Effects of La substitution on the superconducting state of $CeCoIn_5$

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We report the effects of La substitution on the superconducting state of the heavy-fermion superconductor $CeCoIn_5$, as seen in transport and magnetization measurements. As opposed to the case of conventional superconductors, pair breaking by nonmagnetic La results in the depression of T_c and indicates a strong gap anisotropy. The upper critical field H_{c2} values decrease with increased La concentration, but the critical field anisotropy, $\gamma = H^a_{c2}/H^c_{c2}$, does not change in $Ce_{1-x}La_xCoIn_5$ $(x = 0-0.15)$. The electronic system is in the clean limit for all values of x.

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I. INTRODUCTION

The study of heavy fermion superconductors over the past two decades has shown an abundance of new phenomena that are associated with Cooper pair formation.[1] In particular, the competition between magnetic and superconducting interactions among electrons near the Fermi surface has given rise to unconventional superconductivity[2,3] and raised speculations that the spin pairing might be mediated by magnetic interaction.[4] Research in the field has been associated with difficulties in sample preparation, sample to sample variation, experimental conditions and ultimately in the number of examples where relevant physical phenomena can be observed in a clean form. The recently discovered $CeMIn_5$ family (M = Ir, Rh, Co) of heavy-fermion superconductors encapsulates many important aspects of physics in this class of materials. $CeRhIn_5$ (Ref. 5) superconducts under applied pressures above 17 kbar with T_c around 2 K whereas $CeIrIn_5$ (Ref. 6) and $CeCoIn_5$ (Ref. 7) are ambient pressure superconductors. $CeCoIn_5$ offers a clean example of ambient pressure heavy-fermion superconductivity with a remarkably high $T_c = 2.3$ K. The intriguing properties of $CeCoIn_5$ led to the speculation that it may exhibit d-wave superconductivity,[8–10] and the Fulde-Ferrel-Larkin-Ovchinnikov state in high magnetic fields.[11] In order to have more insight into the nature of $CeCoIn_5$ we perturbed its superconducting state by substituting La onto the Ce site. For the purpose of comparing the influence of magnetic and nonmagnetic pair breaking on T_c suppression, we also substituted 5% of Nd on Ce site. We find that the anisotropy in the upper critical field does not change in the whole concentration range and that the decrease of T_c with increased La doping cannot be explained solely with the pressure effects due to the unit-cell expansion. In addition, our results present evidence for an anisotropic order parameter in $CeCoIn_5$.

II. EXPERIMENT

Single crystals of $Ce_{1-x}La_xCoIn_5$ were grown by the self-flux method in a manner previously described.[7] Purity of starting elements (in atomic percent) was: Ce: 99.86, La: 99.8, Nd: 96.9, Co: 99.99, In: 99.999. Crystals grew as thin plates with the c axis perpendicular to the plate. Removal of excess In from the surface was performed by etching in concentrated HCl for several hours followed by a thorough rinsing in ethanol. All samples obtained with this process showed no signs of free In contamination. Powder x-ray patterns showed that the samples crystallized in $HoCoGa_5$ structure without any additional peaks introduced by La alloying. In addition, magnetization measurements provided a more sensitive test of the possible presence of magnetically ordered second phases. Both as grown and etched samples showed no sign of an antiferromagnetic transition of $CeIn_3$. Electrical contacts were made with Epotek-H20E silver epoxy. In-plane resistivity was measured in Quantum Design MPMS and PPMS measurement systems from 0.35 to 300 K and in fields up to 90 kOe applied parallel and perpendicular to the c axis. There is an uncertainty in the nominal resistivity values associated with sample geometry due to the uneven surfaces of etched samples. We measured several samples for each concentration in order to reduce the measurement error, which allowed us to estimate uncertainties in nominal values as well. The dimensions of the samples were measured by a high-precision optical microscope with 10 μm resolution and average values are presented. Randomly chosen samples within each batch had no difference in their $R(T)$ curves. Magnetization measurements were performed in MPMS-7 Quantum Design magnetometer in the magnetic field of 10 kOe, applied parallel and perpendicular to c axis.

