Hidden order in URu_2Si_2

N. Shah
Department of Physics and Astronomy, Rutgers University, 136 Frelinghausen Road, Piscataway, New Jersey 08854-8019

P. Chandra
NEC Research, 4 Independence Way, Princeton, New Jersey 08540

P. Coleman
Department of Physics and Astronomy, Rutgers University, 136 Frelinghausen Road, Piscataway, New Jersey 08854-8019

J. A. Mydosh
Department of Physics and Astronomy, Rutgers University, 136 Frelinghausen Road, Piscataway, New Jersey 08854-8019;
NEC Research, 4 Independence Way, Princeton, New Jersey 08540;
Lucent Bell Laboratories, 100 Mountain Avenue, Murray Hill, New Jersey 07904;
and Kamerlingh Onnes Laboratory, Leiden University, P. O. Box 9506, 2300 RA Leiden, The Netherlands
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We review current attempts to characterize the underlying nature of the hidden order in URu_2Si_2. A wide variety of experiments point to the existence of two order parameters: a large primary order parameter of unknown character which co-exists with secondary antiferromagnetic order. Current theories can be divided into two groups determined by whether or not the primary order parameter breaks time-reversal symmetry. We propose a series of experiments designed to test the time-reversal nature of the underlying primary order in URu_2Si_2 and to characterize its local single-ion physics.

The nature of the hidden order parameter in URu_2Si_2 is a long-standing mystery in heavy fermion physics.[1] At 17.5 K this material undergoes a second-order phase transition characterized by sharp features in bulk properties including specific heat,[2] linear[2,3] and nonlinear[4,5] susceptibilities, thermal expansion,[6] and resistivity.[7] The accompanying gap in the magnetic excitation spectrum,[8,9] also indicated by the exponential dependence of the specific heat below the transition $\Delta C_V \propto e^{-\Delta/T}$, suggests the formation of an itinerant spin-density wave at this temperature. However, the size of the observed staggered moment[10] ($m_0 = 0.03\mu_B$) *cannot* account for the bulk properties, e.g., the entropy loss and the size of the gap which develops at the transition. This mismatch between the tiny ordered moment and the large entropy of condensation indicates the presence of a primary order parameter whose nature remains to be characterized.

Two sets of recent developments provide impetus for a renewed discussion of this material. In particular high-field measurements have emphasized the distinction between the hidden primary and the secondary magnetic order parameters. Though measurements of the high-field resistance,[12] thermal expansion,[13] and specific heat[14] indicate that the primary order parameter is destroyed by a field of 40 T, neutron-scattering results suggest that the magnetic order may disappear at a much lower field strength.[11] On a separate front, measurements of the specific heat, susceptibility, and thermal expansion[15,16] on dilute U in $Th_{1-x}U_xRu_2Si_2$ have provided insight into the uranium single-ion physics of this family of materials. Both of these quantities display a logarithmic dependence on temperature that is suppressed by a magnetic field, features suggesting the presence of a non-Kramers, Γ_5 *magnetic* doublet. Unlike a Kramer's doublet, this ionic ground state can be split by *both* magnetic and strain fields. These two sets of observations motivate us to propose further experiments designed to distinguish between various characterizations of the hidden order.

Many competing theories have been proposed for the primary hidden order in URu_2Si_2. The emphasis of these theoretical proposals has been on the microscopic order parameter. Broadly speaking, these theories divide into two distinct categories. In the first set, from here onwards designated as (A), the primary order parameter *breaks* time-reversal symmetry; proposals include spin-density waves in higher angular momentum channels,[5] three-spin order,[17] valence admixtures,[18] and antiferromagnetic states with strongly renormalized g factors,[19,20] By contrast the primary order parameter in category (B) is *invariant* under time-reversal symmetry, and staggered quadrupolar order[21] and Jahn-Teller distortions[22] are examples in this classification scheme. Unfortunately, experiment has been unable to clearly distinguish between these different microscopic proposals.

