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Probing the magnetism in the $CeMIn_5$ heavy fermion systems by NMR

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Abstract

The $CeMIn_5$ heavy fermion system exhibits antiferromagnetism for both M = Rh and M = Co. $CeRhIn_5$ is antiferromagnetic at ambient pressure and superconducting under hydrostatic pressure, whereas $CeCoIn_5$ is superconducting at ambient pressure, but becomes antiferromagnetic with substitution of In by a few percent of Cd. Pure $CeCoIn_5$ exhibits a field-induced magnetic phase that coexists with the superconductivity. Nuclear magnetic resonance (NMR) and nuclear quadrupolar resonance (NQR) studies of the In(1), In(2) and Co sites reveal similar features in the magnetic states of these systems.

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The unconventional superconductor $CeCoIn_5$ exhibits many features that suggest it is proximate to a quantum critical point (QCP) for antiferromagnetism. Bulk thermodynamic and transport properties reveal non-Fermi liquid behavior in the normal state [1] and comparison of the pressure dependent phase diagram with that of the isostructural antiferromagnet $CeRhIn_5$ reveals that $CeCoIn_5$ is roughly equivalent to $CeRhIn_5$ with an internal chemical pressure of 2.1–2.3 GPa [2]. At these pressures, the Neél temperature of $CeRhIn_5$ vanishes, the Fermi surface undergoes a reconstruction as the Ce 4f electrons become itinerant, and the de Haas–van Alphen (dHvA) frequencies approximately match those of $CeCoIn_5$ [3]. Therefore, it is not surprising that $CeCoIn_5$ is unstable towards antiferromagnetism for small perturbations. Indeed, nuclear magnetic resonance (NMR) spectra of pure $CeCoIn_5$ exhibit features characteristic of field-induced magnetism, and $CeCo(In_{1-x}Cd_x)_5$ shows bulk long range antiferromagnetism for a few percent doping [4–6].

Fig. 1 shows nuclear quadrupolar resonance (NQR) and NMR spectra of the In(2) in the antiferromagnetic states of $CeRhIn_5$ and $CeCo(In_{1-x}Cd_x)_5$, and the field induced B phase of $CeCoIn_5$. The frequency axes have been shifted for comparison. The NQR spectra of the $CeRhIn_5$ and the $CeCo(In_{1-x}Cd_x)_5$ split because the internal hyperfine field at the In(2) sites, $\mathbf{H}_{int}(2)$, lifts the degeneracy (see Inset). $\mathbf{H}_{int}(2)$ is parallel to c-axis, and is given by $\sum_{i\in nn}\mathbb{B}\cdot\mathbf{S}_i \sim 2-2.5$ kOe,where $\mathbf{S}_i$ is the ordered moment at the Ce site, and $\mathbb{B}$ is the transferred hyperfine tensor [7]. $CeRhIn_5$ exhibits a spiral structure with $\mathbf{Q} = (0.5, 0.5, 0.297)$ and an ordered moment of $\sim 0.8\mu_B$, whereas $CeCo(In_{1-x}Cd_x)_5$ exhibits a commensurate structure with $\mathbf{Q} = (0.5, 0.5, 0.5)$ [8,9]. Comparing the spectra of the $CeCo(In_{1-x}Cd_x)_5$ and the $CeRhIn_5$ reveals that $\mathbf{H}_{int}(2)$ is roughly identical in the three materials. This result is surprising, since it implies that (i) the ordered moment in the $CeCo(In_{1-x}Cd_x)_5$ is comparable to that in $CeRhIn_5$, and (ii) the ordered moment in the $CeCo(In_{1-x}Cd_x)_5$ is independent of x. In fact, the $x = 0.10$ material is superconducting at $T_c < T_N$ whereas the $x = 0.15$ is not. A priori, one might expect that if superconductivity and antiferromagnetism competed in $CeCo(In_{1-x}Cd_x)_5$, and the same f-electron degrees of freedom were involved in both phases, then the size of the ordered moment would be reduced in superconducting samples. In all three cases, the spectra reveal a large distribution of $\mathbf{H}_{int}(2)$. In $CeRhIn_5$, this distribution arises because of the incommensurate nature of the long-range

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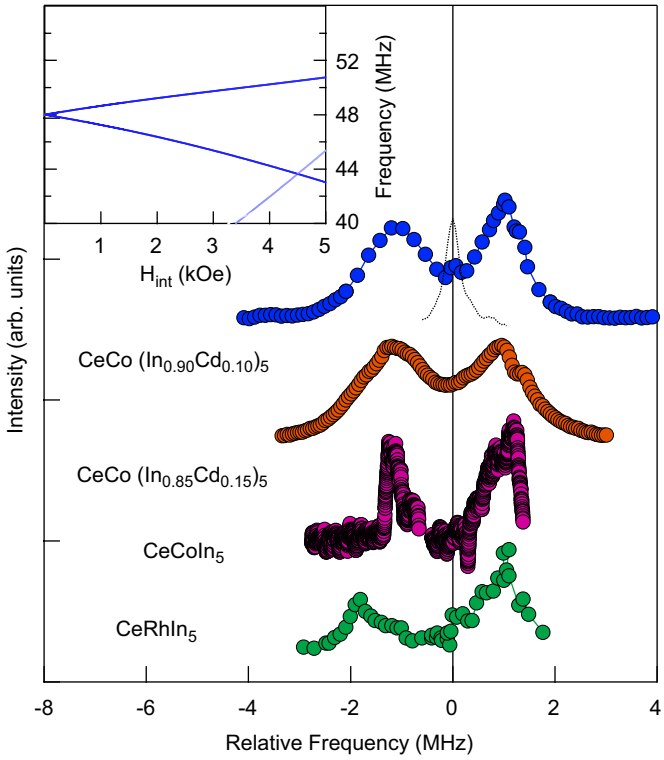

