Implications of the Measured Angular Anisotropy at the Hidden Order Transition of URu$_2$Si$_2$

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The heavy fermion compound URu$_2$Si$_2$ continues to attract great interest due to the long-unidentified nature of the hidden order that develops below 17.5K. Here we discuss the implications of an angular survey of the linear and nonlinear susceptibility of URu$_2$Si$_2$ in the vicinity of the hidden order transition [1]. While the anisotropic nature of spin fluctuations and low-temperature quasiparticles was previously established, our recent results suggest that the order parameter itself has intrinsic Ising anisotropy, and that moreover this anisotropy extends far above the hidden order transition. Consistency checks and subsequent questions for future experimental and theoretical studies of hidden order are discussed.

MOTIVATION

Consensus has not been reached on the nature of the “hidden order” (HO) in URu$_2$Si$_2$ despite several decades of active research. At $T_{HO} = 17.5K$ sharp features in the thermodynamic quantities accompanied by significant entropy loss ($S > \frac{1}{2} R \ln 2$), but to date no associated charge or spin ordering has been directly detected at ambient pressure [2]. The nature of the quasiparticle excitations and the broken symmetries associated with the HO phase are important questions for understanding not only HO but also the exotic superconductivity that develops at low temperatures.

Several measurements on URu$_2$Si$_2$ indicate the importance of Ising anisotropy in the HO phase despite the absence of local moments at these temperatures and pressures. At $T_{HO}$, both the linear ($\chi_1$) and the nonlinear ($\chi_3$) susceptibilities are anisotropic, with $\chi_3$ in the easy axis direction displaying a sharp anomaly $\Delta \chi_3 = \chi_3(T_c^-) - \chi_3(T_c^+)$ that tracks closely with the structure of the specific heat [3, 4]. At lower temperatures, non-spinflip ($\Delta J_z = 0$) magnetic excitations detected by inelastic neutron scattering [5] have Ising character. Quantum oscillations measured deep within the HO region indicate a strongly anisotropic quasiparticle g-factor $g(\theta) \propto \cos \theta$, where $\theta$ is the angle away from the c-axis [6, 7]. This $g(\theta)$ is confirmed by upper critical field experiments [8], indicating that heavy Ising quasiparticles pair to form a Pauli-limited superconductor at low temperatures.

It is thus natural to ask whether the Ising nature of the itinerant quasiparticles has its origin at $T_{HO}$. Support for this idea is suggested by the observation that the Ising anisotropy obtained from dHvA and the superconducting upper-critical field measurements [6, 7] far exceeds the five-fold anisotropy seen in the bulk magnetic susceptibility [3, 4]. However, to confirm this idea, another measurement is needed to probe the quasiparticle g-factors in the vicinity of the hidden order transition.

ANGULAR SURVEY OF THE HIDDEN ORDER TRANSITION WITH A BULK MEASUREMENT

The general expression for the field-dependent part of the free energy in a tetragonal crystal at fixed temperature is

$$F = -\chi_1(\theta) \frac{H^2}{2} - \chi_3(\theta, \phi) \frac{H^4}{4!}$$  \hspace{1cm} (1)$$

where $\theta$ and $\phi$ refer to the angles away from the c-axis and in the basal plane respectively and details of this angular decomposition can be found elsewhere [1]; for
simplicity here we take $\mu_0 = 1$, so that $\mu_0H = H$ is the external field, measured in Tesla. Because $\Delta \chi_3$ [10] is determined by the excitations near the Fermi surface, it is ideally suited as a direct thermodynamic probe of the electronic g-factors at the HO transition [1]. Consistency with the low-temperature $g(\theta) \propto \cos \theta$ results [6–8], requires a $\Delta \chi_3(\theta) \propto \cos^4 \theta$ since $\Delta \chi_3(\theta) \propto (g(\theta))^4$ [11–13].

