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Permalink

<https://escholarship.org/uc/item/1c32k13r>

Journal

Journal of Physics Conference Series, 273(1)

ISSN

1742-6588

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Publication Date

2011

DOI

10.1088/1742-6596/273/1/012066

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A predictive standard model for heavy electron systems

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Abstract. We propose a predictive standard model for heavy electron systems based on a detailed phenomenological two-fluid description of existing experimental data. It leads to a new phase diagram that replaces the Doniach picture, describes the emergent anomalous scaling behavior of the heavy electron (Kondo) liquid measured below the lattice coherence temperature, T^* , seen by many different experimental probes, that marks the onset of collective hybridization, and enables one to obtain important information on quantum criticality and the superconducting/antiferromagnetic states at low temperatures. Because T^* is $\sim J^2\rho/2$, the nearest neighbor RKKY interaction, a knowledge of the single-ion Kondo coupling, J , to the background conduction electron density of states, ρ , makes it possible to predict Kondo liquid behavior, and to estimate its maximum superconducting transition temperature in both existing and newly discovered heavy electron families.

1. Introduction

Although we have yet to obtain a microscopic solution of the Kondo lattice problem that serves as a model for heavy electron materials, thanks to a recent phenomenological analysis [1, 2, 3, 4] of decades of experimental efforts on heavy electron behavior, we now have an experiment-based model for their behavior that explains existing experiments and has predictive power in the exploration of new families of heavy electron materials. Our model provides a unified explanation of the emergence of the heavy electron Kondo liquid that is responsible for the anomalies seen in the Knight shift, magneto-transport, and other experiments, leads to a new phase diagram that replaces the Doniach picture, and has the potential for becoming the standard model of heavy electron physics. It contains several basic aspects:

- There exists a unique temperature scale T^* for each heavy electron material; it marks the emergence of a new state of matter, a coherent heavy electron (Kondo) liquid that is produced by the collective hybridization of the local moments with the conduction electron sea, and exhibits universal behavior that scales with T^* [1].
- T^* is the nearest neighbor RKKY (Ruderman-Kittel-Kasuya-Yosida) local moment interaction; for the local moments it separates their high- T weakly interacting phase from their low- T entangled phase. It is given by $\sim 0.45J^2\rho$, where J is the single-ion Kondo coupling to the background conduction electron density of states ρ [2].
- The entangled phase below T^* is described by a two-fluid model [1, 5, 6] in which an order parameter, $f(T/T^*)$, characterizes the reduction of the overall system entropy brought about by the transfer of spectral weight from the f-electron local moments to the itinerant heavy electron Kondo liquid formed out of the conduction electron sea.

- The quasiparticles of the Kondo liquid exhibit coherent non-Landau Fermi liquid behavior; their spin susceptibility and specific heat vary as $\ln(T^*/T)$ down to a crossover temperature T_0 .
- Their behavior below T_0 can lead to unconventional superconductivity produced by quantum critical fluctuations [4], or an antiferromagnetic ordering of the local moments in which Kondo liquid relocation occurs as a precursor [7].

The position of a number of heavy electron materials on our proposed new phase diagram is given in Fig. 1, where we see that $J\rho \sim 0.15$ roughly separates the antiferromagnets and superconductors. In striking contrast to the conventional Doniach diagram which postulates a competition between collective behavior produced by RKKY interaction and single-ion behavior characterized by the single-ion Kondo temperature, T_K , the latter quantity is found to be much smaller than T^* for almost all the materials considered there. In Fig. 2 we plot the variation of T^* , T_0 , T_K , and the ordering temperatures, T_N and T_c as a function of pressure for the heavy electron material CeRhIn_5 . There one can see how the competition between localization (antiferromagnetic order) and itinerancy (superconductivity) for Kondo liquid quasiparticles plays out as $J\rho$ increases.

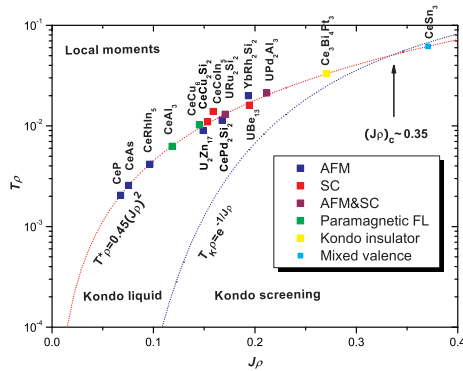


Figure 1. General phase diagram. J is the local Kondo coupling and ρ is the conduction electron density of states. T^* marks the onset of coherence, spin correlations and anomalies.

