Kondo effect in the charge-density-wave state of $Lu_{5-x}Yb_xIr_4Si_{10}$

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$Lu_5Ir_4Si_{10}$ is an established charge-density-wave (CDW) compound (T_{CDW}=83 K) which undergoes superconducting transition (SC) at low temperatures (T_C=3.9 K), leading to the coexistence of CDW and SC. On the other hand, $Yb_5Ir_4Si_{10}$ is a Kondo lattice compound that shows possible CDW transition below 48 K. In this study, we explore the influence of small Yb substitution for Lu in the series $Lu_{5-x}Yb_xIr_4Si_{10}$. Our observations indicate a decrease of both T_{CDW} and T_C with the increase of Yb content, with the evidence of a Kondo-like response below the CDW transition at low temperatures. Thus, one has a unique opportunity to study the Kondo effect in a CDW lattice in this system.

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Rare earth ternary silicides of the type $R_5Ir_4Si_{10}$, which adopt tetragonal structure, have led to a large number of studies due to their unusual ground states.[1–12] Even though the crystal structure does not have any explicit low dimensional features, many members of this family exhibit coexistence of charge-density-wave ordering (CDW) with superconductivity[1,4,5] (SC) or antiferromagnetism (AFM) depending on the nature of the rare earth ion.[2,3,6,7] Recently, it has been established that the Kondo lattice compound $Yb_5Ir_4Si_{10}$ ($Sc_5Co_4Si_{10}$ structure, $P4/mbm$) exhibits possible CDW below 48 K, with antiferromagnetic ordering of Yb^{3+} moments below 2.3 and 1.1 K, respectively.[8] Prior to the above result, investigations[1] revealed the coexistence of CDW transition (T_{CDW}=83 K) with superconductivity (T_{CDW}=3.9 K) in $Lu_5Ir_4Si_{10}$, and coexistence of AFM with CDW in $Er_5Ir_4Si_{10}$.[3] It has been stated in an earlier study[2,3] that the series of $R_5Ir_4Si_{10}$ compounds offers a new and extremely convenient paradigm with which to study strongly coupled CDW and coexisting superconductivity or magnetism.[6–12] Unlike in the established conventional CDW compounds, Fermi surface studies in $R_5Ir_4Si_{10}$ are not successful. However, the presence of a CDW gap seen[13] in $Lu_5Rh_4Si_{10}$ via optical conductivity measurements and the observation of nonlinear transport in incommensurate $Er_5Ir_4Si_{10}$ (Ref. 14) strongly suggest the presence of CDW in $R_5Ir_4Si_{10}$ series.

It is well known that a small amount of disorder suppresses the CDW with the concomitant increase of the SC transition temperature.[9] At the same time, it is also well established that the Kondo effect affects the superconductivity. In order to understand single ion Kondo behavior in superconducting CDW materials, we have investigated the effect of the influence of Yb substitution for Lu in the polycrystalline samples of the series $Lu_{5-x}Yb_xIr_4Si_{10}$. The series were prepared by first making $Lu_5Ir_4Si_{10}$ in an arc furnace. The constituent elements (Lu, Yb, and Ir: 99.9%; Si: 99.999%) were taken in stoichiometric proportion and arc-melted on a water cooled copper hearth under a Ti gettered, high purity argon atmosphere. The resulting ingot was flipped over and remelted 5–6 times to promote homogeneous mixing. The resulting compound was a homogeneous alloy which, again, was remelted with Yb with 5% excess to compensate for the weight loss. We have measured the resistivity, susceptibility, and heat capacity of a number of samples of this series. All the samples (x=0.0, 0.02, 0.05, and 0.07) have the expected $Sc_5Co_4Si_{10}$ structure as determined from powder x-ray diffraction. Our combined bulk studies reveal the decrease of CDW ordering due to the Yb substitution with a Kondo-like response at lower temperatures (<4 K). The superconducting transition temperature is also progressively reduced with the increase of x in $Lu_{5-x}Yb_xIr_4Si_{10}$. The structure of the unit cell of $R_5Ir_4Si_{10}$ is shown in Fig. 1. One of the interesting structural features of this compound is the absence of direct transition-transition metal contacts. The transition metal atoms are connected to each other either through a R or Si atom. One can observe that R has three inequivalent sites.

