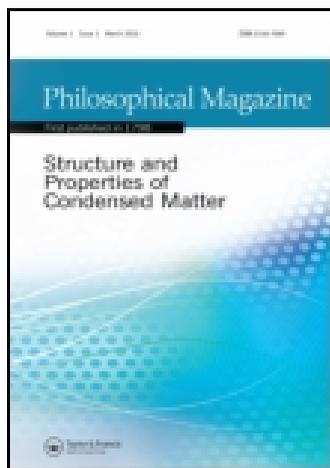


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### Ising quasiparticles and hidden order in $\text{URu}_2\text{Si}_2$

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## Ising quasiparticles and hidden order in URu<sub>2</sub>Si<sub>2</sub>

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The observation of Ising quasiparticles is a signatory feature of the hidden order phase of URu<sub>2</sub>Si<sub>2</sub>. In this paper, we discuss its nature and the strong constraints it places on current theories of the hidden order. In the hastatic theory, such anisotropic quasiparticles are naturally described by resonant scattering between half-integer spin conduction electrons and integer-spin Ising moments. The hybridization that mixes states of different Kramers parity is spinorial; its role as a symmetry-breaking order parameter is consistent with optical and tunnelling probes that indicate its sudden development at the hidden order transition. We discuss the microscopic origin of hastatic order, identifying it as a fractionalization of three body bound-states into integer spin fermions and half-integer spin bosons. After reviewing key features of hastatic order and their broader implications, we discuss our predictions for experiment and recent measurements. We end with challenges both for hastatic order and more generally for any theory of the hidden order state in URu<sub>2</sub>Si<sub>2</sub>.

**Keywords:** Ising models; heavy-fermion metals; order; intermetallic compounds; Kondo effect

### 1. Introduction

We begin by noting that two key developments in heavy fermion physics that relate to the hidden order problem in URu<sub>2</sub>Si<sub>2</sub> were both published in Philosophical Magazine. Forty years ago, Neville Mott [1] pointed out that the development of coherence in heavy electron systems should be understood as a hybridization of f-electrons connected with the Kondo effect. Twenty-five years later, Okhuni et al. [2] discovered that in the hidden order phase, the mobile carriers are Ising quasiparticles. This paper discusses how these two phenomena – the development of an emergent hybridization and the formation of pure Ising quasiparticles – are inextricably linked with the hidden order in URu<sub>2</sub>Si<sub>2</sub>.

There is still no consensus on the nature of the “hidden order” phase in URu<sub>2</sub>Si<sub>2</sub> despite several decades of active theoretical and experimental research [3–5]. At  $T_{HO} = 17.5$  K, there are sharp features in thermodynamic quantities and a sizable ordering entropy ( $S > \frac{1}{3}R \ln 2$ ); however, there is no observed charge order, and spin ordering in the form of antiferromagnetism occurs only at finite pressures [3–8]. At first sight, it seems

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straightforward to link hidden order to the formation of a “heavy density wave” within a pre-formed heavy electron fluid. Since there is no observed magnetic moment or charge density observed in the hidden order (HO) phase, such a density wave must necessarily involve a higher order multipole of the charge or spin degrees of freedom and various theories of this sort have indeed been advanced [9–31]. In each of these scenarios, the heavy electrons develop coherence via a crossover at higher temperatures, and the essential hidden order is then a multipolar charge or spin density wave. However, such multipolar order cannot naturally account for the emergence of heavy Ising quasiparticles, a signature feature of  $\text{URu}_2\text{Si}_2$  that has been probed by two distinct experiments [2,32–34]. The essential point here is that conventional quasiparticles have half-integer spin and are magnetically isotropic; they thus lack the essential Ising protection required by observation. In addition, optical and tunnelling probes [35–39] indicate that the hybridization in  $\text{URu}_2\text{Si}_2$  develops abruptly at  $T_{HO}$  and is thus associated with a global broken symmetry [22,26,40,41]; this is to be contrasted with the usual situation in heavy fermion materials where it is simply a crossover.

Here, we argue that the elusive nature of the “hidden order” in  $\text{URu}_2\text{Si}_2$  is *not* due to its intrinsic complexity but rather that it results from a fundamentally new type of order parameter. In the “hastatic” proposal, [40,41] the observation of heavy Ising quasiparticles [2,32–34] suggests resonant scattering between half-integer spin electrons and integer spin local moments, and the development of a spinorial order parameter. It is perhaps useful to contrast the various staggered multipolar scenarios for the hidden order with the hastatic one proposed here. In the former, mobile f-electrons Bragg diffract off a multipolar density wave (see Figure 1(a)), whereas in the latter, the multipole contains an internal structure, associated with the resonant scattering into an integer spin f-state (Figure 1(b)). Hastatic order can thus be loosely regarded as the “square root” of a multipole order parameter; in other words, we argue that the origin of hidden order is not a complex multipole but instead is an elementary “half-tu-pole” that mediates hybridization between an Ising non-Kramers doublet and the mobile conduction electrons.

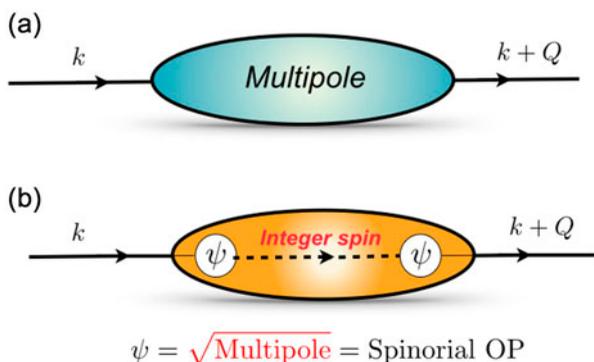


Figure 1. (colour online) Schematic contrasting the multipolar and spinorial theories of Hidden order. (a) in a multipolar scenario, the heavy electrons Bragg diffract off a staggered spin or charge multipole (b) in the hastatic scenario, the development of a spinor hybridization opens up resonant scattering with an integer spin state of the ion. The multipole is generated as a consequence of two spinorial scattering events. In this way, the Hastatic spinor order parameter can be loosely regarded as the square root of a multipole.

