HIDDEN ORDER IN URu_2Si_2 THE NEED FOR A DUAL DESCRIPTION*

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Motivated by experiment, we argue that the enigmatic hidden order in URu_2Si_2 demands a dual description that encompasses both its itinerant and its local aspects. A combination of symmetry considerations and observation allow us to rule out a number of possibilities. The two remaining scenarios, the quadrupolar charge density wave and the orbital antiferromagnet, are discussed and experiments are suggested to select between these proposals.

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The heavy fermion metal URu_2Si_2 undergoes a pronounced second-order phase transition at $T_0 = 17$ K characterized by sharp anomalies in a number of bulk properties including the specific heat [1], the linear [1] and the nonlinear magnetic susceptibilities [2,3], where standard mean-field relations

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between the measured thermodynamic properties are satisfied [4]. Neutron scattering measurements [5–7] indicate gapped, propagating magnetic excitations that suggest the formation of a spin density wave. However the magnitude of the observed moment [7] ($m_0 = 0.03\mu_{\rm B}$) cannot account for the entropy loss at the transition, which has been attributed to the development of an enigmatic hidden order. Furthermore there is strong evidence from muon spin resonance [8] and from pressure-dependent NMR experiments [9] that the magnetic and the hidden ordered phases are phase separated, and thus develop independently [10]. In this paper we review the constraints that recent experimental developments and symmetry arguments place on the nature of the hidden order in URu_2Si_2, and discuss specific theoretical proposals that emerge consistent with known measurements.

It is important to distinguish two distinct aspects of the mysterious phase transition at T_0 in URu_2Si_2. The presence of a Schottky anomaly at 60 K in the specific heat [1] and the development of a dispersing mode at $T \leq T_0$ observed by inelastic neutron scattering [7] both suggest the importance of local crystal-field excitations at the transition. Nevertheless a purely local picture *cannot* provide a straightforward explanation for the observed elastic response [11] at T_0 and yields a field-dependence for the gap [12] that is *distinct* from that associated with observed thermodynamic quantities [13]. Furthermore the clear signatures [1] of Fermi liquid behavior above T_0 and the mean-field nature of the transition [4] suggest that an itinerant density-wave is involved. Thus any microscopic model for the hidden order in URu_2Si_2 must ultimately reconcile the local electronic physics of the strongly interacting uranium ions with the fluid aspects of the heavy-electron phase.

The duality model of Kuramoto and Miyake [14] is a natural way to treat the localized and itinerant aspects of URu_2Si_2 within a single scheme. In this approach, the itinerant excitations of the Fermi liquid are constructed from the low-lying crystal-field multiplets of the uranium ion. The quasiparticles associated with the heavy-electron Fermi liquid in this system are then composite objects formed from the localized orbital and spin degrees of freedom of the U ions and the conduction electron fields [14]. The phase transition at T_0 is a simple Fermi surface instability of the composite mobile f-electrons. This approach was originally adopted by Okuno and Miyake [15] to describe the coexistence of the hidden order with a small moment in URu_2Si_2. With the new understanding that the hidden ordered phase does not contain a staggered magnetization, we revisit this duality scheme and, guided by recent experiment, study its implications for the nature of the enigmatic order that develops at $T = T_0$.

We begin our phenomenological approach to the hidden order in URu_2Si_2 with a consideration of its allowed symmetries, where constraints will be imposed by experimental observation. More specifically, the mean-field character of the transition at T_0 suggests that the itinerant nature of the hidden order can be described by a general density wave whose form factor will yield clues about the underlying local excitations involved; we expect it to be incommensurate due to the fact that the observed entropy loss and the accompanying gap suggest that it results from a Fermi surface instability. We begin by considering a class of density wave with the most general pairing in the particle–hole channel

$$\langle c^{\dagger}_{\boldsymbol{k}+\frac{\boldsymbol{Q}}{2},\sigma} c_{\boldsymbol{k}-\frac{\boldsymbol{Q}}{2},\sigma'} \rangle \;=\; A^{\sigma\sigma'}_{\boldsymbol{k}}(\boldsymbol{Q}), \tag{1}$$

where $\boldsymbol{Q}$ is the incommensurate ordering wave vector, σ is a spin index and $A^{\sigma\sigma'}_{\boldsymbol{k}}(\boldsymbol{Q})$ defines a general function of spin and momentum. A phenomenological Hamiltonian for such an order parameter would take the form