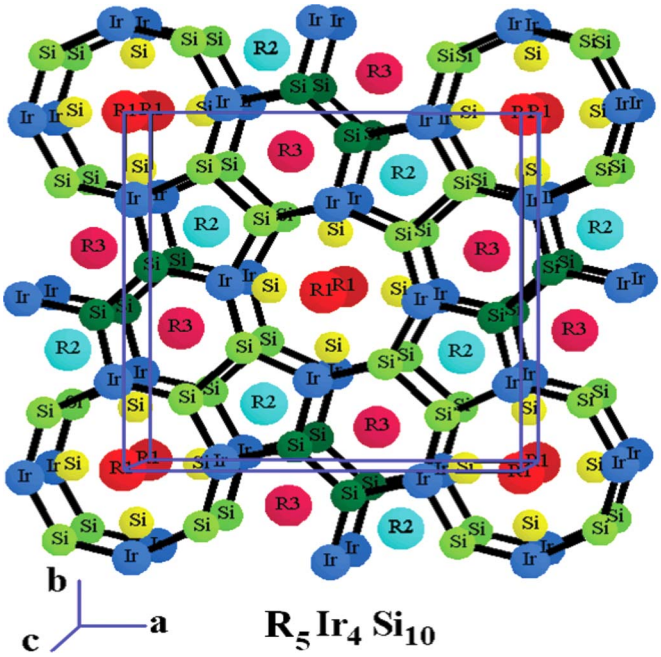

FIG. 1. (Color online) The crystal structure of $R_5Ir_4Si_{10}$ which consists of 2 f.u. with 38 atoms in the unit cell. The unit cell has three types of R atoms with position symmetry 2a, 4h, and 4h three types of Si atoms with position symmetry 4g, 8i, and 8j; and one type of Ir atom with position symmetry 8i. The R1 site (2a symmetry) atoms can be visualized to be forming linear chains along the c axis.

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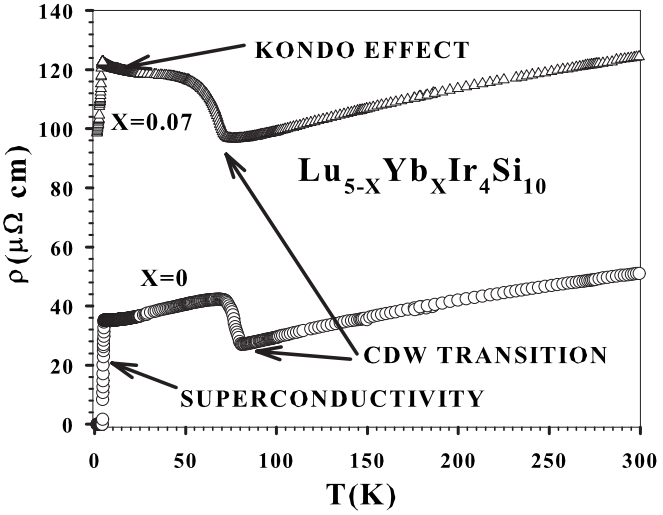

FIG. 2. The temperature vs resistivity $[\rho(T)]$ dependence of $Lu_{5-x}Yb_xIr_4Si_{10}$ samples with $x=0$ and $x=0.07$. The CDW and SC transitions are shown. Presence of possible Kondo-like behavior for $x=0.07$ is also indicated.