Because the observed magnetic anisotropy of the heavy quasiparticles is central to our approach, we will begin by discussing these experiments [2,32–34] in detail. Next we will review “highlights” of the hastatic proposals, [40,41] and the broader implications of an order parameter that transforms under double-group ( $S = \frac{1}{2}$ ) representations. Experimental predictions and recent measurements will be discussed next. We will end with challenges both for hastatic order and more generally for any theory of hidden order in URu<sub>2</sub>Si<sub>2</sub>.

## 2. Ising quasiparticles

Remarkably Fermi surface magnetization experiments in the HO state of URu<sub>2</sub>Si<sub>2</sub> indicate near-perfect Ising anisotropy in the  $g$ -factor ( $g(\theta)$ ) of the quasiparticles [2,33]. Measurements of the bulk susceptibility of URu<sub>2</sub>Si<sub>2</sub> do show a strong Ising anisotropy along the  $c$ -axis (see Figure 2) [3–5,10]; this feature persists in dilute samples (U<sub>*x*</sub>Th<sub>1–*x*</sub>Ru<sub>2</sub>Si<sub>2</sub> with  $x \sim 0.07$ ) suggesting that it is a single-ion effect [42]. However, the Ising anisotropy of the bulk susceptibility is about a factor of five, whereas the anisotropy in the Pauli susceptibility of the heavy Fermi surface in the hidden order phase is in excess of 900.

According to Onsager’s treatment of a Fermi surface, the Bohr-Sommerfeld quantization of quasiparticle orbits leads to a quantization of the area in  $k$ -space according to  $\oint dk_x dk_y = A(\epsilon_n) = (n + \gamma) \left( \frac{(2\pi)^2 eB}{h} \right)$  where  $\gamma$  is a constant Berry phase term and  $\epsilon_n$  is the Kinetic energy of the Bloch waves (i.e. energy without Zeeman splitting) [43]. This condition leads to quantized kinetic energy  $\epsilon_n = \hbar\omega_c(n + \gamma)$ . When the Zeeman spin splitting is included, one finds that the quantized energies are given by [44,45]

$$E_{n\pm} = \overbrace{(n + \gamma)\hbar\omega_c}^{\epsilon_n} \mp \frac{1}{2}g\mu_B B, \tag{1}$$

where  $\omega_c = \frac{eB}{m^*}$  is the cyclotron frequency,

$$m^* = \frac{\hbar^2}{2\pi} \frac{\partial A}{\partial \epsilon}, \tag{2}$$

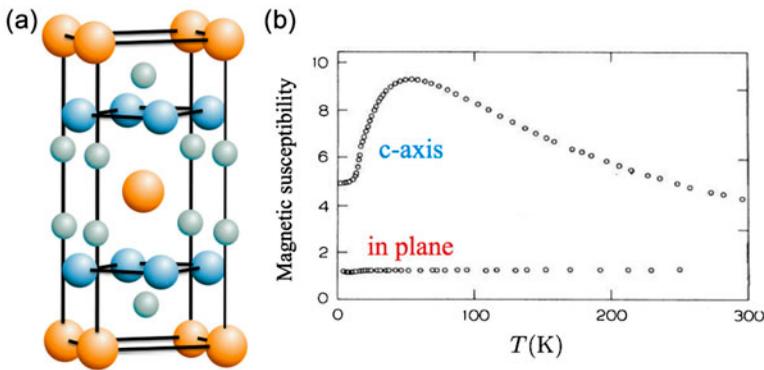


Figure 2. (colour online) (a) Body-centred tetragonal structure of URu<sub>2</sub>Si<sub>2</sub> (b) Measured anisotropic temperature-dependent bulk magnetic susceptibility [3] of URu<sub>2</sub>Si<sub>2</sub>.

is the effective mass and

$$g = \frac{\oint \frac{dk_{\perp}}{v_F} g(\mathbf{k})}{\oint \frac{dk_{\perp}}{v_F}} \quad (3)$$

is the average of the  $g$ -factor over the orbit. Notice that the Onsager quantization condition means that the kinetic energies of the up and down Fermi surfaces are identical with the Zeeman splitting superimposed.

The discrete summation over these quantized energy levels gives rise to an oscillatory component in the magnetization given by [43]

$$M \propto \sum_{\pm} \sin\left(\frac{2\pi\mu_{\pm}}{\hbar\omega_c}\right) = \sum_{\sigma} \sin\left[\frac{2\pi\mu}{\hbar\omega_c} \pm 2\pi\left(\frac{\frac{g}{2}\mu_B B}{\hbar\omega_c}\right)\right], \quad (4)$$

where  $\mu_{\sigma} = \mu + \frac{\sigma}{2}g\mu_B B$  is the Zeeman-split chemical potential. Summing the two terms together

$$M \propto 2 \sin\left(\frac{2\pi\mu}{\hbar\omega_c}\right) \cos \delta \quad (5)$$

where

$$\delta = 2\pi\left(\frac{g\mu_B B}{\hbar\omega_c}\right) = \pi g\left(\frac{m^*}{m}\right) \quad (6)$$

is the phase shift induced by the Zeeman splitting. Notice that  $\delta$  is field independent, so it affects the overall amplitude without changing the dHvA frequencies. In particular in systems where the  $g$ -factor is a strong function of angle, namely in orbits where the Zeeman splitting is a half-integer multiple of the cyclotron energy, the up and down Fermi surfaces destructively interfere to produce a ‘‘spin zero’’; here the dHvA signal identically vanishes when

$$\delta = 2\pi \frac{\text{Zeeman splitting}}{\text{cyclotron energy}} = \pi g(\theta_n) \frac{m^*}{m_e} = \pi \left(n + \frac{1}{2}\right) \quad (7)$$

where  $n$  is a positive integer and  $\theta_n$  is the (indexed) angle with respect to the  $c$ -axis. The observation of spin zeroes in dHvA thus provides a way of detecting the presence of a spin-degenerate Fermi surface and, provided the indexing can be done reliably, enables a direct measurement of the dependence of the  $g$ -factor  $g(\theta)$  on the orientation of the orbit.

