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Occurrence of magnetism in CeMIn$_{5-x}$Hg$_x$ (M = Rh, Ir)

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Abstract

The physical properties of CeM(In$_{1-x}$Hg$_x$)$_5$ (M = Rh, Ir) including specific heat and magnetic susceptibility are reported. Two magnetic phases exist in CeRhIn(In$_{1-x}$Hg$_x$)$_5$ with some evidence of a change from incommensurate magnetic order to a commensurate structure near 10% nominal Hg substitution. In CeIr(In$_{1-x}$Hg$_x$)$_5$, an antiferromagnetic quantum critical point near $x = 3\%$ (followed by robust long-range antiferromagnetism for $x > 5\%$) appears to be separated from superconductivity in CeIrIn$_5$. The multitude of magnetic ground states observed in the CeM(In$_{1-x}$Hg$_x$)$_5$ materials is quite sensitive to doping and magnetic fields.

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The tetragonal CeMIn$_5$ (M = Co, Rh, Ir) heavy-fermion superconductors have attracted interest in recent years due to their high superconducting transition temperatures (e.g., $T_c = 2.3\,K$ in CeCoIn$_5$), unconventional superconductivity, and magnetic-field-induced exotic ground states [1]. In particular, field-induced quantum criticality at the upper critical field $H_{c2} = 5T$ in CeCoIn$_5$ and a possible field-induced magnetic state within the superconducting phase suggests close proximity to antiferromagnetism [2].

Isoelectronic substitution of Co or Ir in antiferromagnetic CeRhIn$_5$ yields coexistence of antiferromagnetism (AFM) and superconductivity over large regions as a function of substituent element [3,4], a result that is difficult to understand within the framework of a single band picture. The exciting discovery [5] of slight changes in the electronic structure of CeMIn$_5$ with Cd substitution at the percent level in CeCoIn$_5$ appears to have “uncovered” the hidden magnetism in this material, which is very different than electron doping with Sn [6], where there is no sign of long-range magnetic order. The substitution of Hg in CeMIn$_5$ offers yet another way to probe the proximity to magnetism in CeCoIn$_5$, the field-induced magnetic state under pressure in CeRhIn$_5$ [7], and the coexistence of magnetism and superconductivity.

Single crystals of CeM(In$_{1-x}$Hg$_x$)$_5$ (M = Rh, Ir) were grown in Hg/In flux. The nominal concentration of the Hg/In ratio is reported; microprobe analysis reveals an actual Hg concentration about 20% of the nominal concentration.

The specific heat, plotted as $C/T$, of CeRhIn(In$_{1-x}$Hg$_x$)$_5$ is shown in Fig. 1a. The cusp-like anomaly at $T_N = 3.8\,K$ in pure CeRhIn$_5$ is initially suppressed with Hg substitution to $T_N \approx 2.6\,K$ and remains sharp until 20% Hg, whereupon the transition broadens and increases to $T_N = 4.5\,K$. The $T-x$ phase diagram of CeRhIn (In$_{1-x}$Hg$_x$)$_5$ is shown in the inset of Fig. 1a. A broad minimum of the $T_N(x)$ curve occurs between 5% and 15% Hg, after which the Néel transition increases rapidly. The shape of the $C(T)$ curves and the evolution of $T_N(x)$ in CeRhIn(In$_{1-x}$Hg$_x$)$_5$ is similar to the CeRh(In$_{1-x}$Cd$_x$)$_5$ system [5]. A qualitative change in the shape of the anomaly at the Néel transition in the magnetic susceptibility (not shown) from a broad maximum followed by a change in slope of $\chi(T)$ for $x < 10\%$, to a cusp-like feature (with no maximum at higher temperatures) for $x > 10\%$, respectively.

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suggests a change in magnetic structure near $x = 10\%$. The mean-field-like anomaly for $x > 10\%$ may reflect an evolution from an incommensurate magnetic structure [8] observed in CeRhIn$_5$ to a simple structure, such as the one found in CeCo(In$_{0.9}$Cd$_{0.1}$)$_5$ [9].

The CeRh(In$_{0.8}$Hg$_{0.2}$)$_5$ sample shows a remarkable sensitivity to magnetic field as displayed in Fig. 1b. The specific heat anomaly at the Néel temperature is reminiscent of a broad second-order transition. With increasing magnetic field up to $H = 5 \text{T}$, the anomaly is suppressed in temperature and the magnitude of $C/T$ decreases slightly. For $H \geq 6 \text{T}$, the transition is roughly constant in temperature at $T_N = 2.7 \text{K}$ and sharpens considerably suggesting a change to a new magnetic structure with field as shown in the inset of Fig. 1b. It is interesting to note that the shape of the anomaly and the value $T_N$ of the 20% Hg sample in field is similar to the transitions for 5% Hg (above 0.4 K). At this concentration, $\Delta C/T$ follows a non-Fermi liquid logarithmic divergence over nearly a decade in temperature from 0.6 to 5.5 K, indicating proximity to an AFM quantum critical point. Similar behavior is found in the CeIr(In$_{1-x}$Cd$_x$)$_5$ compounds, with the non-Fermi liquid behavior extending down to 50 mK for the Cd = 3% material [5,10]. A gap between the superconducting and magnetic regions of the phase diagram has not been observed in heavy-fermion superconductors before. Such a gap may have its origin in the unusual superconductivity in CeIrIn$_5$, since there evidence that this superconducting phase appears to be distinct from another superconducting phase present on the other side of the minimum in $T_c$ near $y = 0.9$ in CeRh$_{1-x}$Ir$_x$In$_5$ [11].

Multiple magnetic instabilities occur in CeMIn$_{5-y}$Hg$_y$ judging from the phase diagrams presented in Figs. 1 and 2, which can be accessed easily with chemical substitution at the percent level or modest magnetic fields. This sensitivity to doping and magnetic field suggests a delicate tuning of the quasi two-dimensional Fermi surface sheets on a global scale and is supported by a rigid band-shift picture implied from the reversibility of Cd (and Hg) doping and pressure in these “115” systems. However, recent Co and In nuclear quadrupole resonance experiments on CeCo(In$_{1-x}$Cd$_x$)$_5$ infer that the Cd ions (and, by analogy, the Hg ions) nucleate magnetic order on a local scale [12]. Resolving this apparent conundrum of Fermi surface effects vs. local tuning of the chemical environment in CeM(In$_{1-x}$Hg$_x$)$_5$ and considering its implications for unconventional superconductivity, quantum criticality,
and the coexistence of superconductivity and magnetism must await further experiments.

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