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Journal
Physical Review Letters, 93(18)

ISSN
0031-9007

Authors
Harrison, N
Alver, U
Goodrich, RG
et al.

Publication Date
2004-10-29

DOI
10.1103/physrevlett.93.186405

4f-Electron Localization in $Ce_xLa_{1-x}MIn_5$ with M = Co, Rh, or Ir

N. Harrison,[1] U. Alver,[2] R. G. Goodrich,[3] I. Vekhter,[3] J. L. Sarrao,[4] P. G. Pagliuso,[4] N. O. Moreno,[4] L. Balicas,[5] Z. Fisk,[5,*] D. Hall,[5,†] Robin T. Macaluso,[6] and Julia Y. Chan[6]

[1]*National High Magnetic Field Laboratory, LANL, MS-E536, Los Alamos, New Mexico 87545, USA*
[2]*Department of Physics, Kahramanmaras Sutcu Imam University, 46100 K. Maras, Turkey*
[3]*Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803, USA*
[4]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[5]*National High Magnetic Field Laboratory, 1800 East Paul Dirac Drive, Tallahassee, Florida 32310, USA*
[6]*Department of Chemistry, Louisiana State University, Baton Rouge, Louisiana 70803, USA*

(Received 4 May 2004; published 29 October 2004)

de Haas–van Alphen measurements on $Ce_xLa_{1-x}MIn_5$ yield contrasting types of behavior that depend on whether M = Co and Ir or M = Rh. A stronger x-dependent scattering in the case of M = Co and Ir is suggestive of a stronger relative coupling, J/W, of the conduction electrons to the 4f electrons, which would then account for the development of a heavy composite Fermi-liquid state as $x \rightarrow 1$. The failure of a composite Fermi-liquid state to form for any x in the case of M = Rh is shown to be inconsistent with theoretical models that propose antiferromagnetism to result from spin-density-wave formation.

DOI: 10.1103/PhysRevLett.93.186405 PACS numbers: 71.45.Lr, 71.18.+y, 71.20.Ps

The behavior of rare earth and actinide compounds depends largely on the extent to which the f electrons participate in chemical bonding. The evolution from localized to itinerant behavior that gives rise to heavy fermions and quantum criticality is the subject of much theoretical debate [1]. The term "itinerant" refers to electrons that are fully hybridized with the conduction band, giving rise to a "large" Fermi surface (FS) that accommodates the f electrons. Strong fluctuations involving the f-electron spin degrees of freedom cause the resulting composite quasiparticles to have large effective masses and a large Pauli paramagnetic susceptibility. Two qualitatively different models attempt to account for the origin of quantum criticality associated with the development of antiferromagnetic (AFM) order in the low temperature limit. One of these assumes the composite quasiparticles to condense upon formation of a spin-density wave (SDW) [2], involving a translation the large FS in k space similar to that which occurs for the d-electron FS of chromium [3] [see schematic in Figs. 1(a) and 1(b)]. The other assumes the heavy quasiparticles to break apart at the quantum phase transition between the AFM and paramagnetic phase [1], giving rise to f electrons that are mostly decoupled from the conduction electrons and therefore effectively "localized." Their localization removes f electrons from the conduction band resulting in a "small" FS of different topology [depicted schematically in Figs. 1(c) and 1(d)].

Given that a RKKY-mediated local moment antiferromagnet and itinerant f-electron SDW each imply different FS topologies for the AFM phase (illustrated in Fig. 1), the appropriate ground state description could, in principle, be identified by performing de Haas–van Alphen (dHvA) measurements in both the AFM and paramagnetic regimes. Several experimental dHvA studies have now been undertaken to explore potential changes of the FS directly in AFM systems such as $CeRhIn_5$ [4] and $CeRh_2Si_2$ [5]. The Néel temperature is tuned to zero by pressure p in both systems, leading to a change from AFM to superconductivity. The effective masses increase on approaching the putative quantum critical point (QCP) with no change in FS topology for $p < p_c$, where p_c is the critical pressure at which the QCP occurs. This result implies that the f electrons remain in the same state, either localized or itinerant, for all pressures $p < p_c$ within the AFM phase, but does not by itself enable one to distinguish between localized f-electron and SDW models. In the $CeRh_2Si_2$ case [5], a new frequency possibly corresponding to a different FS topology is observed at $p > p_c$. This change could be consistent with a picture in which the 4f electrons transform from localized to itinerant behavior at the QCP, although the absence of detailed FS topological study prevents one from making a definitive conclusion, and the general applicability of the SDW models [1,2] has also not been discussed.