Sixteen such spin zeroes are observed (cf. Figure 3) in the HO state of URS, [2,33] in contrast to the one per band seen in the cuprates [46]. At the most elementary level, these results tell us that the heavy  $\alpha$  pocket of the HO state involves quasiparticles that carry spin, with a two-fold degeneracy at each point in  $k$ -space. It is well known that such degeneracies survive strong spin-orbit coupling if there is inversion symmetry combined with time-reversal invariance or a combination of time-reversal and translational invariance as in a commensurate spin density wave. Moreover, we can place stringent bounds on the level of perfection of both the degeneracy and the Ising anisotropy.

The Zeeman splitting scales from more than 15 times the cyclotron frequency along the  $c$ -axis to less than half a cyclotron frequency along the basal plane. This puts a rigorous bound on the  $g$ -factor anisotropy

$$\frac{g_{\perp}}{g_c} < \frac{1}{30} \quad (8)$$

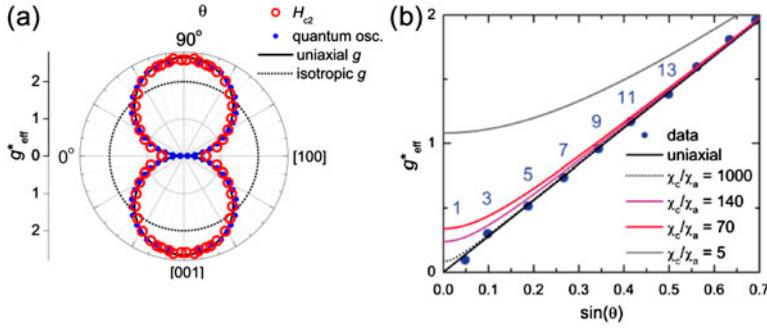


Figure 3. (colour online) Anisotropy of the  $g$ -factor of quasiparticles in  $\text{URu}_2\text{Si}_2$  (a) plotted in polar coordinates derived from spin zeroes in quantum oscillation measurements and the anisotropy of the upper critical field (b) versus sine of the angle out of the basal plane, showing that the data requires a Pauli susceptibility anisotropy in excess of 900 [2,32–34].

where  $g_{\perp} = g(\theta_n \sim \frac{\pi}{2})$  and  $g_c = g(\theta_n = 0)$ , indicating that the splitting energy between the orbits depends *only* on the  $c$ -axis component of the applied magnetic field ( $B_c$ ), namely that

$$g(\theta_n) = g^* \cos \theta_n \tag{9}$$

where  $g^* = 2.6$  in contrast to the isotropic  $g = 2$  for free electrons [2,33]. We note that these dHvA oscillations were generated by the heavy  $\alpha$  pockets of  $\text{URu}_2\text{Si}_2$ , and thus could be argued to come from a select region of its Fermi surface. However, this magnetic anisotropy is also observed in the angular dependence of the upper critical field  $H_{c2}(\theta)$  that is sensitive to the entire heavy fermion pair condensate [32,34]. The  $g(\theta)$  derived from  $H_{c2}(\theta)$  matches that from the dHvA measurements very well for angles near the  $c$ -axis where  $H_{c2}$  is Pauli-limited [34]. However, the anisotropic bound on the  $g$ -factor is less stringent than that found from the quantum oscillation experiments, since the in-plane  $H_{c2}$  is smaller than expected, probably due to orbital contributions. Returning to the bounds placed by the spin-zeroes measurements, we note that since the Pauli susceptibility  $\chi^P$  scales with the *square* of the  $g$ -factor, these resolution-limited measurements of  $\frac{g_c}{g_{\perp}}$  suggest that

$$\chi^P(\theta) = \chi^{P*} \cos^2 \theta \quad \frac{\chi_c^P}{\chi_{\perp}^P} > 900. \tag{10}$$

Such a large anisotropy should be directly observable in electron spin resonance measurements that probe the Pauli susceptibility directly in contrast to bulk susceptibility measurements where Van Vleck contributions are also present.

To our knowledge, this is the largest number of spin zeroes that have ever been observed in any material; furthermore, the Ising nature of the quasiparticles in the hidden order state is a dramatic departure from the usual magnetic isotropy of free conduction electrons. A natural explanation for the quasiparticle Ising anisotropy is that the Ising character of the uranium ions has been transferred to the quasiparticles via hybridization, and this is a key element of the hastatic proposal [40,41]. The giant anisotropy in  $\frac{g_{\perp}}{g_c}$ , places a strong constraint on the energy-splitting  $\Delta$  between the two Ising states. This quantity must be smaller than half a cyclotron frequency, or

$$\Delta < \frac{1}{2} \hbar \omega_c. \tag{11}$$

In the dHvA measurements, the effective mass on the  $\alpha$  orbits is  $m^* = 13m_e$ , and the measurements were made at  $B = 13T$ , giving

$$\frac{\Delta}{k_B} \lesssim \left( \frac{\hbar e B}{2(m^*/m_e)m_e} \right) = 0.67\text{K}. \quad (12)$$

Additional support for a very small  $\Delta$  comes from the dilute limit, [42]  $\text{U}_x\text{Th}_{1-x}\text{Ru}_2\text{Si}_2$  ( $x = .07$ ), where the Curie-like single-ion behaviour crosses over to a critical logarithmic temperature dependence below 10 K,  $\log T/T_K$ , where  $T_K \approx 10$  K. This physics has been attributed to two-channel Kondo criticality, again requiring a splitting  $\Delta \ll 10$  K.

Constrained by the anisotropic bulk spin susceptibility and the quantum spin zeroes, we therefore require the U ion to be an Ising doublet with the form

$$|\Gamma_{\pm}\rangle = \sum_n a_n |\pm (J_z - 4n)\rangle, \quad (13)$$

where the addition and subtraction of angular momentum in units of  $4\hbar$  is a consequence of the four-fold symmetry of the  $\text{URu}_2\text{Si}_2$  tetragonal crystal. However, the presence of a perfect Ising anisotropy requires an *Ising selection rule*

