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Superconducting pairs with extreme uniaxial anisotropy in URu_2Si_2

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We report magnetic field orientation-dependent measurements of the superconducting upper critical field in high quality single crystals of URu_2Si_2 and find the effective g-factor estimated from the Pauli limit to agree remarkably well with that found in quantum oscillation experiments, both quantitatively and in the extreme anisotropy ($\approx 10^3$) of the spin susceptibility. Rather than a strictly itinerant or purely local f-electron picture being applicable, the latter suggests the quasiparticles subject to pairing in URu_2Si_2 to be 'composite heavy fermions' formed from bound states between conduction electrons and local moments with a protected Ising behavior. Non Kramers doublet local magnetic degrees of freedom suggested by the extreme anisotropy favor a local pairing mechanism.

Our understanding of the mechanisms of pairing in superfluids [1, 2] and conventional superconductors [3] is largely contingent upon full characterization of the fermionic excitations within the normal state. Yet such a situation is far from realized in unconventional superconductors in proximity to magnetism [4–7]. At stake is the issue of whether the superconductivity is best described in terms of momentum-space [8, 9] or real-space [10–12] pairing. Complicating matters in rare earth and actinide superconductors, is the propensity for the coupling of the conduction electrons to local magnetic degrees of freedom to cause the elementary excitations to depart significantly from those of regular band electrons [13–15] – a situation which remains poorly understood in actinide materials owing to ambiguity as to the relevant magnetic degrees of freedom [16–21].

In this paper we find that, in spite of the seemingly intractable nature of the electronic structure of URu_2Si_2, the behavior of the superconducting upper critical field in high quality single crystals is decidedly simple. Rather than fitting directly to a model [22–24], we compare the estimated effective g-factor of the paired quasiparticles determined using the Pauli limit [25] against that of the unpaired quasiparticles determined from spin zeroes in magnetic quantum oscillation experiments [24, 26]. We find the two to be in excellent quantitative agreement over a broad angular range, establishing URu_2Si_2 as an ideal example of a Pauli limited heavy fermion superconductor. In doing so, however, we uncover a large effective g-factor with an extreme uniaxial anisotropy characteristic of a local moment with a protected Ising anisotropy [17]. We therefore propose the quasiparticles in URu_2Si_2 to be 'composite heavy fermions' formed from bound states between the conduction electrons and local non Kramers doublets [13–15, 27], having implications both for the nature of the pairing [10–12] and the hidden order phases [28, 29].

Whereas the bulk magnetic susceptibility of heavy fermion compounds typically combines several contributions [15], the heavy fermion state itself is defined only in terms of the spin susceptibility $\chi \propto g^{*2}_{\rm eff}$ of itinerant quasiparicles. Since the composition of the spin degrees of freedom is *a priori* unknown, we treat these as pseudospin $\sigma = \pm\frac{1}{2}$ quasiparticles with an effective g-factor $g^*_{\rm eff}$. Provided these quasiparticles are twofold degenerate and retain their internal structure on pairing, we can use Clogston's expression [25]

$$\mu_0 H_{\rm p} = \frac{2\Delta}{\sqrt{2}\ \mu_{\rm B} g^*_{\rm eff}} \qquad (1)$$

for the Pauli-limited upper critical field, where 2Δ is the superconducting gap (≈ 0.58 meV in URu_2Si_2 [30]), μ_0 is the permeability of free space and $\mu_{\rm B}$ is the Bohr magneton. Figure 1**a** shows the upper critical field of URu_2Si_2 measured in samples with a large residual resistivity ratio (RRR ≈ 400 [26]).

In the case of unpaired quasiparticles in a magnetic field, the same $g^*_{\rm eff}$ introduces a phase difference between magnetic quantum oscillations originating from spin split Fermi surface sheets. Again, provided the quasiparticles are twofold degenerate at zero field (and have effective masses m^* that are independent of spin), the quantum oscillation amplitude is modified by a simple interference term [14]

$$R_{\rm spin} = \cos\left[\frac{\pi g^*_{\rm eff}}{2}\left(\frac{m^*}{m_{\rm e}}\right)\right] \qquad (2)$$

where $m_{\rm e}$ is the mass of the free electron. An anisotropy in $g^*_{\rm eff}$ causes the argument of this term to become magnetic field orientation-dependent, causing the amplitude to oscillate with angle θ (a schematic representation of measured data being shown in Fig. 1**b**), passing through a 'spin zero' each time $g^*_{\rm eff}(m^*/m_{\rm e})$ is an odd integer. A total of 16 spin zeroes are observed on rotating the direction of the field from $\mathbf{H}\|[100]$ to $\mathbf{H}\|[001]$ [24].

