Hybridization Wave as the "Hidden Order" in URu_2Si_2

Yonatan Dubi[1,*] and Alexander V. Balatsky[1,2]

[1]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

[2]*Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

(Received 23 November 2010; published 23 February 2011)

A phenomenological model for the "hidden order" transition in the heavy-Fermion material URu_2Si_2 is introduced. The hidden order is identified as an incommensurate, momentum-carrying hybridization between the light hole band and the heavy electron band. This modulated hybridization appears after a Fano hybridization at higher temperatures takes place. We focus on the hybridization wave as the order parameter in URu_2Si_2 and possibly other materials with similar band structures. The model is qualitatively consistent with numerous experimental results obtained from, e.g., neutron scattering and scanning tunneling microscopy. Specifically, we find a gaplike feature in the density of states and the appearance of features at an incommensurate vector $Q^* \sim 0.6\pi/a_0$. Finally, the model allows us to make various predictions which are amenable to current experiments.

DOI: 10.1103/PhysRevLett.106.086401 PACS numbers: 71.27.+a, 75.20.Hr, 75.30.Mb

The heavy Fermion material URu_2Si_2 exhibits an enigmatic hidden order (HO) transition at a temperature $T_{HO} = 17.5$ K [1–3]. Although being more than two decades old, the nature of the transition (which is a typical mean-field second order transition) and the order parameter which describe it are still unknown [4,5]. Various theories have been put forward to explain the transition, some of them describing it as a localized one [6–11] and some describe it as an itinerant phenomenon [12–16].

In the last few years various new experimental techniques, previously unavailable, were presented, and now provide additional valuable hints as to the origin of the Hidden Order transition. The key experimental observations relevant for our discussion are: (i) In inelastic neutron-scattering measurements [17] (as well as other measurements [18]) enhanced scattering is observed at the incommensurate wave vectors $Q^* \sim 0.6,\ 1.4\pi/a_0$ below the HO transition. A spin-gap-like feature of $\Delta \sim 5$ meV is observed, as extracted from the heat capacity measurements. In many heavy-Fermion systems the spin gap is indicative of the accompanying charge gap [19]. (ii) Angle-resolved photoemission spectroscopy measurements indicate that the heavy f band is very weakly dispersive, crosses the Fermi energy at the HO transition, and settles slightly below it [20]. (iii) A Fano line shape in the density of states (DOS) which starts to form at $\sim$120 K has been seen in recent scanning tunneling microscopy (STM) measurements [21,22]. This Fano line shape is fully developed at $\sim$20 K [inset of Fig. 2(a)]. (iv) At the HO transition temperature, the bottom of the Fano line shape develops a gaplike feature which evolves with temperature [21,22] [inset of Fig. 2(b)]. Both the Fano parameters and the gap structure depend on the STM tip position and modulate in space with the positions of the different atoms. (v) The hole band develops a hybridization feature below the HO transition, corresponding to momentum $Q \approx 0.3\pi/a_0$, close to where the heavy Fermion band crosses it [21].

These observations seem to suggest that a description of the HO in terms of reorganization of the localized degrees of freedom may be incomplete. Guided by the experimental findings, we construct a phenomenological theory for the HO transition. A one-dimensional model system is taken for simplicity, which contains a light hole (d) band and a heavy electron (f electrons, dashed line) band, depicted in the inset of Fig. 1. We also performed the same calculation using bands generated by local density approximation (LDA) as input (to be shown elsewhere). Both from the model and from LDA based bands we take the bands crossing at momentum points $Q \approx \pm 0.3$ (along (1,0), (0,1) in units of π/a_0, where a_0 is the lattice spacing, in which all momenta are measured hereafter). The holes first hybridize with the local part of the f electrons,

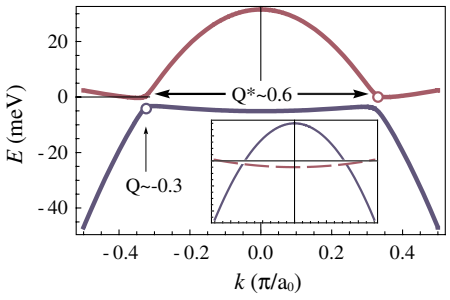

FIG. 1 (color online). Schematic representation of the band structure assumed in the model and the incommensurate scattering. The Heavy electron f band and the light hole d band cross at $Q \approx \pm 0.3$ (inset). The resulting scattering hybridizes electrons with momentum Q and holes with momentum $-Q$ (and vice versa), generating a hybridization wave which is the HO order parameter. The $Q^* = 2Q \approx 0.6$ momentum transfer is in accord with Neutron-scattering experiments [17].