$$\langle \Gamma_{\pm} | J_{\pm} | \Gamma_{\mp} \rangle = 0 \quad (14)$$

that, in the absence of fine-tuning of the coefficients  $a_n$ , leads to the condition that  $-(J_z + 4n') \neq (J_z + 4n) \pm 1$ , or  $J_z \neq 2(n - n') \pm \frac{1}{2}$ , requiring  $J_z \in \mathbb{Z}$  must be an integer. For any generic half-integer  $J_z$ , corresponding to a Kramers doublet, the selection rule is absent so that crystal fields mix the  $J_z$  states leading to isotropic magnetic properties. Within the five-parameter crystal-field Hamiltonian of  $\text{URu}_2\text{Si}_2$ , a simulated annealing search yielded just one finely tuned  $5f^3$  (Kramers) state with nearly zero transverse moment, but the fit to single-ion bulk properties was poor [47]. In the tetragonal crystalline environment of  $\text{URu}_2\text{Si}_2$ , such Ising anisotropy is most natural in a  $5f^2$  ( $J = 4$ ) configuration of the uranium ion, but doublets with integer  $J$  in general do not enjoy the symmetry protection of their half-integer (Kramers) counterparts. However in  $\text{URu}_2\text{Si}_2$ , a combination of tetragonal and time-reversal symmetries protects a non-Kramers doublet

$$|\Gamma_{5\pm}\rangle = \alpha |J_z = \pm 3\rangle + \beta |J_z = \mp 1\rangle \quad (15)$$

that is quadrupolar in the basal plane and magnetic along the  $c$ -axis, and it has been proposed as the origin of the magnetic anisotropy in both the dilute and the dense  $\text{URu}_2\text{Si}_2$  [40–42]; this can be checked with a direct benchtop test [47]. In the hastatic proposal, the Ising anisotropy of the U  $5f^2$  ions is transferred to the quasiparticles via hybridization between integer  $J$  local moments and half-integer  $J$  conduction electrons, and this mixing of Kramers parity ( $K = (-1)^{2J}$ ) has important symmetry implications [40,41].

Conventionally in heavy fermion materials, hybridization involves valence fluctuations between a ground-state Kramers doublet and an excited singlet (cf. Figure 4); in this case, hybridization is a scalar that develops via a crossover leading to mobile heavy quasiparticles. However, if the ground-state is a non-Kramers doublet, the Kondo effect will involve an excited Kramers doublet (cf. Figure 4). The quasiparticle hybridization now carries a global spin quantum number and has two distinct amplitudes that form a spinor defining the hastatic order parameter

$$\Psi = \begin{pmatrix} \psi_{\uparrow} \\ \psi_{\downarrow} \end{pmatrix}. \quad (16)$$

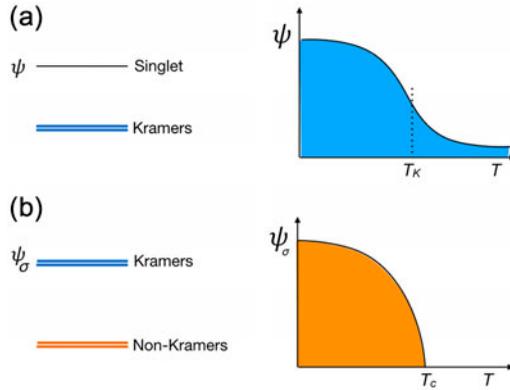


Figure 4. (colour online) Schematic of (a) conventional (scalar) vs. (b) spinorial hybridization where the hybridization is (a) a crossover and (b) breaks spin-rotational and time-reversal symmetries and thus develops discontinuously as a phase transition.

The onset of hybridization must break spin rotational invariance in addition to single- and double time-reversal invariances via a phase transition; we note that optical, spectroscopic and tunnelling probes [35–39] in URu<sub>2</sub>Si<sub>2</sub> indicate the hybridization occurs abruptly at the hidden order transition in contrast to the crossover behaviour observed in other heavy fermion systems (cf. Figure 4).

### 3. Hastatic order “Highlights”

We next summarize the main points of the hastatic proposal, [40,41] noting that the interested reader can find further discussion with more details elsewhere. Hastatic order captures the key features of the observed pressure-induced first-order phase transition in URu<sub>2</sub>Si<sub>2</sub> between the hidden order and the Ising antiferromagnetic (AFM) phases [7,48–51]. The most general Landau functional for the free energy density of a hastatic state with a spinorial order parameter  $\Psi$  as a function of pressure and temperature is

$$f[\Psi] = \alpha(T_c - T)|\Psi|^2 + \beta|\Psi|^4 - \gamma(\Psi^\dagger \sigma_z \Psi)^2 \tag{17}$$

and  $\gamma = \delta(P - P_c)$  where  $P$  is pressure and the term  $\gamma(\Psi^\dagger \sigma_z \Psi)^2$  determines whether the direction of the spinor, either along the  $c$ -axis or in the basal plane (cf. Figure 5(a)).

Experimentally the  $T_{AFM}(P)$  line is almost vertical, indicating by the Clausius–Clapeyron relation that there is negligible change in entropy between the HO and the AFM states. Indeed these two phases share a number of key features, including common Fermi surface pockets; this has prompted the proposal that they are linked by “adiabatic continuity”, associated by a notational rotation in the space of internal parameters [20,48]. This is easily accommodated with a spinor order parameter; for the AFM phase ( $P > P_c$ ), there is a large staggered Ising f-moment with

$$\Psi_A \propto \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \Psi_B \propto \begin{pmatrix} 0 \\ 1 \end{pmatrix} \tag{18}$$

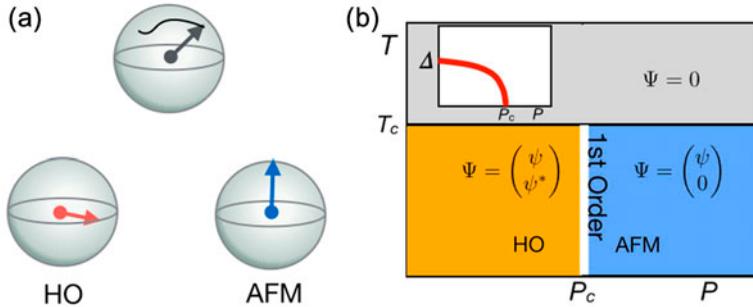


Figure 5. (colour online) (a) The hastatic (hybridization) spinor disordered (at high temperatures) and ordered along the  $c$ -axis (Antiferromagnet) and in the basal plane (hidden order) (b) Temperature-Pressure Phase Diagram and the pressure-dependence of the gap to longitudinal predicted by the hastatic theory.