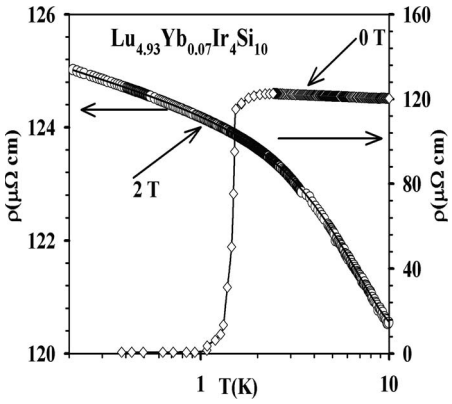

FIG. 3. The low temperature resistivity $[\rho(T)]$ dependence of $Lu_{5-x}Yb_xIr_4Si_{10}$ samples with $x=0.07$ at zero applied magnetic field (Y1 axis, right hand side) and the same sample at 2 T (Y axis, left hand side) are shown. The superconductivity at 1.5 K in the absence of field is quenched at 2 T, and the Kondo-like behavior is evident. The solid line in the $\rho(T)$ data for 2 T is a fit to the equation described in the text. The line connecting the superconducting transition to the right is a guide for the eye.

The Ir and Si atoms form planar nets of pentagons and hexagons which are stacked parallel to the basal plane and connected along the c axis via Ir-Si-Ir zigzag chains. The pentagon and hexagon layers are separated by layers of R. The distances between Ir and Si as well that of Ir and Ir are short, indicating strong covalent interaction. Preliminary band structure studies suggest that zigzag chains and linear chains (formed by rare earth atoms occupying R1 sites with position symmetry $2a$) probably play a crucial role in the formation of the CDW state in these compounds. Figure 2 shows the typical temperature vs resistivity $[\rho(T)]$ dependence of $Lu_{5-x}Yb_xIr_4Si_{10}$ samples with $x=0$ and $x=0.07$. The sample with $x=0$ displays the CDW transition below 83 K and superconductivity at 3.9 K, which are in agreement with earlier data.[1] In the case $x=0.07$, one observes the presence of the CDW and superconductivity at lower temperatures compared to those of the $x=0$ sample. The resistivities of other Yb substituted samples have similar temperature dependence as the $x=0.07$ sample, with T_{CDW} and T_C decreasing with increase of x in $Lu_{5-x}Yb_xIr_4Si_{10}$. In the case of $x=0.07$, both CDW and SC are decreased (T_{CDW} from 83 to 69 K, and T_C from 3.9 to 1.5 K). Though the amount of substitution is significant (>1 at. % of Lu by Yb), the reduction in the transition temperatures is not large. In the case of CDW, the small reduction in T_{CDW} with substitution of the rare earth ion has been a feature of this series (possibly the chain sites responsible for the CDW are not directly affected by the substitution).[9,15,16] In the case of SC, the Kondo impurity ($T_K \approx 5$ K) should have reduced T_C considerably since values of T_K and T_C are quite close. In general, magnetic impurities in the Kondo limit cause spin flip scattering of conduction electrons through antiferromagnetic coupling. This scattering breaks the copper pairs in conventional superconductors and even suppresses the superconductivity in the presence of small concentration of magnetic impurities.[17] This suppression of superconductivity takes place due to the reduction of the electronic density of states at the Fermi level. If one uses the standard Mueller-Hartman–Zittartz model,[18] the estimated T_C would be well below 1 K even for $x=0.02$. However, the system under study has an underlying CDW state. We wish to recall here that the decrease of T_{CDW} would result in the increase of T_C since both phenomena compete for the same Fermi surface. In the present system, the Kondo effect due to Yb reduces the electronic density of states, thereby reducing T_C, while the reduction in T_{CDW} enhances T_C. Therefore, in the present scenario, Yb impurities are not very effective in the reduction of T_C in $Lu_{5-x}Yb_xIr_4Si_{10}$. The low temperature resistivity data (shown in Fig. 3) of the sample with $x=0.07$ down to 0.2 K in a field of 2 T (to quench the superconductivity) clearly shows a Kondo-like response in ρ, with an increase of $\rho(T)$ down to 0.2 K as compared to normal state behavior of $\rho(T)$ of $Lu_5Ir_4Si_{10}$ (data not shown). The low temperature resistivity could be fitted to a simple expression $\rho(T)=\rho_0[1-(T/T_K)^2]+a\ln(T)+bT^n$, where T_K is the Kondo temperature, and a, b, and n are constants. The last term for which we obtained $n=2$ reflects the lattice contribution. Similar power law dependence of ρ at low temperature for the normal state has been observed in A-15 compounds[19] due to anomalous electron-phonon interaction. It is pertinent to recall here that A-15 compounds also have linear chains and exhibit anomalous low temperature properties.[20] For the sample with $x=0.07$, we obtained $\rho_0=124.1$ $\mu\Omega$ cm, $a=-0.5$ $\mu\Omega$ cm/K, and $b=3.0$ $\mu\Omega$ cm/K^2. We also observed that the resistivity at 8 K for the doped sample at ambient field decreased by about 5% when a magnetic field of 2 T was applied. This behavior clearly points to the Kondo behavior of substituted Yb ions in the lattice.