III. RESULTS

The results of powder x-ray diffraction measurement taken at room temperature are summarized in Table I and shown in Fig. 1, together with the unit-cell volume of $LaCoIn_5$. As expected, the La-doped samples have a larger unit-cell volume. The volume increase in the concentration range $x = 0-0.175$ is consistent with the expansion of the unit cell as La substitutes Ce in accordance with Vegard's law.

Figure 2 shows the magnetic susceptibility for $Ce_{0.95}Nd_{0.05}CoIn_5$, $Ce_{0.85}La_{0.15}CoIn_5$, and $CeCoIn_5$, taken in the applied field of 10 kOe. In the whole temperature range above T_c, the substitution of magnetic Ce^{3+} by nonmagnetic La^{3+} reduces the susceptibility values in the La-doped sample when compared with undoped $CeCoIn_5$. Comparison of high-temperature moments through Curie-Weiss analysis

TABLE I. Properties of $Ce_{1-x}La_xCoIn_5$ doping series: T_c, lattice parameters, unit-cell volumes, $H'_{c2}(T)$, calculated $H_{c2o}(0)$ from WHH model and approximate chemical pressure $P_{chemical}$ due to La alloying. Final row: properties of $Ce_{0.95}Nd_{0.05}CoIn_5$.

x	T_c (K)	a(Å)(±0.007 Å)	c(Å)(±0.007 Å)	V(Å)^3	$-\frac{dH_{c2}}{dT}$ (kOe/K)	H_{c2o} (0)(kOe)	$P_{chemical}$ (kbar)
0	2.3	4.613	7.542	160.49±0.4	240(a),110(c)	370(a),170(c)	0
0.02	2.0	4.613	7.551	160.65±0.53	170±23(a),86±3(c)	235(a),119(c)	−0.6
0.05	1.68	4.614	7.551	160.76±0.2	190±19(a),95±7(c)	214(a),107(c)	−1.1
0.075	1.31	4.615	7.551	160.86±0.23	207±27(a),98±2(c)	188(a),89(c)	−1.5
0.1	1.22	4.615	7.557	160.97±0.35			−2
0.125	0.86	4.623	7.546	161.27±0.1			−3.1
0.15	0.78	4.619	7.563	161.35±0.4	236±27(a),103±2(c)	127(a),55(c)	−3.5
0.175	-	4.619	7.567	161.48±0.1			
1.0	-	4.638	7.612	163.74±0.1			
0.05(Nd)	2.0	4.601	7.546	160.37±0.3			0.5

of the polycrystalline susceptibility average at high temperatures shows that approximately 14% of the Ce ions were substituted with La. Low-temperature magnetic susceptibility of $Ce_{0.85}La_{0.15}CoIn_5$ does not reveal any difference in Curie tail from the pure material, thus ruling out Kondo-hole interpretation of La dilution (Fig. 2 inset).[12] We also see broadening of the plateaulike feature in χ_c in $Ce_{0.85}La_{0.15}CoIn_5$ ascribed[9] to thermal depopulation of Ce $4f$ levels. On the other hand, Nd impurities contribute to a pronounced Curie tail at low temperatures. Subtraction of magnetic susceptibility of $CeCoIn_5$ from $Ce_{0.95}Nd_{0.05}CoIn_5$ in the normal state below 10 K is consistent with approximately 8% of Nd^{3+} paramagnetic moment, a result close to nominal stoichiometric value and within rough approximation of our analysis.