The surprising result here is that by making rather simple assumptions [implicit in Equations (1) and (2)], the estimates for $g^*_{\rm eff}$ (shown in Fig. 2) made using two independent experimental methods are quantitatively consistent over a broad angular range. The comparability of these estimates both establishes the twofold degeneracy

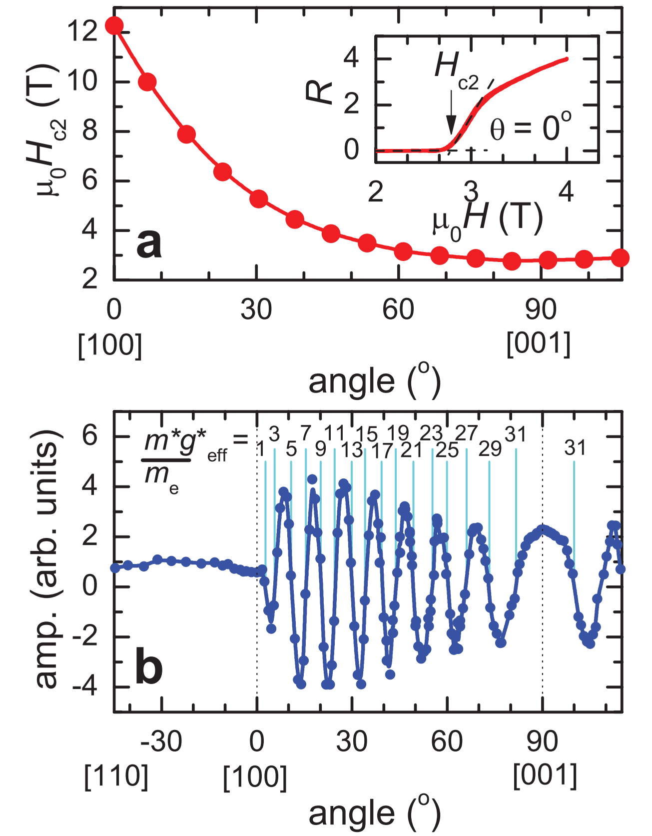

FIG. 1: Data used for determining the effective g-factor. (**a**) shows the upper critical field H_{c2} of the superconducting state in URu_2Si_2 determined from the projected onset of resistivity at $\approx$ 30 mK (similar to the method adopted by Ohkuni *et al.* [24]). An example trace is shown in the inset. (**b**) shows a schematic representation of the angle-dependent magnetic quantum oscillations adapted from Fig. 18 of reference [24], with the indices of the spin zeroes indicated. The plot pertains to the dominant α frequency [24], which can be followed uninterrupted over the entire angular range. In order to show the oscillatory behavior, the the amplitude here is multiplied by -1 on crossing each spin zero.

