SC-to-AFM transition in CeCo(In$_{1-x}$Cd$_x$)$_5$: de Haas-van Alphen Measurements


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Abstract. The results of de-Haas-van-Alphen (dHvA) measurements on Cd doped CeCoIn$_5$ in the high magnetic field paramagnetic phase are reported. The replacement of trivalent In by divalent Cd is known to induce an antiferromagnetic order coexisting with superconductivity in this heavy fermion superconductor. We find a small but systematic change in the dHvA frequencies with Cd doping, reflecting the chemical potential shift due to the removal of conduction electrons. The frequencies and effective masses are close to those found in the undoped compound. We observe no abrupt change in the electron Fermi surface (FS) volume in the high field paramagnetic phase for $x > x_c$ corresponding to the onset of antiferromagnetic ordering at zero magnetic field in CeCo(In$_{1-x}$Cd$_x$)$_5$. Our results show that no significant change of the Fermi surface volume occurs to cause the AFM onset inside the SC state in CeCoIn$_5$ with Cd doping. Only slight changes causing the Band 15 electron FS to be more cylindrical are observed making nesting more likely.

1. Introduction

CeCoIn$_5$ is a heavy fermion superconductor in which Cooper pairs are formed out of a non-Fermi liquid metallic state. The ability to grow sizable, high quality, single crystals enables detailed investigations of the effect of chemical doping in this and other 115 compounds. While Sn substitution for In has been found to suppress $T_c$ without revealing any incipient magnetism$^1$, Cd replacing In induces an antiferromagnetic ground state in CeCoIn$_5$$^2$. Because Sn and Cd are next to In in the periodic table, their substitutions for In result in electron and hole doping, respectively. The effect of Cd doping is quite unusual in that it takes a very small amount of Cd in CeCoIn$_5$ to induce a change in the ground state from paramagnetic to antiferromagnetic (AFM) that can be reversed with the application of pressure$^2$. How Cd induces long range AFM order with a large ordered magnetic moment$^3$ (0.7µB per Ce) in CeCoIn$_5$ remains an open question. One possible mechanism is the formation of antiferromagnetic droplets at the Cd sites, as was inferred from NMR measurements$^3$. Long range AFM order occurs once the density of such droplets reaches a percolation threshold for nearest neighbor interactions. However, the density of Cd necessary to induce ordering is well below the percolation threshold. Thus the ordering at such a small Cd concentration requires interactions having a longer interaction range. Since the ordered moments are likely local moments on the Ce sites,
one way to account for the reversibility of Cd doping with pressure is for the change in carrier density and disorder caused by Cd substitution to affect the valence of nearby Ce atoms. Application of pressure to metals with localized f-electrons (corresponding to Ce$^{3+}$) tends to increase their hybridization with the conduction band and promote a mixed valence regime. Another possibility is that the AFM state is due to a Fermi Surface (FS) instability, which is the well known explanation in the case of elemental Cr. Recent neutron scattering results on Cd doped CeCoIn$_5$[4] have demonstrated that the AFM ordering has a wavevector, $Q$, of (1/2,1/2,1/2) suggesting that if the AFM is nesting-driven, the nesting wavevector is commensurate with the lattice. This is a plausible, but unusual, situation that occurs, for example, in Mn doped Cr[5]. One possibility in the case of Cd doping is that in the doping region intermediate between local and itinerant magnetism the magnetic ordering is driven by the unscreened component of the spin. The improved near-nesting with Cd doping could lower the energy necessary to open the gap for itinerant electrons at the magnetic Brillouin Zone boundary and this would enabling the long-range order to appear. One of the most surprising features of this ordered state is that the ordering wavevector[6] coincides with the wavevector at which an inelastic neutron scattering resonance[7] is observed in the superconducting state of pure CeCoIn$_5$. The origin of this resonance has been attributed to AFM magnons[8]. A similar AFM state can be induced with Rh substitution for Co in CeCoIn$_5$ for Rh concentrations greater than $\cong$ 25%. Here, the AFM wavevector is identical to that found for Cd doped CeCoIn$_5$, which coexists with superconductivity for Rh concentrations of less than $\cong$ 60%. For larger Rh concentrations the AFM wavevector becomes (1/2,1/2,$\cong$ 0.3) and superconductivity is suppressed. The change in wavevector and the loss of superconductivity at Rh concentrations above 60% suggest that the AFM states in Rh (for $x_{Rh} < 0.6$) and Cd doped CeCoIn$_5$ have a different mechanism from the incommensurate antiferromagnetic order found in CeRhIn$_5$. In order to understand more fully the superconducting state in CeCoIn$_5$, the relevance of the coexisting AFM, the character of the AFM state will require further investigations. In this paper, we report one such investigation by specifically exploring the evolution of the FS in the high magnetic field paramagnetic phase of CeCoIn$_5$ as a function of Cd doping via de Haas van Alphen (dHvA) measurements.

**II. Experimental Details**

The single crystals of CeCo(In$_{1-x}$Cd$_x$)$_5$ used in these measurements were grown from high purity starting materials, as described elsewhere[9]. The values of $x$ we give are the nominal flux concentrations and it has been found that the actual Cd concentration is consistently 10% of the nominal flux concentration across the entire range of flux compositions[9]. The lattice parameters were determined by using both powder and single crystal X-ray diffraction and the lattice constants were found to change by < 0.1 % across the doping range indicating that FS volume changes due to changes in the BZ dimensions are insignificant. Extensive measurements previously have shown that the Cd atoms are distributed uniformly throughout the samples at each concentration[10].