corresponding to time-reversed spin configurations on alternating layers  $A$  and  $B$ . For the HO state ( $P < P_c$ ), the spinor points in the basal plane

$$\Psi_A \propto \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\phi/2} \\ e^{i\phi/2} \end{pmatrix}, \quad \Psi_B \propto \frac{1}{\sqrt{2}} \begin{pmatrix} -e^{-i\phi/2} \\ e^{i\phi/2} \end{pmatrix} \quad (19)$$

and there is no Ising  $f$ -moment, consistent with experiment, but Ising fluctuations do exist. From this perspective, the transition from HO to AFM corresponds to a spin-flop of the two-component hybridization order parameter from the basal plane to the  $c$ -axis, and the resulting temperature–pressure phase diagram is displayed in Figure 5. Generalizing this Landau theory to study soft modes of the hastatic order, we find that even though the transition at  $P = P_c$  is first order, the gap for longitudinal spin fluctuations decreases continuously as

$$\Delta \propto |\Psi_0| \sqrt{P_c - P}.$$

Since  $dP_c/dT_c$  is finite, close to the transition,  $\sqrt{P_c - P} \approx \sqrt{dP_c/dT_c(T - T_c)}$ , and  $\Delta \propto \sqrt{T - T_c}$ . Inelastic neutron scattering experiments can measure this gap (at the commensurate  $\mathbf{Q}$ ) as function of temperature at a fixed pressure where there is a finite-temperature first-order transition, but to our knowledge a detailed study of this gap behaviour has not yet been performed. The iron-doped compound,  $\text{URu}_{2-x}\text{Fe}_x\text{Si}_2$  can provide an attractive alternative to hydrostatic pressure, as iron doping acts as uniform chemical pressure and tunes the hidden order state into the antiferromagnet [51]. The Landau theory can also be generalized to include coupling to an applied magnetic field  $B$ , predominantly to  $B_z = B \cos \theta$  due to the Ising nature of the non-Kramers doublet; this then leads to an explanation of the observed large  $c$ -axis non-linear susceptibility [10,52] anomaly ( $\Delta\chi_3$ ) in  $\text{URu}_2\text{Si}_2$ , and a prediction of a large  $\Delta\chi_3$  anisotropy,  $\Delta\chi_3 \propto \cos^4 \theta$  where  $\theta$  is the angle from the  $c$ -axis and the coupling coefficient must be determined from a microscopic approach [40,53].

We use a two-channel Anderson lattice model to link hastatic order to the valence fluctuation physics of non-Kramers doublets in  $\text{URu}_2\text{Si}_2$ . The  $5f^2$  Ising  $\Gamma_5$  ground-state configuration [42] fluctuates to an excited  $5f^3$  or  $5f^1$  state via valence fluctuations. The lowest lying excited state is most likely the  $5f^3$  ( $J = 9/2$ ) state, but for simplicity we take it to be the  $5f^1$  state, as the Kramers doublets have the same symmetry, and assume

that fluctuations to the  $5f^3$  are suppressed; in this sense, we take an infinite-U two-channel Anderson model.  $\Gamma_7^+$  is taken to be the lowest energy doublet of the  $5f^1$  state, and then the form of the valence fluctuation Hamiltonian is determined by the orbital structure of the  $\Gamma_5$  doublet. Valence fluctuations occur in two orthogonal conduction electron channels,  $\Gamma_7^-$  and  $\Gamma_6$ , and we find

$$H_{VF}(j) = V_6 c_{\Gamma_6^\pm}^\dagger(j) |\Gamma_7^\pm\rangle \langle \Gamma_5 \pm | + V_7 c_{\Gamma_7^\mp}^\dagger(j) |\Gamma_7^\mp\rangle \langle \Gamma_5 \pm | + \text{H.c.} \quad (20)$$

where  $\pm$  denotes the ‘‘up’’ and ‘‘down’’ states of the coupled Kramers and non-Kramers doublets. The field  $c_{\Gamma_\sigma}^\dagger(j) = \sum_{\mathbf{k}} \left[ \Phi_{\Gamma}^\dagger(\mathbf{k}) \right]_{\sigma\tau} c_{\mathbf{k}\tau}^\dagger e^{-i\mathbf{k}\cdot\mathbf{R}_j}$  creates a conduction electron at uranium site  $j$  with spin  $\sigma$ , in a Wannier orbital with symmetry  $\Gamma \in \{6, 7\}$ , while  $V_6$  and  $V_7$  are the corresponding hybridization strengths. The full model is then written

$$H = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_j [H_{VF}(j) + H_a(j)] \quad (21)$$

where  $H_a(j) = \Delta E \sum_{\pm} |\Gamma_7^\pm, j\rangle \langle \Gamma_7^\pm, j|$  is the atomic Hamiltonian.

Hastatic order is revealed by factorizing the Hubbard operators

$$X_{\sigma\alpha} = |\Gamma_7^+\sigma\rangle \langle \Gamma_5\alpha| = \hat{\Psi}_\sigma^\dagger \chi_\alpha. \quad (22)$$

Here  $|\Gamma_5\alpha\rangle = \chi_\alpha^\dagger |\Omega\rangle$  is the non-Kramers doublet, represented by the pseudo-fermions  $\chi_\alpha^\dagger$ , while  $\hat{\Psi}_\sigma^\dagger$  are slave bosons [54] representing the excited  $f^1$  doublet  $|\Gamma_7^+\sigma\rangle = \hat{\Psi}_\sigma^\dagger |\Omega\rangle$ . Hastatic order is the condensation of this bosonic spinor (cf. Figure 6)

$$\Psi_\sigma^\dagger \chi_\alpha \rightarrow \langle \hat{\Psi}_\sigma^\dagger \rangle \chi_\alpha. \quad (23)$$

This may be viewed as a symmetry-breaking Gutzwiller projection. The resulting quadratic Hamiltonian involves a symmetry-breaking hybridization between the conduction electrons and the pseudofermions. Because experimentally the HO and the AFM share a single commensurate wavevector [49,50]  $Q = (0, 0, \frac{2\pi}{c})$ , we use this wavevector in the description of the HO state where  $\langle \Psi_\pm \rangle = |\Psi| \exp \left[ \pm \frac{i(\vec{Q}\cdot\vec{R}_j + \phi)}{2} \right]$ , where the internal angle  $\phi$  rotates the hastatic spinor in the basal plane. Exploiting the gauge symmetries of the problem, we

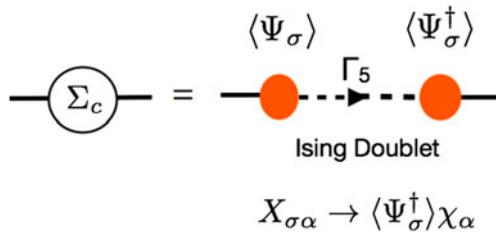


Figure 6. (colour online) The conduction electron self-energy  $\Sigma_c$ . Hybridization with spinorial order parameter  $\langle \Psi_\sigma \rangle$  permits the development of a  $\Gamma_5$  Ising resonance inside the conduction sea, represented by the above Feynman diagram.