To this end, the $CeMIn_5$ isostructural series of compounds provide an alternative tuning parameter for investigating the factors responsible for quantum criticality. $CeMIn_5$ can be made to lie on either side of the QCP depending on whether M = Co and Ir or M = Rh [6], hence M behaves like a tuning parameter in a qualitatively similar manner to p. In the case of M = Co and Ir, a heavy composite Fermi-liquid ground state is thought to exist, which is then unstable to superconductivity at low temperatures. In the case of M = Rh, AFM order is observed, with the magnitude of the moment $\mu \approx (0.75\text{–}0.8)\mu_B/\text{Ce}$ [7] being slightly smaller than that $\mu = 0.92\mu_B/\text{Ce}$ predicted by the crystal field theory of local-

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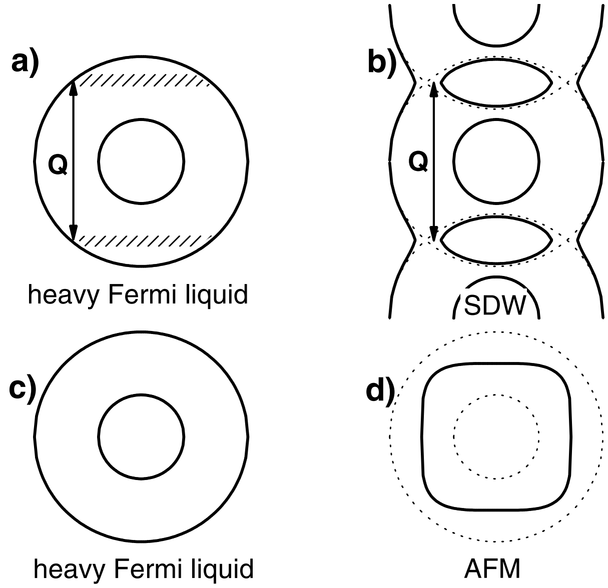

FIG. 1. Schematic of the modification of the FS upon formation of AFM order. (a) represents a notional heavy composite FS constructed from two bands, the larger of which may be subject to quantum fluctuations with respect to the translational vector **Q** on approaching a SDW phase transition at zero temperature. The process of translation (b) causes the larger section to be fragmented, leaving the smaller section intact. Quasiparticles may be able to tunnel through the SDW gaps in a magnetic fields, enabling the orbits of the original unreconstructed FS (dotted lines) to be observed. (c) shows the same FS as in (a) in the absence of SDW fluctuations. Instead, the entire FS may be subject to quantum fluctuations, with an entirely different FS in (d) being created on entering the AFM phase, for which Bragg reflection effects are less significant.

ized $4f$ electrons [8]. This difference is assumed to be caused by partial Kondo screening, a view that is indirectly supported by the observation that not all of the $R\ln 2$ entropy per spin is released above the Néel temperature [9]. However, direct measurements of the FS topology [10], using the dHvA effect, show no significant change in the FS topology or volume over the entire range of x in $Ce_xLa_{1-x}RhIn_5$, showing conclusively that the f electrons remain localized (in contrast to the compounds containing Co and Ir).

Here, we report the results of a dHvA study on the alloy series $Ce_xLa_{1-x}MIn_5$, that reveals notable differences between M = Co and Ir and M = Rh that occur over the entire range $0 < x \leq 1$. We show that while the $4f$ electrons do not contribute to the FS volume in *all* cases for $x \lesssim 0.3$, significant differences in the quasiparticle scattering emerge. The $4f$ electrons reduce the quasiparticle mean free path much more severely for M = Co and Ir than for M = Rh, finally becoming itinerant as $x \rightarrow 1$ only for M = Co and Ir. We then argue that these findings are inconsistent with SDW formation [1], which is a popular scenario for quantum criticality in heavy fermion materials.