The evolution of the FS of the same crystals used for the single crystal X-ray diffraction measurements was investigated via the dHvA oscillations measured using a torque magnetometer[11]. Single crystals were mounted on a Cu-Be cantilever in a dilution refrigerator equipped with a sample rotator. The dHvA measurements were made at the National High Magnetic Field Laboratory in magnetic fields of up to 45 T, and for temperatures down to 0.05 K. The susceptibility of single crystals of CeCo(In$_{1-x}$Cd$_x$)$_5$ was measured at $H = 0.1$ T applied perpendicular to [001] axis for temperatures ranging from 1.8 to 400 K using a Superconducting Quantum Interference Device (SQUID) magnetometer. For the dHvA data analysis it is assumed that $H \cdot B$ and no demagnetizing factor corrections were made, since we estimate from the susceptibility measurements the magnetization of CeCo(In$_{1-x}$Cd$_x$)$_5$ to be $\cong$0.4% of the applied field. The
in-plane susceptibility (measured down to 1.8 K) is extrapolated with a power law fit to \( \chi_\perp = 0.0143 \) emu/mol to \( T = 50 \) mK in 5% Cd, which corresponds to \( \chi_{||} = 0.0286 \) emu/mol for \( H \parallel [001] \) (with a magnetic anisotropy of 2) and to a volume magnetization of \( 4 M = 0.0126 \) T at an applied field of 35 T.

III. Experimental Results

Figure 2 shows the magnetization as a function of applied field for the 5% and 7.5% Cd samples. In the 7.5% sample both the superconducting transition \( H_{C2} \) and the AFM to paramagnetic transition, \( H_m \), can be seen, while in the 5% sample only \( H_{C2} \) is observed. All of the dHvA signals reported here are from the paramagnetic phase at fields greater than either \( H_{C2} \) or \( H_m \). In Fig. 3 the dHvA signal in the 5% sample at base temperature is shown. Fourier transforms (FTs) of data such as this were performed to obtain the dHvA frequencies (proportional to \( 1/H \)) in all of the samples. The angular dependence of the frequencies is shown in Figs. 4 and 5 for the 2.5% and 7% Cd samples. The three frequencies, \( F_3, F_4, \) and \( F_5 \) arise from the Band 15 undulating cylindrical shaped electron sheets located at the corners of the BZ, see Fig. 7.

As can be seen from the data from the two Cd concentrations, the Band 15 surface becomes more cylindrical as the amount of Cd is increased causing the \( F_3, F_4, \) and \( F_5 \) frequencies to merge into one frequency. The frequencies for \( H \) along [001] are given in the Table below.

III. Conclusions

We find a small but systematic change in the dHvA frequencies with Cd doping in CeCo(In, Cd), reflecting the chemical potential shift due to the removal of conduction electrons. The frequencies and effective masses of the Band 15 electron portions of the FS are close to those found in the undoped compound (see Fig. 7)[12]. We note that in the doped compounds we only observe signals from the Band 15 electron sheet. We observe no abrupt change in the Band 15 electron Fermi surface volume.
in the high field paramagnetic state for \( x > x_c \), corresponding to the onset of antiferromagnetic ordering that occurs at \( H = 0 \) in \( \text{CeCo(In}_{1-x}\text{Cd}_x)\). Our results show that no significant change of the Fermi surface volume in the paramagnetic state occurs to cause the AFM onset inside the SC state in CeCoIn\(_5\) with Cd doping. Only slight changes causing the Band 15 electron FS to be more cylindrical (note that a \( 1/\cos \theta \) angular dependence indicates a cylindrical surface) are observed making nesting more likely. In another paper [13] we show that the changes that occur with Cd doping in

Table of dHvA frequencies and \( m^* \)

<table>
<thead>
<tr>
<th>( F(T) )</th>
<th>( m^* )</th>
<th>( F(T) )</th>
<th>( m^* )</th>
<th>( F(T) )</th>
<th>( m^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( F_1 )</td>
<td>267</td>
<td>215</td>
<td>13.8±3.0</td>
<td>350</td>
<td></td>
</tr>
<tr>
<td>( F_2 )</td>
<td>411</td>
<td>8.7±2.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( F_3 )</td>
<td>4566</td>
<td>11.2±0.2</td>
<td>4390</td>
<td>8.5±0.8</td>
<td>4347</td>
</tr>
<tr>
<td>( F_4 )</td>
<td>5161</td>
<td>20.3±0.7</td>
<td>4928</td>
<td>14.9±1.4</td>
<td></td>
</tr>
<tr>
<td>( F_5 )</td>
<td>5401</td>
<td>5353</td>
<td>16.7±2.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( F_6 )</td>
<td>7535</td>
<td>8651</td>
<td>15.2±---</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

LaCoIn\(_5\) and CeCoIn\(_5\) compounds suggests that Cd does not induce any valence fluctuation on neighboring Ce atoms, as originally assumed in order to explain the reversible tuning. The high field Fermi surface in antiferromagnetic \( \text{CeCo(In}_{1-x}\text{Cd}_x)\) is large, i.e. of a size similar to the FS of CeCoIn\(_5\), in contrast to the small, f electron localized, FS found in CeRhIn\(_5\), suggesting a separate mechanism for the antiferromagnetism. It follows that the commensurate AFM order in \( \text{CeCo(In}_{1-x}\text{Cd}_x)\) could be due to an itinerant, SDW-type, mechanism which relies on FS nesting. This would make the current situation similar to the example of SDW ordering that is commensurate with the underlying lattice in the elemental antiferromagnet Cr which, although incommensurate when pure, evolves to a commensurate state with small amounts of Mn doping[5].

References