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Anomalous effect of doping on the superconducting state of $CeCoIn_5$ in high magnetic fields

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We investigated the effect of electron and hole doping on the high-field low-temperature superconducting state in $CeCoIn_5$ by measuring specific heat of $CeCo(In_{1-x}M_x)_5$ with M=Sn, Cd and Hg and x up to 0.33% at temperatures down to 0.1 K and fields up to 14 T. Although both Cd- and Hg-doping (hole-doping) suppresses the zero-field T_c monotonically, H_{c2} increases with small amounts of doping and has a maximum around x=0.2% (M=Cd). On the other hand, with Sn-doping (electron-doping) both zero-field T_c and H_{c2} decrease monotonically. The critical temperature for the high-field low-temperature superconducting state correlates with H_{c2} and T_c, which we interpret in support of the superconducting origin of this state.

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Magnetic field's coupling to spins of the electrons of a superconducting pair (Cooper pair) destroys superconductivity via the Pauli limiting effect [1]. In the normal state, magnetic field splits the spin-up and spin-down states (Zeeman splitting), and the system can lower its free energy by preferentially populating the lower energy level (spin-up band). This also gives rise to the Pauli susceptibility. On the other hand, the energy of a spin-singlet SC state can not be affected by Zeeman effect, because electrons pair up with opposite spins, i.e. there is an equal number of superconducting spin-up and spin-down electrons. When the energy gain from Zeeman effect in the normal state exceeds the SC condensation energy with increasing magnetic field, superconductivity is destroyed at a Pauli-limiting field H_P. The field's coupling to the orbital motion of the superconducting electrons also can destroy superconductivity at a characteristic field H^0_{c2}. For strongly Pauli limited superconductors (when the Maki parameter $\alpha = \sqrt{2}H^0_{c2}/H_P$ is large) a number of peculiar properties were anticipated theoretically in 1960's. The SC transition was predicted to become first order at high fields, when T_c is suppressed below $0.56T_c(H{=}0)$ [2, 3]. The Zeeman effect was also expected to lead to a spatially inhomogenious SC state, stable at high fields and low temperatures, proposed by Fulde and Ferrell [4] and Larkin and Ovchinnikov [5] (FFLO).

The discovery of a strongly Pauli-limited superconductor $CeCoIn_5$ has triggered numerous investigations of its peculiar SC properties [6, 7]. $CeCoIn_5$ is SC below T_c=2.25 K with a large Maki parameter α, which is anisotropic with respect to the direction of the magnetic field and ranges between 3.5 ($H \parallel [001]$) and 4.5 ($H \parallel [100]$) [8]. $CeCoIn_5$ shows a first order SC transition in a bulk superconductor via specific heat and magnetization anomalies at high fields [8, 9], in accord with the above mentioned theoretical expectations. When a high magnetic field close to H_{c2} is applied within the basal (a-b) plane of the tetragonal crystal structure of $CeCoIn_5$, specific heat shows an additional anomaly within the superconducting state into a High Field Low Temperature (HFLT) phase, originally proposed to be a realization of a long-searched-for FFLO state [10, 11].

In this Rapid Communication, we show that very small amounts of both magnetic (Cd and Hg) and non-magnetic (Sn) doping have dramatic effects on both gross superconducting properties (H_{c2}) and the evolution and stability of the HFLT phase in $CeCoIn_5$. Our data, in particular the broadening of the specific heat anomaly, suggest that the underlying origins of this phase are indeed of the FFLO nature.

The HFLT transition itself has been confirmed by penetration depth [12], thermal conductivity [13], ultrasound velocity [14], magnetostriction [15], and nuclear magnetic resonance (NMR) [16–18] measurements (for a recent review, see Ref. [19]). Importantly, NMR investigation [20] revealed a long range antiferromagnetic (AFM)order within the HFLT state [20]. Further, magnetic Bragg peaks with an incommensurate propagation vector $\vec{Q} = (0.44, 0.44, 0.50)$, was found within the HFLT SC phase by neutron scattering [21], and that phase was named by the authors Q-phase, in reference to the observed $\vec{Q}$. The presence of the long range AFM order revealed by the NMR and neutron scattering experiments gave rise to an alternative possibility of magnetism being the driving force of the phase transition into the HFLT state. Our present doping studies test this hypothesis.