Temperature-dependent electrical resistivities normalized to their value at 300 K for $Ce_{1-x}La_xCoIn_5$ and $Ce_{0.95}Nd_{0.05}CoIn_5$ are presented in Fig. 3(a). There are several key features to notice. Resistivities of all samples are weakly temperature dependent at high temperatures, and they pass through a maximum as the temperature is decreased. This behavior is traditionally interpreted as a crossover from incoherent Kondo scattering to coherent Bloch states of heavy electrons in the Kondo lattice. In the case of $CeCoIn_5$ this drop, at least partially, could be attributed to a depopulation of crystalline electric field levels.[13] We observe a decrease of T_{max} for higher La concentrations [Fig. 3(a) inset]. At low temperatures, there is a clear suppression of T_c as more Ce ions are replaced by La [Fig. 3(b)]. The increase of the normal state residual resistivity ρ_0 is probably due to a disorder that contributes to an increased conduction electron scattering. On the other hand, the resistive transition width sharpens with La alloying. It is interesting to note that $Ce_{1-x}La_xCoIn_5$ is not in the well-defined Fermi liquid regime above T_c. The $\rho(T)$ curves above T_c do not show signs of T^2 dependence, as it has been reported for $CeCu_2Si_2$.[14] Depression of T_c in $CeCoIn_5$ seems to scale with the ρ_0 values for both magnetic and nonmagnetic dopants, as seen by a comparison of the $\rho(T)$ data of $Ce_{0.95}Nd_{0.05}CoIn_5$ with $Ce_{0.98}La_{0.02}CoIn_5$.

Figure 4 shows the anisotropic upper critical field for $Ce_{1-x}La_xCoIn_5$, normalized to the transition temperature in zero field for each value of x (values for $x=0$ were taken from previous report, Ref. 15). The H_{c2} data were determined as a midpoint between onset of superconductivity and zero resistivity from $\rho(T)$ curves at a constant field and $\rho(H)$ curves at a constant temperature. Adding La impurities results in a depression of H_{c2}. The anisotropy γ

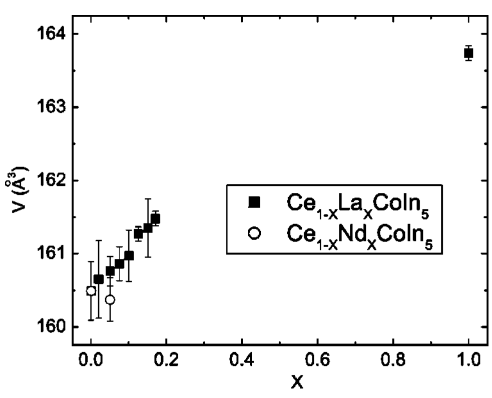

FIG. 1. Unit-cell volume of $Ce_{1-x}La_xCoIn_5$ ($x=0$–0.175,1) shown together with unit-cell volume of $Ce_{0.95}Nd_{0.05}CoIn_5$.

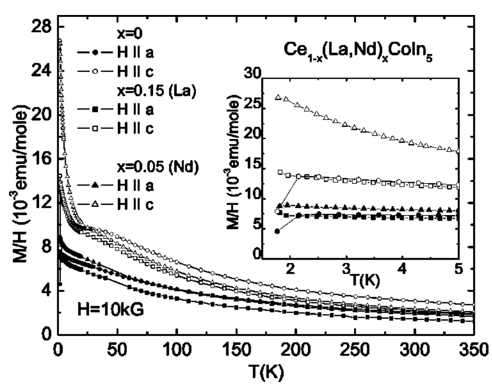

FIG. 2. Magnetic susceptibility of $Ce_{0.85}La_{0.15}CoIn_5$, $Ce_{0.95}Nd_{0.05}CoIn_5$, and $CeCoIn_5$. Low-temperature susceptibility (inset) shows pronounced Curie tail with 5% of Nd substitution but no difference for 15% La substitution.

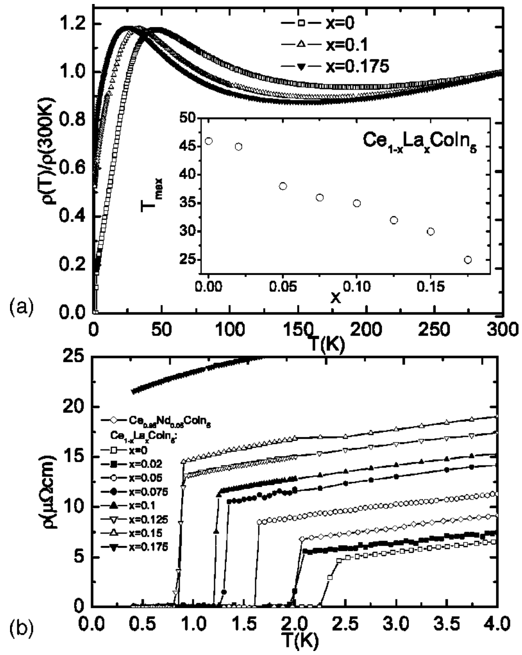

FIG. 3. (a) Electrical resistivity ρ normalized to its value at 300 K vs temperature for $Ce_{1-x}La_xCoIn_5$ for $x=0$, 0.1 and 0.175. T_{max} is shifted to lower temperatures with increased La substitution (inset). (b) Low-temperature resistivity shows depression of T_c and increase in ρ_0.

