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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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Keywords: Heavy fermion; Magnetic order; Superconductivity; Quantum critical point

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 $H_{c2} = 5$ T 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 $T_N \sim 2.6$ 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 $x > 10\%$,

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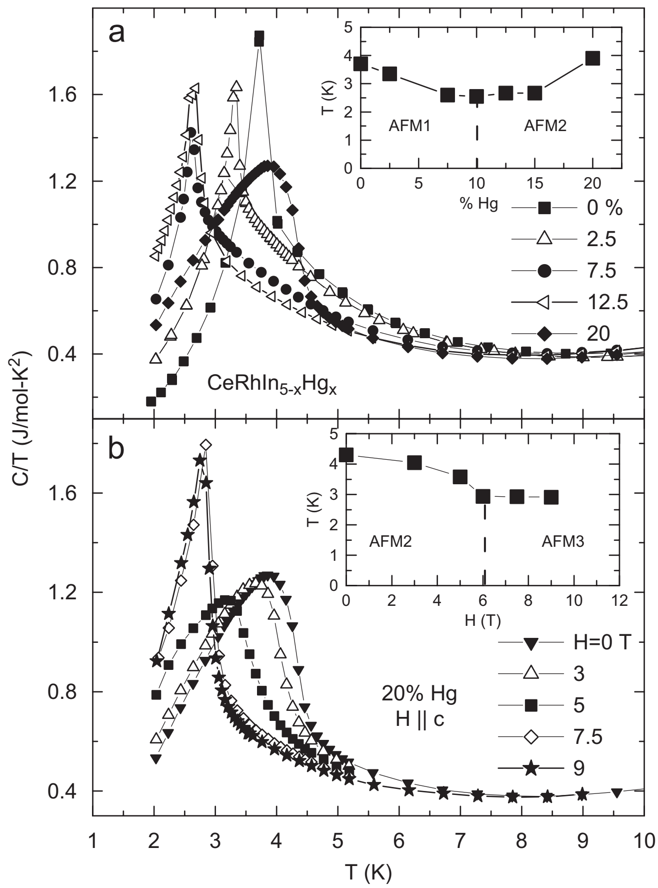

Fig. 1. (a) Specific heat C/T of CeRhIn$(In_{1-x}Hg_x)_5$ for $0 \leqslant x \leqslant 20\%$. Inset: T–x phase diagram of CeRhIn$(In_{1-x}Hg_x)_5$. (b) C/T of Hg = 20% in magnetic fields up to 9 T ($H \| c$). Inset: H–T phase diagram of the Hg = 20% sample.

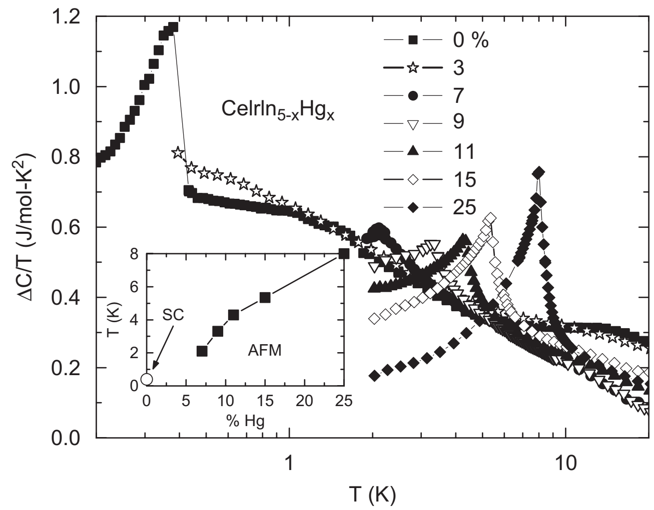

Fig. 2. (a) Magnetic contribution to the specific heat $\Delta C/T$ of CeIr$(In_{1-x}Hg_x)_5$ for $0 \leqslant x \leqslant 25\%$ on a semi-log scale. Inset: T–x phase diagram of CeIr$(In_{1-x}Hg_x)_5$.

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 $H = 5$ T, the anomaly is suppressed in temperature and the magnitude of C/T decreases slightly. For $H \geqslant 6$ T, the transition is roughly constant in temperature at $T_N = 2.7$ 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 $5\% \leqslant x \leqslant 15\%$ in zero field where $T_N(x)$ is a minimum.

The magnetic contribution to the specific heat $\Delta C/T$ of CeIr$(In_{1-x}Hg_x)_5$ for $0 \leqslant x \leqslant 25\%$ is shown in Fig. 2 on a semi-log scale, after subtraction of the nonmagnetic contribution of $LaIrIn_5$. Superconductivity is observed at $T_c = 0.4$ K for $CeIrIn_5$. Peaks in $\Delta C/T$ reveal AFM order for $x > 5\%$, reaching $T_N = 8$ K at 25% Hg. Of all the CeM$(In_{1-x}Hg_x)_5$ (M = Co, Rh, Ir) systems, the CeIr$(In_{1-x}Hg_x)_5$ materials show the largest values of the Néel temperature. The absence of superconductivity or long-range order is found in a narrow concentration range near 3% 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 $y \sim 0.9$ in $CeRh_{1-y}Ir_yIn_5$ [11].

Multiple magnetic instabilities occur in $CeMIn_{5-x}Hg_x$ 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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