The temperature dependence of the susceptibility (shown in Fig. 4) exhibits a modified Curie-Weiss dependence $[\chi(T)=\chi_0+C/(T+\theta_P)]$, with $\mu_{eff}=4.52\mu_B$ which is close to the free ion value of the Yb^{3+} moment. The value of the Curie-Weiss temperature (θ_P) is found to be 20 K, which is much smaller than the value ($\theta_P \approx 61$ K) reported for the

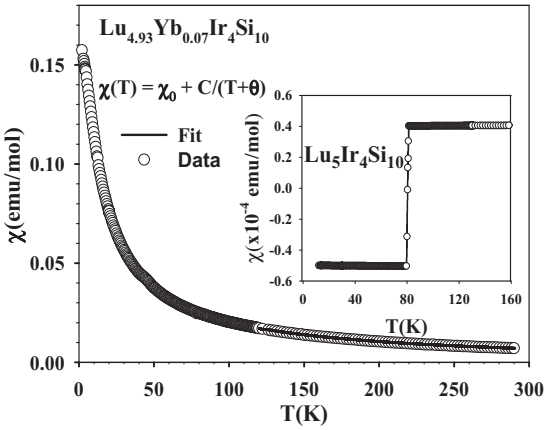

FIG. 4. The temperature dependence of the normal state susceptibility $[\chi(T)]$ of $Lu_{4.97}Yb_{0.03}Ir_4Si_{10}$ in applied field of 2 T is shown in the figure. High temperature data ($125\ \mathrm{K} < T < 300\ \mathrm{K}$) is fitted to the Curie-Weiss expression (see text). The solid line is the fit. The inset shows the $\chi(T)$ of the parent sample $Lu_5Ir_4Si_{10}$, which displays a diamagnetic transition due to the CDW transition.

pure $Yb_5Ir_4Si_{10}$.[8] The reason for this discrepancy could be related to the possibility that intersite interactions could raise the T_K similar to the one observed in some of the Ce based alloys.[21] The moderately large value of θ_P is frequently observed in Kondo systems. Within the Kondo impurity picture, one can estimate the Kondo temperature using the relation $T_K \approx \theta_P/4$ as suggested in an earlier review.[22]

The temperature dependence of the heat capacity (C_p) for the doped sample $Lu_{4.93}Yb_{0.07}Ir_4Si_{10}$ from 1.9 to 110 K is shown in Fig. 5(c). Figures 5(a) and 5(b) display C_p/T vs T data at high and low temperatures. From Fig. 5(c), one can see that the value of the C_p does not change under the application of 2 T at high temperatures. However, at low temperatures, one sees a significant change [see Fig. 5(b)]. Figure 5(a) shows the reduction in T_{CDW} as Yb is substituted for Lu, and also we see a significantly large smearing of the CDW transition unlike the reduction of the jump at T_{CDW} seen with the other dopants.[9,15,16] Notice the jump at the T_{CDW} is also small in $Yb_5Ir_4Si_{10}$,[8] which is attributed to the correlated behavior of the Yb ion. Figure 5(c) depicts the C_p data from 1.9 to 110 K at ambient and applied field of 2 T. One can clearly see the absence of any field effect until one reaches a temperature of 6 K. Below 6 K, there is a significant build up of the Sommerfeld coefficient γ when a magnetic field of 2 T is applied. Even in zero field, the value of the γ is larger ($40\ \mathrm{mJ/mol\,K^2}$ as compared to $23\ \mathrm{mJ/mol\,K^2}$ of $Lu_5Ir_4Si_{10}$).[10,23] It is well established that Kondo impurities contribute to the heat capacity, which causes the upturn in C_p at low temperatures. One can calculate the excess specific heat in the presence of magnetic field since the field does not affect the C_p in the normal state. By estimating the excess specific heat (after subtracting the heat capacity due to lattice contribution) as a function of temperature at various fields, T_K as function of x in $Lu_{5-x}Yb_xIr_4Si_{10}$ is listed in Table I using the models.[24,25] The estimated values of T_K from three measurements are in good agreement, which is consistent with our assumption of Kondo effect in $Lu_{5-x}Yb_xIr_4Si_{10}$. As emphasized before, both T_{CDW} and T_C decrease with x in