$=H^a_{c2}/H^c_{c2}$, however, remains at the same value of $\gamma\approx 2$ (inset in Fig. 4). Uncertainty in our estimate of γ decreases for higher-field data, away from $H=0$ transition ($T/T_c\approx 1$).

Assuming that the Fermi surface properties of the doped material do not change substantially in the dilute La limit,[16] it is reasonable to assume an inverse proportionality between ρ and l, and therefore values of l_0 could be estimated from ρ_0 for the whole doping series ($l_0=A/\rho_0$) using the value of constant A from reported l_0 and ρ_0 values for a pure material.[17] We obtain $l_0\approx 540$Å for $CeCoIn_5$ without La impurities. Figure 5 shows the ratio of mean free path l_0 to

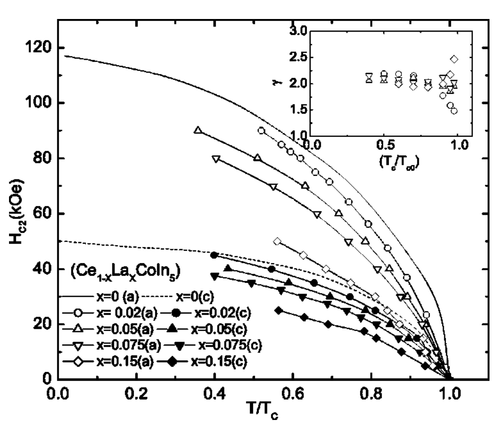

FIG. 4. Anisotropy in the upper critical field H_{c2} for $Ce_{1-x}La_xCoIn_5$ ($x=0$–0.15). Inset shows value of $\gamma=H^a_{c2}/H^c_{c2}$ vs T_c/T_c ($H=0$) for varous La concentrations: $x=0.02$ (circles), $x=0.05$ (up triangles), $x=0.075$ (down triangles), $x=0.15$ (diamonds).

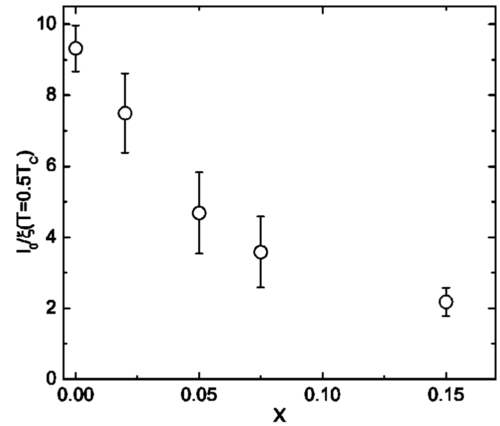

FIG. 5. Ratio of mean free path (l) to coherence length (ξ) for $Ce_{1-x}La_xCoIn_5$. Electronic system is in the clean limit already at $T=T_c/2$ for La concentrations $x=0$–0.15.

in-plane superconducting coherence length ξ $[\xi^2(T)=\Phi_0/2\pi H_{c2}(T)]$ for $Ce_{1-x}La_xCoIn_5$ obtained at $T=T_c/2$. In the whole doping range the electronic system is in the clean limit which could explain a nearly constant value of $\gamma=H^a_{c2}/H^c_{c2}$.