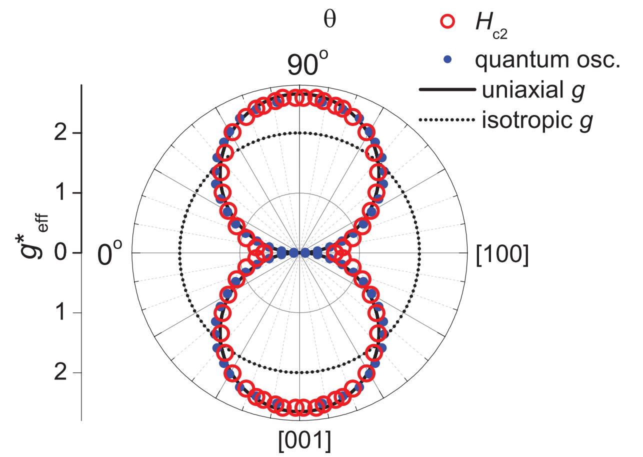

FIG. 2: A polar plot of the field orientation-dependence of $g^*_{\rm eff}$. The values are estimated using Equations (1) and (2) represented by open and filled circles respectively. Also shown, is a fit (solid line) of Equation (3) to $g^*_{\rm eff}$, and the isotropic $g^* \approx 2$ (dotted line) expected for conventional band electrons. In Fig. 1**a** we assume $H_{c2} \approx H_{\rm p}$. In extracting $g^*_{\rm eff}$ from the index assignments of $g^*_{\rm eff}(m^*/m_{\rm eff})$ in Fig. 1**b**, the weakly angle-dependent m^* is interpolated from the measured values in Reference [24].

of the quasiparticles and shows that the superconducting critical field of URu_2Si_2 corresponds to that of a Pauli limited paired fermion condensate [25] for all orientations of the magnetic field – the exception being a narrow range of angles within $\sim 10°$ of the [100] axis in Fig. 2 (likely associated with the dominant role of diamagnetic screening currents once $g^*_{\rm eff}$ is strongly suppressed [23]).

The field orientation-dependence of $g^*_{\rm eff}$ in Fig. 2 is notably different from the usual isotropic case of $g^* \approx 2$ for band electrons (dotted line), indicating the spin susceptibility of the quasiparticles in URu_2Si_2 to differ along the two distinct crystalline axes. Since the Zeeman splitting of the quasiparticles is given by the projection $\mathbf{M} \cdot \hat{\mathbf{H}}$ of the spin magnetization $\mathbf{M} = \rho \frac{\mu_{\rm B}^2}{2}(g_a^2 \cos\theta, 0, g_c^2 \sin\theta)H$ along $\mathbf{H} = H(\cos\theta, 0, \sin\theta)$ [where ρ is the electronic density-of-states], setting $\mathbf{M} \cdot \hat{\mathbf{H}} = \rho \frac{\mu_{\rm B} g^*_{\rm eff}}{2} H$ defines an effective g-factor

$$g^*_{\rm eff} = \sqrt{g_c^2 \sin^2\theta + g_a^2 \cos^2\theta} \qquad (3)$$

that (in the case of a strong anisotropy) traces a figure of '8' in polar coordinates. A fit to Equation (3) in Fig. 2 (solid line) yields $g_c = 2.65 \pm 0.05$ and $g_a = 0.0 \pm 0.1$, implying a large anisotropy in the spin susceptibility $\frac{\chi_c}{\chi_a} = \left(\frac{g_c}{g_a}\right)^2$.

To obtain a lower bound for the anisotropy, we plot $g_{\rm eff}$ (circles) in Fig. 3 extracted from quantum oscillation experiments [24] versus $\sin\theta$ (in the vicinity of the cusp in Fig. 2) together with the prediction (lines) for different values of $\frac{\chi_c}{\chi_a} = \left(\frac{g_c}{g_a}\right)^2$ made using Equation (3). The observation of a spin zero in Fig. 1 at angles as small as 3° implies a lower bound $\frac{\chi_c}{\chi_a} \gtrsim 1000$. A smaller anisotropy would be expected to lead to the observation of fewer spin zeroes and nonlinearity in the plot with an upturn in $g_{\rm eff}$ at small values of $\sin\theta$ (see Ref. [31] and Fig. 4).

A large anisotropy in the magnetic susceptibility is the behavior expected for local magnetic moments of large angular momenta whose confinement within a crystal lattice gives rise to an Ising anisotropy. Kondo coupling provides one possible means by which such an anisotropy can be transferred to itinerant electrons [15]. In the case of an isolated magnetic impurity (i.e. an isolated magnetic moment), Kondo singlets can be considered the result of an antiferromagnetic coupling between the impurity and conduction electron states expanded as partial waves of the same angular momenta [32]. A Fermi liquid composed of 'composite heavy quasiparticles' with heavy