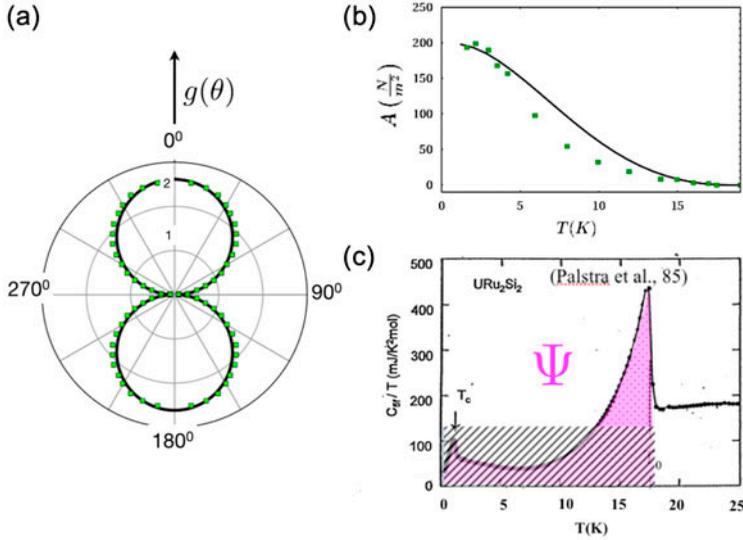


Figure 7. (colour online) Consistency calculations from the hastatic theory indicating good agreement with experiment for (a) the anisotropic  $g$ -factor of the quasiparticles (b) the anisotropic susceptibility  $\chi_{xy}$  and the (c) entropy associated with the hidden order transition.

can simplify the valence-fluctuation Hamiltonian to read

$$H_{VF} = \sum_{\mathbf{k}} c_{\mathbf{k}}^{\dagger} \mathcal{V}_6(\mathbf{k}) \chi_{\mathbf{k}} + c_{\mathbf{k}}^{\dagger} \mathcal{V}_7(\mathbf{k}) \chi_{\mathbf{k}+\mathbf{Q}} + \text{h.c.} \quad (24)$$

where the hybridization form factors are  $\mathcal{V}_7(\mathbf{k}) = V_7 \Phi_7^{\dagger}(\mathbf{k}) \sigma_1$  and  $\mathcal{V}_6(\mathbf{k}) = V_6 \Phi_6^{\dagger}(\mathbf{k})$  and there is uniform ( $\Gamma_6$ ) and staggered ( $\Gamma_7^-$ ) hybridization in the two channels.

This mean-field hastatic model can be used to calculate observable quantities, both to check consistency with known measurements and also to make predictions for future experiment. The full anisotropic  $g$ -factor is a combination of  $f$ -electron and conduction electron contributions and the result for the Fermi-surface averaged  $g$ -factor as a function of field-angle to the  $c$ -axis is displayed in Figure 3, demonstrating good consistency with previous experiment. Magnetometry measurements indicate the development of an anisotropic basal-plane spin susceptibility,  $\chi_{xy}$  at the HO transition, [55] and this result is interpreted as resonant scattering off the Ising  $U$  moments and calculated  $\chi_{xy}$  within our model; the result compares well with experiment as displayed in Figure 7. The development of hastatic order in the lattice at the HO transition liberates a large entropy [56] of condensation,  $\frac{S}{N} \sim \frac{1}{2} k_B \ln 2$  a natural consequence of a Majorana zero-mode in two-channel Anderson impurity physics.

Having established consistency, we now discuss the resulting predictions. The gap to longitudinal spin fluctuations in the hastatic state, and the highly anisotropic non-linear susceptibility anomaly has been discussed earlier (Figure 8a). The detailed microscopic model can be used to determine the magnitude of this quantity. Within the hastatic theory, there is time-reversal breaking in both the HO and the AFM phases and there must be some physical manifestation of this phenomenon in the HO state. Below  $T_{HO}$ , this theory predicts a small conduction electron and  $f$ -electron moment in the basal plane; this moment

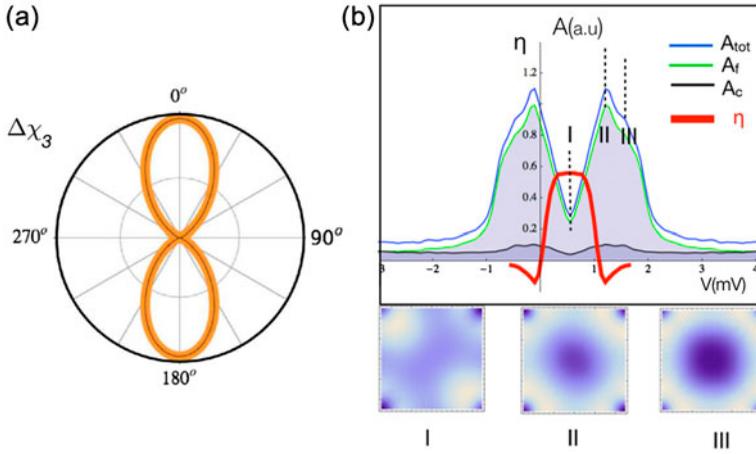


Figure 8. (colour online) Predictions from the hastatic theory for the (a) anisotropy of the  $\chi_3$  anomaly and the (b) energy-dependent resonant nematicity.

is distinct from the extrinsic, inhomogeneous  $c$ -axis moment found in all samples, and the smallness of the transverse moment,  $O(T_{HO}/D)$  is guaranteed by its Kondo origin. This prediction will be discussed further when we review recent experiment. The hastatic theory also predicts a hybridization gap that breaks tetragonal symmetry below  $T_{HO}$ . The resonant scattering via this hybridization leads to a resonant nematicity in the local density of states that is predicted to be a maximum at energies corresponding to the Kondo resonance: this signal should be observable in STM and ARPES measurements (Figure 8b).

