

## Interplay between Fermi Surface Topology and Ordering in URu<sub>2</sub>Si<sub>2</sub> Revealed through Abrupt Hall Coefficient Changes in Strong Magnetic Fields

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Temperature- and field-dependent measurements of the Hall effect of pure and 4% Rh-doped URu<sub>2</sub>Si<sub>2</sub> reveal low density (0.03 hole/U) high mobility carriers to be unique to the “hidden order” phase and consistent with an itinerant density-wave order parameter. The Fermi surface undergoes a series of abrupt changes as the magnetic field is increased. When combined with existing de Haas–van Alphen data, the Hall data expose a strong interplay between the stability of the “hidden order,” the degree of polarization of the Fermi liquid, and the Fermi surface topology.

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When observed, quantum oscillatory effects such as the de Haas–van Alphen (dHvA) effect provide a definitive measure of the Fermi surface topology in metals. There are some situations, however, in which quantum oscillatory effects lose their sensitivity—at very low magnetic fields, through a thermally driven phase transition, or, more recently, at a quantum phase transition. As the predictive power of Hall theory improves, the Hall effect is becoming increasingly recognized as a viable alternative for understanding Fermi surface changes in *f*-electron antiferromagnets and ferromagnets tuned close to quantum criticality [1,2]. Any knowledge of the extent to which the *f* electrons contribute to the Fermi surface topology is of crucial importance for understanding the nature of the ordering and the fate of the heavy quasiparticles.

In URu<sub>2</sub>Si<sub>2</sub>, Fermi surface measurements have the potential to assist in the identification of the “hidden order” phase that forms below  $T_0 \approx 17.5$  K [3,4]. Even if the order parameter cannot be seen directly, its effect on modifying the Fermi surface topology could provide valuable information on its symmetry breaking properties. Strong correlations make this all the more challenging by both restricting the observable temperature ( $T$ ) range of quantum oscillations to  $T \ll T_0$  [5] and inhibiting the predictive ability of band structure calculations. Under strong magnetic fields ( $B \approx \mu_0 H$ ), the HO phase is destabilized through a cascade of first order phase transitions between consecutive field-induced phases (see Fig. 1) [6]. A strengthening of the correlations in the vicinity of a putative field-tuned quantum critical point at  $B_m \approx 37$  T [7] is identified as a likely factor, although the changes in the Fermi surface topology have not been addressed.

In this Letter, we show that Hall effect measurements extended to high magnetic fields reveal the anomalous contribution to become weak at low  $T$ . The remaining orbital Hall effect uncovers an intricate level of interplay between the Fermi surface topology and the stability of the various phases of pure and 4% Rh-doped URu<sub>2</sub>Si<sub>2</sub> [7]. At

low  $H$  and  $T$ , the enhancement of the Hall coefficient and Hall angle [8–10] shows that the otherwise large Fermi surface is reconstructed into small high mobility pockets below  $T_0$  in URu<sub>2</sub>Si<sub>2</sub>: a finding that is ubiquitous among