Figure 1 shows evolution of T_c and T_N for Cd-doped and T_c for Sn-doped [22] $CeCoIn_5$ at zero-field. Both Cd- and Sn-doping suppress the SC state monotonically and Cd-doping induces AFM order above 0.5 %. Hg-doping has essentially the same effect as Cd-doping. [23]

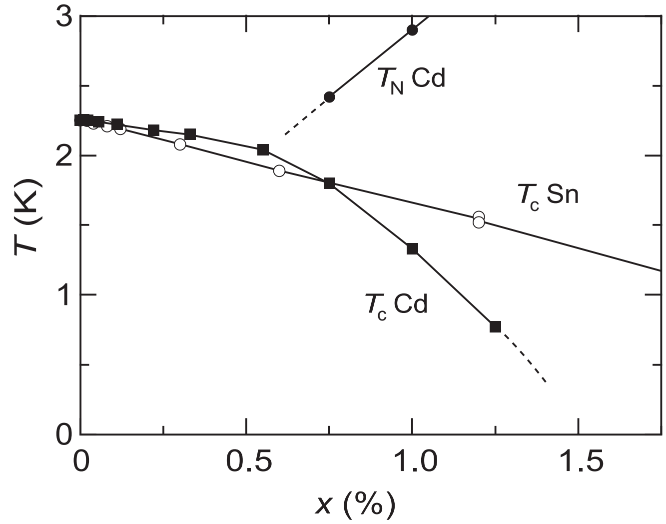

FIG. 1: Zero-field superconducting transition temperature, T_c, of $CeCo(In_{1-x}Cd_x)_5$ and $CeCo(In_{1-x}Sn_x)_5$ [22] as a function of concentration, together with antiferromagnetic transition temperature, T_N, of $CeCo(In_{1-x}Cd_x)_5$.

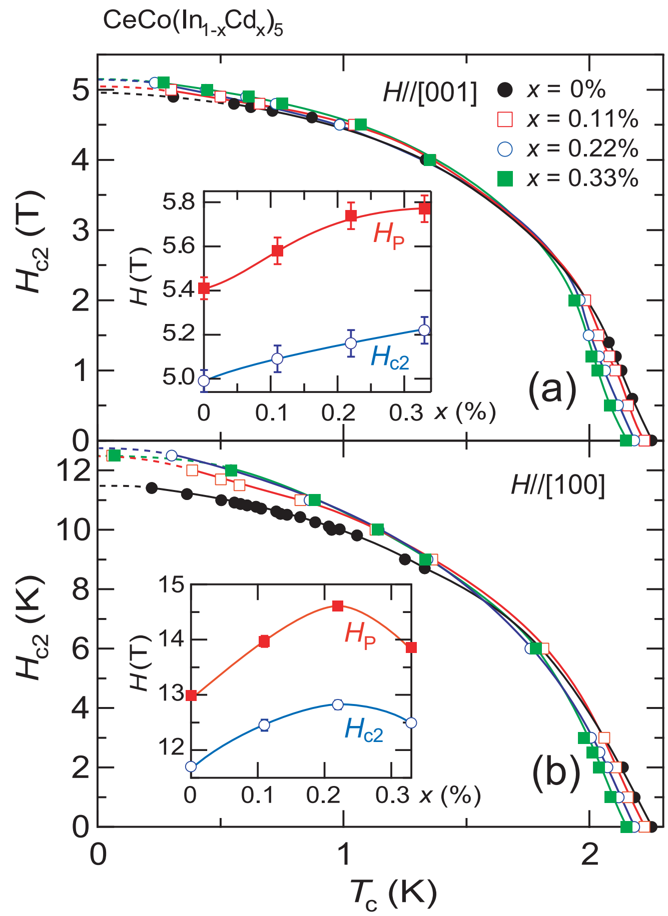

FIG. 2: (Color online) H-T phase diagrams of $CeCo(In_{1-x}Cd_x)_5$ with x=0, 0.11, 0.22 and 0.33 % for H ∥[001] (a) and H ∥[100] (b). Solid lines are guides to eye. Insets: Experimental upper critical field H_{c2} and deduced Pauli-limiting field H_P as a function of Cd-doping concentration.

Since Cd-doping induces AFM-order, one would naively expect it to also stabilize the HFLT phase with its long range AFM order. However, the HFLT phase was found to be extremely fragile with respect to Cd and Hg impurities [24]. Only 0.05% of Hg on In-sites destroys the HFLT phase. Furthermore, Fig. 1 shows that the AFM order speeds up the suppression of T_c with a steeper slope above 0.5 %, implying a competition between the two states, while the magnetically ordered HFLT phase appears to need superconductivity for its existence.