giving rise to a Fano line shape in the DOS [11,23–26] at temperatures that are much higher then the HO transition. The band structure gives rise to enhanced hybridization between electrons with momentum Q and holes with momentum $-Q$ (and vice versa, Fig. 1) at specific points along the Fermi surface (''hot spots''). The resulting electron-hole coherence (which may be considered as an *indirect exciton*) is the HO order parameter. Since the ''hot spots'' are located at the band crossing, this incommensurate scattering generates a gap structure in the DOS with a line shape similar to that observed in STM experiments [21,22].

The starting point of the model is a two-band Hamiltonian,

$$\mathcal{H} = \sum_k \varepsilon_k^{(c)} c_k^\dagger c_k + \sum_k \varepsilon_k^{(f)} f_k^\dagger f_k, \tag{1}$$

where $c_k^\dagger$ ($f_k^\dagger$) creates a hole (electron) with momentum k. For simplicity (and as it plays no important role in the physics we describe) the electronic spin is omitted. In real materials the d-f hybridization sets in at much higher temperatures then HO transition and hence the remnant heavy band and light hole bands we use here are already hybridized. We assume that lighter band is more d character and heavier band has more f character near the Γ point where we concentrate.

The origin of the Fano line shape has been a subject of recent study [24–27]. In the absence of either translation invariance breaking or level broadening, a Fano line shape should not appear [28]. A simple mechanism for its appearance is based on local scattering between the d electrons and the f electrons under the STM tip (the effect of the tip may not be negligible, [29]). The Hamiltonian is then given by $\mathcal{H}_F = V_0 \sum_k c_k^\dagger f_{r=0} + \text{H.c.}$, where $f_{r=0}$ creates an f electron on the lattice site $r = 0$, i.e., under the tip. Alternatively, a similar result as below can be obtained by assuming full decoherence between the f orbitals which lie on different lattice sites [28].

It is now straightforward to write the Dyson's equations, with the solutions for the Green's functions for the f- and d-electrons $f = \frac{1}{\omega - \varepsilon_0 - V_0^2 \chi_0}$, $g_{k,k'} = g_k^{(0)} \delta_{k,k'} + \frac{V_0^2}{\omega - \varepsilon_0 - V_0^2 \chi_0} g_k^{(0)} g_{k'}^{(0)}$. Here $g_k^{(0)}$ is the bare d-electron propagator, and its susceptibility $\chi_0 = \sum_k g_k^{(0)} = -\Gamma_0 (i - q)$, which defines Γ_0 (proportional to the bare d-band DOS, which we assume constant for simplicity, and set it as $\Gamma_0 = 1$ hereafter) and q (which describes the ratio between the real and imaginary part of the bare self-energy, and is known as the Fano factor). The level ε_0 is not the bare f level, but rather a renormalized value, which may arise due, e.g., to hybridization with other bands, inelastic scattering effects, interaction renormalization, etc., ε_0 can be lower than the original f level [27], and therefore, a negative value of ε_0 does not imply an assumption that the f level lies below the Fermi energy. The local DOS is given by $\rho = -\frac{1}{\pi} \Im \sum_{k,k'} g_{k,k'}$. To be specific in comparing our results with experiments we fix parameters used as $\Gamma_1 = V_0^2 \Gamma_0 = 5$ (all energy scales are taken in meV), the bottom of the f band $\varepsilon_0 = -5$ (shifting this energy does not alters the line shape) and the Fano factor $q = 1.5$ (these numerical parameters are maintained hereafter unless stated otherwise). This line shape is to be compared with that obtained from STM measurements, shown in the inset of Fig. 2 [21,22]. In our treatment, temperature only enters via the temperature dependence of the couplings. The hybridization feature is of $\Delta \sim 5$ meV ~ 60 K, and the HO transition is at 17.5 K, therefore the finite temperature should smear the DOS line shape, but not change its features.