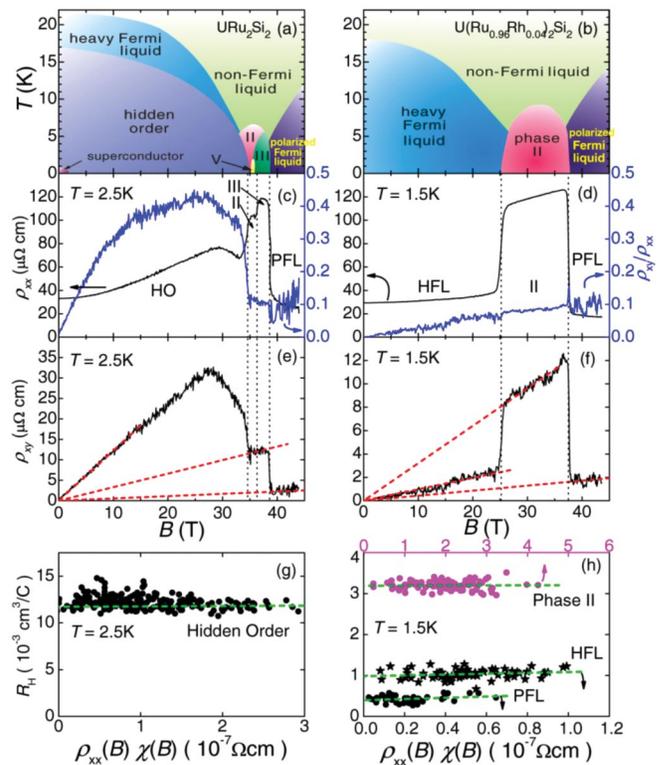


FIG. 1 (color). Left axes: (a),(b) Phase diagram [6,7], (c),(d) longitudinal resistivity  $\rho_{xx}$ , and (e),(f) Hall resistivity  $\rho_{xy}$  (g),(h) linear extrapolations to estimate the anomalous Hall contribution of URu<sub>2</sub>Si<sub>2</sub> at  $T = 2.5$  K and U(Ru<sub>0.96</sub>Rh<sub>0.04</sub>)<sub>2</sub>Si<sub>2</sub> at  $T = 1.5$  K, respectively. Red dotted linear lines passing through the origin are guides for the eye. Green dotted lines are linear extrapolations as explained in texts. Right axes: (c),(d)  $\rho_{xy}/\rho_{xx}$  for both compounds.

imperfectly nested itinerant forms of broken translational symmetry ground states [11–14] and consistent with low  $T$  dHvA measurements on  $\text{URu}_2\text{Si}_2$  [5]. This ground state is then destabilized when a magnetic field causes two of the high mobility pockets to become spin polarized, ultimately leading to its destruction at  $\approx 35$  T. Intermediate larger and strongly polarized Fermi surfaces appear in phases II, III, and V before a fully polarized unreconstructed Fermi surface is achieved beyond  $\approx 39$  T with 1 hole/U and  $1.5\mu_B/\text{U}$ .

Single crystals of  $\text{URu}_2\text{Si}_2$  and  $\text{U}(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$  (Rh 4%) are grown using the Czochralski method and cut into standard Hall bar geometries with 6-wire contacts in the tetragonal  $ab$  plane. The longitudinal and transverse voltage signals are detected simultaneously using a pulsed magnetic field of  $\sim 100$  ms duration applied along  $c$  axis up to 45 T. Special care is taken to obtain Hall coefficient data under isothermal conditions during the pulse by confirming consistency of the resistivity and phase boundaries with measurements made in the static magnet [7]. The ability to reverse the polarity of the pulsed magnetic field *in situ* between pulses enables the diagonal  $\rho_{xx}$  and off-diagonal (Hall)  $\rho_{xy}$  to be determined accurately. Resistivities measured at positive and negative  $B$  are subtracted and added to extract the two components.

Figures 1(c)–1(f) shows typical  $\rho_{xx}$  and  $\rho_{xy}$  data for both pure and 4% Rh-doped  $\text{URu}_2\text{Si}_2$  obtained at low temperatures. The  $\rho_{xx}$  data are qualitatively similar to published  $\rho_{zz}$  data [6,7]. In contrast to the original Hall measurements of Bakker *et al.* up to 40 T [15], the present  $\rho_{xy}$  data are linear in  $B$  within each high magnetic field phase, and also within the HO phase for  $B \lesssim 26$  T (we discuss the region  $26 \lesssim B \lesssim 35$  T below). This is true at all temperatures  $\lesssim 5$  K and for both samples in Figs. 2(a) and 3(a), enabling the Hall coefficient  $R_H \equiv \rho_{xy}/B$  to be reliably extracted in Figs. 2(b) and 3(b). Temperature control and isothermal conditions are important factors in ensuring the reliability of the present measurements. The ability to stabilize  $T$  over a considerable range also enables us to ascertain the anomalous Hall coefficient  $R_{aH}$  as a relatively weak effect when compared to the orbital contribution in the low  $T$  limit.