The dHvA oscillations are measured using a combination of pulsed and static magnetic fields, using signal detection and ^{3}He refrigeration techniques described elsewhere [10]. In all experiments, the magnetic field $H \approx B/\mu_0$ is applied along the c axis of the tetragonal unit cell, where a potential change in electronic structure due to metamagnetism is not observed for $B \lesssim 50$ T [11]. Figure 2 shows the dependence of dHvA frequencies F_i on concentration x for several different FS sheets for M = Co, Rh, and Ir. Each frequency F_i corresponds to a distinct extremal cross section A_i of the FS in k space, as determined by the Onsager relation $F_i = (\hbar/2\pi e)A_i$ [12]. In all cases, F_i can be seen to depend only very weakly on x for $x \lesssim 0.3$. This implies that the $4f$ electrons provided by the Ce atoms do not contribute significantly to the FS volume at low concentrations ($x \lesssim 0.3$), and remain essentially localized. In the case of M = Co and Ir, however, all of the dHvA frequencies undergo a significant change between $x \sim 0.3$ and $x = 1$. These changes indicate that the FS topology undergoes a transformation between $x \sim 0.3$ and $x = 1$, corresponding to an expansion of the FS volume so as to accommodate $4f$ electrons that have now become itinerant [13,14]. In order to enable frequencies from each sheet of the FS to be followed through the range of concentrations where the dHvA effect cannot be observed, additional field-orientation (θ-dependent) data are shown for pure $CeMIn_5$ and $LaMIn_5$ with M = Co and Ir in Fig. 3: data of Haga *et al.* [15] are used for the case where M = Ir and $x = 1$, while similar data for the other limits are published in Refs. [13,16]. While the sizes of each FS sheet vary between the end limits of x, the overall shapes remain unchanged, giving rise to clearly identifiable angular dependences of F_i.

Let us focus initially on the dilute Ce limit: The x dependence of the effective mass for $x \lesssim 0.3$ is seen to be similar for all M in Figs. 4(a)–4(c) (though slightly heavier for M = Co), being consistent with a scenario in which the Ce atoms behave primarily as isolated Kondo

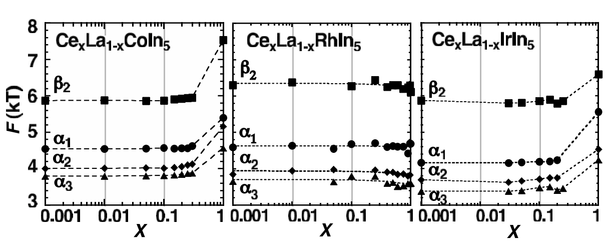

FIG. 2. A plot of the various frequencies F_i (in tesla) versus the substituted concentration x of Ce, for M = Co, Rh, and Ir in order of increasing atomic weight. The actual FS cross-sectional areas in k space can be obtained by applying the Onsager relation. The nomenclature introduced by Shishido *et al.* [13] is used for the subscripts i.

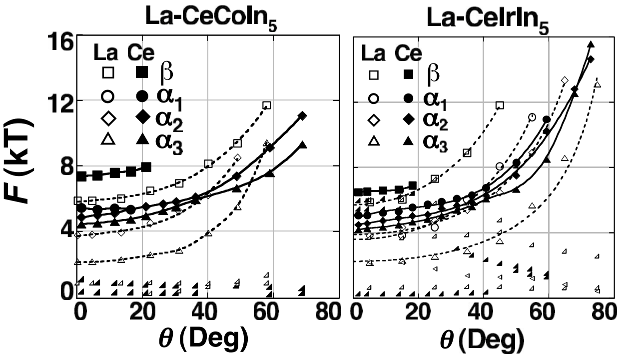

FIG. 3. A comparison of the FSs of the pure La (open symbols) and Ce (filled symbols) compounds for $M = \mathrm{Co}$ (a) and $M = \mathrm{Ir}$ (b), made by means of angle-dependent studies, where θ is angle between of **H** and the tetragonal c axis. Data for $M =$ Ir and $x = 1$ are taken from Haga *et al.* [15].

impurities in the dilute limit [17]. We note that the single-ion Ce Kondo temperature is estimated to be $T_K \approx 2$ K in $\mathrm{LaCoIn_5}$ [18]. Consequently, in magnetic fields above $\sim$10 T, where the effective masses are measured, the degeneracy of the ground state doublet is lifted, causing the Kondo screening to be mostly destroyed. Weakened resonant scattering therefore causes Ce to act mostly as a magnetic impurity for all M in the very dilute limit [17], causing the dHvA oscillations to be mostly damped by a Dingle scattering term $R_D = \exp(-\pi/\omega_c\tau) \equiv \exp(-\pi l_c/l)$, where $l_c = \sqrt{2\hbar F/eB^2}$ is the cyclotron length [12]. Figure 4(d) shows the mean free path l obtained from an analysis of the field dependence of this damping. Whereas weak scattering in the case of $M = \mathrm{Rh}$ enables the dHvA effect to be observed for all x [10], it is sufficiently strong in the case of $M =$ Co and Ir for l to become less than the perimeter $2\pi l_c$ of the cyclotron orbit when $x \gtrsim 0.3$. This causes the Landau levels to become incoherent, rendering the dHvA effect unobservable [Fig. 4(d)].