The AFM order in $CeCoIn_5$, induced by Cd doping above 0.5 %, potentially may be related to the AFM order within the HFLT phase. To untangle the effect of suppression of the HFLT phase at low impurities concentration from that of inducing long range magnetic order at higher levels of Hg and Cd doping, it is important to perform complimentary investigations of the response of the HFLT phase to a non-magnetism-inducing impurity. Doping with Sn achieves just that, as Sn-doping was shown to suppress SC without driving the system into an AFM ordered state [25].

Specific heat measurements were performed on single plate-like samples with a typical weight of 1-3 mg. Initial sample characterization via micro-probe analysis, using wavelength dispersive spectroscopy, showed uniform distribution of the dopants. Specific heat was measured in a dilution refrigerator with a superconducting magnet, employing the quasi-adiabatic heat pulse method.

We first address the effects of doping on the gross features of the phase diagram of $CeCoIn_5$. In zero field (see Fig. 1) the low doping behavior for Cd and Sn doping is similar, reflecting impurity pair breaking effects. At high field the response of $CeCoIn_5$ to small amounts of hole and electron doping is strikingly different. Figure 2 shows H-T phase diagrams of Cd-doped $CeCoIn_5$ for the field along [001] and [100]. H_{c2} increases for both H ∥ [001] and [100]. The zero-field T_c monotonically decreases with Cd doping, and increase of H_{c2} leads to the crossing of SC phase boundaries for the undoped and Cd-doped $CeCoIn_5$ (Fig. 2b), indicating more stable SC state at high fields in the Cd-doped compounds. Extrapolated H_{c2} to zero temperature is shown in the insets. H_{c2} exhibits a maximum at x = 0.22 % for H ∥[100], while it increases monotonically up to x = 0.33 % for H ∥[001].

The orbital limiting field, $H^0_{c2} = 0.7T_c dH_{c2}/dT|_{T=T_c}$, for H ∥ [100] is 37, 37, 36 and 39±1 T for x=0, 0.11, 0.22 and 0.33 %, respectively. It is rather independent of Cd concentration, and can not be responsible for the increase in $H_{c2}(0)$. The Pauli-limiting field is estimated from the orbital limiting field and experimental H_{c2}, using the results of the numerical calculations in Ref [26]. The resulting H_P is shown in the insets of Fig. 2, and it follows the same

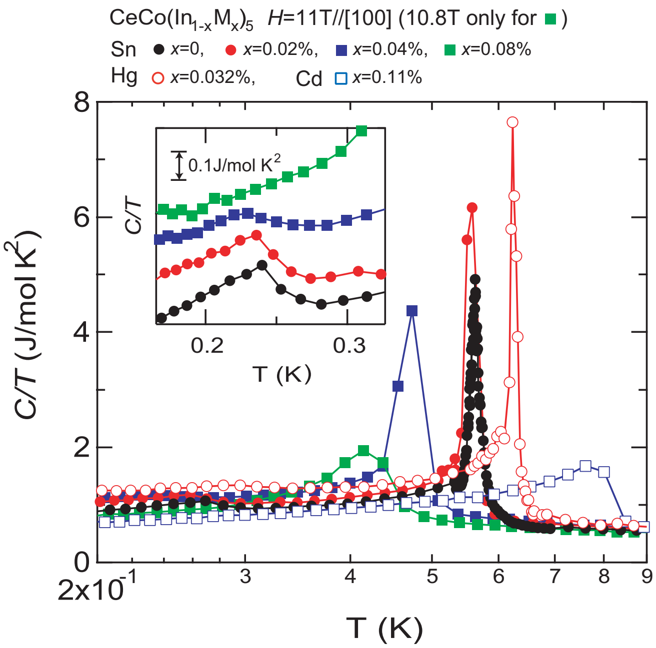

FIG. 3: (Color online) Specific heat divided by temperature, C/T, for $CeCo(In_{1-x}T_x)_5$ (T=Sn, Hg and Cd) as a function of temperature at 11 T and at 10.8 T for T=Sn and x=0.08 %. Inset: Expanded plot of C/T around HFLT phase transition for T=Sn. Data are shifted vertically for clarity.