Empirically,  $R_{aH}(T) \approx C_a \rho_{xx}(T) \chi(T)$  in heavy fermion metals [2,16], where  $C_a$  is a constant. The intrinsically narrow quasiparticle bandwidth approaching  $B_m$  implies that even the orbital contribution will be  $T$  dependent (i.e., see Figs. 2 and 3), thus requiring a modified analysis. Here, we evaluate the anomalous contribution by plotting  $R_H(B)$  versus  $\rho_{xx}(B) \chi(B)$  at fixed values of  $T$  when  $\chi(B) = \partial M(B)/\partial H(B)$  and  $B \approx \mu_0 H$ , yielding a constant slope  $C_a$  within each phase as shown in Figs. 1(g) and 1(h). Within the HO phase at  $T = 2.5$  K,  $C_a \approx 0.0420 \text{ T}^{-1}$  and the resultant  $R_{aH}$  is less than 2% of the orbital contribution.  $C_a$  cannot be extracted reliably for the other phases of pure  $\text{URu}_2\text{Si}_2$ . However, for the 4% sample at

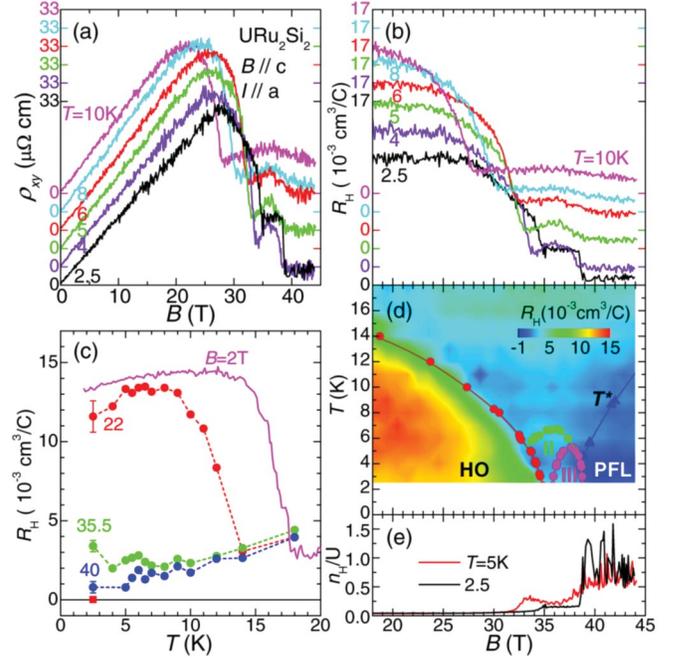


FIG. 2 (color). (a)  $\rho_{xy}$  and (b)  $R_H \equiv \rho_{xy}/B$  of  $\text{URu}_2\text{Si}_2$  at selected temperatures. (c) Temperature dependence of  $R_H$  at selected  $B$  and typical error bars of  $R_H$  in each phase region.  $R_H$  at 2 T (magenta line) is obtained from the temperature sweep using a superconducting magnet. The red square represents the estimated anomalous contribution in the HO state, which is less than 2% of the measured  $R_H$ . (d) The  $B$ - $T$  contour plot of  $R_H$  and published phase boundaries (solid symbols) [6]. (e) The effective carrier density,  $n_H \equiv -1/eR_H$  at  $T = 2.5$  and 5 K.

$T = 1.5$  K, we obtain  $C_a \approx 0.1013$ ,  $0.0027$ , and  $0.1473 \text{ T}^{-1}$  for the heavy Fermi liquid (HFL), phase II and polarized Fermi liquid (PFL) regimes, respectively, implying that  $R_{aH}$  contribution is always less than 7% of  $R_H$  for  $T \lesssim 3$  K. This serves as an upper limit for the error involved in extracting values of  $n_H$  from  $R_H$  at the lowest temperatures.