Details of the linear and the nonlinear susceptibility measurements as a function of angle can be found elsewhere [1]. The linear susceptibility displayed in figure 1b is characterized by the form

$$\chi_1(\theta, T) = \chi_1(0) + \chi_{Ising}(T) \cos^2 \theta,$$  \hspace{1cm} (2)

where the isotropic (Van Vleck) component $\chi_1(0)$ of the susceptibility displays no discernible temperature dependence. Whereas $\chi_1(\theta)$ varies as $\cos^2 \theta$ at $T = 18K$, in Fig. 2 a) $\Delta \chi_3$ has a distinctive $\cos^4 \theta$ dependence

$$\Delta \chi_3(\theta, \phi) = \Delta \chi_{Ising} \cos^4 \theta$$  \hspace{1cm} (3)

without any Van Vleck (constant) terms, consistent with the low-temperature $g(\theta)$ measurements. In Figure 2c the robustness of the Ising anisotropy is codified [1] by considering an angle-dependent coupling between the hidden order parameter and the magnetic field that results in

$$\Delta \chi_3(\theta) \propto (\cos^2 \theta + \Phi \sin^2 \theta)^2.$$  \hspace{1cm} (4)

where $\Phi$ quantifies the fidelity of Ising behavior. Our measurements indicate a very small $\Phi = 0.036 \pm 0.021$, shown in figure 2c (inset), that could be due to an angular offset of only one degree; details of the fitting procedure can be found elsewhere [1].

These results, at the very least, indicate that the free energy of URu$_2$Si$_2$ only depends on the $z$ component of the magnetic field, namely $F[H] = F[H_z]$. This in turn implies that the Zeeman term in the microscopic Hamiltonian $H_{Zeeman} \propto -J_zB_z$ is coupled to the single-ion properties of the $U$ ions in URu$_2$Si$_2$ via hybridization with the conduction electrons. The observed Ising anisotropy also suggests an integer spin 5/2 U ground-state. This point of view is further supported by both dynamical mean-field theory [14] and high-resolution RIXs measurements [15].

However this picture is incomplete, for the sharpness of the specific-heat anomaly, the sizable entropy and the gapping of two-thirds of the Fermi surface associated with the hidden order transition[2] indicate an underlying itinerant ordering process, as if the hybridization itself is the order parameter[11]. An intriguing feature of these results is that the jump $\Delta \chi_3$ that reflects the itinerant ordering process also exhibits a strong Ising anisotropy. Furthermore, as shown in Figure 3d, there is a positive anisotropic $\chi_3$ to temperatures well above the hidden order transition and this cannot be explained with single-ion physics. We therefore must consider the strong Ising character of the hidden order parameter, now that we have established that we have heavy Ising quasiparticles at the hidden order transition. The reconciliation of the single-ion and the itinerant perspectives, both supported by experiment, presents a fascinating challenge in URu$_2$Si$_2$.

**CONSISTENCY CHECKS**

It is important to cross-check these susceptibility results with other experimental measurements on URu$_2$Si$_2$. At the HO transition, our results can be analyzed within a minimal Landau free energy density of the form

$$f[T, \psi] = a[T - T_c(H)]\psi^2 + \frac{b}{2}\psi^4,$$  \hspace{1cm} (5)

where $\psi$ is the hidden order parameter, we continue to take $\mu_0 = 1$ for simplicity, and

$$T_c(H) = T_c - \frac{1}{2}Q_{ab}H_aH_b + O(H^4)$$  \hspace{1cm} (6)

defines the leading field-dependent anisotropy in the transition temperature, where $Q_{ab}$ is a tensor describing the coupling of the order parameter to the magnetic field. The quantity $\Delta \chi_{ab} = -a(T_c - T)Q_{ab}(T)\psi^2 = \chi_{ab}(T^-) - \chi_{ab}(T^+)$ is the (reduction) in the magnetic susceptibility tensor associated with the hidden order. To explore the non-linear susceptibility in a given direction $\hat{n}$ of the magnetic field, we write $H_a = \hat{n}_aH$, so that $T_c(H) = T_c - \frac{1}{2}Q(\theta, \phi)H^2$, where $Q(\theta, \phi) = \hat{n}_aQ_{ab}\hat{n}_b$. Using thermodynamic arguments, we can explore the experimental consequences of Equation (5) [1]. Solving for $\psi$, we can rewrite this free energy below $T_c$ as

$$f[T] = -\frac{a^2}{2b}[T_c(H) - T]^2.$$  \hspace{1cm} (7)

so that, taking appropriate derivatives in zero field [1, 4], we find that $\Delta C_V/T_c = \frac{a^2}{T}$, and