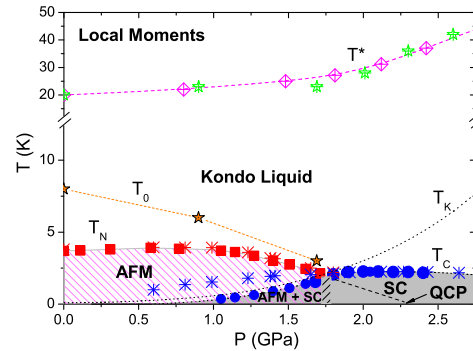


Figure 2. New phase diagram of CeRhIn_5 [8]. T^* is from resistivity peak and Hall anomaly. T_0 is from Hall anomaly [9]. T_K is estimated from $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$.

2. The physical origin and determination of the fundamental temperature scale T^*

The Kondo coupling J between a localized f-moment and conduction electron spins gives rise to two competing characteristic temperature scales in a Kondo lattice:

$$k_B T_K = \rho^{-1} e^{-1/J\rho}, \quad (1)$$

$$k_B T_{RKKY} = cJ^2\rho, \quad (2)$$

where ρ , the density of states of conduction electrons, can be determined by the Sommerfeld coefficient γ of the corresponding nonmagnetic compound (e.g., LaCoIn_5 for CeCoIn_5), $\gamma = \pi^2 k_B^2 \rho / 3$. After a correction to account for change of chemical pressure upon doping, the value of J for the Kondo lattice material can be determined by measurements of T_K in the diluted compound (e.g., $\text{Ce}_{1-x}\text{La}_x\text{CoIn}_5$), where it is given by $\pi^2 k_B^2 / 3\gamma \ln(\pi^2 k_B / 3\gamma T_K)$. An analysis [2] of dozens of heavy electron materials and the pressure data of CeRhIn_5 showed that in all cases of interest, T_K was quite small compared to T^* , so that it was natural to inquire whether T^* might be given by the RKKY interaction, Eq. (2), with a near-universal value of c . As may be seen in Fig. 1, this is indeed the case, with a best fit analysis yielding $c \sim 0.45$, so that

$$k_B T^* = 0.45 J^2 \rho. \quad (3)$$

3. The emergent heavy electron liquid

Although T^* marks the onset of coherent collective hybridization, the process is not instantaneous; the Kondo liquid emerges over a broad temperature range from T^* to a much lower cut-off temperature, T_0 , and throughout this temperature region, one therefore expects two components: the emergent Kondo non-Landau Fermi liquid composed of conduction electrons made heavy by their collective hybridization with the local moments and a local moment spin liquid formed by the substantial fraction of local moments that have not hybridized.

One is thus led to postulate a two-fluid model [5] with an order parameter $f(T/T^*)$ that denotes the fraction of the f-moments that participate in the collective hybridization; both $f(T/T^*)$ and the scaling properties of the Kondo liquid can be determined directly from experiments displaying the various heavy electron anomalies. Consider, for example, Knight shift measurements [6]. Since the f-moments are fully localized above T^* , the Knight shift of a given probe nucleus measures directly their susceptibility. Below T^* , it probes both the local moment and Kondo liquid contributions to the total magnetic susceptibility, χ . But since it couples differently to each, it will no longer be proportional to the total susceptibility, hence the appearance of the anomaly seen beginning at T^* . The two-fluid model expressions are thus [1]:

$$\chi = f(T)\chi_h + [1 - f(T)]\chi_l, \quad (4)$$

$$K = K_0 + Af(T)\chi_h + B[1 - f(T)]\chi_l, \quad (5)$$

where χ_h and χ_l are the intrinsic susceptibility of the heavy electrons and the unhybridized local moments, respectively. A and B are their hyperfine couplings. Since the Knight shift anomaly below T^* is given by, $K_a = K - K_0 - B\chi = (A - B)f(T)\chi_h$, we see that it acts as a direct probe of the emergent Kondo liquid susceptibility. A careful analysis of the Knight shift and other experiments on CeCoIn₅ shows that $f(T/T^*) = f_0(1 - T/T^*)^{3/2}$, while if one requires that the intrinsic entropy of the Kondo liquid approaches the free moment entropy $R \ln 2$ at T^* ,

$$\chi_h = R_W \frac{R \ln 2}{2T^*} \left(1 + \ln \frac{T^*}{T} \right), \quad (6)$$

where R_W is the Wilson ratio. Knight shift measurements on other heavy electron materials and fits to the anomalous behavior seen in magneto-transport, point contact spectroscopy, and Raman spectroscopy show that these expressions are universal [1]. A comparison to the recent state-of-the-art DMFT calculations [10] shows good agreement with the Kondo liquid quasi-particle density of states determined from Eq. (6), and demonstrates that that these contain the feedback needed to achieve collective hybridization.

