_\alpha \langle f_{r,\alpha}^r f_{r',\alpha}^\dagger \rangle. \]

A non-zero hybridization $s(r)$ between the $c$- and $f$-electron states describes the screening of a magnetic moment, and the bond variable $\chi(r, r')$ represents the antiferromagnetic (spin-liquid) correlations [12] between nearest-neighbor sites. For a translationally invariant system, $s(r) = s$, $\chi(r, r') = \chi_0$ and $\chi_1$ for nearest and next-nearest-neighbor sites, respectively. Adding the term $\sum_{r, r', \alpha} \varepsilon_{f} f_{r, \alpha}^r f_{r', \alpha}^\dagger$ to the Hamiltonian [16, 18] allows one to fix the $f$-electron occupancy, $\langle \hat{n}_f \rangle$, by adjusting the on-site energy $\varepsilon_f$. To solve the self-consistency equations, Eq.\,(2), for finite lifetimes of the $f$- and $c$-electron states, we rework them in the form:

\[ s(r) = \frac{-J}{\pi} \int_{-\infty}^{\infty} d\omega \, n_F(\omega) \, \text{Im} G_{fc}(r, r, \omega); \]

\[ \chi(r, r') = \frac{-I_{r, r'}}{\pi} \int_{-\infty}^{\infty} d\omega \, n_F(\omega) \, \text{Im} G_{ff}(r, r', \omega), \]

where $G_{\gamma\zeta}(r, r', \tau) = -\langle T_{\tau} \gamma_{\zeta}^\dagger(\tau) \zeta_{\gamma}^0(0) \rangle$ ($\gamma, \zeta = c, f$, spin indices are omitted) are the full Green’s function describ-
ing the hybridization of the c- and f-electron bands, with

\[
G_{\text{ff}}(k, \omega) = \left[ (G_{\text{ff}}^0(k, \omega))^{-1} - s^2 G_{\text{cc}}^0(k, \omega) \right]^{-1};
\]

\[
G_{\text{cc}}(k, \omega) = \left[ (G_{\text{cc}}^0(k, \omega))^{-1} - s^2 G_{\text{ff}}^0(k, \omega) \right]^{-1};
\]

\[
G_{\text{cf}}(k, \omega) = G_{\text{cc}}^0(k, \omega) s G_{\text{ff}}(k, \omega),
\]

where \( G_{\text{ff}}^0 = (\omega + i \Gamma - \chi_k)^{-1}, G_{\text{cc}}^0 = (\omega + i \Gamma_c - \varepsilon_k)^{-1}, \) and \( \Gamma_c^{-1} \) and \( \Gamma_f^{-1} \) are the lifetimes of the c- and f-electron states, respectively. For \( \Gamma_c = \Gamma_f = 0^+ \), the poles of the above Green’s functions yield two energy bands

\[
E_{\pm}^c = \frac{\varepsilon_k + \chi_k}{2} \pm \sqrt{\left(\frac{\varepsilon_k - \chi_k}{2}\right)^2 + s^2},
\]

with \( \chi_k = -2 \chi_0 (\cos k_x \cos k_y - 4 \chi_1 \cos k_x \cos k_y + \varepsilon_f). \)

To compute the differential conductance, \( dI/dV \) [19–21], measured in STS experiments [8, 9], we define the spinhor \( \Psi_k^\dagger = (c^\dagger_k f^\dagger_k) \) and the Green’s function matrix \( \hat{G}(k, \tau) = -(\hat{T}_c \Psi_k^\dagger(\tau) \Psi_k(0)) \). With \( t_c \) and \( t_f \) being the tunneling amplitudes into the c- and f-electron bands, respectively, one has in the weak-tunneling limit [20]

\[
\frac{dI(r, \omega)}{dV} = -\frac{2e}{h} \hat{N}_i \sum_{i,j=1}^2 \left[ i \text{Im} \hat{G}(r, r, \omega) \hat{t}_{ij} \right], \tag{6}
\]

where \( \hat{t} = \begin{pmatrix} t_c & 0 \\ 0 & t_f \end{pmatrix} \), and \( \hat{N}_i \) is the STS tip’s density of states. To gain insight into the momentum resolved electronic structure of URu$_2$Si$_2$, Schmidt et al. [8] performed a quasi-particle interference (QPI) analysis via the substitution of U by Th atoms. The measured QPI intensity, \( S(q, \omega) \), is given by the Fourier transform of \( dI/dV \) into \( q \)-space, yielding

\[
S(q, \omega) = \frac{dI(q, \omega)}{dV} = \frac{2\pi e}{h} N_i \sum_{i,j=1}^2 \left[ i \hat{N}(q, \omega) \hat{t}_{ij} \right];
\]

\[
\hat{N}(q, \omega) = -\frac{1}{\pi} \text{Im} \int \frac{d^2k}{(2\pi)^2} \hat{G}(k, \omega) \hat{U} \hat{G}(k + q, \omega). \tag{7}
\]

\( \hat{U} = \begin{pmatrix} U_c & 0 \\ 0 & U_f \end{pmatrix} \) and \( U_c \) and \( U_f \) are the Th atoms’ scattering potential in the c- and f-electron bands, respectively.