In this paper, we should like to turn the debate in a more phenomenological direction. We argue that as a necessary prelude to the development of a theory for the microscopic order parameter in URu_2Si_2, we need to ask two key questions:

(i) Does the primary order parameter break time-reversal symmetry?

(ii) What single-ion physics governs the low-energy behavior in stoichiometric URu_2Si_2?

At present neither question has been definitively answered, and to this end we propose a set of experiments designed to address these issues.

The ideal framework for our phenomenological discussion about the order parameter is Landau-Ginzburg theory. In this context, the distinction between theories in categories (A) and (B) lies in the allowed couplings between the primary and the secondary order parameters. Let us denote the primary and secondary order parameters by ψ and m, respectively. Quite generally the Landau-Ginzburg free energy must contain three terms:

$$\mathcal{F}[\psi,m]=\mathcal{F}_1[\psi]+\mathcal{F}_2[m]+\mathcal{F}_c[\psi,m]. \tag{1}$$

A number of experiments suggest that the hidden order is staggered.[5] Uniform order parameters tend to couple directly to macroscopic properties, e.g., the uniform magnetization and thus cannot be easily hidden. For simplicity, we assume that the primary (hidden) and secondary order parameters are commensurate; in this case, the free energy must satisfy

$$\mathcal{F}[\psi,m]=\mathcal{F}[-\psi,-m]. \tag{2}$$

Since antiferromagnetism seems to develop simultaneously with the hidden order, it is natural to consider coupling terms of the form

$$\mathcal{F}_c^{(A)}(\psi,m)=g_A m\psi. \tag{3}$$

As magnetization breaks time reversal symmetry, and is of odd parity under time reversal, such a term is only permitted if ψ is also odd under time reversal, and thus also breaks time-reversal symmetry. Such terms can only occur in models of type (A) where ψ *breaks* time-reversal symmetry. In theories of type (B) where ψ is even under time-reversal invariance the simplest coupling consistent with both time-reversal symmetry and translational invariance takes the form

$$\mathcal{F}_c^{(B)}(\psi,m)=g_B m^2\psi^2. \tag{4}$$

Note that terms of the form $m^2\psi$ and $m\psi^2$ are ruled out if the hidden order is secondary and is invariant under time-reversal symmetry.[23] These two types of coupling, Eqs. (3) and (4), lead to very different predictions for the $H-T$ phase diagram.

In order to understand these distinctions, let us write the separate free energies for the secondary and primary order parameters. For both categories of theory, the primary free energy takes the form

$$\mathcal{F}_1[\psi]=-\alpha t\psi^2+\beta\psi^4+\alpha h^2\psi^2, \tag{5}$$

where $t=(T_c-T)/T_c$ is the reduced temperature, measuring the deviation from the transition temperature T_c of the primary order parameter and $h=H/H_c$ is the ratio between the external magnetic field and the measured critical field at zero temperature ($H_c=40T$). Translational invariance is enough to rule out a linear coupling between h and ψ in both categories of theory. This form of the free energy is broadly consistent with many of the observed phenomenon. We can rewrite $\mathcal{F}_1$ in the form

$$\mathcal{F}_1[\psi]=\beta[\psi^2-\psi_0^2(h,t)]^2+F_1, \tag{6}$$

where

$$\psi_0(h,t)=\left[\frac{\alpha}{2\beta}(t-h^2)\right]^{1/2} \tag{7}$$

is the equilibrium value of the primary order parameter, and

$$F_1=-\beta\psi_0^4(h,t)=-\frac{\alpha^2}{4\beta}(t-h^2)^2 \tag{8}$$

is the equilibrium free energy.