$$\frac{d\Delta \chi_1[\theta, \phi]}{dT} = \frac{\Delta C_V}{T_c}Q[\theta, \phi]$$  \hspace{1cm} (8)

leading to the relationship

$$\frac{\Delta C_V}{T} \Delta \chi_3(\theta, \phi) = 3\left(\frac{d\chi_1(\theta, \phi)}{dT}\right)^2$$  \hspace{1cm} (10)

that has been previously checked for URu$_2$Si$_2$ along the c-axis [16]. Equation (10) holds for all orientations of the
applied magnetic field. From (9) we can estimate $Q_{zz}$:

$$
\Delta \chi_3 = 0.18 \text{ emu/mol T}^3 = 0.18 \text{ mJ/mol T}^4 \quad [1] \text{ (where we note that 1 emu = 1 mJ/T)}
$$

and $\Delta \chi_{zz} = 300 \text{ mJ/mol K}^3 \quad [17]$ so we find that

$$
Q_{zz} = \sqrt{\frac{\Delta \chi_3 T_{HO}}{3 \Delta C_V}} = 0.014 \text{ K/T}^2. \quad (11)
$$

This value of $Q_{zz}$ suggests that $T_c$ vanishes at fields of $\mu_0 H_c \sim \sqrt{2 T_{HO}/Q} = 50 \text{T}$, a number that is roughly consistent with the experimental value \cite{18}, particularly as our extrapolated phase boundary from small fields is expected to overshoot the measured one.

In-plane anisotropy has been reported in torque magnetometry \cite{19}, cyclotron resonance \cite{20}, x-ray diffraction \cite{21} and elastoresistivity measurements \cite{22} though NMR and NQR studies suggest that this nematic signal decreases with increasing sample size and also depends on sample quality, suggesting that the bulk is tetragonal \cite{23, 24}. In principle inter-domain fluctuations of the basal plane susceptibility contribute to an in-plane $\chi_3$ below $T_c$, and so again experimental consistency with these different measurements must be checked.

For a single domain, broken tetragonal symmetry-breaking manifests itself through the development of a finite off-diagonal component of the magnetic susceptibility $\chi_{xy}^D \sim \left(\frac{V_D}{V_c}\right) \left(\frac{m_a \langle m_y \rangle}{V} \right)$, where $V_D$ is the volume of the domain, $V_c$ is the volume of a unit cell and $m_a = M_a/N_{cells}$ is the magnetization per cell. The bulk off-diagonal magnetic susceptibility involves an average over many different domains that is zero, namely $\chi_{yy}^D \sim 0$ where the over-bar denotes a domain average. However domain fluctuations in the susceptibility remain finite, given by

$$
(\Delta \chi_{xy})^2 \sim \left(\frac{V}{V_D}\right) \left(\chi_{xy}^D\right)^2, \quad (12)
$$

where $V$ is the total volume of the sample. The change in the bulk basal-plane nonlinear susceptibility in the hidden order phase is then given by

$$
\Delta \chi_{3.1} \sim -\left(\frac{V}{V_c}\right) \left(\frac{m_a \langle m_y \rangle}{V} \right)^2 \frac{1}{T^3}. \quad (13)
$$

Putting in numbers, the anisotropy in the linear susceptibility is at least five, $\frac{\chi_{xx}}{\chi_{yy}} \sim 14 \text{ (where we use that}\langle m_y \rangle = m_a)$, while the error bounds on the measurement of the in-plane nonlinear susceptibility are given by $\frac{\Delta \chi_{3.1}}{\chi_{3.1} zzzz} \leq 14 \text{ so that}$

$$
\frac{\chi_{xy}^D}{\chi_{xx}} \leq 5 \times \sqrt{0.14} \sim 1.9 \quad (17)
$$

which sets a bound that is two orders of magnitude larger than the anisotropy measured by torque magnetometry in micron-sized tiny samples. Thus there is no inconsistency between our nonlinear susceptibility measurements and previous torque magnetometry measurements. We also see that an order of magnitude improvement in the nonlinear susceptibility measurements would make it possible to observe the probe the reported in-plane anisotropy with a bulk measurement.

**OPEN QUESTIONS FOR FUTURE WORK**

We next turn to the many open questions, both for experiment and for theory motivated these angle-dependent susceptibility measurements.

