Strong suppression of superconductivity by divalent ytterbium Kondo holes in $CeCoIn_5$

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To study the nature of partially substituted Yb ions in a Ce-based Kondo lattice, we fabricated high quality $Ce_{1-x}Yb_xCoIn_5$ epitaxial thin films using molecular beam epitaxy. We find that the Yb substitution leads to a linear decrease of the unit cell volume—indicating that Yb ions are divalent, forming Kondo holes in $Ce_{1-x}Yb_xCoIn_5$—and leads to a strong suppression of the superconductivity and Kondo coherence. These results, combined with measurements of the Hall effect, indicate that Yb ions act as nonmagnetic impurity scatterers in the coherent Kondo lattice without serious suppression of the antiferromagnetic fluctuations. These results are in stark contrast to previous studies performed using bulk single crystals, which claim the importance of valence fluctuations of Yb ions. The present work also highlights the suitability of epitaxial films in the study of the impurity effect on the Kondo lattice.

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The impurity effect has been one of the key issues in the study of unconventional superconductors including cuprates,[1] iron-pnictides,[2] and heavy fermion compounds.[3–9] In these strongly correlated electron systems, the superconducting state emerges from the competition between different phases, such as a magnetically ordered phase. Understanding the impurity effect is important because it is intimately related to the superconducting pairing interaction and competing electronic correlations. In heavy fermion compounds, it was generally found that the substitution of magnetic sites with nonmagnetic impurities yields a substantial reduction in both the onset of coherence temperature T_{coh} at which the formation of heavy fermions occurs, and the superconducting transition temperature T_c. Recently, however, a highly unusual impurity effect has been reported in the heavy fermion compound $CeCoIn_5$, which is in sharp contrast to the above tendency.[6–8]

$CeCoIn_5$ with tetragonal symmetry[10] is a very clean superconductor with a large mean free path and is known to exhibit a plethora of fascinating electronic properties. The unconventional superconductivity with d-wave symmetry appears in the vicinity of the magnetic ordered phase.[11–13] The normal state exhibits pronounced non-Fermi-liquid behaviors, which are believed to be due to the proximity of an antiferromagnetic (AF) quantum critical point.[14,15] Recent studies revealed that the superconductivity occurs in a two-dimensional Kondo lattice composed of a square lattice of Ce atoms.[16] Thus $CeCoIn_5$ may provide an ideal playground for investigating the impurity effect on the unconventional superconductivity in strongly correlated f-electron systems.

The impurity effect on $CeCoIn_5$ has been studied extensively by investigating $Ce_{1-x}R_xCoIn_5$ (R = rare earth) and $CeCo(In_{1-y}M_y)_5$ (M = Hg, Sn, and Cd).[3,4,17,18] Two notable features have been pointed out. First, nonmagnetic impurities locally suppress superconductivity, generating an inhomogeneous electronic "Swiss cheese,"similar to that observed in cuprates.[5] Second, several groups have reported more striking and unexpected results in the case of Yb substitution on the Ce site: the superconductivity in $Ce_{1-x}Yb_xCoIn_5$ is robust against Yb substitution, and T_c decreases linearly with x towards 0 K as $x \to 1$, while T_{coh} remains essentially unaffected with Yb doping.[6–8] $YbCoIn_5$ is a conventional nonmagnetic metal with no superconducting transition down to 20 mK, indicating that Yb is close to divalent and is in the nonmagnetic closed shell $4f^{14}$ configuration.[16,19] Therefore, the robustness of the superconductivity upon Yb substitution in $CeCoIn_5$ with unconventional pairing symmetry is extraordinary. Moreover, according to Refs. 6 and 7, the lattice parameters remain roughly constant as x changes until the phase separation occurs at $x \sim 0.8$, indicating a violation of Vegard's law.

These results of Yb substitution are markedly different from other R-substituted $CeCoIn_5$, in which R substitutions suppress the superconductivity at approximately $x = 0.2$–0.3.[17,20] These anomalous behaviors have been discussed in the light of the valence fluctuations of Yb ions.[6] However, the electronic state of Yb in the Kondo lattice of $CeCoIn_5$ is poorly understood.