$$\begin{aligned} H \;=\; & \sum_{\vec{k}\sigma} \epsilon_{\vec{k}} c^{\dagger}_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma} \\ & + \sum_{\boldsymbol{k}} \left[\Delta(\boldsymbol{Q}) c^{\dagger}_{\boldsymbol{k}+\boldsymbol{Q}/2,\sigma} f_{\sigma\sigma'}(\boldsymbol{k}) c_{\boldsymbol{k}-\boldsymbol{Q}/2,\sigma} + \text{H.c.} \right] + \mathcal{N} \frac{|\Delta(\boldsymbol{Q})|^2}{g}\,, \end{aligned} \tag{2}$$

where $\mathcal{N}$ is the number of sites in the lattice and $f_{\sigma\sigma'}$ is a form factor associated with the incommensurate spin density wave; a mean-field treatment yields the expression

$$-gA^{\sigma\sigma'}_{\boldsymbol{k}}(\boldsymbol{Q}) = \Delta(\boldsymbol{Q}) f_{\sigma\sigma'}(\boldsymbol{k})\,. \tag{3}$$

Of course, a true microscopic approach must account for how such a coupling emerges from the residual interactions amongst the heavy electrons.

We now follow the approach of Halperin and Rice [16], categorizing the possible particle–hole pairings in URu_2Si_2. Assumingthee hidden order develops between U atoms in each basal plane, we restrict our attention to nearest-neighbor pairings on a two-dimensional square lattice [10] and display the five resulting possibilities in Table I. All of these pairings lead to a gapping of the Fermi surface, accounting for the large entropy loss and the observed anomalies in various bulk properties. For example, let $\Delta\chi$ be the reduction in the Pauli susceptibility due to this gap, then we can describe the hidden order phase by a Landau–Ginzburg free energy

$$F = a(T - T_{\rm c})\,\Psi^2 + b\Psi^4 + \tfrac{1}{2}\Delta\chi B^2\,\Psi^2. \tag{4}$$

such that the last term is responsible for the strong field-dependence of the transition temperature ($T_{\rm c}(B) = T_{\rm c} + \frac{1}{2a}\Delta\chi B^2$) in this material [17]; it is also responsible to why χ_3 has the same temperature-dependence as the specific

TABLE I

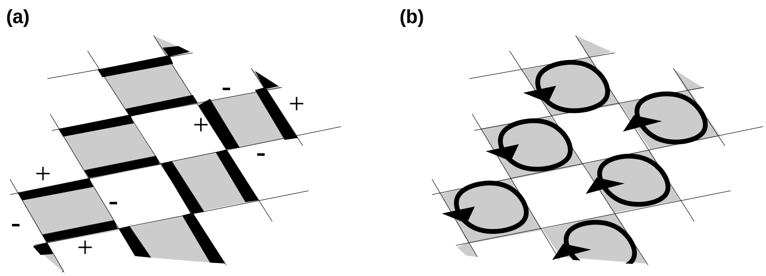
Possible symmetries for particle–hole pairing.

Name	$f_{\sigma\sigma'}(\boldsymbol{k})/\delta_{\sigma\sigma'}$	T–reversal -invariance	Local fields
SDW (isotropic SDW)	σ	no	yes
CDW (isotropic CDW)	const.	yes	no
d-SDW	$\sigma\,(\cos(k_x a) - \cos(k_y a))$	no	no
Q-CDW (quadrupolar CDW)	$\cos(k_x a) - \cos(k_y a)$	yes	no
OAFM (orbital antiferromagnet)	$i\,(\sin(k_x a) - \sin(k_y a))$	no	yes

heat [3, 4]. Unfortunately this aspect of the problem does not discriminate between the possible density wave order parameters, and we are forced to make more microscopic considerations.

Because of large Coulomb repulsion between the quasiparticles in URu_2Si_2, we rule out isotropic pairing in the charge density wave channel. Furthermore, an s-wave charge density wave would result in an accompanying lattice distortion, but none is observed [11]. Similarly neutron scattering does not support the presence of a spin density wave in the hidden ordered phase [7, 9]. Thus, due to the incompressibility of the heavy Fermi liquid, we are left with the three remaining anisotropic pairing states (see Table I). We recall that URu_2Si_2 undergoes a transition to a d-wave superconducting state at $T = 1.2$ K, suggesting the importance of antiferromagnetic fluctuations in the associated normal state; by contrast in a d-SDW scenario [3] we would expect ferromagnetic fluctuations to be favored.