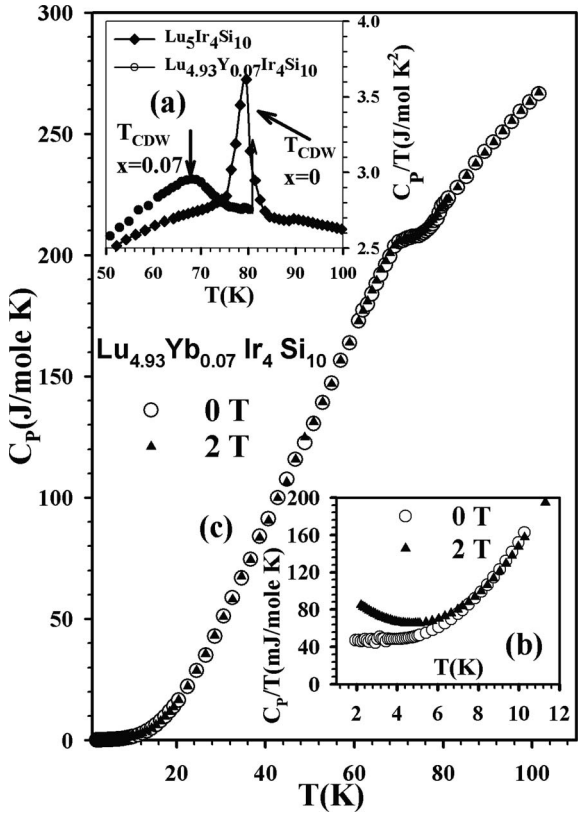

FIG. 5. The temperature dependence of the heat capacity of $Lu_{5-x}Yb_xIr_4Si_{10}$ from 2 to 100 K in fields of 0 and 2 T. (a) shows the high temperature data of $x=0.0$ and $x=0.07$, which illustrate the smearing and reduction of the CDW peak with decrease of T_{CDW} as a function of x. (b) displays the low temperature data, which show an increase of C_p with magnetic field, possibly implying the presence of Kondo effect in the Yb substituted sample.

$Lu_{5-x}Yb_xIr_4Si_{10}$, which is also illustrated in Table I.

To conclude, we have shown that dilute Yb behaves like a Kondo impurity in the $Lu_5Ir_4Si_{10}$ lattice, and to understand the interplay of T_{CDW}, T_C, and T_K, more studies spanning the entire range ($x=0-5$) in $Lu_{5-x}Yb_xIr_4Si_{10}$ are required.

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TABLE I. Values of T_{CDW}, T_C, and T_K as function of x in $Lu_{5-x}Yb_xIr_4Si_{10}$. The values of T_K have been estimated from magnetic susceptibility (χ), electrical resistivity (ρ), and heat capacity (C_p) studies.

Concentration of Yb	T_{CDW} (K)	T_C (K)	T_K (χ) (K)	T_K (ρ) (K)	T_K (C_p) (K)
$x=0.0$	83	3.9			
$x=0.02$	78	3.0	4.7	5	4.8
$x=0.05$	73	2.0	4.8	5.5	5.0
$x=0.07$	69	1.5	5.0	5.9	5.4

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