Interacting Antiferromagnetic Droplets in Quantum Critical $CeCoIn_5$

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The heavy fermion superconductor $CeCoIn_5$ can be tuned between superconducting and antiferromagnetic ground states by hole doping with Cd. Nuclear magnetic resonance data indicate that these two orders coexist microscopically with an ordered moment $\sim 0.7\mu_B$. As the ground state evolves, there is no change in the low-frequency spin dynamics in the disordered state. These results suggest that the magnetism emerges locally in the vicinity of the Cd dopants.

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The discovery of superconductivity in the layered $CeMIn_5$ system has reignited interest in the low temperature physics of the Kondo lattice. The $CeMIn_5$ materials, with M = Rh, Co, or Ir, exhibit antiferromagnetism, superconductivity, or the coexistence of these two orders, depending on the external hydrostatic pressure and the particular alloy content of the M element [1]. $CeCoIn_5$ is particularly interesting as it has the highest T_c for a Ce-based heavy fermion superconductor, and the normal state exhibits non-Fermi liquid behavior that may be associated with a quantum critical point (QCP) [2]. Recently Pham and co-workers discovered that the ground state of $CeCoIn_5$ can be tuned reversibly between superconducting and antiferromagnetic by substituting Cd for In, with a range of coexistence for intermediate dopings [3]. Although other materials exhibit coexisting antiferromagnetism and superconductivity, the $CeMIn_5$ system is unique in that it can be continuously tuned by hole doping.

In the Doniach model of a Kondo lattice, localized spins interact with conduction electron spins via an exchange interaction, J, and the ground state depends sensitively on the product $JN(0)$, where $N(0)$ is the density of conduction electron states at the Fermi level [4]. For $JN(0) \gg 1$, Kondo screening of the local moments by the conduction electron spins dominates, resulting in a spin liquid ground state. At the other extreme, where $JN(0) \ll 1$, the indirect exchange (Ruderman-Kittel-Kasuya-Yosida) between local spins mediated by the conduction electrons dominates, and the ground state is antiferromagnetic. When $JN(0) \sim 1$ there is a QCP where $T_N \to 0$. Superconductivity typically emerges in this regime where competing interactions lead to complex behavior and small perturbations can drastically alter the ground state. In $CeCoIn_5$ Cd doping introduces holes that may modify the Fermi surface(s) of the conduction electrons, changing $N(0)$ and hence the ground state. As the Fermi surface evolves with hole doping, either superconductivity or antiferromagnetism may emerge, depending on the quantity $JN(0)$ in much the same way that pressure changes the ground state by modifying J [1]. An alternative to this global interpretation is that the Cd acts as a local defect that nucleates antiferromagnetism in a quantum critical system [5,6]. In a system close to a QCP, a local perturbation can induce droplets of local antiferromagnetic order. As the correlation length grows and reaches the scale of the distance between impurities, the system can undergo long-range order.

In this Letter we report nuclear magnetic resonance (NMR) data in $CeCo(In_{1-x}Cd_x)_5$ that support the latter scenario. We report data for $x = 0.10$ and $x = 0.15$, where x is the nominal concentration as reported in Ref. [3] [Fig. 1(a)]. At $x = 0.10$, the superconducting transition