If we ignore the coupling to the secondary order parameter, then by reading off the various derivatives with respect to temperature and field, we are able to deduce that

$$\Delta\left(\frac{C_v}{T}\right)=-\frac{1}{T_c^2}\frac{\partial^2F_1}{\partial t^2}=\frac{1}{2T_c^2}\zeta,$$

$$\Delta\left(\frac{d\chi}{dT}\right)=-\frac{1}{H_c^2T_c}\frac{\partial^3F_1}{\partial t\,\partial h^2}=-\frac{1}{H_c^2T_c}\zeta,$$

$$\Delta\chi_3=-\frac{1}{H_c^4}\frac{\partial^4F_1}{\partial h^4}=\frac{6}{H_c^4}\zeta, \tag{9}$$

where $\chi_3=-\partial^4\mathcal{F}_1/\partial H^4$ is the nonlinear susceptibility and we have denoted $\zeta=\alpha^2/\beta$. From these three results, we can obtain the relationship

$$\Delta\left(\frac{C_v}{T}\right)\Delta\chi_3=3\left[\Delta\left(\frac{d\chi}{dT}\right)\right]^2. \tag{10}$$

This result is in good accord with the measured anomalies in this material.[24] This agreement indicates that the phase transition is well described by mean-field theory, though it does not reveal any specifics about the nature of the hidden order. As an aside, we note that if the transition were associated with a conventional spin-density wave, this expression would become

$$\Delta\left(\frac{C_v}{T}\right)\Delta\chi_3=3(m_0^4), \tag{11}$$

where m_0 is the staggered moment; this relation is clearly *not* obeyed[5] in URu_2Si_2 where the anomalies in the specific heat and the nonlinear susceptibility are large and $m_0=0.03\mu_B$.

Let us now consider the way in which theories of type (A) and (B) differ. In type (A) theories, the quartic terms in $\mathcal{F}_2$ may be neglected, and it is sufficient to take

$$\mathcal{F}_2^{(A)}[m]=a(h)m^2+O(m^4), \tag{12}$$

where $a(h)$ is positive. Now since a magnetic field always raises the energy of an antiferromagnet, we may write

$$a(h)=a[1+\delta h^2]. \tag{13}$$

This means that at reduced fields above the scale $h\sim 1/\sqrt{\delta}$ ($H\sim H_c/\sqrt{\delta}$), the energy of the induced order parameter $\mathcal{F}_2^{(A)}=a(h)m^2$ is dominated by its coupling to the external magnetic field. Furthermore in these theories in category (A) where ψ breaks time-reversal symmetry, the linear coupling between the primary and secondary order parameters means that the primary order parameter always induces

a staggered magnetic moment. If we assume g_A is small, then by minimizing $\mathcal{F}=\mathcal{F}_1+\mathcal{F}_2+\mathcal{F}_c^A$, we obtain

$$m=-\frac{g_A}{2a(h)}\psi_0(h,t). \tag{14}$$

The small magnitude of the magnetic order parameter in scenario (A) arises naturally from the assumed small magnitude of g_A. From the high-field experiments,[11–14] it is known that the field dependence of m at low temperatures is more rapid than that of the primary order parameter. Using Eqs. (13) and (14), at $T=0$,

$$m[h]=m_0\frac{[1-h^2]^{1/2}}{1+\delta h^2}, \tag{15}$$

where $m_0=-(g_A/2a)\sqrt{\alpha/2\beta}$. We see that the staggered magnetization is then a product of a Lorentzian times the field dependence of the hidden order parameter ψ. For small fields

$$m=m_0\left[1-\frac{1}{2}\left(\frac{h}{h_m}\right)^2\right], \tag{16}$$

where

$$h_m=\frac{1}{\sqrt{1+2\delta}}, \tag{17}$$

h_m sets the magnitude of the field scale where the secondary order vanishes, based on a low-field extrapolation of the magnetization. Since the magnetization is always finite for $\psi\neq 0$, scenario (A) necessarily implies that there will be a point of inflection in the field dependence of the staggered magnetization around the field value $H_m\sim H_c h_m$; at field strengths greater than H_m, the energy of the secondary order is dominated by its coupling to the external magnetic field, but the secondary order is prevented from going to zero by its coupling to the hidden primary order. In Fig. 1 we show a typical curve for $m(h)/m_0$. The absence/presence of a point of inflection in $m(h)$ is a key experimental test for scenario (A).