In this respect, the 4% Rh-doped sample serves as a “reference” sample for which there is no HO at low fields—the only phase that survives Rh doping is phase II. The high sensitivity of the Fermi surface transitions to perturbations in the electronic structure may explain the complete suppression of the HO phase in the Rh 4% sample at low fields as shown in Fig. 1(b). 4% Rh-doped  $\text{URu}_2\text{Si}_2$  nevertheless has the advantage of having its unreconstructed Fermi surface probed by Hall effect measurements at the lowest temperature ( $T \ll T_{\text{HO}}$ ) where the anomalous contribution is smallest. The low magnetic field value of  $R_H$  corresponds to  $n_H \approx 0.5$  holes per U, which is significantly larger than the additional  $\approx 0.08$   $4d$ -electrons per U introduced by Rh substitution on the Ru sites. This greatly reinforces previous assumptions that low  $T$  value of  $n_H \equiv -1/R_H e \approx 0.03/\text{U}$  within the HO phase of pure  $\text{URu}_2\text{Si}_2$  is primarily due to the opening of a gap over the majority of the Fermi surface

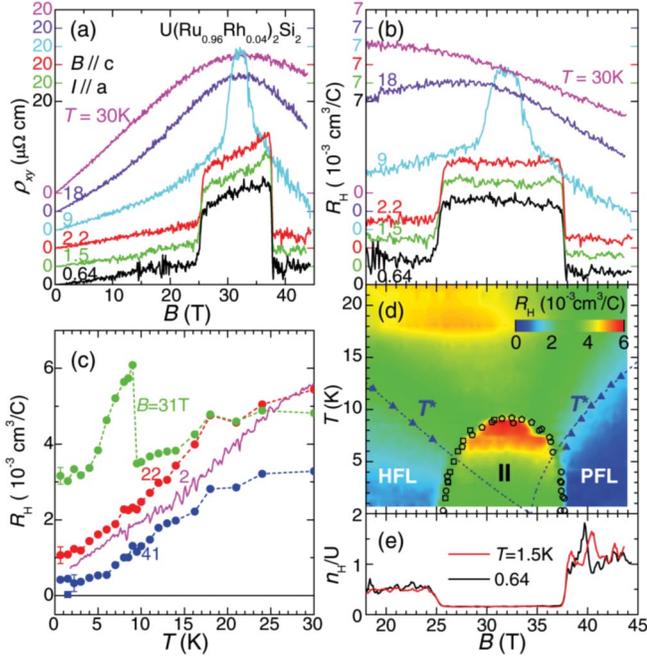


FIG. 3 (color). (a)  $\rho_{xy}$  and (b)  $R_H$  of  $U(Ru_{0.96}Rh_{0.04})_2Si_2$  at selected temperatures. (c) Temperature dependence of  $R_H$  at selected  $B$  and typical error bars of  $R_H$  in each phase region. The blue square represents the estimated anomalous contribution in the PFL region, which is less than 7% of the measured  $R_H$ . (d) The  $B$ - $T$  contour plot of  $R_H$  and published phase boundaries (open circles) [7]. (e) The effective carrier density,  $n_H \equiv -1/eR_H$  at  $T = 0.64$  and  $1.5$  K.

[8–10]. This result is robust against differences in residual resistivity and sample quality.

