

Superconducting Pairs with Extreme Uniaxial Anisotropy in URu₂Si₂

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We report magnetic field orientation-dependent measurements of the superconducting upper critical field in high quality single crystals of URu₂Si₂ 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 URu₂Si₂ 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.

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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 that remains poorly understood in actinide materials owing to ambiguity as to the relevant magnetic degrees of freedom [16–21].

In this Letter, we find that, in spite of the seemingly intractable nature of the electronic structure of URu₂Si₂, 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 by using the Pauli limit [25] against that of the unpaired quasiparticles determined from spin zeros in magnetic quantum oscillation experiments [24,26]. We find the two to be in excellent quantitative agreement over a broad angular range, establishing URu₂Si₂ 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 URu₂Si₂ to be “composite heavy fermions” formed from bound states between the conduction electrons and local non-Kramers doublets [13–15,27], having implications for both 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 $\chi \propto g_{\text{eff}}^{*2}$ of itinerant quasiparticles. 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_{eff}^* . Provided these quasiparticles are twofold degenerate and retain their internal structure on pairing, we can use Clogston’s expression [25]

$$\mu_0 H_p = \frac{2\Delta}{\sqrt{2}\mu_B g_{\text{eff}}^*} \quad (1)$$

for the Pauli-limited upper critical field, where 2Δ is the superconducting gap (≈ 0.58 meV in URu₂Si₂ [30]), μ_0 is the permeability of free space, and μ_B is the Bohr magneton. Figure 1(a) shows the upper critical field of URu₂Si₂ 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_{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]

$$R_{\text{spin}} = \cos\left[\frac{\pi g_{\text{eff}}^*}{2} \left(\frac{m^*}{m_e}\right)\right], \quad (2)$$

where m_e is the mass of the free electron. An anisotropy in g_{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

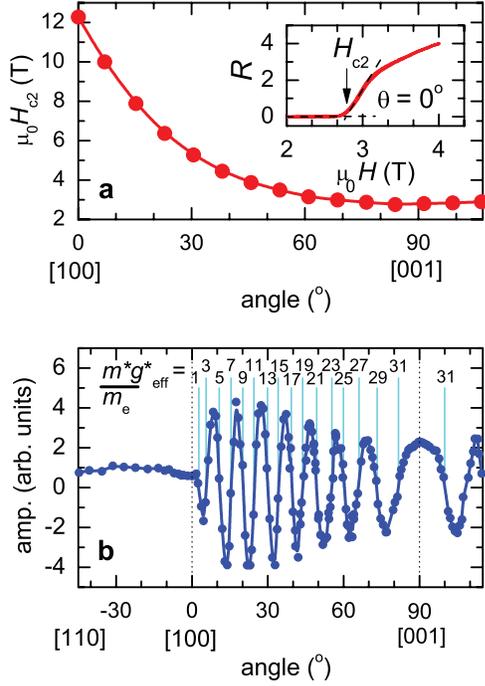


FIG. 1 (color online). Data used for determining the effective g factor. (a) shows the upper critical field H_{c2} of the superconducting state in URu_2Si_2 determined from the projected onset of resistivity at ≈ 30 mK (similar to the method adopted by Ohkuni *et al.* [24]). An example trace is shown in the inset. (b) shows a schematic representation of the angle-dependent magnetic quantum oscillations adapted from Fig. 18 of Ref. [24], with the indices of the spin zeros 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 amplitude here is multiplied by -1 on crossing each spin zero.

a “spin zero” each time $g_{\text{eff}}^*(m^*/m_e)$ is an odd integer. A total of 16 spin zeros are observed on rotating the direction of the field from $\mathbf{H} \parallel [100]$ to $\mathbf{H} \parallel [001]$ [24].

The surprising result here is that, by making rather simple assumptions [implicit in Eqs. (1) and (2)], the estimates for g_{eff}^* (shown in Fig. 2) made by using two independent experimental methods are quantitatively consistent over a broad angular range. The comparability of these estimates both establishes the twofold degeneracy 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 $\sim 10^\circ$ of the [100] axis in Fig. 2 (likely associated with the dominant role of diamagnetic screening currents once g_{eff}^* is strongly suppressed [23]).

The field orientation dependence of g_{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

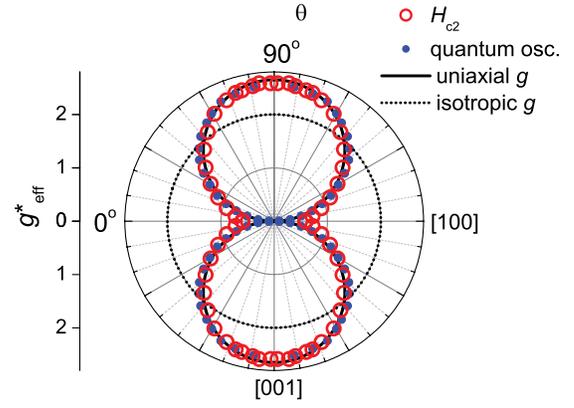


FIG. 2 (color online). A polar plot of the field orientation dependence of g_{eff}^* . The values are estimated by using Eqs. (1) and (2) represented by open and filled circles, respectively. Also shown is a fit (solid line) of Eq. (3) to g_{eff}^* and the isotropic $g^* \approx 2$ (dotted line) expected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