We begin by discussing the STS results of Schmidt et al. [8] and present in Fig. 1(a) the experimentally measured change in \( dI/dV \) below the HOT \( T = 1.9 K \) data of Fig. 3(b) in Ref. [8] together with a theoretical fit, \( \delta(dI/dV) = dI/dV(T < T_0) - dI/dV(T = T_0) \) (with \( s = 0 \) at \( T = T_0 \)) obtained from Eq.(6). For the same parameter set as in Fig. 1(a), we present in Figs. 1(c) and (e) [(d) and (f)] a contour plot of the QPI intensity, \( |S(q, \omega)| \), and the maxima in \( |S(q, \omega)| \) (i.e., the QPI dispersion), respectively, along \( q_\parallel = 0 \) [\( q_\parallel = q_z \)]. Also shown are the experimental QPI dispersions (black lines) of Figs. 5(c) and (d) in Ref. [8]. The experimental \( dI/dV \) and QPI data were obtained on a U-terminated surface of a 1% Th-doped sample. The very good quantitative agreement between the theoretical and experimental \( dI/dV \) and QPI dispersions (arising from a unique set of parameters) strongly suggests that their origin lies in the emergence of a coherent Kondo lattice below the HOT, in agreement with the conclusions by Schmidt et al. [8] (similar STS signatures of a coherent Kondo lattice were recently also reported in YbRh$_2$Si$_2$ [22]).

The QPI pattern is determined by scattering of electrons both within and between the two electronic bands, \( E^\pm_k \). Intraband scattering [see Fig. 1(b)] gives rise to the \( q_1 \) and \( q_2 \) branches in \( |S(q, \omega)| \) shown in Figs. 1(c) and
The main contribution to these branches arises from $2k_F$-scattering [Fig. 1(b)], such that their dispersion is approximately described by $E_{q}^{\pm}$ as shown in Figs. 1(c) and (f). Moreover, for $-1 \text{ meV} \lesssim \omega \lesssim 1.5 \text{ meV}$, $E_{q}^{\pm}$ both possess equal energy surfaces, giving rise to interband scattering with wave-vector $q_3$, and a corresponding $q_3$ branch in $S(q, \omega)$ [see Figs. 1(c) and (d)]. The $q_3$ branch has also been seen experimentally along $q_y = 0$, where the experimental and theoretical QPI results are in very good agreement, but is absent along $q_y = q_x$. The latter could arise from the smaller gap along $q_y = q_x$ which might make it difficult to resolve the $q_1$ and $q_3$ branches. Additional support for this conclusion comes from the experimental $dI/dV$ data in Fig. 1(a). Here, the upper band edge of $E_{k}^{c}$ leads to a sharp decrease in $dI/dV$ [see arrow (2)] which occurs around $\omega \approx 1 \text{ meV}$. This energy is consistent with the extrapolation of the experimental $q_1$ dispersion along $q_y = 0$ [Fig. 1(c)], where the $q_1$ and $q_3$ branches are well resolved, but inconsistent with the extrapolation along $q_y = q_x$. Note that the peak in $dI/dV$ at $\omega = -2 \text{ meV}$ [see arrow (1) in Fig. 1(a)] arises from the van Hove singularity of the $f$-electron band.

The experimental QPI pattern implies that the doped Th atoms scatter electrons in both the $c$- and $f$-electron bands [9]. To support this conclusion, we present in Fig. 2(a) the QPI pattern, $|S(q, \omega)|$, for $U_f = 0$. It is similar to that of the unhybridized $c$-electron band [see Fig. 2(a)] since its dominant contribution arises from scattering between states where the coherence factors of the $c$-electrons are large. Conversely, for $U_c = 0$, $|S(q, \omega)|$ is determined by scattering between states with large $f$-electron weight. Both cases are inconsistent with the experimental QPI results, whose description requires $U_f/U_c \approx 0.6$, as shown in Fig. 4.