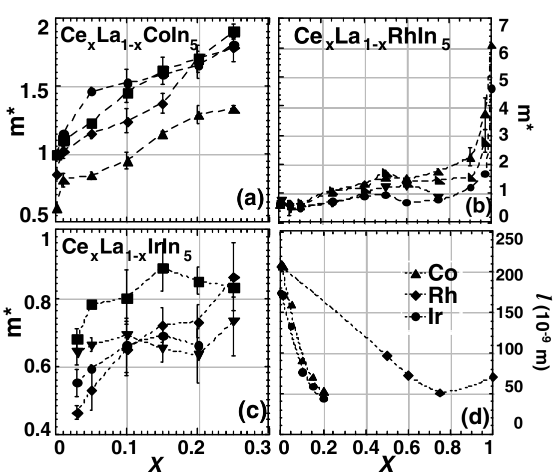

FIG. 4. A plot of the dependence of the quasiparticle effective masses m_i^* on x for the various FS cross sections observed in $M = \mathrm{Co}$ (a), Rh (b), and Ir (c), obtained from fitting the T dependence of the dHvA amplitudes for $20 < B < 40$ T, $10 < B < 20$ T, and $25 < B < 45$ T, respectively. The symbols refer to the different frequencies identified in Figs. 2 and 3. (d) l for the $F_{\beta 2}$ frequency as a function of x for $M = \mathrm{Co}$, Rh, and Ir.

Scattering in the dilute limit depends on the coupling J of the $4f$ electrons to the conduction electrons, of bandwidth W, and the density of states $N(\varepsilon_F) \simeq 1/W$, which is thought to be similar for all $\mathrm{La}M\mathrm{In_5}$ compounds. The above differences between $M = \mathrm{Co}$ and Ir and $M = \mathrm{Rh}$ must be a direct consequence of differences in J, which exist in the single impurity limit. Adopting a simple qualitative analysis based on Doniach's phase diagram [19], a smaller J/W in the case of $M = \mathrm{Rh}$ favors a stronger RKKY coupling strength, $T_{\mathrm{RKKY}} \simeq J^2/W$, compared to the Kondo temperature, $T_K \simeq W\exp(-W/J)$, which is consistent with the formation of AFM order for $x \gtrsim 0.6$ [10,20]. A larger J/W in the case of $M =$ Co and Ir favors the formation of a heavy composite Fermi-liquid. According to simple percolation arguments based a square lattice of randomly arranged Ce and La atoms [18,21], however, composite quasiparticles cannot move freely throughout the bulk for $x \lesssim 0.4$; possibly explaining why the $4f$ electrons appear to remain localized for $x \lesssim 0.3$ in the case of $M = \mathrm{Co}$ and Ir. The region $0.3 \lesssim x \lesssim 1$ therefore likely corresponds to a highly disordered intermediate state in which the composite Fermi-liquid Landau levels are incoherent, which overlaps with the region of non-Fermi-liquid behavior reported in the electrical transport by Nakatsuji *et al.* [18] at zero B. After the dHvA signal is lost above $x = 0.3$, fully coherent Landau levels are not recovered until $0.99 < x \leq 1$, whereupon the $4f$ electrons have become itinerant [13,14]. The composite heavy Fermi-liquid state realized as $x \rightarrow 1$ is much more susceptible to disorder than the weakly correlated state that exists in the dilute limit. The dHvA effect has thus far not been observed in $\mathrm{Ce_{0.99}La_{0.01}}M\mathrm{In_5}$ for $M = \mathrm{Co}$ and Ir (i.e., with only 1% of the Ce atoms replaced by La).

Given that the compounds with $M = \mathrm{Co}$ and Ir and $M = \mathrm{Rh}$ occur on opposite sides of the QCP that can be tuned by p and M [6], the results of the present study are consistent with the model illustrated in Figs. 1(c) and 1(d), in which the composite fermions break up into their constituent conduction electron and localized $4f$-electron components [1], i.e., the electronic structures are significantly different on either side. The arguments against the AFM forming as the consequence of a SDW instability of the composite quasiparticle FS are therefore rather straightforward.