Let us now turn to scenario (B). In this case, it is necessary to assume that the system is close to an antiferromagnetic instability, so that

$$\mathcal{F}_2^{(B)}+\mathcal{F}_c^{(B)}=-a(T_m-T)m^2+bm^4+g_B m^2\psi^2. \tag{18}$$

We can rewrite this in the form

$$\mathcal{F}_2^{(B)}+\mathcal{F}_c^{(B)}=-a(T_m[\psi]-T)m^2+bm^4, \tag{19}$$

where $T_m[\psi]=T_m-(g_B/a)\psi^2$. Clearly at temperatures close to T_c where ψ is small the renormalization of T_m is negligible, so that the coupling between the two order parameters can be effectively neglected. Within scenario (B) the coupling between the order parameters does not contribute towards linking the two transitions and they are therefore truly independent as displayed in Fig. 1(b). Experimentally the transition temperatures, T_m and T_c, associated with the development of the primary and secondary order parameters are roughly comparable,[11–14] as discussed below. We note that staggered quadrupolar order is an example of a primary order parameter in class (B); all known systems with *continuous* double quadrupolar-magnetic transitions have a separation in the two temperature scales.[25] If the primary phase transition of URu_2Si_2 had been *discontinuous* (e.g., first-order) then this requirement ($T_m\approx T_c$) could have been relaxed, and indeed there is such an example of a first-order quadrupolar-magnetic transition[26] in $U_2Rh_3Si_5$. However, field-dependent measurements in URu_2Si_2 clearly indicate that the primary order parameter grows continuously as the temperature is reduced, ruling out this first-order possibility.[11–14]

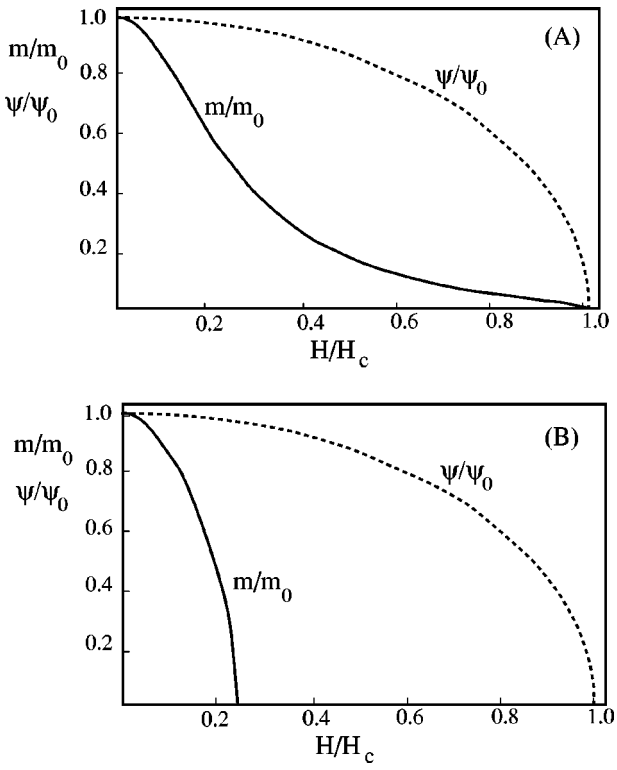

FIG. 1. Field dependence of the primary order parameter and the staggered magnetization in scenarios (A) and (B). When the primary order parameter breaks time-reversal symmetry, the staggered magnetic order remains finite so long as the primary order is present, leading to a point of inflection in the magnetization versus field.

A second aspect of scenario (B) concerns the size of the staggered magnetization. In order to account for the small size of the staggered moment, we require that

$$m_0=\sqrt{\frac{aT_m}{2b}} \tag{20}$$

is naturally small. A microscopic theory would have to account for the magnitude of this parameter. In scenario (B) the field-dependence of the secondary order parameter is then entirely independent of the primary order parameter.