To obtain further insight into the nature of Yb ions in the Ce-based Kondo lattice, we fabricated $Ce_{1-x}Yb_xCoIn_5$ thin films using molecular beam epitaxy (MBE). This technique, recently advanced by our group, enables the growth of uniform heavy fermion thin films, as demonstrated by the successful growths of $CeIn_3/LaIn_3$[21] and superconducting $CeCoIn_5/YbCoIn_5$[16] superlattices with one-unit-cell thick $CeIn_3$ and $CeCoIn_5$ layers, respectively. Since the epitaxial growth occurs in nonequilibrium conditions at temperatures much lower than the temperature used for single-crystal growth by the flux method, MBE is suitable for the preparation of systems with homogeneously distributed Ce and Yb atoms. Moreover, owing to the ability to evaporate Ce and Yb atoms simultaneously, the ratio of Ce to Yb can be controlled precisely. Using our thin films, we find that the Yb substitution leads to a strong suppression of T_c, in accordance with Abrikosov-Gorkov (AG) theory. These results are in sharp contrast to previous studies performed using bulk single crystals.[5–8]

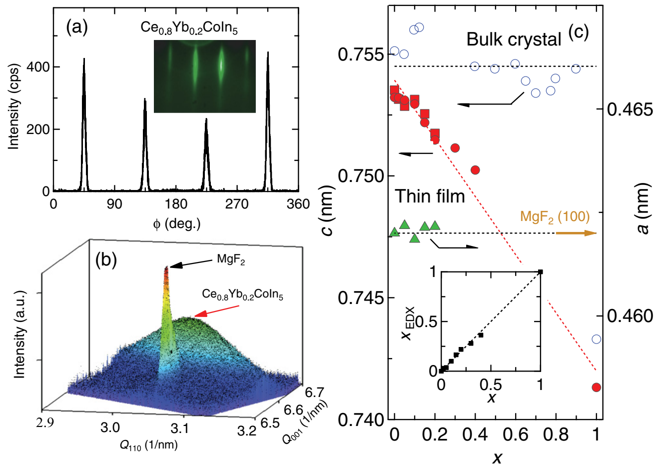

FIG. 1. (Color online) (a) X-ray diffraction ϕ-scan data of the (115) peak for $Ce_{0.8}Yb_{0.2}CoIn_5$. Inset: Streak patterns of the RHEED image during the crystal growth. (b) X-ray reciprocal lattice mapping for $Ce_{0.8}Yb_{0.2}CoIn_5$ near the (115) peak. (c) Lattice parameters a (filled green triangles) and c (filled red squares and circles) for $Ce_{1-x}Yb_xCoIn_5$ films. Here c is determined by reciprocal mapping (squares) and θ–2θ scan (circles). Open blue circles represent c of bulk crystals reported in Ref. 6. The dashed lines are guides for the eyes. The inset shows x determined by EDX, x_{EDX}, vs x determined by the evaporation ratio of Ce to Yb atoms.

The c-axis oriented epitaxial $Ce_{1-x}Yb_xCoIn_5$ films were grown by MBE. The (001) surface of MgF_2 with rutile structure ($a = 0.462$ nm, $c = 0.305$ nm) was used as a substrate. The substrate temperature was kept at 530–550 °C, depending on the composition. Each metal element was evaporated from an individually controlled Knudsen cell. The typical deposition rate was 0.01–0.02 nm/s. The area and thickness of the films were 5.0×10.0 mm^2 and 120 nm, respectively. The epitaxial growth of each layer with atomic flatness was carefully checked by monitoring the streak patterns of the reflection high-energy electron diffraction (RHEED) during the deposition [see inset of Fig. 1(a)]. The x-ray diffraction ϕ scan with fourfold peaks shown in Fig. 1(a) also indicates the epitaxial growth of the film, which was further confirmed by the x-ray reciprocal lattice mapping as shown in Fig. 1(b).[22] Surface roughness detected by atomic force microscopy is approximately 1 nm, which is less than 2 unit cells thick. The energy dispersive x-ray spectroscopy (EDX) analysis shows that the distribution of x is within 2%, demonstrating an excellent chemical homogeneity across the whole sample area. We could not grow the epitaxial films for $x > 0.4$ without buffer layers. To grow $YbCoIn_5$ epitaxial films, we used $CeIn_3$ as a buffer layer on the substrate. As shown in the inset of Fig. 1(c), the ratio of Ce to Yb of the measured films, determined with EDX, coincides well with the evaporation ratio of Ce to Yb atoms.