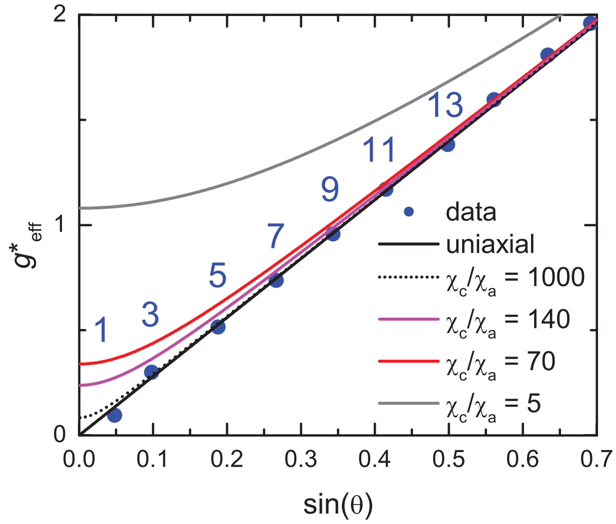

FIG. 3: A plot of $g_{\rm eff}$ versus $\sin\theta$ (circles) in the vicinity of the cusp in Fig. 2. Lines correspond to the expectation for different values of $\frac{\chi_c}{\chi_a} = \left(\frac{g_c}{g_a}\right)^2$ in Equation (3). The straight line fit corresponds to $\frac{\chi_c}{\chi_a} \to \infty$, while that with $\frac{\chi_c}{\chi_a} = 1000$ is the lower bound compatible with the data. Values 140, 70 and 5 correspond to quoted estimates from the dilute limit [33], from fits only to $H_{\rm p}$ [23] and to the measured susceptibility of pure URu_2Si_2 [4] (which likely includes non-itinerant contributions to the susceptibility).

effective masses and local angular momentum quantum numbers is one of the anticipated outcomes in a lattice of moments should such partial states overlap and satisfy Bloch's theorem at low temperatures [13, 27]. The finding of a large anisotropic impurity susceptibility ($\frac{\chi_c}{\chi_a} \sim 140$) in the dilute limit of $U_xTh_{1-x}Ru_2Si_2$ [33] supports the applicability of the Kondo lattice model to URu_2Si_2, as does the observation of a Fano lineshape in scanning tunneling microscopy experiments [34].

While magnetic moments in uranium heavy fermion compounds are generally regarded to be close to the $5f^2$ electronic configuration [11, 16–21] (i.e. with 2 f-electrons per site constituting the moment), $5f^1$, $5f^2$ and $5f^3$ are all capable of producing magnetically anisotropic low lying doublets in the tetragonal crystal environment of URu_2Si_2. The $5f^3$ configuration can yield a vanishing a-axis susceptibility for a precisely tuned combination of parameters [33]. However, only the $5f^2$ configuration can yield non-Kramers doublets in which a vanishing a-axis susceptibility is protected by a large difference ($\Delta J_z = 2$) between J_z angular momentum quantum numbers [17, 29]. A protected anisotropy can also explain why $\frac{\chi_c}{\chi_a} \gg 100$ in both the dilute and lattice limits [33].

On equating the product of the pseudospin and effective g-factor with the product of the J_z quantum numbers and Landé g-factor $g_{\rm L} = \frac{4}{5}$ of a $5f^2$ non Kramers Γ_5 doublet, we arrive at

$$\pm\frac{1}{2}g^*_{\rm ff} = (\cos\alpha|J_z = \pm 3> + \sin\alpha|J_z = \mp 1>)g_{\rm L} \quad (4)$$

(neglecting any possible additional enhancement of $g^*_{\rm eff}$ by many body effects). The solid line in Fig. 2 is produced by setting $\cos\alpha = 0.8$, which is comparable to that ($\cos\alpha \approx 0.9$) obtained from fits in the dilute limit [33].

Our study implies that neither a strictly localized or itinerant picture applies to the state of the $5f$-electrons in URu_2Si_2, which is likely to impact the origin of both the superconducting pairing and hidden order phases [28, 29]. While band structure calculations often treat the $5f$-electrons as itinerant [35], we find the quasiparticles to strongly reflect the anisotropy of the local moments determined by the crystal electric field environment. The nature of the crystal fields in URu_2Si_2 have been difficult to pin down using other spectroscopic tools. One way to understand the quasiparticles phenomenologically is through the formation of ‘composite heavy fermions,’ which constitute bound states between the conduction electrons and local moments [27]. The spin degrees of freedom of the moments become incorporated into the Fermi surface volume in such a picture, possibly giving rise to a Fermi surface topologically similar to that found in itinerant f-electron band structure calculations. The discovery of such behavior in URu_2Si_2 suggests the composite heavy fermion picture has a broader range of applicability than originally envisaged [14, 15].