One remarkable feature of the data presented in Fig. 1 is that the tangent of the Hall angle estimated from  $\tan\theta = \rho_{xy}/\rho_{xx}$  is strongly enhanced only within the HO phase, indicating the existence of high mobility carriers which cause the product  $\omega_c\tau$  (where  $\omega_c$  is the cyclotron frequency and  $\tau$  is the relaxation time) to become much larger than elsewhere in the phase diagram. The development of high mobility carriers is a ubiquitous feature of itinerant order parameters that break translational symmetry (e.g., spin- and charge-density waves) in which imperfect nesting leads to the creation of small pockets [11–14]. On considering the mean free path  $l = \tau v_F$ , where  $v_F = \hbar k_F/m^*$  is the orbitally averaged Fermi velocity,  $k_F = \sqrt{2eF/\hbar}$  is the mean Fermi radius and  $F$  is the dHvA frequency, the carrier mobility becomes  $\omega_c\tau/B \equiv e\tau/m^* = el/\hbar k_F$ . Thus, high mobilities within the HO phase can be explained simply by the creation of small pockets with small  $k_F$  values, without needing to consider changes in  $\tau$  or  $l$  at  $T_0$ , or  $d$ -wave order parameters [17].

dHvA experiments within the HO phase at  $T \ll T_0$  provide direct evidence for small high mobility pockets [5]. The largest of these ( $\alpha$ ) has double the mobility  $e\tau/m^* \approx 0.12$  T $^{-1}$  of the others ( $\beta$  and  $\gamma$ ) and a volume

that agrees with  $n_H$  obtained from the Hall effect measurements. Such small pockets, and subsequently the stability of order parameter responsible for their creation, are vulnerable to perturbations. The intrinsically large effective masses in itinerant  $f$ -electron metals enables such pockets to be easily polarized by a magnetic field, as shown schematically in Fig. 4(c). The polarization field  $B_p$  can be estimated directly from the dHvA data by equating the Fermi liquid,  $\varepsilon_F = \hbar eF/m^*$  and Zeeman  $h = g_z\sigma\mu_B B$  (band splitting) energy scales. Here,  $F$  is proportional to the cross-sectional area of an orbit  $A_k$  in reciprocal space by way of the Onsager relation  $F = \hbar A_k/2\pi e$  while  $g_z\sigma \approx 1.3$  is the product of the  $g$  factor and spin determined from field orientation-dependent experiments [18]. This yields  $B_p \approx 120$ , 25, and 35 T for the known  $\alpha$ ,  $\beta$ , and  $\gamma$  carrier pockets, respectively [5]. Thus, one spin component of the heaviest  $\beta$  pocket is expected to become depopulated well within the HO phase, while that of the  $\gamma$  pocket is depopulated at the same field  $B_c \approx 35$  T where the transition out of the HO phase into phase II is observed [see Fig. 1(a)]. As a consequence of such a depopulation, the chemical potential  $\mu$  must reequilibrate itself among the remaining pockets, leading to changes in their sizes and net observable changes in  $R_H$ . Indeed, as observed in Figs. 1(e) and 2(b), once one spin component of the heaviest  $\beta$  pocket becomes depopulated beyond  $\approx 25$  T,  $\rho_{xy}$ , and consequently,  $R_H$  starts a downward trend. Since  $R_H$  is primarily due to the  $\alpha$  pocket of holes, the downward trend suggests that it is the up spin component of a holelike  $\beta$  pocket that becomes depopulated contributing its carriers to  $\alpha$ . This downward trend then rapidly accelerates beyond  $\approx 34$  T as  $B_p$  for the  $\gamma$  pocket is approached, indicating it to be another hole pocket that becomes depopulated. Close inspection of Fig. 2(b) reveals that the drop in  $R_H$  overshoots that subsequently observed within phases II and III, suggesting that depopulation of the  $\beta$  and  $\gamma$  pockets factors into the destruction of the HO phase. If stability of the HO phase requires an optimum value of translational vector  $\mathbf{Q}$  that gaps most of the Fermi surface, this  $\mathbf{Q}$  will no longer be optimal once  $\mu$  begins to change, exacerbating the destabilization of the HO phase by the Zeeman effect.