The next step is to define the effective interaction between the f electrons and the d holes which gives rise to the HO. The central part of our proposal for the HO is to assume that the most important electron correlations affect matrix element between d-band holes with momentum $-Q$ and f electrons with momentum Q (and vice versa), with $Q \approx 0.3$ (the position where the bands cross, Fig. 1). This assumption is consistent with the observed sharp onset of the hybridization feature at $Q^* = 2Q$ seen in quasiparticle

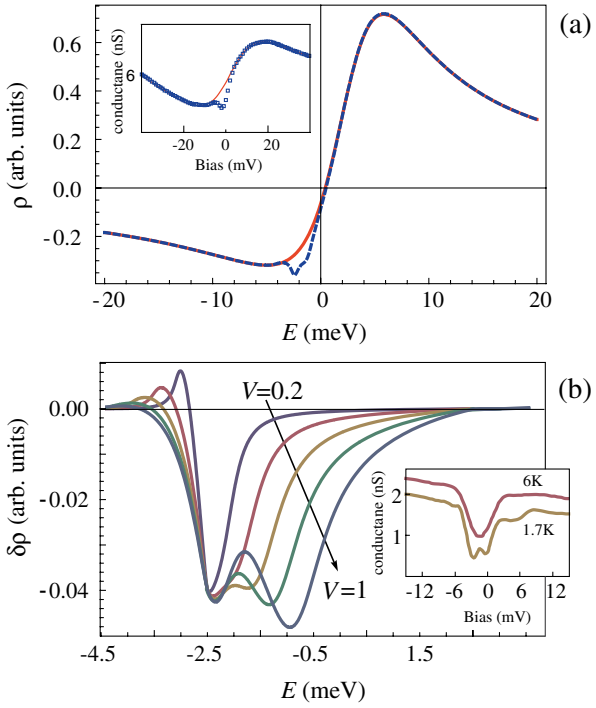

FIG. 2 (color online). (a) Local DOS for a d band coupled to the localized f electrons, exhibiting an asymmetric Fano line shape (solid line, see text for parameters). The dashed line is the local DOS in the presence of the hidden order. These are in good agreement with the DOS (proportional to conductance) seen in STM experiments, shown in the inset (extracted with permission from [21]). (b) Change in DOS due to the HO coupling, $\delta\rho = \rho(V) - \rho(V = 0)$, for $V = 0.2, 0.4, \ldots, 1$. The main features are the appearance of a gap structure (which scales with V, see Fig. 3), and a peak structure within the gap, appearing for large enough V (corresponding to low enough temperature). Both these features have been observed experimentally [21,22], as seen in the inset (extracted with permission from [21]).

interference [21]. Thus, we take the interaction Hamiltonian as

$$\mathcal{H}_I = \sum_{kk'} U_{kk'} c^\dagger_{k-Q} f^\dagger_{k'-Q} f_{k'+Q} c_{k+Q} + (Q \to -Q), \quad (2)$$

where U_k is strongly peaked around $k = 0$ (the width of U_k determines the range of the effective d-f interactions). To continue analytically, we set $k = k'$ and $U_k \sim U\delta_{k,0}$, resulting in the effective Hamiltonian $\mathcal{H}_I \approx U_{Q,-Q} c^\dagger_{-Q} f^\dagger_{-Q} f_Q c_Q + (Q \to -Q)$. The physical meaning of these assumptions is that (i) the most important scattering is with momentum transfer Q [16,17,30], and (ii) the electron interactions are long-ranged [31]. Since the HO transition seems like a mean-field transition (based on the shape of the heat capacity [6] and the dependence of the gap on temperature [22]), it is natural to decouple $\mathcal{H}_I$ in a mean-field way. We thus identify the HO order parameter V as

$$V = V_{\mathrm{HO}} = U_{-Q,Q} \langle c^\dagger_{-Q} f_Q \rangle. \quad (3)$$

The hidden nature of this excitonic order is evident from the observation that the expectation value of the spin and of the charge (density waves) would be zero in this state hence there is no primary single spin and or charge order in this hybridization wave state. We do expect a charge density modulation at momentum $Q^* = 2Q$ as a secondary effect, which can be however very small [32]. In the mean-field approximation, V depends on temperature as $V \propto (T_{\mathrm{HO}} - T)^{1/2}$. The interaction Hamiltonian in the mean-field approximation takes the form $\mathcal{H}_{I,\mathrm{MF}} = V c^\dagger_{-Q} f_Q + \mathrm{H.c.}$