In our epitaxial thin films ($a = 0.462$ nm, $c = 0.753$ nm), the lattice parameter a is dictated by the substrate lattice parameter: the misfit strain slightly enlarges a from that of the bulk single crystal value ($a = 0.461$ nm, $c = 0.755$ nm). Figure 1(c) shows the doping evolution of the lattice parameters determined by x-ray diffraction. Because of the epitaxial strain effect, a is independent of x and coincides well with a of the MgF_2 substrate. On the other hand, c (and hence unit-cell volume) decreases linearly with increasing x and lies on the line connecting $x = 0$ and 1. According to the Vegard's law, the unit-cell volume should decrease linearly with Yb substitution, if there are no changes in the valence of Ce and Yb ions. Therefore the observed linear relation implies that the Ce and Yb ions in the present $Ce_{1-x}Yb_xCoIn_5$ films retain the valences of the end member compounds, i.e., the Yb ion is divalent in $Ce_{1-x}Yb_xCoIn_5$, forming nonmagnetic "Kondo holes" in the $4f^1$ lattice. This result is in sharp contrast to the results using bulk single crystals, in which c is x independent [open circles in Fig. 1(c)].[6,7]

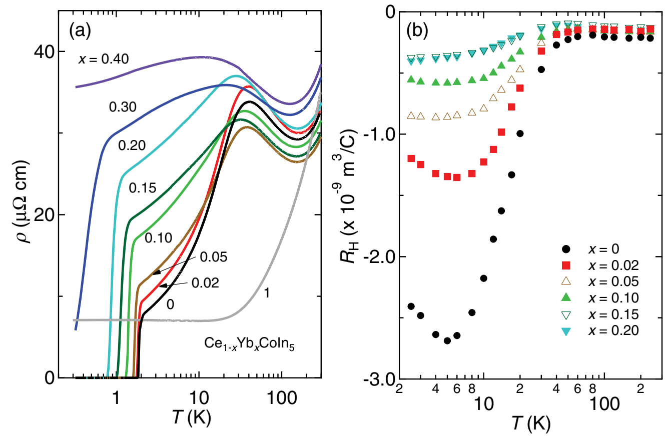

FIG. 2. (Color online) Temperature dependence of the (a) resistivity and (b) Hall coefficient for $Ce_{1-x}Yb_xCoIn_5$.

Figures 2(a) and 2(b) depict the temperature dependence of the in-plane resistivity ρ and Hall coefficient R_H for $\boldsymbol{H} \parallel c$, which is defined as the field derivative of the Hall resistivity at the zero-field limit. First we compare the transport properties of the pure $CeCoIn_5$ thin film with those of the single crystal. The temperature dependence of both the resistivity and Hall coefficient in the thin film are essentially the same as those in bulk single crystals.[15] Associated with an incoherent-coherent crossover, ρ shows a maximum at T_{coh} at around 40 K. The absolute value of ρ of the $CeCoIn_5$ thin film is close to that of the bulk single crystal. In particular, $\rho \simeq 6$ $\mu\Omega$ cm at the superconducting onset is close to the bulk single crystal value, which ranges from 3 to 8 $\mu\Omega$ cm.[3,23,24] The resistivity below T_{coh} in the bulk single crystal exhibits a T-linear dependence, which is a hallmark of non-Fermi-liquid behavior. In our thin film, the resistivity exhibits a T dependence with an exponent slightly below unity.