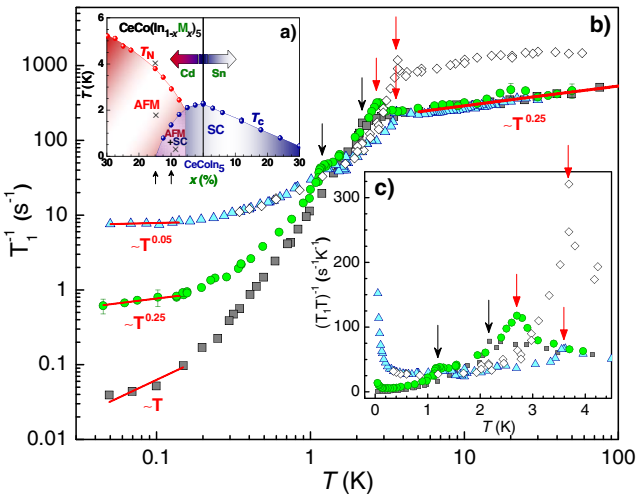

FIG. 1 (color online). (a) T_c (blue) and T_N (red) versus nominal doping in $CeCo(In_{1-x}Cd_x)_5$ and $CeCo(In_{1-x}Sn_x)_5$ [3,26]. Arrows show the concentrations reported in this study, and crosses show the points corresponding to the spectra in Fig. 2. (b) T_1^{-1} versus temperature in $CeCo(In_{1-x}Cd_x)_5$ with $x = 0.15$ (blue, ▲), $x = 0.10$ (green, ●), $x = 0$ (gray ■, from [18]), and $CeRhIn_5$ (◇). $T_N(x)$ is indicated by red arrows (at peaks of T_1^{-1}) and $T_c(x)$ by black arrows (drops in T_1^{-1}). (c) $(T_1T)^{-1}$ versus temperature.

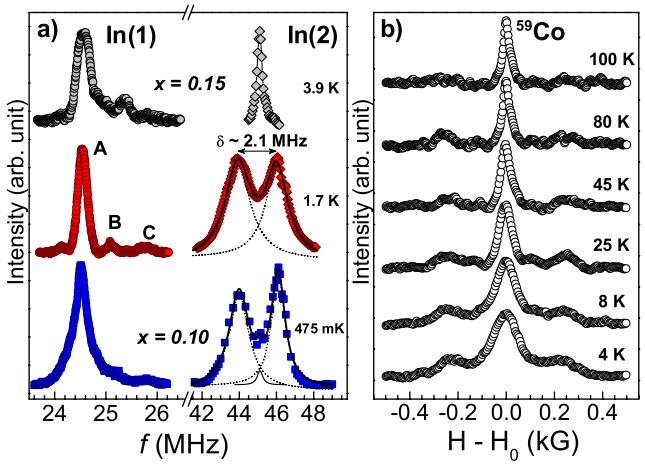

FIG. 2 (color online). (a) ^{115}In NQR spectra of the In(1) and In(2) sites at 3.9 K ($x = 0.15$, gray), at 1.7 K ($x = 0.15$, red), and at 475 mK ($x = 0.10$, blue). The A, B, and C lines of the In(1) are shown. The In(2) spectrum is split by the internal field below T_N. Dotted lines are Gaussian fits, as described in the text. (b) The central transition of the ^{59}Co at $H_0 = 33.824$ kG for the $x = 0.15$ sample. The first satellite transitions are also seen at ± 220 G.

has been suppressed from 2.3 K to $T_c = 1.2$ K, and antiferromagnetism emerges at $T_N = 2.8$ K; for $x = 0.15$, $T_c = 0$ K and $T_N = 3.7$ K. We find that the spin-lattice relaxation rate, T_1^{-1}, falls at both T_N and T_c, in agreement with the thermodynamically determined values [3], and the spectra reveal a homogeneous internal field below T_N. These results imply that both the antiferromagnetism and superconductivity coexist microscopically throughout the bulk of the material. In the disordered state, the NMR spectra are inhomogeneously broadened and the second moment of the Co resonance is strongly temperature dependent, suggesting that the electronic response of the sample depends on the proximity to a doped Cd site. The Knight shift, K, is nearly identical to pure $CeCoIn_5$, with a strong anomaly at $T^* \sim 60$ K, and T_1^{-1} exhibits an unusual $\sim T^{1/4}$ variation that is doping independent above 5 K. These results are surprising, as they indicate that the low energy spin dynamics are nearly independent of the ground state order and that the fate of the $4f$ degrees of freedom are determined only below $T \approx 2T_c$.