We now write the Dyson Equation for the Green's function in the presence of $\mathcal{H}_I$. We point that if $\mathcal{H}_I$ would operate on the bare Hamiltonian, then no correction (other than at the points $k = \pm Q$) would be observed. However, the d band is already dressed by the Fano interaction described above, and thus the correction is to all momenta. Since the bands are symmetric in k, we can treat only the $-Q \to Q$ (the full calculation which includes also $Q \to -Q$ scattering gives similar results). The Dyson equations are

$$\begin{aligned} G_{k,k'} &= g_{k,k'} + V^2 g_{k,-Q} F_{Q,Q} G_{-Q,k}, \\ F_{Q,Q} &= f_{Q,Q} + V^2 f_{Q,Q} G_{-Q,-Q} F_{Q,Q}, \end{aligned} \quad (4)$$

which are easily solved by simple algebra.

The resulting local DOS $\rho = \sum_{k,k'} G_{k,k'}$ is plotted in Fig. 2(a) (dashed line) for $V = 0.8$ and $\varepsilon_Q = -2.5$. in Fig. 2(b) we plot the change in DOS due to the HO coupling, $\delta\rho = \rho(V) - \rho(V = 0)$, for $V = 0.2, 0.4, \ldots, 1$. As seen, a gaplike feature develops, its width and position depending on the value of V (which in turn depends on temperature). Note also the additional peak at the bottom of the gap, which was observed experimentally [21,22]. For comparison, in the inset of Fig. 2(b) we plot the experimental DOS taken from [21]. The qualitative agreement between the data and theory is evident. A quantitative fit would require additional parameters to account for the complicated background DOS seen in the experiments.

From the DOS one can extract the size of the energy gap Δ as a function of V (for instance, by taking the distance between the peak just left of the gap and the point where the curvature is maximal just right of the gap). The dependence of Δ on $V^2 \propto (T_{\mathrm{HO}} - T)$ is plotted in Fig. 3(a). We find that $\Delta \propto (T_{\mathrm{HO}} - T)^\nu$, with $\nu \approx 0.4$ [solid line in Fig. 3(a)], in good agreement with the exponent extracted from the experimental data [22] and in agreement with the mean-field treatment assumed.

A pronounced feature of the DOS is the appearance of an additional peak structure within the gap in the DOS [Fig. 2(b)]. In the experiment [21,22] this feature only appears at $T \ll T_{\mathrm{HO}}$ which we interpret as large enough V, as it grows below T_{HO}. In Fig. 3(b) we have plotted the position of the minimal point of the DOS (E_{min}, empty circles) and the position of the peak within the gap (E_{peak}, full circles) as a function of the HO order parameter V (which, in experiment, corresponds to temperature). The peak structure only appears for large enough V, and scales with temperature with the same exponent as the energy gap itself, $E_{\mathrm{peak}} \propto \Delta \propto (T_{\mathrm{HO}} - T)^{0.4}$. E_{min}, on the other hand, quickly saturates to become temperature-independent.