the quasiparticles is given by the projection $\mathbf{M} \cdot \hat{\mathbf{H}}$ of the spin magnetization $\mathbf{M} = \rho \frac{\mu_B}{2} (g_a^2 \cos\theta, 0, g_c^2 \sin\theta)H$ along $\mathbf{H} = H(\cos\theta, 0, \sin\theta)$ [where ρ is the electronic density of states], setting $\mathbf{M} \cdot \hat{\mathbf{H}} = \rho \frac{\mu_B g_{\text{eff}}^*}{2} H$ defines an effective g factor

$$g_{\text{eff}}^* = \sqrt{g_c^2 \sin^2\theta + g_a^2 \cos^2\theta} \quad (3)$$

that (in the case of a strong anisotropy) traces a figure “8” in polar coordinates. A fit to Eq. (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_{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 by using Eq. (3). The observation of a spin zero in Fig. 1 at angles as small as 3° implies a lower bound $\frac{\chi_c}{\chi_a} \geq 1000$. A smaller anisotropy would be expected to lead to the observation of fewer spin zeros and nonlinearity in the plot with an upturn in g_{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

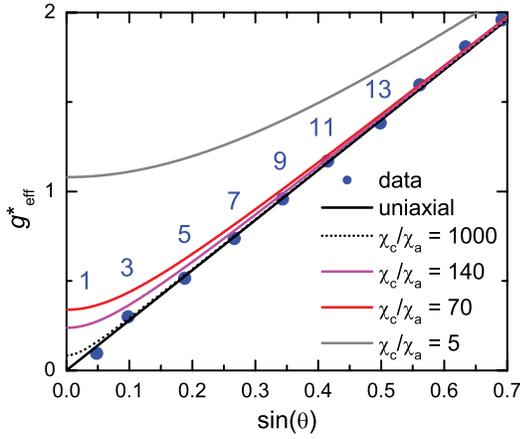


FIG. 3 (color online). A plot of g_{eff}^* versus $\sin\theta$ (circles) in the vicinity of the cusp in Fig. 2. Lines correspond to the expectation for different values of $\frac{\chi_c}{\chi_a} = \left(\frac{g_c}{g_a}\right)^2$ in Eq. (3). The straight line fit corresponds to $\frac{\chi_c}{\chi_a}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

conduction electron states expanded as partial waves of the same angular momenta [32]. A Fermi liquid composed of composite heavy quasiparticles with heavy 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 $\text{U}_x\text{Th}_{1-x}\text{Ru}_2\text{Si}_2$ [33,34] supports the applicability of the Kondo lattice model to URu_2Si_2 , as does the observation of a Fano line shape in scanning tunneling microscopy experiments [35].

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 $g_L = \frac{4}{5}$ of a $5f^2$ non-Kramers Γ_5 doublet, we arrive at

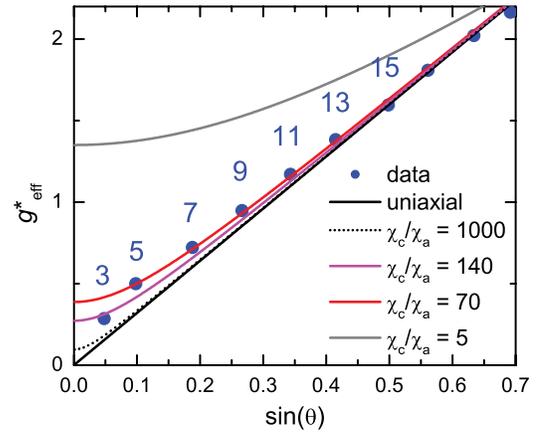


FIG. 4 (color online). As a consistency check, we plot g_{eff}^* versus $\sin\theta$ (circles) in the vicinity of the cusp in Fig. 2 where the assignment of $g^*\left(\frac{m^*}{m_c}\right)$ indices begins at 3 instead of 1 [31], causing all points to be shifted upwards relative to Fig. 3. In such a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

$$\pm \frac{1}{2}g_{\text{eff}}^* = (\cos\alpha|J_z = \pm 3\rangle + \sin\alpha|J_z = \mp 1\rangle)g_L \quad (4)$$

(neglecting any possible additional enhancement of g_{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 [28,29,36]. While band structure calculations often treat the $5f$ electrons as itinerant [37], 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 has been difficult to pin down by 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 that 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 URu₂Si₂ at T_c includes the large value of $\frac{\Delta}{k_B T_c} = 4.5$ that exceeds the weak coupling value of 3.5 [30] (where $T_c = 1.5$ K is the superconducting transition temperature), the pseudogap observed above T_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_c evidenced by the climb in both quantities with decreasing temperature [38,39].