R_H is negative across the whole temperature range. The temperature dependence of R_H is closely correlated with that of the resistivity.[15,25] At high temperatures $T > T_{coh}$, R_H is nearly temperature independent. Below T_{coh}, R_H decreases rapidly as the temperature is lowered. Further reduction of the temperature increases R_H after showing a minimum at around 5 K. The magnitude of R_H above T_{coh} matches well with that of the bulk single crystal.[15] The enhancement of the absolute value of R_H below T_{coh} is nearly 15 times that at above T_{coh}, which is nearly half of that seen in the bulk single crystal. The superconducting transition temperature of the $CeCoIn_5$ thin

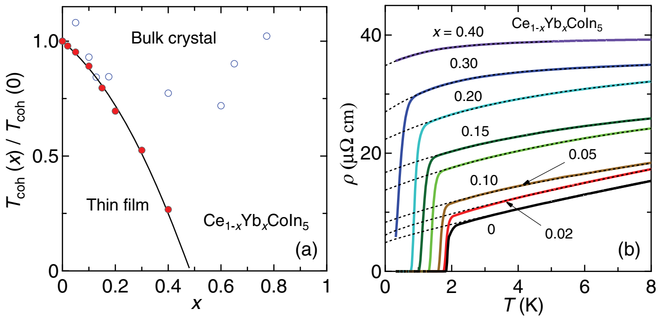

FIG. 3. (Color online) (a) x dependence of T_{coh} normalized by the value at $x = 0$ for thin films (filled red circles) and bulk crystals reported in Ref. 6 (open blue circles). (b) Temperature dependence of the resistivity at low temperatures. The dashed lines are the extrapolation above T_c.

film is 1.95 K, which is slightly lower than $T_c = 2.3$ K of the bulk single crystal. Since the ρ value at the superconducting onset (and residual resistivity which will be discussed later) in our thin film is comparable to the bulk single crystal value, this T_c reduction is likely to be due to the strain effect arising from the substrate. In fact, it has been reported that the negative pressure in $\mathrm{CeCoIn_5}$ reduces T_c.[26] Based on these results, we conclude that the quality of our epitaxial thin films grown by MBE is comparable to that of high quality bulk single crystals.

Next we discuss the effect of Yb substitution on the normal state transport properties. As shown in Fig. 2(a), a broad maximum of $\rho(T)$ associated with the formation of Kondo coherence is observed at $T_{\mathrm{coh}}(x)$ in Yb-substituted $\mathrm{CeCoIn_5}$. Figure 3(a) depicts the x dependence of T_{coh}, which is simply defined as the peak position of $\rho(T)$. Yb substitution seriously reduces the Kondo coherence and T_{coh} appears to go to zero at around $x = 0.5$. Since it is highly unlikely that the Fermi temperature is dramatically suppressed by Yb substitution, the observed reduction of T_{coh} arises from the destruction of the Kondo coherence by the Yb^{2+} Kondo holes in the Ce^{3+} f-electron lattice. In Yb-doped compounds $\rho(T)$ exhibits sub-T-linear behavior below T_{coh}. We fit the resistivity to a power law $\rho(T) = \rho_0 + AT^{\alpha}$ [Fig. 3(b)]. The resistivity can be fitted very well by this equation for $x = 0$ and 0.02 with the residual resistivity $\rho_0 = 4.8$ and 6.0 $\mu\Omega$ cm and $\alpha = 0.88$ and 0.80, respectively, in a wide temperature range below $\sim T_{\mathrm{coh}}/2$ down to T_c. For $x \geqslant 0.05$, we cannot fit $\rho(T)$ to this equation in a wide temperature range and therefore we estimate ρ_0 by a polynomial fitting from T_c to $T_{\mathrm{coh}}/2$ [see the dashed lines in Fig 3(b)]. As shown in Fig. 4(a), the residual resistivity nearly linearly increases with x. The present results are very different from those reported in bulk single crystals where T_{coh} is nearly x independent, as shown with open symbols in Fig. 3(a), and α barely changes with x up to $x = 0.1$.[6]