#### 4. Can Landau order parameters fractionalize?

A broader implication of hastatic order is the possibility of a new type of Landau order parameter, one that transforms under double-group (half-integer spin) group representations. Conventionally Landau theory in electronic systems is based on the formation and condensation of two-body bound-states, described by a Wick contraction of two electron field operators. The resulting order parameter carries an integer spin. For example in magnetism, the development of a magnetic order parameter  $\vec{M}(x)$  is given by the contraction

$$\overline{\psi_\alpha^\dagger(x)\psi_\beta(x)} = \vec{\sigma}_{\alpha\beta} \cdot \vec{M}(x) \tag{25}$$

By contrast, s-wave superconductivity is based on the formation of spinless bosons given by the contraction

$$\overline{\psi_\uparrow(1)\psi_\downarrow(2)} = -F(1-2), \tag{26}$$

where  $F(1-2) = -\langle T\psi_\uparrow(1)\psi_\downarrow(2) \rangle$  is the anomalous Gor'kov Greens function which breaks the gauge system of the underlying system (Figure 9a). The take-home message from conventional two-body condensation is that when the two-body bound-state wavefunction carries a quantum number (e.g. charge or spin), a symmetry is broken. However under this scheme, all order parameters are bosons that carry integer spin.

Hastatic order carries half-integer spin and cannot develop via this mechanism. We are then led to the question of whether it is possible for Landau order parameters to transform

under half-integer representations of the spin rotation group. At first sight, this is impossible as all order parameters are necessarily bosonic, and bosons carry integer spin. However, the connection between spin and statistics is strictly a relativistic idea that depends on the full Poincare invariance of the vacuum. This invariance is lost in non-relativistic condensed matter systems suggesting the possibility of order parameters with half-integer spin that transform under double-group representations of the rotation group. Spinor order parameters involving “internal” quantum numbers are well known in the context of two-component Bose-Einstein condensates. The Higgs field of electroweak theory is also a two-component spinor. However in neither case does the spinor transform under the physical rotation group. Moreover it is not immediately obvious how such bound-states emerge within fermionic systems.

In the mean-field formulation of hastatic order, [40] a spin-1/2 order parameter develops as a consequence of a factorization of a Hubbard operator that connect the Kramers and non-Kramers states; it is a tensor operator that corresponds to the three-body combination

$$X_{\alpha\sigma}(R) \equiv |f^2\alpha\rangle\langle f^1\sigma| = \Lambda_{\alpha\sigma}^{abc}(R; 1, 2, 3)\psi_a^\dagger(1)\psi_b^\dagger(2)\psi_c(3), \quad (27)$$

where we have used the short-hand notation  $1 \equiv R_1$  etc. and

$$\Lambda_{\alpha\sigma}^{abc}(R; 1, 2, 3) = \langle R_1, a; R_2, b | \hat{X}_{\alpha\sigma}(R) | R_3, c \rangle \quad (28)$$

defines the overlap between the Hubbard operators and the bare electron states. In a simple model, this three-body wavefunction is local,  $\Lambda_{\alpha\sigma}^{abc}(R; 1, 2, 3) = \Lambda_{\alpha\sigma}^{abc}\delta(R-1)\delta(R-2)\delta(R-3)$ . The factorization of the Hubbard operator into a spin-1 fermion and a spin-1/2 boson

$$X_{\alpha\sigma}(R) \rightarrow \chi_\alpha^\dagger(R) \langle \Psi_\sigma(R) \rangle, \quad (29)$$

then represents a “fractionalization” of the three-body operator. Written in terms of the microscopic electron fields, this becomes

$$\Lambda_{\alpha\sigma}^{abc}(R; 1, 2, 3) \overbrace{\psi_a^\dagger(1)\psi_b^\dagger(2)\psi_c(3)} = \chi_\alpha^\dagger(R) \langle \Psi_\sigma(R) \rangle. \quad (30)$$

This expression can be inverted to give the three-body contraction

$$\overbrace{\psi_a^\dagger(1)\psi_b^\dagger(2)\psi_c(3)} = \sum_R G_{abc}^{\alpha\sigma}(1, 2, 3; R) \chi_\alpha^\dagger(R) \langle \Psi_\sigma(R) \rangle, \quad (31)$$

where  $G_{abc}^{\sigma\alpha}(1, 2, 3; R) = [\Lambda_{\sigma\alpha}^{abc}(R; 1, 2, 3)]^*$  (Figure 9b). The asymmetric decomposition of a three-body Fermion state into a binary combination of boson and fermion is a fractionalization process. If the boson in binary carries a quantum number, when it condenses we have the phenomenon of “order parameter fractionalization”.

Fractionalization is well established for excitations of low-dimensional systems, such the one-dimensional Heisenberg spin chain and the fractional quantum Hall effect, [57–60] however order parameter fractionalization is a new concept. The hastatic ordering process involves the order parameter fractionalization into binary combination of a condensed half-integer spin boson and an integer spin fermion. Unlike pair or exciton condensation, the order parameters formed by this mechanism transform under double group representations

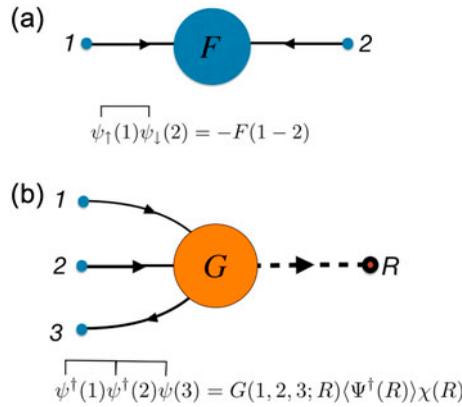


Figure 9. (colour online) Schematic Feynman diagrams indicating (a) two-body (b) and three-body electronic bound-states where in the latter case spin indices have been suppressed for pedagogical simplicity.

of the underlying symmetry groups, and thus represent a fundamentally new class of broken symmetries. We look forward to investigating this “order parameter fractionalization” well beyond the realm of URu<sub>2</sub>Si<sub>2</sub>. The proposed three-body bound-state has a non-local order parameter, and it may be possible to identify a dual theory with a local order parameter that breaks a global symmetry.