All measurements were made using crystals of $CeCo(In_{1-x}Cd_x)_5$ grown from indium flux as described in [3]. Single crystal were used with the field $\mathbf{H}||\hat{c}$ for the Knight shift and linewidth measurements. T_1^{-1} was measured in zero field using nuclear quadrupolar resonance (NQR) at the high symmetry In(1) site in powdered samples, using conventional inversion recovery pulse sequences [7]. Below 1 K, the rf power was reduced sufficiently to prevent heating during the time scale of the measurement. The In NQR spectra are broadened relative to the pure compound, and two new features emerge in the In(1) spectra, which we term the B and C sites [Fig. 2(a)]. The origin of these sites is unclear, but we speculate that they are related to nearest-neighbor sites of the Cd dopants. The T_1^{-1} data reported here were obtained at the A site, which we associate with the bulk In(1) sites.

In the antiferromagnetic state, the In(1) NQR resonance moves down by ~ 25 kHz, whereas the In(2) resonance splits into two lines separated by ~ 2.1 MHz [see Fig. 2(a)]. This response is similar to that of the $CeRhIn_5$, where the internal field at the In(1) lies in the ab plane and the internal field at the In(2) lies along the c axis [7]. The internal field at the In(1) nearly vanishes, even though the ordered moment remains finite, which is analogous to the response of $CeRhIn_5$ under pressure [8,9]. Recent neutron scattering experiments suggest that the ordering wave vector in $CeCo(In_{1-x}Cd_x)_5$ is $\mathbf{Q} = (\pi/a, \pi/a, \pi/c)$ [10]. The splitting of the In(2) spectrum reveals $H_{\rm int}(2) \approx 2.3$ kOe along the c axis, nearly identical to that observed at the same transition in $CeRhIn_5$ (2.7 kOe) and in the field-induced magnetic phase of pure $CeCoIn_5$ [7,11]. Assuming the hyperfine interaction is the same in both materials, this result suggests that the ordered moment is $\sim 0.7\mu_B$ in the $CeCo(In_{1-x}Cd_x)_5$ system. Surprisingly, we find that the internal field is identical in both the $x = 0.10$ and $x = 0.15$ materials, suggesting that the degrees of freedom involved with the long-range magnetic order are independent of the superconducting degrees of freedom. The spectra also reveal approximately 3% of the sample volume with no internal field in the $x = 0.10$ sample (solid line in Fig. 2). One explanation for this result is that the sample is inhomogeneous, so that some fraction of the sample sees magnetic order while the other fraction sees superconductivity. However, if this were the case then the jump in the specific heat at T_c would be $\Delta C \sim 0.03\gamma T_c \approx 13$ mJ/mol K (using $\gamma \approx 350$ mJ/mol K^2), about a factor of 6 smaller than observed [3]. Therefore, the anomalous 3% may be due to doping inhomogeneity on a macroscopic scale in the powdered sample. The remaining 97% of the sample volume shows microscopic coexistence of antiferromagnetism and superconductivity.

Figure 3 shows the temperature dependence of the Knight shift of the In(1) A line and the Co [12]. Like the pure compound, there is a large Knight shift anomaly that develops below a temperature $T^* \sim 60$ K [13]. In fact, both T^* and the temperature dependence of K remain almost doping independent for both sites. This result suggests that the spin response at the bulk A sites remains unaffected by the Cd doping and that the difference in bulk susceptibility, $\chi_c(T)$, may arise solely in the vicinity of the Cd. This scenario is further supported by the enhanced magnetic broadening as seen in Fig. 4. Doping broadens the In(1) NQR spectra by a factor of 10 due to the distribution of local electric field gradients (EFGs). On the other hand, the EFG at the Co is nearly 2 orders of magnitude smaller than that at the In(1) site, so it is less sensitive to the distribu-