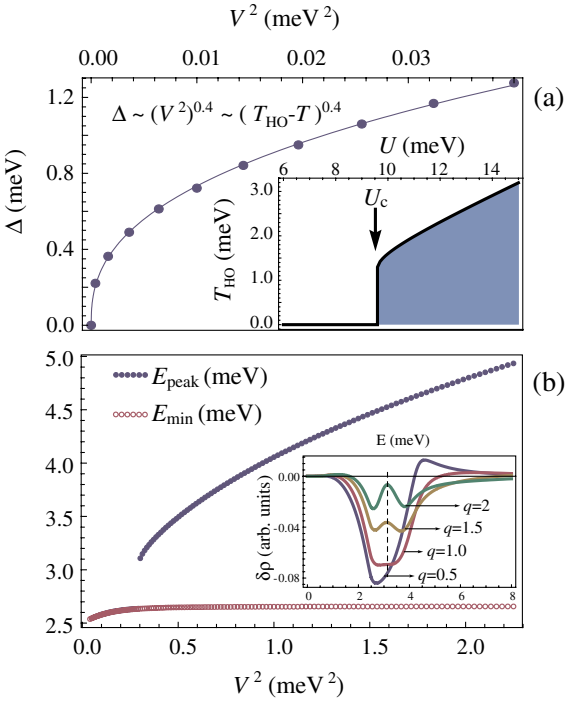

FIG. 3 (color online). (a) The energy gap, as obtained from the DOS (Fig. 2), as a function of the HO order parameter V squared, which is proportional to $(T_{\mathrm{HO}} - T)$. The solid line is the function $\Delta \propto (T_{\mathrm{HO}} - T)^{0.4}$. Inset: the dependence of T_{HO} on the interaction strength U, indicating a possible first order transition at $U = U_c$. (b) Position of the minimal point of the DOS (E_{min}, empty circles) and the position of the peak within the gap (E_{peak}, full circles) as a function of the HO order parameter V^2 (which corresponds to temperature). While E_{min} saturates at large V, $E_{\mathrm{peak}} \sim V \sim |T - T_{\mathrm{HO}}|^{0.4}$ in the same way as the gap Δ (Fig. 3). Inset: change in the DOS, $\delta\rho$ for various values of the Fano factor q (for $V = 0.5$).

Both these quantities could be examined in a detailed STM experiment.

In the inset of Fig. 3(b) we have plotted the DOS change $\delta\rho$ for different values of the Fano factor, $q = 0.5, 1, 1.5, 2$ (for $V = 0.5$). In the STM experiments, the value of the Fano factor changes, depending on whether the tip is positioned above a Si site or a uranium site. Presumably, when the tip is above the Si site it has better coupling to the d band, which effectively increases the Fano factor. Regardless of the reason for the q-factor modulation, we predict that the height of the peak structure within the gap will also modulate (and will be commensurate with the modulation of q), but its position will not change [dashed line in the inset if Fig. 3(b)].

The order parameter we discuss here $V_{\mathrm{HO}} = U_{-Q,Q}\langle c^{\dagger}_{-Q} f_Q\rangle$ also carries a nonzero expectation value for the dipole moment modulation at Q^*. In the coherent excitonic state $|\Psi\rangle = \prod_k (u_k c^{\dagger}_{-k} + v_k f^{\dagger}_k)|0\rangle$ there is a nonzero expectation value for the dipolar moment at momentum Q^*: $\langle |P_{2Q}|\rangle = \langle\Psi|\hat{r}e^{i2Qr}|\Psi\rangle$, as long as the parity of d and f orbitals is different. This modulation of the dipole moment could be observed with probes that couple to electric density modulation, for example, an STM with a ferroelectric (or hybrid ferroelectric-metallic) tip.

The formalism described above allows us to calculate the critical temperature of the HO transition. Working in a Nambu-like space mixing d and f electrons and using Eq. (3), we identify the self-consistency equation for the HO,

$$-\frac{1}{U_{-Q,Q}} = T_{\mathrm{HO}} \sum_{i\omega_n} f_Q(i\omega_n, Q) g_{-Q}(i\omega_n), \qquad (5)$$

where $f_Q(i\omega_n)$ and $g_{-Q}(i\omega_n)$ are the Matsubara Green's function in the presence of the Fano interaction, evaluated at momentum Q, and $i\omega_n$ are Matsubara frequencies. We have calculated the critical temperature T_{HO} for the parameters above as a function of U, shown in the inset of Fig. 3(a). We find that for U below a certain value U_c, Eq. (5) does not have a solution, due to the compact nature of the interaction in momentum space. This feature may explain why the hidden order has not been universally observed in other heavy-Fermion materials, even ones with a similar band structure. It may also be relevant to the destruction of the HO and appearance of magnetic ordering with applied pressure, with a 1st order transition between them [33,34] A possible scenario for the connection between the HO and magnetic order (clearly avoided in the present study which discusses spinless electrons) is the following. As pressure is applied the interaction strength decreases, until finally it becomes lower than U_c. The magnetic interaction between the localized f electrons, previously screened by the conduction electron which are coupled to the f electrons in the HO state, becomes significant and gives rise to magnetic ordering. This scenario will be studied in detail in the future.