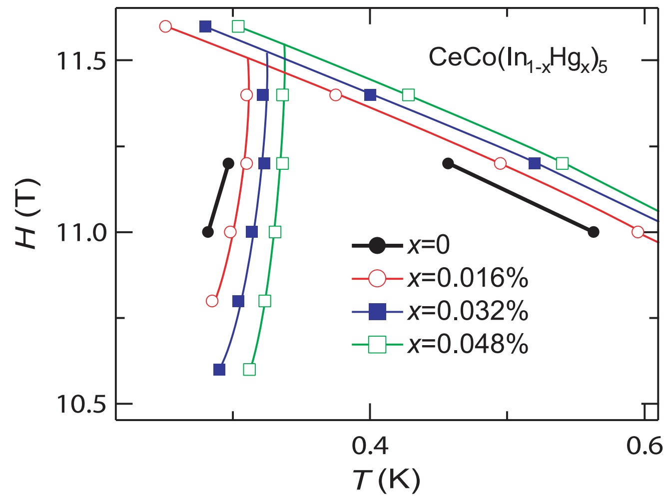

FIG. 4: (Color online) H-T phase diagram of $CeCo(In_{1-x}Hg_x)_5$ for the HFLT phase region of SC state. Solid lines are guides to eye.

trend as H_{c2}. The Pauli-limiting field $H_P = \sqrt{2}\Delta/g\mu_B$, where Δ is the SC energy gap at zero temperature and g is the gyromagnetic ratio. Assuming that Δ is proportional to zero-field T_c, the relative change of g as a function of doping is $g(x)/g(0)=[T_c(x)/H_P(x)]/[Tc(0)/H_P(0)]$ (not shown). As H_P exhibits a maximum at 0.22 % for $H \parallel [100]$, g has a minimum, with the reduction of 14 % from $g(0)$. The g-factor as a function of pressure was found previously to increase from 0.632 at ambient pressure to 0.6365 at 0.45 GPa (a change of less then a percent), and then drop to 0.554 at 1.34 GPa. [27, 28]. In the earlier zero-field study [29], it was concluded that Cd-doping acts as negative pressure, based on the finding that Cd-doping suppress SC and induces AFM order, while pressure suppresses AFM and can drive AFM ordered $CeCo(In_{1-x}Cd_x)_5$ back to non-magnetic SC ground state. Here we note that Cd doping of 0.22 % corresponds to an effective negative pressure of only 0.28 GPa [29]. Therefore, the relative change of the g-factor with Cd doping is at least an order of magnitude greater than can be accounted for by the pressure effect alone.

We now address the effect of small levels of electron and hole doping on the HFLT phase. We will show that Sn (electron) doping has both similar and opposite effects, to those of Cd and Hg (hole) doping, on the superconducting state of $CeCoIn_5$. However, the data are consistent and can be understood within one picture.

Figure 3 shows specific heat divided by temperature, C/T, of $CeCo(In_{1-x}T_x)_5$ (T=Cd, Hg and Sn) at 11 T, and for T=Sn at x=0.08 % at 10.8 T, for doping concentrations up to 0.11 %. The HFLT-phase anomaly, displayed in the inset of Fig. 3, broadens with increasing concentration (as in the case of Cd and Hg doping) and disappears at x=0.08 %. The critical Sn-concentration to destroy the specific heat anomaly associated with the HFLT phase, 0.08 % at most, is similar to 0.05 % for Hg doping. These results show that the HFLT phase is extremely fragile with respect to impurities, regardless of whether the same impurities at higher doping levels stabilize AFM ground state or not. The inter-impurity distance at the critical doping concentrations of 0.05% (the characteristic length scale of the HFLT state) is ∼40 Å, which is comparable to the SC coherence length (70 Å), supporting the SC origin of the HFLT state, instead of an AFM one.

The effects on the normal-SC phase boundary at high fields for the two types of dopants in the low-doping regime are dramatically different. For Sn-doping, $T_c(H{=}11\,T)$ shifts monotonically to lower temperature, contrary to the effect of Cd and Hg doping discussed above. Electron doping (with Sn) suppresses H_{c2} roughly by the same amount that Cd and Hg increase it for the same amount of doping (see Fig. 5), supporting the picture of charge doping (Fermi surface effect) being the dominant driver of the change of H_{c2}.