In summary, we find surprisingly excellent quantitative agreement between the spin susceptibility of the paired quasiparticles in URu₂Si₂ 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 by using two independent measurement techniques also reveals URu₂Si₂ 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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- [1] A. J. Leggett, *Rev. Mod. Phys.* **71**, S318 (1999).
 - [2] M. W. Zwierlein *et al.*, *Science* **311**, 492 (2006).
 - [3] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1996).
 - [4] T. T. M. Palstra *et al.*, *Phys. Rev. Lett.* **55**, 2727 (1985).
 - [5] C. Geibel *et al.*, *Z. Phys. B* **84**, 1 (1991).
 - [6] N. D. Mathur *et al.*, *Nature (London)* **394**, 39 (1998).
 - [7] S. S. Saxena *et al.* *Nature (London)* **406**, 587 (2000).

- [8] M. Sigrist and K. Ueda, *Rev. Mod. Phys.* **63**, 239 (1991).
- [9] P. Monthoux, D. Pines, and G. G. Lonzarich, *Nature (London)* **450**, 1177 (2007).
- [10] P. Coleman *et al.*, *Phys. Rev. B* **60**, 3608 (1999).
- [11] D. L. Cox and A. Zawadowski, *Adv. Phys.* **47**, 599 (1998).
- [12] R. Flint, M. Dzero, and P. Coleman, *Nature Phys.* **4**, 643 (2008).
- [13] D. M. Newns and N. Read, *Adv. Phys.* **36**, 799 (1987).
- [14] J. W. Rasul, *Phys. Rev. B* **39**, 663 (1989).
- [15] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1993).
- [16] P. Santini and G. Amoretti, *Phys. Rev. Lett.* **73**, 1027 (1994).
- [17] F. J. Ohkawa and H. Shimizu, *J. Phys. Condens. Matter* **11**, L519 (1999).
- [18] A. Grauel *et al.*, *Phys. Rev. B* **46**, 5818 (1992).
- [19] G. Zwicknagl, A. N. Yaresko, and P. Fulde, *Phys. Rev. B* **65**, 081103 (2002).
- [20] B. R. Trees, A. J. Fedro, and M. R. Norman, *Phys. Rev. B* **51**, 6167 (1995).
- [21] H. Ikeda and K. Miyake, *J. Phys. Soc. Jpn.* **66**, 3714 (1997).
- [22] W. K. Kwok *et al.*, *Phys. Rev. B* **41**, 11649 (1990).
- [23] J. P. Brison *et al.*, *Physica (Amsterdam)* **250C**, 128 (1995).
- [24] H. Ohkuni *et al.*, *Philos. Mag. B* **79**, 1045 (1999).
- [25] A. M. Clogston, *Phys. Rev. Lett.* **9**, 266 (1962).
- [26] M. M. Altarawneh *et al.*, *Phys. Rev. Lett.* **106**, 146403 (2011).
- [27] P. Coleman *et al.*, *J. Phys. Condens. Matter* **13**, R723 (2001).
- [28] J. A. Mydosh and P. M. Oppeneer, *Rev. Mod. Phys.* (to be published).
- [29] R. Flint, P. Chandra, and P. Coleman (unpublished).
- [30] F. Morales and R. Escudero, *J. Low Temp. Phys.* **154**, 68 (2008).
- [31] A consistency check is required to ensure that the indices of $g^*(\frac{m^*}{m_c})$ in Figs. 1 and 3 begin at 1 (at $\theta \approx 3^\circ$) rather than a higher odd integer. In Fig. 4, we plot the same data points as in Fig. 3 but with the indices reassigned beginning at 3 instead of 1. In this case, no single line given by Eq. (3) can go through all the points. The curve that goes through most points (corresponding to $\chi_c/\chi_1 \approx 70$) is incompatible with the observation of a spin zero at $\theta \approx 3^\circ$. Furthermore, the assignment of $g^*(\frac{m^*}{m_c}) = 3$ for the first spin zero requires that $g_{\text{eff}}^* \approx 2.9$ at the last spin zero (or that $g_c \approx 3.2$ on fitting), which is too large an effective g factor to account for the observed critical fields in Fig. 1(a). Consistency between data sets (as shown in Fig. 2) and with an anisotropic spin susceptibility is therefore achieved only when the spin zero indices begin at $g^*(\frac{m^*}{m_c}) = 1$, as shown in Fig. 3.
- [32] B. Coqblin and J. R. Schrieffer, *Phys. Rev.* **185**, 847 (1969).
- [33] H. Amitsuka and T. Sakakibara, *J. Phys. Soc. Jpn.* **63**, 736 (1994).

- [34] It is necessary to subtract off the background ThRu_2Si_2 contribution to the susceptibility to obtain the extremely anisotropic component originating solely from U substitution.
- [35] A. R. Schmidt *et al.*, *Nature (London)* **465**, 570 (2010).
- [36] In the case of “hastatic order,” the hybridization between the conduction electrons and non-Kramers doublets is a two-component spinor that plays the role of an order parameter [29].
- [37] P. M. Oppeneer *et al.*, *Phys. Rev. B* **82**, 205103 (2010).
- [38] C. Pfleiderer, J. A. Mydosh, and M. Vojta, *Phys. Rev. B* **74**, 104412 (2006).
- [39] W. Schlabitz *et al.*, *Z. Phys. B* **62**, 171 (1986).