Fig. 1. Spectra of the In(2) in several $CeMIn_5$ systems. The data are plotted versus frequency relative to f_0, the frequency in the absence of a hyperfine field. For $CeRhIn_5$ $f_0 = 48.0055$ MHz [13] and the data were obtained at ($H_0 = 0, T = 1.4$ K); for $CeCo(In_{1-x}Cd_x)_5$ $f_0 = 45.0909$ MHz and ($H_0 = 0, T = 1.4$ K (0.45 K) for $x = 0.15$ (0.10)) [6]. For the field induced B phase of $CeCoIn_5$ $f_0 = 118.4$ MHz, and ($H_0 = 11.1$ T along *ab*-axis, $T = 0.05$ K) [4]. The dotted line is the spectrum of the $x = 0.10$ sample above T_N. Inset: The frequencies of the In(2) resonances versus internal hyperfine field along $\hat{c}$. The third line starting at 3.4 kOe arises from the NQR transition at 30.9 MHz that is split by H_{int}.

order. In $CeCo(In_{1-x}Cd_x)_5$, the spectra may be broadened by disorder induced by the Cd dopants.

For the field induced B phase of $CeCoIn_5$, the degeneracy is already lifted. The spectrum splits because there are an equal number of In(2) sites where $\mathbf{H}_{int}(2)$ is either parallel or antiparallel to the applied external field ($\mathbf{H}_{ext} = 11.1$ T along $\hat{a}$). At present there are no neutron scattering data that can provide information about the ordering wavevector. However, a plausible magnetic structure is $\mathbf{Q} = (0.5 + \delta, 0.5, 0.5)$, where δ is an incommensuration along the field direction, and the Ce moments are (anti)parallel to $\hat{a}$ [4]. In this case, $H_{int}(2) \sim \delta$, so it is impossible to determine the size of the ordered moment from the splitting of the spectra.

The B phase of $CeCoIn_5$ has been proposed as a possible Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) inhomogeneous superconducting phase. In this state, the superconducting order parameter, $\psi(\mathbf{r})$, is spatially modulated and vanishes periodically at nodes. The incommensurate magnetism revealed in the NMR spectra is clearly inconsistent with the long-wavelength modulation of the magnetization that should exist in an FFLO state, since the response at In(1) and Co are different than that at In(2). Any spatial variations of the superconductivity must occur over length scales larger than the coherence length, which is several unit cell lengths. Therefore, this phase must consist of coexisting local moment magnetism and superconductivity. However, it is conceivable that the magnetism is stabilized by the FFLO phase, where the nodes of $\psi(\mathbf{r})$ occupy a greater measure of space than a vortex phase.

In fact, the emergence of magnetism, with order parameter $\phi(\mathbf{r})$, at zeros of $\psi(\mathbf{r})$ or at local defects in a system close to a QCP is a natural consequence of competing order parameters [10–12]. Demler has shown that in the core of a vortex, $\phi(\mathbf{r})$ can be finite and extend over a length scale much larger than the core radius, ξ. If the B phase of $CeCoIn_5$ is a true FFLO state, then it is reasonable to expect that $\phi(\mathbf{r})$ may extend over length scales much larger than the effective width of the normal domain walls. Indeed, our measurements indicate that $\phi(\mathbf{r})$ remains finite everywhere. We cannot, however, determine the topology of the zeros of $\psi(\mathbf{r})$, nor whether it changes sign across a domain wall, as predicted in an FFLO state.

The In(1) and Co sites also reveal information about the magnetic structure in these systems. In $CeCo(In_{1-x}Cd_x)_5$ and the field induced phase of $CeCoIn_5$, these resonances are unaffected by the onset of antiferromagnetism. The symmetry of In(1) and Co sites in the unit cell (Wyckoff positions 1*c* and 1*b*, respectively) are such that the hyperfine field cancels. Surprisingly, however, the hyperfine field at In(1) site, $\mathbf{H}_{int}(1)$, does not vanish in $CeRhIn_5$ at ambient pressure [13]. Under hydrostatic pressure $\mathbf{H}_{int}(1)$ gradually decreases to zero, even though the ordered moment remains unchanged [14]. One way to understand these results is that the transferred hyperfine coupling is anisotropic in the $CeRhIn_5$ [7]. This coupling is probably mediated by orbital overlaps between 4f orbitals of the Ce and 5p orbitals of the In(1). The dHvA frequencies and effective masses of the $CeRhIn_5$ evolve with pressure; so it is plausible that the orbital overlaps, and hence the anisotropy of the hyperfine couplings, may change with pressure. Thus the difference in $\mathbf{H}_{int}(1)$ in the $CeRhIn_5$ and the $CeCo(In_{1-x}Cd_x)_5$ may reflect the degree to which 4f electrons are fully hybridized.

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