In Fig. 2 we contrast the phase diagrams expected in the two different scenarios (A) and (B). The qualitative distinction is quite striking and immediately suggests a ''tie-breaking'' experiment. If the underlying order parameter is indeed of type (A), then high-field neutron-scattering experiments should observe a marked inflection in the field depen-

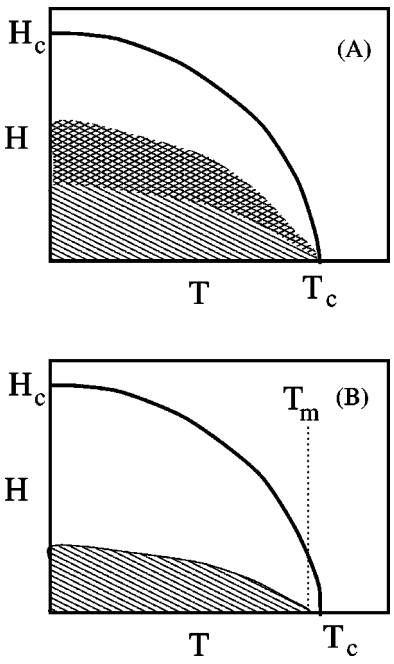

FIG. 2. The contrasting phase diagrams for scenarios (A) and (B). In (A), where the primary order parameter has broken time-reversal symmetry, the staggered magnetic order remains finite so long as the primary order is present. The cross-hatched area refers to where $m(h,t)$ has a region of inflection (see Fig. 1). In (B), there is a sharp phase transition at a finite field and T_m and T_c match up only by coincidence.

dence of the staggered magnetization; this should occur long before the upper critical field ($H \approx 40T$) of the primary order parameter is reached.

For the sake of completeness, we note that there are unresolved issues regarding the experimental determination of T_m. More specifically elastic neutron-scattering intensity persists for several degrees above $T_c = 17.5$ K, thus making it very difficult to extract a precise onset temperature T_m associated with the development of the staggered moment.[11,27] This additional intensity could be ascribed to sample quality, instrumental resolution, or possibly to quasi-elastic contributions to the Bragg peak. Current experimental results suggest that T_m and T_c are not identical within experimental accuracy. If it can be shown conclusively that $T_m > T_c$ this would *rule* out type (A) theories where m is induced by ψ. By contrast this possibility could be accommodated within type (B) scenarios, since here the two transitions are essentially independent. We note that if ψ is incommensurate we expect the transitions associated with the two order parameters to occur at different temperatures.

We now turn to the second part of our discussion and consider the nature of the single ion physics. Any microscopic theory is critically dependent on this physics. For example, Santini and Amoretti[21] have proposed that the key physics of URu_2Si_2 is governed by the mixing of two non-degenerate singlet ground states leading to a staggered quadrupolar ground state. On the other hand, Amitsuka *et al.*[15] suggest that a different ground state is relevant to dilute concentrations of uranium in $ThRu_2Si_2$, involving a magnetic non-Kramers doublet of a type considered by Cox and Makivic.[28] Were such a ground state to survive to the dense system, it would lead to a magnetic two-channel Kondo lattice. This immediately suggests three distinguishing experiments:

(i) A definitive test of the proposal by Amitsuka and co-workers[15,16] for dilute U concentrations has not yet been performed. Theory predicts that if the ground state is that of a two-channel Kondo model, then at finite magnetic fields the logarthmic divergence of $\gamma = C_v(T)/T$ will be cutoff by a Schottky anomaly with an associated entropy of $\frac{1}{2}\ln 2$. This fractional entropy is distinctive of the two-channel Kondo model and heuristically arises from the partial quenching of the fermionic degrees of freedom in the system. The degeneracy of the proposed non-Kramers doublet should also be lifted with application of a uniaxial strain; again the signatory entropy associated with the two-channel Kondo model should be observed.