The response to charge doping of the superconducting critical field correlates well with that of the HFLT phase. A recent study has shown that T_{HFLT} increases slightly with Hg-doping, while the specific heat anomaly associated with the transition into the HFLT state broadens and is quickly destroyed [24]. Fig. 4 shows that both HFLT and the homogenous SC states expand in H-T phase space with Hg doping. For non-magnetic Sn-doping, on the other hand, both SC and HFLT states

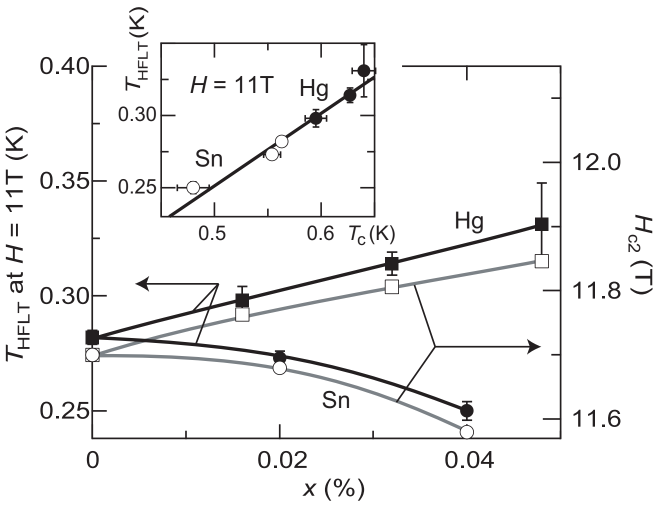

FIG. 5: Doping dependence of HFLT-phase transition temperature T_{HFLT} at 11 T for $CeCo(In_{1-x}Hg_x)_5$ and $CeCo(In_{1-x}Sn_x)_5$ (solid symbols, left axis), together with superconducting upper critical field, H_{c2}, of $CeCo(In_{1-x}Hg_x)_5$ and $CeCo(In_{1-x}Sn_x)_5$ (open symbols, right axis). (■, □ - $CeCo(In_{1-x}Hg_x)_5$; ●,○ - $CeCo(In_{1-x}Sn_x)_5$. Solid lines are guides to eye. Inset: T_{HFLT} vs T_c at 11 T with doping concentration x as an implicit parameter. Data for both M=Hg and Sn are plotted. Solid straight line goes through the origin.

contract, and both T_c and T_{HFLT} at 11 T reduce with doping, as shown in Fig. 3.

The correlation between H_{c2} and T_{HFLT} is clearly seen in Fig. 5, where these quantities are plotted as functions of both Sn- and Hg-doping. Both H_{c2} and T_{HFLT} increase with Hg-doping, while they decrease with Sn-doping. We also plot T_{HFLT} against T_c at 11 T in the inset of Fig. 5, with the doping concentration x as an implicit parameter. Remarkably, a line, which goes through the origin, gives a good fit (solid line).

The observed strong correlation between $T_{HFLT}(H)$ and $T_c(H)$ should not be surprising, since the magnetic HFLT phase only exists within the superconducting state, and therefore its properties should be strongly linked to the superconducting properties of $CeCoIn_5$. The broadening and suppression of the anomaly, however, is much more significant with respect to the possible origin of the HFLT phase. Recent theoretical work [30] on the effect of impurities on both FFLO state and the magnetic-origin scenario [21] showed that the FFLO state is much more fragile to impurities, a result of the softness of the nodal planes due to a strong Pauli depairing effect in $CeCoIn_5$. Contrary, for a low temperature state with magnetism as a dominant driving force, one would expect suppression of T_c to zero, without observed strong broadening and suppression on the specific heat anomaly itself [30].

In conclusion, we have studied the effect of magnetic (Cd and Hg) and non-magnetic (Sn) doping effects on the high field - low temperature superconducting HFLT phase of $CeCoIn_5$ by means of specific heat measurements. H_{c2} increases anomalously with hole doping, while it decreases with electron doping. The change of H_{c2} is much greater than can be expected to be due to an effective negative pressure effect, and is dominantly an effect of carrier doping on the Fermi surface. The HFLT phase is extremely sensitive to Sn-doping with a critical concentration of 0.08 %, at most, and is similar to 0.05 % for Hg-doping. We note here that, despite the existence of the long range magnetic order in the HFLT state, it is destroyed by both the AFM- and non-AFM-inducing impurities. The scaling of T_{HFLT} against T_c at 11 T indicates a close correlation between the HFLT and the homogenous SC states. More significantly, the broadening of the HFLT specific heat anomaly and its suppression with minute amount of impurities supports the FFLO origin of the HFLT state.

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