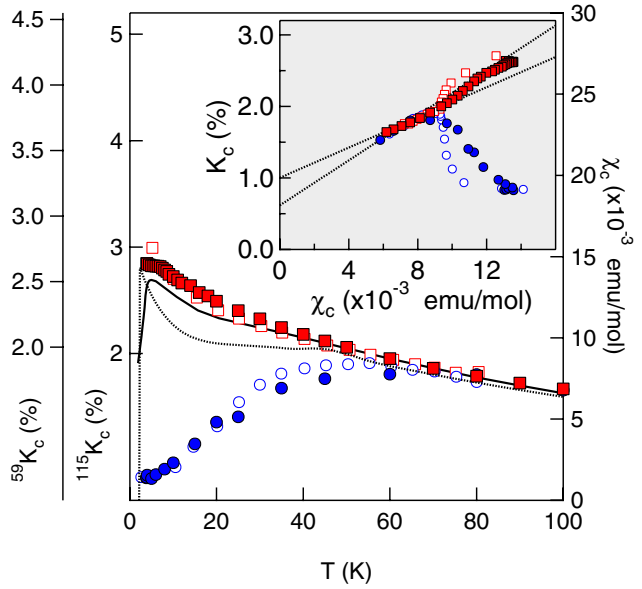

FIG. 3 (color online). The Knight shift of the ^{115}In(1) (■, $x = 0.15$ and □, $x = 0$) and ^{59}Co (●, $x = 0.15$ and ○, $x = 0$) versus temperature. The shift of the $x = 0.10$ sample (not shown) is qualitatively similar. The solid (dotted) line is the bulk susceptibility for $x = 0.15$ ($x = 0$) in the c direction [12]. Inset: The Knight shifts versus the bulk susceptibilities. The dotted lines are fits to the high temperature data.

tion of local EFGs and more sensitive to distributions of local magnetic fields. As observed at the central transition ($+\frac{1}{2} \leftrightarrow -\frac{1}{2}$) [Figs. 2(b) and 4] the magnetic broadening is significantly larger in the doped samples and is strongly temperature dependent. The broadening is not directly

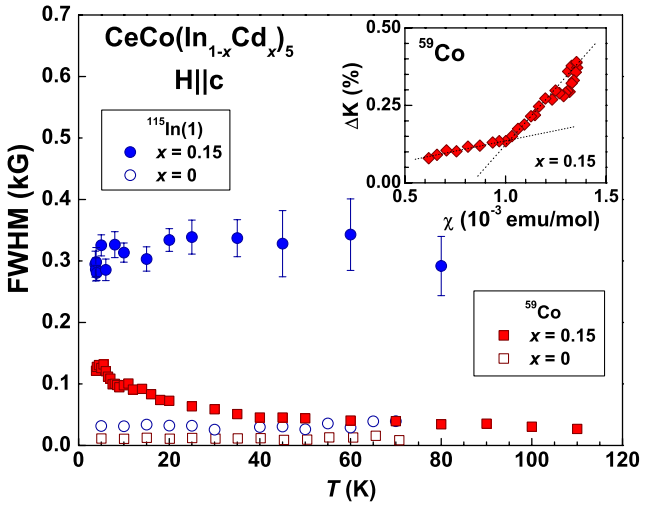

FIG. 4 (color online). The linewidths (full width half maxima) of the Co (□, $x = 0$ at 38 kOe, and ■, $x = 0.15$ at 33.824 kOe) and In(1) (○, $x = 0$ at 38 kOe, and ●, $x = 0.15$ at 33.824 kOe) spectra. The linewidth of the $x = 0.10$ sample (not shown) is qualitatively similar. Inset: The Co Knight shift distribution (FWHM/γH_0) versus the bulk susceptibility.

proportional to $\chi_c(T)$ but shows a change in behavior below 60 K (Fig. 4 inset). This result implies that the magnetic broadening mechanism does not arise from a distribution of demagnetization fields but rather has microscopic origin, possibly due to a variation in the local staggered magnetization in the vicinity of the Cd [14].