First, the differences in behavior between $M = \mathrm{Co}$ and Ir, on one hand, and $M = \mathrm{Rh}$, on the other, in the limit $x \rightarrow 1$ are consistent with different bare interaction strengths in the dilute limit. This suggests that differences in the degree of hybridization between the $4f$- and conduction electrons in the AFM state is predetermined by the differences in J in the dilute limit.

Second, a SDW in $CeRhIn_5$ would have to condense from a composite Fermi-liquid state of the type that exists in $CeCoIn_5$ and $CeIrIn_5$. There is, however, no evidence that such a state is ever realized to any extent in $Ce_xLa_{1-x}RhIn_5$, with or without AFM order (which is realized for $x \gtrsim 0.6$ [10,20]). The SDW would cause such a composite quasiparticle FS to become fragmented by Bragg reflection upon formation of the AFM Brillouin zone in the manner depicted schematically in Fig. 1(b): orbits smaller than the AFM Brillouin zone would remain mostly unaffected while larger orbits could be recovered upon the tunneling of quasiparticles through the SDW gap, Δ, in a magnetic field (dotted line). This tunneling (often referred to as magnetic breakdown [12]) can occur provided $\hbar\omega_c\varepsilon_F \gtrsim \Delta^2$; here, $\hbar\omega_c = \hbar eB/m^*$ is the cyclotron energy and $\varepsilon_F = \hbar eF/m^*$ is the effective Fermi energy. This inequality would have to be satisfied in $CeRhIn_5$ to account for the observation of dHvA frequencies (e.g., from the β orbit) [13] that exceed the frequency $F_{MBZ} \approx 4780$ T corresponding to the cross-sectional area of the AFM Brillouin zone [22,23]. The SDW model is disproved in $Ce_xLa_{1-x}RhIn_5$ by virtue of the fact that all of the observed dHvA frequencies are consistent with a FS in which the $4f$ electrons remain localized. The AFM order therefore cannot form as a consequence of a SDW instability of a composite heavy Fermi-liquid in the manner depicted in Fig. 1(b).

Any Bragg reflection that occurs must therefore occur between conduction electrons and a localized lattice of $4f$ electrons. Uncoupled conduction electrons with lighter effective masses, $m^* = eB/\omega_c$, and therefore larger kinetic energies, ε_F, cause the weaker Bragg reflection in this case to be more vulnerable to magnetic breakdown, i.e., $\hbar\omega_c\varepsilon_F \gg \Delta^2$. This perhaps explains why evidence for Bragg reflection is seldom observed in dHvA experiments on AFM ordered heavy fermion materials [5,23,24], in contrast to *genuine* SDW materials where such evidence is abundant [3,25].

The picture that emerges is therefore one in which a weak relative coupling, J/W, predisposes the $4f$ electrons in $Ce_xLa_{1-x}RhIn_5$ to be localized for all $x > 0$, including the dilute limit where AFM order cannot form. The AFM order must therefore be driven by RKKY exchange interactions between $4f$ electrons that are predisposed to be localized by a small J/W, causing $T_K/T_{RKKY} \approx (W/J)^2 \exp(-W/J)$ to be small. The present results further suggest that $4f$-electron localization is the primary driving factor for quantum criticality in the p, M phase diagram of $Ce_xLa_{1-x}M In_5$ rather than the AFM order parameter. The close correspondence between antiferromagnetism and f-electron localization appears to be qualitatively similar to that observed between ferromagnetism and f-electron localization reported in systems such as $CeRu_2Si_2$ [26], where ferromagnetism is induced by a magnetic field by way of metamagnetism. Metamagnetism has also been proposed as a source of quantum critical behavior [27].

This work was supported in part by the DOE under Grant No. DE-FG02-01ER45860 (R. G. G.), the NSF under Grant No. DMR-0203214 (Z. F.). A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement No. DMR-9527035 and by the State of Florida. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy. One of the authors (N. H.) would like to thank Qimiao Si for useful discussions.

*Present address: Department of Physics, University of California, Davis, CA 95916.

†Present address: American Physical Society, 1 Research Road, P.O. Box 9000, Ridge, NY 11961.

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