4. Superconducting condensation and antiferromagnetic relocation

The low temperature cut-off at T_0 in the measured scaling behavior of the Kondo liquid marks the onset of new physical processes that take precedence over its growth due to collective hybridization. The two-fluid model continues to apply in this region, and provides important information about both the unconventional superconducting states and the onset of antiferromagnetism observed in heavy electron materials. Knight shift measurements are especially informative, as these provide direct information on changes in the Kondo liquid as the material becomes superconducting or antiferromagnetic. An analysis of CeCoIn₅ confirms the universal behavior of the Kondo liquid [4]: it follows two-fluid prediction above T_0 ($\sim T_c$) and, upon becoming superconducting, follows the BCS d-wave prediction below T_c , independent of the probe nuclei. Important information on the pairing mechanism for the measured d-wave superconductivity can be obtained from the NQR spin-lattice relaxation rate T_1^{-1} by using the two-fluid formula, $T_1^{-1} = f(T)T_{1h}^{-1} + [1 - f(T)]T_{1l}^{-1}$, where the local moment T_{1l}^{-1} is given by the high temperature fit. A simple two-fluid analysis reveals quantum critical fluctuations in the Kondo liquid, and yields [4],

$$T_{1h}T \propto (T + T_{QCP}), \quad (7)$$

where T_{QCP} is the distance from the quantum critical point.

A similar two-fluid analysis applied to heavy electron antiferromagnets CeRhIn₅ and CePt₂In₇ has recently led to the discovery that at a temperature T_0 well above the Néel temperature, the Kondo liquid begins to relocalize, with a corresponding decrease in $f(T)$, confirming the important role played by the Kondo liquid in bringing about the antiferromagnetic order of both it and the residual local moments present at T_N .

5. Toward a standard predictive model of heavy electron materials

By analogy to the idea of a standard model based on a well-defined microscopic theory, we have proposed here an experiment-based standard phenomenological model for heavy electron materials that derives from the successful systematic examination of existing experiments on a broad spectrum of heavy electron materials. Its predictions for heavy electron emergence at T^* accompanied by anomalous scaling behavior can easily be checked for any newly-discovered heavy electron family, and have recently been confirmed by experiments on the 127 family.

Of particular interest are the correlations in our model between measured values of $J\rho$ and the appearance of antiferromagnetism and superconductivity at low temperatures. As we have noted, materials with $J\rho < 0.15$ all appear to be antiferromagnets, but can easily be made into superconductors by applying pressure to increase $J\rho$, as Fig. 2 demonstrates vividly for the Rh-115 family. Moreover, since the superconducting transition temperature seems to scale with T^* , a strategy for finding a higher T_c family of heavy electron materials might begin by searching for materials with large values of T^* , a strategy that proved successful in the discovery of the PuGa members of the 115 family. We note this can be achieved either by finding materials with values of $J\rho$ that significantly exceed 0.15, or, with $J\rho \sim 0.15$, finding materials that possess a comparatively low value of ρ , or equivalently a large conduction electron bandwidth, and a correspondingly high value of J . Moreover, to the extent that T_c scales with T^* , for a fixed value of $J\rho$, one would have T_c scaling with J or the conduction electron bandwidth $W \sim \rho^{-1}$.

Our model is based on phenomenology, and represents a summary of our efforts to understand what experiment is telling us about the physics of Kondo lattice systems. It is incomplete, in that it poses the questions to be answered by microscopic theory but does not answer them. The remarkably successful calculations for CeIrIn₅ using DMFT represent a significant first step toward finding those answers [10]. As noted above, this is not a single site calculation; it yields a quantitative account of collective hybridization in this material by incorporating the feedback of the hybridization at one Kondo lattice site on the hybridization at its neighboring site. But what is needed are transparent microscopic calculations, beginning with perturbation theory, that point the way to the eventual renormalization group analysis needed to provide the desired microscopic theory of the collective hybridization that experiment suggests is brought about by the nearest neighbor coupling between the localized spins.

Acknowledgments

Y.Y. thanks H.-O. Lee and T. Park for experimental data of CeRhIn₅. Y.Y. and D.P. acknowledge ICAM support. Z.F. acknowledges support through NSF grant DMR-0801253. Work at Los Alamos was performed under the auspices of the US Department of Energy.

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