Finally, we discuss possibilities for the nature of the superconducting pairing in URu_2Si_2. One popular notion is that the composite heavy fermions pair in momentum space in precisely the same way as ordinary spin $\frac{1}{2}$ electrons, with the local magnetic degrees of freedom having little impact on the symmetry of pairing [8, 9]. The finding of non-kramers degrees of freedom and composite heavy fermions, however, lends itself favorably to an alternative scheme involving local magnetic degrees of freedom [10–12]. Evidence supporting strong coupling in URu_2Si_2 at $T_{\rm c}$ includes the large value of $\frac{\Delta}{k_{\rm B}T_{\rm c}} = 4.5$ that exceeds the weak coupling value of 3.5 [30] (where $T_{\rm c} = 1.5$ K is the superconducting transition temperature), the pseudogap observed above $T_{\rm c}$ in point contact spectroscopy experiments [30], and the existence of residual magnetic entropy contributions to the susceptibility and Sommerfeld coefficient at temperatures above $T_{\rm c}$ – evidenced by the climb in both quantities with decreasing temperature [36, 37].

In summary, we find surprisingly excellent quantitative agreement between the spin susceptibility of the paired quasiparticles in URu_2Si_2 and that obtained from quantum oscillations of the unpaired fermions over a broad angular range, providing unambiguous evidence for a Pauli limited heavy fermion superconductor. The extreme anisotropy of the spin susceptibility found using two independent measurement techniques also reveals

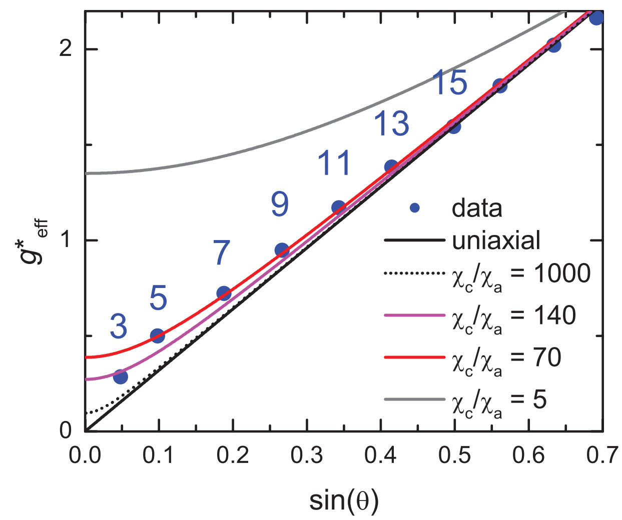

FIG. 4: As a consistency check, we plot $g_{\rm eff}$ versus $\sin\theta$ (circles) in the vicinity of the cusp in Fig. 2 where the assignment of $g^*\left(\frac{m^*}{m_{\rm e}}\right)$ indices begins at 3 instead of 1 [31], causing all points to be shifted upwards relative to Fig. 3. In such a case, one no longer obtains consistent values for g_c. From reading off the last index [at which now $g^*\left(\frac{m^*}{m_{\rm e}}\right) = 33$] we obtain $g_c \approx 2.9$. On fitting Equation (3) through the data points we obtain $g_c \approx 3.2$. Neither value is consistent with $g^*_{\rm eff} = 2.5 \pm 0.1$ estimated from H_{c2} at $\theta \approx 90°$ in Fig. 2 [31].

URu_2Si_2 to be a likely example of a system in which the magnetic properties of the itinerant carriers is determined entirely by local non Kramers doublet magnetic degrees of freedom, whose extreme Ising anisotropy is protected within the tetragonal lattice. A Fermi liquid composed of unusual heavy composite quasiparticles is therefore suggested, with the non Kramers doublets being conducive to a local superconducting pairing mechanism.

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