The measurements of the Hall effect provide vital information regarding the nature of the Yb ion in the Ce-Kondo lattice. As shown in Fig. 2(b), the Hall effect is very sensitive to the Yb substitution. At $T > T_{\mathrm{coh}}$, the absolute value of R_{H} is small and nearly temperature independent for all x. This is consistent with Vegard's law which suggests that Yb is divalent in a trivalent Ce lattice. Below T_{coh}, the enhancement of $|R_{\mathrm{H}}|$ is strongly suppressed by the Yb doping. The low-temperature enhancement of $|R_{\mathrm{H}}|$, which is significantly larger than $|1/ne|$ where n is the carrier concentration, has also been reported in other strongly correlated electron systems including cuprates,[27] iron-pnictides,[28] κ-(BEDT-TTF)$_2$Cu(NCS)$_2$,[29] and $\mathrm{V_{2-y}O_3}$.[30] It has been shown that the striking enhancement of $|R_{\mathrm{H}}|$ at low temperatures can be accounted for in terms of a "hot spot"on the Fermi surface (FS) and backflow effect, both of which originate from strong AF fluctuations.[31] The hot spot is a FS region where the electron lifetime is unusually short, which appears at the positions where the AF Brillouin zone boundary intersects with the FS. Since the hot spot does not contribute to electron transport, the effective carrier concentration is reduced, which results in the enhancement of $|R_{\mathrm{H}}|$ from $|1/ne|$.[32] However, this effect is not enough to explain the dramatic enhancement of $|R_{\mathrm{H}}|$ below T_{coh}. Another important effect is the backflow effect, which when accompanied by the anisotropic scattering, is called the "current vertex correction."[31] Backflow is a polarized current caused by a quasiparticle excitation inherent in the Fermi liquid. In the presence of the backflow effect, the total current is not parallel to the Fermi velocity, and then the Hall coefficient is seriously modified from the Boltzmann value derived from the curvature of the FS. It has been shown that the backflow effect can largely enhance $|R_{\mathrm{H}}|$ in CeMIn$_5$ (M = Co, Rh, and Ir).[15,25]

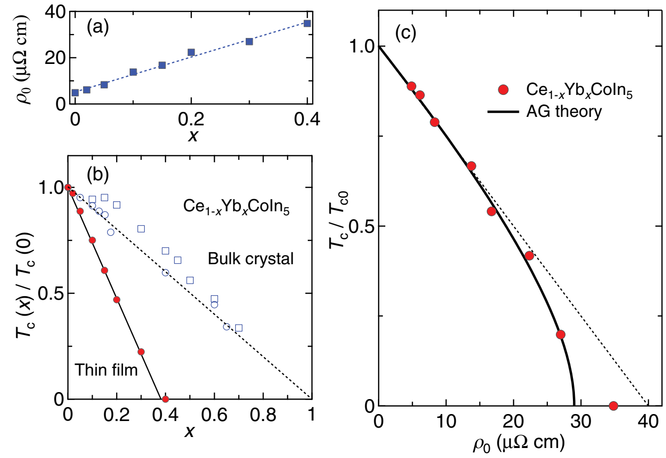

FIG. 4. (Color online) (a) x dependence of the residual resistivity. The dotted line is a guide for the eyes. (b) x dependence of the superconducting transition temperature normalized by the value at $x = 0$ for thin films (filled red circles) and bulk crystals [open blue squares (Ref. 6) and open circles (Ref. 8)]. (c) T_c/T_{c0} plotted as a function of residual resistivity. The dotted line is the linear extrapolation from the low ρ_0 region. The solid curve represents the result fitted by Abrikosov-Gorkov (AG) theory.