We thank P. Wölfle, M. Graf, A. Chantis, K. Haule, T. Durakiewicz, G. Kotliar, and J.-X. Zhu for valuable discussions, and A. Schmidt, M. Hamidian and C. S. Davis for stimulating discussions and for sharing with us their STM data. This work was supported by BES, UCOP-TR-01 and in part, by grant No. DE-AC52-06NA25396 and

*Present address: The School of Physics and Astronomy, Tel-Aviv University, Israel.

[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] V. Tripathi *et al.*, Nature Phys. **3**, 78 (2007).
[5] N. Shah *et al.*, Phys. Rev. B **61**, 564 (2000).
[6] P. Chandra *et al.*, Nature (London) **417**, 831 (2002).
[7] P. Santini, Phys. Rev. B **57**, 5191 (1998).
[8] T. Kasuya, J. Phys. Soc. Jpn. **66**, 3348 (1997).
[9] A. E. Sikkema *et al.*, Phys. Rev. B **54**, 9322 (1996).
[10] F. Cricchio *et al.*, Phys. Rev. Lett. **103**, 107202 (2009).
[11] K. Haule and G. Kotliar, Nature Phys. **5**, 796 (2009).
[12] H. Ikeda and Y. Ohashi, Phys. Rev. Lett. **81**, 3723 (1998).
[13] C. M. Varma and L. Zhu, Phys. Rev. Lett. **96**, 036405 (2006).
[14] S. Elgazzar *et al.*, Nature Mater. **8**, 337 (2009).
[15] H. Harima *et al.*, J. Phys. Soc. Jpn. **79**, 033705 (2010).
[16] A. V. Balatsky *et al.*, Phys. Rev. B **79**, 214413 (2009).
[17] C. R. Wiebe *et al.*, Nature Phys. **3**, 96 (2007).
[18] C. Broholm, Phys. Rev. Lett. **58**, 1467 (1987).
[19] For recent reviews see, e.g., P. S. Riseborough, Adv. Phys. **49**, 257 (2000); D. T. Adroja *et al.*, J. Optoelectron. Adv. Mater. **10**, 1564 (2008).
[20] A. F. Santander-Syro, Nature Phys. **5**, 637 (2009).
[21] A. R. Schmidt, Nature (London) **465**, 570 (2010).
[22] P. Aynajian *et al.*, Proc. Natl. Acad. Sci. U.S.A. **107**, 10 383 (2010).
[23] V. Madhavan *et al.*, Phys. Rev. B **64**, 165412 (2001).
[24] Y.-F. Yang, Phys. Rev. B **79**, 241107 (2009).
[25] M. Maltseva, M. Dzero, and P. Coleman, Phys. Rev. Lett. **103**, 206402 (2009).
[26] J. Figgins and D. K. Morr, Phys. Rev. Lett. **104**, 187202 (2010).
[27] M. Fogelström *et al.*, Phys. Rev. B **82**, 014527 (2010).
[28] P. Wolfle, Y. Dubi, and A. V. Balatsky, Phys. Rev. Lett. **105**, 246401 (2010).
[29] P. Cheng *et al.*, Phys. Rev. Lett. **105**, 076801 (2010).
[30] This assumption is consistent with the appearance of superconductivity at lower temperatures, since it implies the Fermi surface is only partially gapped, allowing for SC correlations to develop.
[31] The fact that the interaction is limited to $k = k' = 0$ implies infinite-long-ranged electron interactions. While this is clearly an approximation (taken to allow for an analytic solution) the fact that the interactions might not be short-ranged is quite reasonable, noting that the screening length in this system could be reasonably long.
[32] J.-J. Su *et al.*, J. Phys. Condens. Matter (to be published).
[33] E. Hassinger *et al.*, Phys. Rev. B **77**, 115117 (2008).
[34] P. G. Niklowitz *et al.*, Phys. Rev. Lett. **104**, 106406 (2010).