### 5. Discussion of recent experiments ... with specific requests

Let us now return to the situation in URu<sub>2</sub>Si<sub>2</sub>. We mentioned earlier that hastatic order leads to a prediction of a basal-plane moment of order  $T_K/D$ , [40,41] where  $T_K$  and  $D$  are the Kondo temperature and band-width, respectively. The transverse moment in our mean-field treatment includes both conduction and f-electron contributions which point in perpendicular directions. The ratio  $T_K/D$  is very sensitive to the degree of mixed valence of the  $5f^2$  state. Our original calculation assumed a 20% mixed valence, leading to a basal plane moment of order  $0.01\mu_B$ .

Past experiments on URu<sub>2</sub>Si<sub>2</sub> had exhaustively demonstrated that there is no longitudinal moment along the  $c$ -axis in the hidden order phase. However, these earlier neutron measurements had chosen a choice of momentum transfer  $\mathbf{Q}$  in the basal plane, where they are maximally sensitive to  $c$ -axes moments, but unable to filter out a small transverse moment. Recent high-resolution neutron experiments [61–63] with momentum transfer along the  $c$ -axis designed to detect the predicted transverse moment have however failed to observe a transverse moment of this magnitude, and have placed a bound  $\mu_\perp < 0.0011\mu_B$  on the ordered transverse moment of the uranium ions. Paradoxically various other probes including X-rays,  $\mu$ -spin resonance and NMR [64–67] have detected the presence of static basal moments on the order of  $0.005\mu_B$  that would be consistent with a more integral valent scenario for the  $U$  ions.

These remaining ambiguities suggest that we need to reconsider the calculation of the transverse moment and understand why it is so small if not absent. There are a number of interesting possibilities:

- *Fluctuations.* The hastatic theory, in its current version, ignores fluctuations of the spinor order that will reduce the transverse moment. Gaussian fluctuations of the corresponding Schwinger boson field are needed to describe the development of the incoherent Fermi liquid observed to develop at  $T > T_{HO}$  in optical, tunnelling and thermodynamic measurements [36–38,68].
- *Uranium Valence.* As mentioned already, the predicted transverse moment is sensitive to the 5f valence, and would be much reduced by a vicinity to integral valence. Moreover, it should be proportional only to the change in valence between  $T_{HO}$  and the measurement temperature, which will be significantly smaller than the high-temperature mixed valency. It would be very helpful to have low temperature probes of the 5f-valence.
- *Domain Size.* The X-ray, [64] muon, [65] torque magnetometry [55] and NMR measurements [66,67] that indicate either a static moment or broken tetragonal symmetry are all carried out on small samples, whereas the neutron measurements involve large ones [61–63]. The discrepancy between the two classes of measurement may indicate the formation of small hidden order domains. Such domain structure might be the result of random pinning [69] of the transverse moment by defects of random strain fields. The situation in URu<sub>2</sub>Si<sub>2</sub> is somewhat analogous to that in Sr<sub>2</sub>RuO<sub>4</sub>, where there is evidence for broken time-reversal symmetry breaking with a measured Kerr effect and  $\mu$ SR to support chiral p-wave superconductivity, but no surface currents have yet been observed [70]. Domains are an issue in this system too.
- *Continuous versus discrete order.* The current mean-field theory has the transverse hastatic vector  $\Psi^\dagger \vec{\sigma} \Psi$  pointing in one of four possible directions at each site, corresponding to a four-state clock model. The tunnelling barrier between these configurations is very small, leaving open the possibility that at long distances the residual physics is that of an  $xy$  order parameter. Such  $xy$  order would then give rise to a kind of spin-superfluid, in which the persistent spin currents avoid the formation of a well-defined static staggered moment.

There are a number of important measurements that would help to resolve some of the current uncertainties and test some of the outstanding predictions:

- (1) *Giant Anisotropy in  $\Delta\chi_3 \propto \cos^4 \theta$ .* This measurement is important to confirm that that the Ising quasiparticles are associated with the development of the hidden order.
- (2) *dHvA on all the heavy Fermi surface pockets.* We expect that the heavy quasiparticles on the  $\alpha$   $\beta$  and  $\gamma$  orbits will all exhibit the multiple spin zeros of Ising quasiparticles. At present, only the  $\alpha$  orbits have been measured as a function of field orientation.
- (3) *Spin zeros in the AFM phase? (Finite pressure)* If the AFM is also hastatic, then we expect the spin zeros to persist into the finite pressure AFM phase.

## 6. The challenges ahead

The observation of Ising quasiparticles in the hidden order state [2,32–34] represents a major challenge to our understanding of URu<sub>2</sub>Si<sub>2</sub>; to our knowledge this is only example

of such anisotropic mobile electrons. It plays a central role in the hastatic proposal, and a key question is whether this phenomenon can be accounted for in other HO theories:

- (1) *Can band theory account for the  $g(\theta)$  observed in URu<sub>2</sub>Si<sub>2</sub>?* Recent advances in the understanding of orbital magnetization [71–73] suggest it may be possible to compute the  $g$ -factor associated with conventional Bloch waves; in a strongly spin-orbit coupled system, the orbital contributions to the total energy in a magnetic field are significant. It would be particularly interesting to compare the  $g(\theta)$  computed in a density functional treatment of URu<sub>2</sub>Si<sub>2</sub> with that observed experimentally.
- (2) *Can other  $5f^2$  theories account for the multiple spin zeroes and the upper bound  $\Delta < 1K$  on the spin degeneracy of the heavy fermion bands?* In particular, is it possible to account for the observed spin zeros without invoking a non-Kramers  $5f^2$  doublet?

In summary, any theory of hidden order has to be able to explain the giant Ising quasiparticle anisotropy in URu<sub>2</sub>Si<sub>2</sub>. The smooth pressure-dependence of the Fermi surfaces between the Hidden Order and the Antiferromagnetic states is also mysterious [50]; it is as if the differences between the two order parameters are “invisible” to the two Fermi surfaces! Finally, there is the key question of why superconductivity only emerges from the hidden order state.

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