The observed strong suppression of $|R_{\mathrm{H}}|$ by Yb substitution indicates that the backflow effect is seriously reduced. There are two possible origins for this. The first is the reduction of AF fluctuations and the second is the increase of isotropic impurity scattering by nonmagnetic Yb ions. In the former case, since the system is tuned away from the quantum critical point, the T-linear resistivity is expected to change to T^2 dependence accompanied by a reduction of ρ_0.[15] However, such a trend is not observed here. Instead, the T-linear resistivity becomes sub-T-linear and ρ_0 increases with Yb substitution. This increase of ρ_0 is consistent with the increase of isotropic scattering. In addition, it has been shown that, in the presence of strong AF fluctuations, sub-T-linear dependence of $\rho(T)$ appears with increasing impurity scattering,[33] which is again consistent with the present results. Thus the normal state transport properties suggest that Yb ions are divalent and act as impurity scattering centers, rather than a suppressor of AF fluctuations.

Next we discuss the influence of Yb substitution on the superconducting properties. As shown in Fig. 3(b), sharp resistive transitions are observed for $x \leqslant 0.20$, but the transition is slightly broader for $x = 0.30$. No superconducting transition is observed for $x = 0.40$ down to 0.3 K. Figure 4(b) displays the x dependence of T_c, which is defined as the midpoint of the resistive transition, normalized by T_c of $x = 0$. T_c decreases linearly with x and goes to zero at around $x = 0.40$. We stress that the observed Yb-doping evolution of T_c is again in marked contrast to that reported in bulk single crystals shown by open symbols in Fig. 4(b). In Fig. 4(c) we plot T_c/T_{c0} (see below) against ρ_0. In the low-ρ_0 range, T_c/T_{c0} decreases linearly as shown by the dotted line. When ρ_0 exceeds $\sim 18\ \mu\Omega$ cm, T_c begins to deviate from linearity. We analyze this trend of T_c/T_{c0} in accordance with the AG theory of nonmagnetic impurity effects in d-wave superconductors.[34] The fitting parameters used to compare experiment to theory are the slope in the low-ρ_0 region and T_{c0}, where T_{c0} is the transition temperature with no pair breaking. The solid line in Fig. 4(c) is the result of fitting obtained by using $T_{c0} = 2.20$ K and the initial slope $-0.025\ (\mu\Omega\,\mathrm{cm})^{-1}$. As shown in Fig. 4(c), the suppression of T_c is well reproduced by the AG pair breaking curve in the whole ρ_0 range.

The fact that our data can be well described by the AG theory immediately implies that the Yb ions are randomly distributed with weak inter-ion correlation. Moreover, the "Swiss cheese"model[5] is inadequate because such a model will give a $T_c(\rho_0)$ curve that lies above the dotted line for high ρ_0 in Fig. 4(c).[35] The effect of Yb substitution on $CeCoIn_5$ bears striking resemblance to other rare-earth substitutions, although Yb is divalent while other rare earths are trivalent. This suggests that these ions act as impurity centers with unitary scattering, regardless of their valence. Furthermore, the valence fluctuations of Yb ions, if present at all, do not appear to play an important role on the physical properties in the normal state as well as the superconducting state. At the present stage, the essential difference of the Yb-substitution effect between thin films and bulk single crystals remains an open question. A possible origin for this may be that bulk crystals contain some regions where inter-Yb-ion correlation is important.[36] Further studies are required to clarify this issue.

To summarize, in high quality $Ce_{1-x}Yb_xCoIn_5$ epitaxial thin films, Yb ions are divalent and no signature of the valence fluctuation is observed. The Yb substitution leads to a strong suppression of the superconductivity and Kondo coherence. The suppression of T_c can be well described by AG theory. These results indicate that Kondo holes created by Yb ions act as nonmagnetic impurity scatterers in the Kondo lattice with no serious reduction of AF fluctuations. These results are in sharp contrast to previous studies performed using bulk single crystals, which claim the importance of valence fluctuations of Yb ions. The present work also emphasizes the uniqueness of the epitaxial films in the study of the impurity effect on the f-electron Kondo lattices, due to the prospect of preparing highly homogeneous doped systems.

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