We next discuss the STS results by Aynajian et al. [9], and present in Figs. 2(a) and (b) the experimental $dI/dV$ data obtained on a U-terminated surface of pure URu$_2$Si$_2$ for $T = 2K$ and $4K$, respectively [Fig. 4(b) in Ref. 9], together with the theoretical results obtained from Eq. (3). The theoretical $dI/dV$ curves reproduce the salient features of the experimental results: the asymmetry and magnitude of the gap in $dI/dV$ as well as the peak at $\omega \approx -0.8 \text{ meV}$ [see arrows in Figs. 2(a) and (b)] which arises from the van Hove singularity of the $f$-electron band (a similar peak and gap magnitude were also reported for a Si-terminated surface in Ref. 9). Both, the gap asymmetry and the peak, are characteristic signatures of the Kondo lattice bandstructure, and thus suggest the existence of a coherent Kondo lattice below the HOT. To gain further insight into the microscopic origin of the CKL, we note that the changes in $dI/dV$ between $T = 2K$ [Fig. 2(a)] and $T = 4K$ [Fig. 2(b)] can be solely attributed to an increase in the damping of the $f$-electron states from $\Gamma_f = 0.013t$ at $T = 2K$ to $\Gamma_f = 0.02t$ at $T = 4K$, and the resulting changes in $s$, $\chi_0$ and $\varepsilon_f$ which are self-consistently computed from Eq. (3) for fixed $J, I$ and $n_f$. Increasing $\Gamma_f$ even further (while self-consistently computing $s$, $\chi_0$ and $\varepsilon_f$) yields the evolution of $dI/dV$ shown in Fig. 2(c) which possesses the same characteristic signatures as those observed by Aynajian et al. [9] with increasing temperature. In particular, the gap in $dI/dV$ is filled in, its magnitude remains approximately constant (until close to the HOT), and the center of the gap shifts to larger energies with increasing temperature or $\Gamma_f$. Both $s$ and $\chi_0$ decrease with increasing $\Gamma_f$ (not shown) since the increased decoherence of the $f$-electron states necessarily suppresses coherent Kondo screening and magnetic correlations. The results in Figs. 2(a)-(c) suggest that the formation of a coherent Kondo lattice below the HOT is driven by a drastic increase in the $f$-electron lifetime. In contrast, increasing solely the hybridization, $s$, below the HOT leads to an evolution of $dI/dV$ (not shown) that is inconsistent with the experimental observations.

In Fig. 2(d), we present the bandstructure, $E_{k}^{\pm}$, obtained from the fits to the STS data by Schmidt et al. for the 1% Th-doped sample [Fig. 1] and by Aynajian et al. [9] for pure URu$_2$Si$_2$ [Fig. 2(a)]. In the Th-doped sample, the hybridization ($s = 0.06t$) is smaller while $\Gamma_f$ is larger than in the undoped compound ($s = 0.32t$). These results are consistent with a suppression of the HOT (and thus $s$) and an increased decoherence by Th-doping, and thus suggest that both groups probe the same heavy and light bands (while irrelevant for our study, we note that the groups disagree on the identification of the cleaved surfaces). Further support for this conclusion comes from the extracted $f$-electron density, which is quite similar in
both cases with $n_f = 1.59$ in the pure and $n_f = 1.52$ in the Th-doped sample. While these values deviate from the constraint $n_f = 1$ usually enforced in the Kondo model [12], the good agreement between the theoretical and experimental results suggest that corrections due to valence fluctuations are small.

Finally, we discuss the $dI/dV$ Fano-lineshape observed above the HOT [9] and its relation to the conduction band observed in QPI [8]. To this end, we extend our analysis of the STS results by Schmidt et al. [8] by assuming a vanishing hybridization, $s = 0$, for $T > T_0$. The resulting QPI intensity $|S(q, \omega)|$ shown in Fig. 3(c) reproduces well the experimental QPI dispersion (black line) of Fig. 5(b) in Ref. [8]. Due to the smallness of $t_f/t_c = 0.0075$, and since $s = 0$, the contribution of the heavy $f$-electron band to $|S(q, \omega)|$ and $dI/dV$ is negligible, thus explaining its absence in the experimental QPI data above the HOT [8]. However, the resulting $dI/dV$ lineshape does not exhibit the characteristic Fano form, implying that its origin resides in electronic bands not yet seen in QPI (the sharp drop in $dI/dV$ at $\omega \approx 40$ meV signifies the upper band edge of $\varepsilon_k$). To further investigate this possibility, we present in Fig. 3(c) a theoretical fit using Eq. [6] to the experimental $dI/dV$ data of Ref. [8] above the HOT on a Si-terminated surface in a pure sample, which are similar to those of Ref. [9] on a U-terminated surface. The resulting bands, $E_k$, shown in Fig. 2(d), are not only significantly different from the ones seen in QPI below the HOT, but also exhibit much larger quasi-particle dampings, $\Gamma_{f,c}$, thus representing an incoherent Kondo lattice. As a result, $|S(q, \omega)|$, shown in Fig. 3(d), exhibits very little $q$-structure (for fixed $\omega$), thus explaining the difficulty in detecting these bands in QPI. We therefore conclude that an explanation of the STS data above and below the HOT requires multiple sets of $c$- and $f$-electron bands.