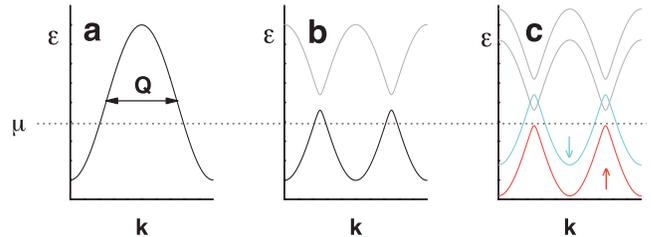


FIG. 4 (color). Schematic cross section in  $k$  space through a hypothetical hole band. Broken translational symmetry ( $\mathbf{Q}$ ) can cause such a band (a) to be reconstructed into small pockets with a Dirac-like dispersion (b) which are then easily polarized by a magnetic field (c).

The destruction of the HO phase is accompanied by an increase in  $n_H$  from  $\sim 0.03/U$  inside HO to  $\sim 0.15/U$  inside phase II, suggesting that a significant fraction of the unreconstructed Fermi surface is recovered. The appearance of such a large Fermi surface will cause the Hall effect to become dominated by lower mobility carriers, thus explaining the restoration of  $\tan\theta$  to values comparable to that of the complete unreconstructed Fermi surface. Surprisingly, neither the additional 0.08 electrons per U nor the disorder introduced by charged impurities in 4% Rh-doped URu<sub>2</sub>Si<sub>2</sub> appears to have a detrimental effect on the stability of phase II. The collapsing energy scale of the Fermi liquid temperature of the unreconstructed Fermi surface associated with the field-induced correlations could be playing a role in making this phase more robust to disorder [6,7,19]. This would be especially true were the phase to involve local moment ordering of XY electric quadrupolar degrees of freedom [20,21]. Similarities in the value of both  $R_H$  [Figs. 2, 3(b), and 3(c)] and  $M_z$  within phase II [20,22] in pure and Rh-doped URu<sub>2</sub>Si<sub>2</sub> suggest that the precise degree of polarization of the Fermi liquid is an intrinsic property of the order parameter.

In both pure and 4% Rh-doped URu<sub>2</sub>Si<sub>2</sub>, the destruction of the field-induced phases is followed by a complete polarization of the unreconstructed quasiparticle bands [19] whereupon ordering is no longer possible. The saturation of the magnetization at  $\sim 1.5\mu_B/U$  within the polarized Fermi liquid (PFL) state is accompanied by an increase in  $R_H$  to a value corresponding to  $0.9 \pm 0.3/U$  and  $1.1 \pm 0.2/U$  in pure and 4% Rh-doped URu<sub>2</sub>Si<sub>2</sub> respectively. The realization of a carrier density of  $\approx 1/U$  is consistent with the entropy recently observed in heat capacity measurements of the polarized bands [19].

In conclusion, a clear picture of the interplay between the Fermi surface topology, degree of polarization of the Fermi liquid, and the stability of the HO and field-induced order parameters is now starting to emerge in URu<sub>2</sub>Si<sub>2</sub>. At low magnetic fields,  $T_0$  is accompanied by a significant gapping of the majority of the Fermi surface, leading to a reduction in the electronic heat capacity and Hall number. An itinerant density-wave order parameter of the type inferred from thermal conductivity measurements [23] would provide a mechanism by which small pockets of highly mobile carriers could be produced in Fig. 4. Evidence for a low density of highly mobile carriers appears not just in the measurements of the Hall angle, but also in the Nernst [10] and dHvA effects [5]. The intrinsically heavy masses of these pockets (compared to regular band electrons) makes them vulnerable to polarization by a magnetic field [i.e., as in Fig. 4(c)], which subsequently destabilizes the HO phase. The polarization of at least two of the three pockets can both be predicted from existing dHvA measurements [5,18] and observed directly in the

Hall effect, ultimately factoring into the destruction of the HO phase at  $\approx 35$  T. II, III, and V involve some form of intermediate ordering that accommodates a partial polarization of the  $f$  electrons and partially gapped quasiparticle bands. Finally, ordering of any type is prohibited once the unreconstructed quasiparticle bands are completely polarized [19], yielding a simple polarized  $\approx 1$  hole/U metal at  $B \gtrsim 39$  T.

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