A comparison of the effects of La substitution on T_c in $CeCoIn_5$ and $CeCu_{2.2}Si_2$ is shown in Fig. 6.[18] Doping results in a depression of T_c in both cases but $CeCoIn_5$ is more robust to pair breaking arising from La impurities. The initial rate of T_c suppression is smaller than the rate seen in $CeCu_{2.2}Si_2$: $[(0.056T_c)/(1\%$ of La substitution) in $CeCoIn_5$ vs $(0.085T_c)/(1\%$ of La substitution in $CeCu_{2.2}Si_2)]$. La doping in $CeCoIn_5$ is associated with only a modest increase in nominal residual resistivity values ρ_0, shown in the Fig. 6 inset. The ρ_0 values for $x=0$ (~ 5 $\mu\Omega$ cm) in our experiment are in between values reported previously in the literature [3.1 $\mu\Omega$ cm (Ref. 17) and ~ 7 $\mu\Omega$ cm (Ref. 19)].

IV. DISCUSSION AND CONCLUSIONS

The slope of H_{c2} vs T curve at T_c can be used to estimate zero-temperature orbital critical field $H_{c2o}(0)$ using the

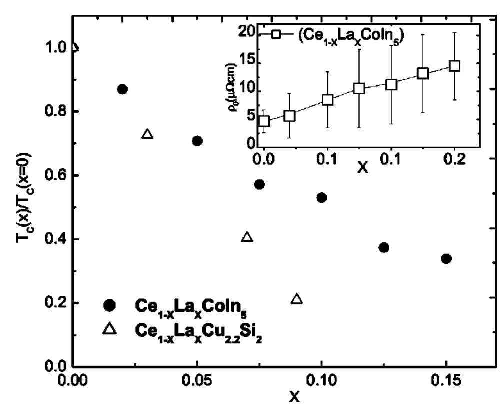

FIG. 6. Comparison of La doping on T_c of $CeCoIn_5$ (this work) and $CeCu_{2.2}Si_2$ (Ref. 19). Inset shows increase in ρ_0 of $Ce_{1-x}La_xCoIn_5$ caused by La substitution.

weak-coupling formula for conventional superconductors in Werthamer-Helfand-Hohenberg (WHH) model: $H_{c2o}(0) \approx 0.7H'_{c2}(T_c)T_c$.[20] Table I shows estimates of H'_{c2} near T_c for doped samples, together with the previously reported value for $x=0$ for both crystalline directions.[21] All investigated samples have high initial slopes, as expected in the case of heavy-fermion superconductors.[22,23] Values of $H_{c2o}(0)$ decrease with introduction of La impurities (Table I). The paramagnetic limiting field $H_p(0)=\Delta_0/\mu_B\sqrt{2}$ (where Δ_0 is the energy gap at $T=0$ and μ_B is the Bohr magneton) for pure $CeCoIn_5$ ($T_c=2.3$ K) is well below the orbital critical field $H_{c2o}(0)$ for either *s*-wave ($\Delta_0=3.52k_BT_c$),[24] or *d*-wave pairing state ($\Delta_0=2.14k_BT_c$),[25] and our results indicate that this unusual situation is valid for the investigated La-doping range. We note that the experimental values of the upper critical field for $Ce_{1-x}La_xCoIn_5$ ($x=0-0.15$) samples are most likely below the values obtained by applying the WHH model (Table I), probably due to the polarization of the magnetic sublattice due to an enhanced internal field along both crystalline axes.

It has recently been reported that T_c in $CeCoIn_5$ increases under applied pressure.[19] Negative chemical pressure should cause some decrease in T_c. In the lack of better approximation, we take bulk modulus of $CeCoIn_5$ to be the same as the one for $CeIn_3$ (650 kbar),[26] and we calculate approximate chemical pressure ($P_{chemical}$) for each La concentration using $-V_0\partial P/\partial V\approx 650$ kbar. The results are shown in Table I. The depression of T_c occurs at the rate of $\partial T_c/\partial P\approx -0.43$ K/kbar—a slope that is an order of magnitude larger than reported for the increase of T_c under hydrostatic pressure. An order of magnitude difference from pure pressure effect on T_c is likely to exceed error in the estimation of bulk modulus, and therefore points to the conclusion that the pair-breaking mechanisms that enter through disorder due to La alloying and increased scattering of Cooper pairs are dominant in $CeCoIn_5$. In contrast to the conventional superconductors where nonmagnetic impurities have small effect on T_c, Cooper pairs formed in $CeCoIn_5$ are rather sensitive to La doping: 2% of La depresses T_c to the same value as $\sim$5% of Nd.