This leaves us with two remaining options: the quadrupolar charge density wave [18] (Fig. 1(a)) and the orbital antiferromagnet (Fig. 1(b)) [19]. Both of these scenarios are consistent with the current picture of URu_2Si_2 as an incompressible Fermi liquid with strong antiferromagnetic fluctuations. They each have nodes in the order parameter, so that neither couple directly to the local charge density. Furthermore we expect both incommensurate density waves to couple weakly to uniform strain, and thus are both consistent with the observed insensitivity of the elastic response [11] at T_0. Recent uniaxial stress measurements suggest that the hidden order is sensitive to the presence of local tetragonal symmetry [20], a feature that is consistent with both scenarios for completely different reasons. In the orbital antiferromagnet the currents are equal in each basal direction [19], whereas in the quadrupolar charge density wave it is known that some of the singlet crystal-field states with tetragonal symmetry are quadrupolar [12]. We note that the diamagnetic response of the orbital antiferromagnet is small compared



$f(\boldsymbol{k}) = \cos(k_x) - \cos(k_y)$ $f(\boldsymbol{k}) = i[\sin(k_x) - \sin(k_y)]$

Fig. 1. (a) Incommensurate quadrupolar density wave (Q-CDW). In two dimensions, the form factor $\cos(k_x) - \cos(k_y)$ leads to a an incommensurate density wave with a quadrupolar charge distribution, the CDW analog of a d-wave superconductor. (b) Incommensurate orbital antiferromagnet. Here currents circulate around square plaquettes defined by nearest-neighbor uranium ions.

to that associated with the gapping of the Fermi surface, $(\frac{\chi_{\rm Pauli}}{\chi_{\rm diamagnetic}} \sim 100)$, so that the field-dependence of the hidden order parameter [17] cannot be used to discriminate between these two scenarios.

At present, the key factor distinguishing the orbital antiferromagnet from the quadrupolar charge-density wave scenarios is the presence or absence of time-reversal breaking. Orbital antiferromagnetism is consistent with the isotropy and magnitude of local magnetic fields [19] measured at ambient pressure by nuclear magnetic resonance [21]. Such current-carrying states have also been proposed for one-dimensional ladders [22]. In URu_2Si_2 the line-broadening of the central silicon NMR peak is the only direct evidence for broken time-reversal symmetry in the hidden order phase [19]. Recent muon spin measurements by Amitsuka and coworkers [18] support the emergence of local magnetic fields with the temperature dependence seen in the NMR data [21], but the overall amplitude is two orders of magnitude less than that seen in NMR measurements. This has led to the proposal that these features may be an artifact of a minority ferromagnetic phase with dipolar interactions. In our opinion, an equally feasible explanation is that charged muons destroy the hidden order in their immediate surroundings. This would explain the similar but dramatically reduced anomalous magnetic field measured by the μSR measurements. Finally there have been attempts to test predictions for neutron scattering but so far no clear conclusion has been reached [23].

There are three types of experiments that would be very helpful in resolving the nature of the hidden order in URu_2Si_2:

- Probes of the nodal quasiparticle structure. Both Q-CDW and OAFM have nodes in the gap. These nodes would show marked features in the optical conductivity and the scanning tunneling spectroscopy; at present these nodes are inferred but have not been observed directly.

- Probes of broken time-reversal symmetry. Broken time-reversal symmetry is clearly a critical discriminating feature between the OAFM and Q-CDW. The existing NMR measurements strongly support the idea of broken time reversal symmetry breaking in this material, but further confirmation is vital. Further NMR measurements, especially on alternative crystal sites, would provide an important way of confirming whether there are local fields developing at T_0. Careful comparison between the μSR and NMR signals is crucial for determining whether these two techniques are measuring similar or different phenomena.

- Revisit old scattering measurements. There exist a number of scattering experiments, including polarized neutron and resonant X-ray, that are suggestive of broken time-reversal symmetry; these should be redone, given what is now known about phase-separation [9].

In summary, we believe that a dual description of the hidden order in URu_2Si_2 is necessary for capturing its coexisting itinerant and local features. The latter is necessary for explaining the mysterious dispersing mode at $T \leq T_0$, whereas the former is crucial for understanding the mean-field nature of the transition. At present there are two competing scenarios which differ by a form factors. We propose a number of experiments which could resolve the dispute.

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