**Experiment**

Can this angular anisotropy in the hidden order parameter be probed by further spectroscopic measurements at temperatures in the vicinity of $T_{HO}$?
• Does the Knight shift display a similar angular anisotropy?

The NMR Knight shift may be a useful additional tool to probe the g-factor anisotropy at the hidden order transition. In URu$_2$Si$_2$ the Knight shift closely tracks with the bulk susceptibility and thus can be used as a cross-check of the angular anisotropy at the hidden order transition [25]. NMR measurements [26, 27] suggest that the spin contribution to the Knight shift has an anisotropy in excess of 25. It would be interesting to follow this detailed anisotropy both as a function of angle and as a function of pressure in the vicinity of the transition from HO to antiferromagnetism.

• Does Raman probe Ising spin fluctuations?

Recent Raman measurements indicate that the most significant temperature-dependent response is in the $A_{2g}$ channel [28, 29] where the measured Raman response function

$$\chi_{A_{2g}}(\omega, T) = \int_0^\infty dt \langle O_{A_{2g}}(t), O_{A_{2g}}(0) \rangle e^{i\omega t}$$

(18)

closely resembles the inelastic neutron scattering signal at small wavevector [5]. Furthermore, the static Raman susceptibility

$$\chi_{A_{2g}}(T) = \frac{2}{\pi} \int_{-\infty}^{\infty} \frac{\text{Im} \chi_{A_{2g}}(\omega, T)}{\omega} d\omega$$

(19)

tracks the c-axis magnetic susceptibility [28, 29]. If we expand the crystal-field Hamiltonian of tetragonal URu$_2$Si$_2$ to linear order in the electromagnetic stress-energy tensor, the the $A_{2g}$ component of the coupling takes the form

$$H = \hat{H}_0 + \hat{O}_{A_{2g}}(A_x A'_y - A_y A'_x),$$

(20)

where unprimed and primed vector potentials refer to in and outgoing fields, respectively, while the operator

$$\hat{O}_{A_{2g}} = \left[ a(\omega)(J_2^2 - J_y^2) J_x J_y + b(\omega) J_y \right].$$

(21)

Here the first term derives from the oscillatory electric field components ($E_x E'_y - E_y E'_x$) of the stress-energy tensor while the second term derives from the Poynting vector $\hat{z} \cdot (\hat{E} \times \hat{B} + \hat{E}' \times \hat{B})$. The close resemblance between the Raman signal and the measured spin fluctuations [28, 29] suggests that this second magnetic $J_y$ term is dominant.

More work is needed to determine the relative importance of $a(\omega)$ and $b(\omega)$, particularly for strongly spin-orbit coupled materials.

**Theory**

Some argue that the hidden order parameter is elusive because it is fundamentally complex. In this approach a performed band of Ising quasiparticles with half-integer angular momentum form a multipolar density wave. However because URu$_2$Si$_2$ is tetragonal, $J_z$ is conserved (mod 4). This angular momentum exchange of $\pm 4\hbar$ implies mixing of states, for example of the form

$$|k\pm\rangle = \alpha|k, \pm\frac{5}{2}\rangle + \beta|k, \pm\frac{3}{2}\rangle$$

(22)

that will lead to a finite transverse coupling ($\Phi \propto |\alpha\beta|^2$) that is ruled out by the observed Ising anisotropy observed in experiment. How to reconcile this approach with experiment?

Another tack is to argue that the hidden order parameter is elusive because it is a fundamentally novel nonlocal order parameter as occurs in superconductivity [30]. In particular it could be the case of a fractionalized order parameter, for example the square root of a multipole. One such proposal [11–13] suggests that the itinerant f quasiparticles have integer angular momentum due to a coherent, symmetry-breaking hybridization of the conduction electrons with integer spin f-states. In this case the Ising anisotropy is preserved since the up- and down-spin configurations differ by at least two units of angular momentum. This approach predicted [11–13]

$$\Delta \chi_3 \propto \cos^4 \theta$$

(23)

but the microscopic theory needs revision, partially due to the absence of the predicted transverse moment. There we treated the hybridization of $f$-moments with a simplified s-conduction band; we now know, due to interest in topological Kondo insulators, that with this approach $SmB_6$ is a metal. Instead it is crucial that we consider p-wave hybridization [31] and this will surely affect the microscopics and the gap structure leading to new, verifiable predictions for experiment.

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