The T_c suppression induced by the nonmagnetic La substitution in $Ce_{1-x}La_xCoIn_5$ is reminiscent of the pair-breaking effect by magnetic impurities.[27] Although various factors may suppress T_c (an anisotropic scattering, for example),[28] we focus here on the scenario of $CeCoIn_5$ having an anisotropic gap $\Delta(\vec{k}_F)$ at the Fermi surface. This scenario is quite likely to occur given the unconventional nature in many heavy-fermion materials.

It is known[29] that if Δ depends on the position at the Fermi surface, the critical temperature is suppressed by nonmagnetic scattering according to

$$\ln\frac{T_{c0}}{T_c}=\alpha\left[\psi\left(\frac{1+\mu}{2}\right)-\psi\left(\frac{1}{2}\right)\right],\quad \mu=\frac{\hbar}{2\pi k_B\tau T_c}. \qquad (1)$$

Here T_{c0} is the critical temperature of the material in the absence of all scattering, τ is the scattering time by nonmagnetic impurities, and $\alpha=1-\langle\Delta\rangle^2/\langle\Delta^2\rangle$ characterizes the gap anisotropy, $\langle\cdots\rangle$ stands for averaging over Fermi surface, and ψ is the digamma function. For a weak gap anisotropy, this result is due to Hohenberg,[29] see also later publications.[30,31] It can be shown that in fact Eq. (1) holds for an arbitrary gap anisotropy.[32] For isotropic Δ, $\alpha=0$, and we come to Anderson's theorem: $T_c=T_{c0}$. For pure *d*-wave order parameter, $\langle\Delta\rangle=0$, and Eq. (1) describes the *d*-pair breaking by nonmagnetic scattering (which differs from the Abrikosov-Gor'kov result only by a factor of 2 in the definition of the parameter $\mu_m=\hbar/\pi k_BT_c\tau_m$).

To analyze the $T_c(x)$ data shown in Fig. 6, one has to relate x to the scattering time τ, a nontrivial connection. We avoid this difficulty by assuming that the residual resistivity ρ_0 is proportional to $1/\tau$. Further, we exclude parameter T_{c0} from Eq. (1) by writing it for two values of x and subtracting the results,

$$\ln\frac{T_2}{T_1}=\alpha\left[\psi\left(\frac{1+\mu_1}{2}\right)-\psi\left(\frac{1+\mu_2}{2}\right)\right],\quad \mu_{1,2}=\beta\frac{\rho_{1,2}}{T_{1,2}}, \qquad (2)$$

where $T_{1,2}=T_c(x_{1,2})$ and β is a constant to be determined. Writing this equation for two different pairs $x_{1,2}$ one can determine the unknown α and β. This procedure yields values scattered around $\alpha=0.5$ and $\beta=0.2$ K/$\mu\Omega$ cm.

Hence, we find $\alpha=\langle\Delta\rangle^2/\langle\Delta^2\rangle\approx 0.5$ which implies a strongly anisotropic gap. Knowing the value of β we can estimate the scattering time using measured resistivities; for $x=0$ we obtain $\tau=\hbar/2\pi k_B\beta\rho\approx 1.3\times 10^{-12}$ s. With the electronic-specific-heat coefficient[17] $\gamma=290$ mJ/mol K^2 we roughly estimate the Fermi velocity $v_F=\pi k_B/e\sqrt{\gamma\tau\rho_0}\approx 2\times 10^6$ cm/s. This would correspond to the mean-free path $l\approx 260$ Å, a value smaller than expected but within a factor of 2 of our determination of mean-free path which is reasonable given the assumptions of average Fermi velocity and isotropic scattering.

In summary, diamagnetic pair-breaking effect in $CeCoIn_5$ is consistent with the picture of a strongly anisotropic order parameter. Anisotropy in the upper critical field $\gamma=H^a_{c2}/H^c_{c2}$ does not change for $x=(0-0.15)$ in $Ce_{1-x}La_xCoIn_5$, indicating an electronic system in the clean limit.

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