We have shown that the STS results by Schmidt et al. [8] and Aynajian et al. [9] are consistent with the emergence of a coherent Kondo lattice below the HOT in URu$_2$Si$_2$. While it is not expected that the CKL is the primary origin of the HOT [13], it could be a result of the HOT. In particular, one might speculate that the emergence of a yet unknown order parameter at $T_0$ could lead to a significant decrease in the quasi-particle decoherence, for example, through the gapping of low-energy excitations, and thus induce the formation of a coherent Kondo lattice, as described above. Clearly, further studies are required to investigate this possibility.

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[1] M.B. Maple et al., J. Low Temp. Phys. 99, 223 (1995); A. Schroder et al. Nature (London) 407, 351 (2000); G.R. Stewart, Rev. Mod. Phys. 73, 797 (2001); J. Custers, et al. Nature (London) 424, 524 (2003); H. von Lohneysen et al., Rev. Mod. Phys. 79, 1015 (2007); P. Gegenwart, Q. Si, and F. Steglich, Nature Physics, 4, 186 (2008).

[2] J. Kondo, Prog. Theor. Phys. 32, 37 (1964).

[3] S. Doniach, Physica B 91, 231 (1977).

[4] T. T. M. Palstra et al. Phys. Rev. Lett. 55 2727 (1985).

[5] M.B. Maple, et al. Phys. Rev. Lett. 56, 185 (1986).

[6] J. Schoenes et al., Phys. Rev. B 35, 5375 (1987); D.A. Bomn et al., Phys. Rev. Lett. 61, 1305 (1988); S.V. Dordevic, et al., Phys. Rev. Lett. 86, 684 (2001); J.G. Rodrigo et al. Phys. Rev. B. 55, 14318 (1997); R. Escudero, F. Morales, and P. Lejay, Phys. Rev. B 49, 15271 (1994); J.D. Denlinger et al., J. Elec. Spectrosc. 117/118, 347 (2001).

[7] D.L. Cox, Phys Rev Lett 59, 1240 (1987); P. Chandra et al., Nature 417, 831 (2002); C.M. Varma and L. Zhu, Phys. Rev. Lett. 96, 036405 (2006); K. Haule, and G. Kotliar, Nature Phys. 5, 796 (2009); A.V. Balatsky et al., Phys. Rev. B 79, 214413 (2009); S. Elgazzar, et al., Nature Mater. 8, 337 (2009); P. M. Oppeneer et al., Phys. Rev. B. 82, 205103 (2010); C. Pepin et al., arXiv:1010.1237; Y. Dubi and A.V. Balatsky, arXiv:1011.5315.

[8] A.R. Schmidt et al. Nature 465, 570 (2010).

[9] P. Aynajian et al., PNAS 107, 10383 (2010).

[10] U. Fano, Phys. Rev. 124, 1866 (1961).

[11] Q.M. Si et al., Nature 413, 804 (2001).

[12] T. Senthil, S. Sachdev, and M. Vojta, Phys. Rev. Lett. 90, 216403 (2003); I. Paul, C. Pepin, and M.R. Norman, Phys. Rev. Lett. 98, 026402 (2007).

[13] P. Coleman, Handbook of Magnetism and Advanced Magnetic Materials (Wiley and Sons, 2007).

[14] N. Read and D. M. Neumns, J. Phys. C 16, 3273 (1983); P. Coleman, Phys. Rev. B 28, 5255 (1983); N. E. Bickers, Rev. Mod. Phys. 59, 845 (1987); A.J. Millis and P.A. Lee, Phys. Rev. B 35, 3394 (1987).

[15] A. D. Hewson, The Kondo Problem to Heavy Fermions (Cambridge University Press, Cambridge, 1997).

[16] J.R. Iglesias, C. Lacroix, and B. Coqblin, Phys. Rev. B 56, 11820 (1997).

[17] R.K. Kaul and M. Vojta, Phys. Rev. B 75, 132407 (2007).

[18] J.Figgins and D.K. Morr, preprint, arXiv:1001.3875.
[19] M. Maltseva, M. Dzero, and P. Coleman, Phys. Rev. Lett. 103, 206402 (2009).
[20] J. Figgins, and D.K. Morr, Phys. Rev. Lett. 104, 187202 (2010).
[21] P. Wölfle, Y. Dubi, and A.V. Balatsky, Phys. Rev. Lett. 105, 246401 (2010).
[22] S. Ernst et al., preprint.