(ii) The crystal-field schemes proposed by Amitsuka *et al.*[15] and by Santini and Amoretti[21] are for the dilute and the dense limits, respectively. Qualitatively they are very different; more specifically the lowest lying state is a doublet in the scheme of Amitsuka *et al.*[15] whereas it is a singlet in the other proposed scenario.[21] If indeed there is such a dramatic shifting of the crystal-field levels as a function of uranium density it should lead to observable nonlinearities in the lattice parameters, deviations from Vegart's law and dramatic changes in the nonlinear susceptibility[30,31] as a function of uranium doping. By contrast if the lattice parameters grow monotonically with doping levels, we can conclude that the single-ion physics of the dilute system and the lattice are qualitatively similar.

(iii) If the underlying physics of the dense system involves a non-Kramer's magnetic doublet, then we expect that a uniaxial strain and magnetic field will split this doublet in precisely the same way, up to a scale constant that can be deduced from the dilute limit. In this situation, the phase diagram as a function of uniaxial strain will look *identical* to the phase diagram as a function of field. This is the definitive test of whether a non-Kramer's magnetic doublet underpins the physics of the dense lattice.

Summarizing the discussion so far, we have presented some simple experimental probes of time-reversal violation and the local single-ion physics that, if observed, will substantially advance our basic understanding of the underlying order in URu_2Si_2. We should now look ahead to the constraints that our discussion imposes on any future microscopic theories in URu_2Si_2. Such theories must provide:

(i) A description of the local single-ion physics that is consistent with the heavy fermion behavior.

(ii) A description of how the hidden order emerges from the local ion physics. Clearly, the character of the theory depends critically on an experimental test of whether the primary order breaks time-reversal symmetry.

It is important to remember in this discussion that URu_2Si_2 is a heavy fermion compound, both before and after the hidden order develops. In the low-temperature phase, the size of $C_V/T \sim 65$ mJ mol^{-1} K^{-1} puts this material into the category of intermediate heavy fermion behavior. The superconducting transition at 1.7 K also has a large specific-heat anomaly characteristic of heavy fermion superconductivity.

The dramatic contrast with $ThRu_2Si_2$, which is a normal, low-mass metal, serves to emphasize that it is the local *f*-electron physics of the uranium atom which drives the unusual properties in URu_2Si_2. The large values of γ derive from the quenching of the local ionic degrees of freedom. Any microscopic theory of the hidden order in URu_2Si_2 must respect these essential observations.

For example, let us suppose that the local-ion physics suggested by Koga and Shiba[29] for dilute U in $ThRu_2Si_2$ persists to stoichiometric URu_2Si_2, involving a magnetic non-Kramers doublet. Such a state has the capacity to provide the required low-lying degrees of freedom for heavy fermion behavior, but now we must address the second point above. One of the interesting questions here is how the two-channel physics of the single ion might play a role in the hidden order. In the dense lattice, there is the possibility of constructive interference between the Kondo effect in the two channels that hypothetically couple to each uranium ion. This has the potential to produce composite orbital order that breaks time-reversal symmetry and thus is of type (A); such order parameters that combine aspects of the Kondo effect and orbital magnetism have been proposed for heavy fermion superconductivity.[32–35] Should experiments confirm the equivalence of field and uniaxial strain on the primary order parameter, then this composite approach might be an appealing one to describe the underlying hidden order.

At present the only scenario which addresses how the hidden order might emerge from the local ion physics is the quadrupolar theory of Santini and Amoretti,[21] one that is of type (B). However if the single-ion physics of the uranium in URu_2Si_2 is described by a nondegenerate singlet state which mixes with higher-lying singlets to produce a quadrupole, it is very difficult to see how this picture can provide the necessary degrees of freedom for the heavy electron behavior below the transition without the addition of local spin excitations.[9] The formation of the heavy fermion state at temperatures $T > 17.5$ K remains to be addressed by this approach.

In conclusion, we have contrasted two classes of theory for the hidden order in URu_2Si_2 and have proposed measurements designed to test (i) whether the order breaks time-reversal symmetry and (ii) whether the local physics is described by a non-Kramers magnetic doublet. The results of these experiments would considerably further our understanding of this fascinating heavy-fermion superconductor.

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