Figure 1(b) shows the temperature dependence of T_1^{-1} at the In(1) site for several different samples. In the disordered state, T_1^{-1} is independent of doping above 5 K and shows an unusual $T^{1/4}$ dependence. This behavior contrasts with the evolution of T_1^{-1} with pressure in pure $CeCoIn_5$, $CeRhIn_5$, $CeIn_3$, and $CeCu_2(Si, Ge)_2$, where the magnitude of T_1^{-1} in the disordered state changes dramatically as the ground state evolves from antiferromagnetic to superconducting [15,16]. Pressure tuning modifies the Kondo interaction, J, and changes in the ground state are reflected in T_1^{-1} through modifications of the spin fluctuation spectrum. If the Cd doping induces changes to the Fermi surface, modifying either $N(0)$ or J, and hence the Ruderman-Kittel-Kasuya-Yosida interaction, then similar modifications will be evident in the spin fluctuation spectra of the doped materials. On the other hand, if the Cd doping acts as a local seed for antiferromagnetic order, the majority of the In sites will not sense any change in the spin fluctuations until the correlation length, ξ, reaches a value on the order of the dopant spacing (~4 lattice spacings). Neutron scattering studies suggest that ξ is indeed of this order at T_N [17]. This latter scenario offers an explanation not only for the T_1^{-1} and K data but also for the sensitivity of the ground state to only a few percent of dopants.

Below T_N, T_1^{-1} in the $x = 0.15$ sample agrees with that in $CeRhIn_5$ down to 300 mK. Surprisingly, T_1^{-1} remains constant down to 45 mK. This result suggests that even in the antiferromagnetically ordered state, sufficient fluctuations remain in some channel to relax the nuclei. It is not clear if these fluctuations arise from excitations of the ordered magnetic structure, possibly excited by interactions with the heavy electrons, or from some other remaining degree of freedom, possibly as a precursor to the onset of superconductivity at a lower temperature.

In the $x = 0.10$ sample, the onset of superconductivity is clearly reflected in T_1^{-1}, which drops by more than an order of magnitude below T_c. However, the temperature dependence of T_1^{-1} below T_c is not simply T^3 as observed in pure $CeCoIn_5$ [18]. We find that $T_1^{-1} \sim T^{\alpha}$ below 200 mK, where $\alpha = 0.05(3)$ for $x = 0.15$, $\alpha = 0.25(3)$ for $x = 0.10$, and $\alpha = 1.0(2)$ for $x = 0$, so that $T_1^{-1}(x, T \rightarrow 0)$ increases with x. It is not clear if the unusual sublinear behavior in the doped samples is related to the normal state relaxation or is part of a crossover to a constant at lower temperature. In a d-wave superconductor $T_1^{-1} \sim T^3$ in the clean limit, but impurity scattering can give rise to an enhanced spin-lattice relaxation that varies linearly with temperature for sufficiently low temperatures [8,19–24]. In superconductors with coexisting antiferromagnetism,

the low energy density of states is even more sensitive to the presence of impurity scattering [25]. The Cd dopants in $CeCo(In_{1-x}Cd_x)_5$ certainly provide a source for impurity scattering, but the sublinear behavior cannot be explained by conventional in-gap states. Indeed, if the Cd served as a pair-breaking impurity, then superconductivity could not be recovered by applying pressure [3]. One possibility is that the density of states has an unusual energy dependence, which may arise as a result of interactions between the nodal quasiparticles and the low energy excitations of the antiferromagnetic structure. $CeRh_{0.5}Ir_{0.5}In_5$ has coexisting antiferromagnetic and superconducting order, and both In sites show enhanced T_1^{-1} in the superconducting state. However, the enhanced relaxation turns on at the In(2) site at a higher temperature than for the In(1) [21]. If the origin of the enhanced T_1^{-1} were purely from impurity scattering, then the onset temperature should not depend on the particular site. This result suggests instead that the enhanced T_1^{-1} is dominated by fluctuations of the antiferromagnetic order, since the static hyperfine field at the In(2) is larger than that at the In(1). It is possible that the unusual temperature dependence of the enhanced relaxation in $CeCo(In_{0.9}Cd_{0.1})_5$ is driven by such a mechanism, but it is not clear what the temperature dependence should be in this case.

In summary, we have measured the NMR response of the $CeCo(In_{1-x}Cd_x)_5$ and found evidence that Cd doping does not modify the Fermi surface but rather nucleates local antiferromagnetic droplets. When the droplets overlap the system undergoes long-range order and the antiferromagnetism and superconductivity coexist microscopically.

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