

Competition between superconductivity and charge-density waves in the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$

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The variation of superconducting transition temperature (T_c) with hydrostatic pressure up to 21 kbar is reported for compounds in the pseudoternary systems $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ ($0.05 \leq x \leq 0.95$) and $(\text{Lu}_{0.9}\text{R}_{0.1})_5\text{Ir}_4\text{Si}_{10}$ ($R = \text{Dy, Ho, Er, Tm, Sc, Y, and Th}$). Measurements of the temperature dependence of the electrical resistivity and ac magnetic susceptibility for $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ ($0.005 \leq x \leq 0.02$) are made to show the correlation between the decrease of the charge-density-wave formation temperature (T_0) and the increase of T_c . $(\text{Lu}_{0.98}\text{Sc}_{0.02})_5\text{Ir}_4\text{Si}_{10}$ and $\text{Lu}_5(\text{Ir}_{0.78}\text{Rh}_{0.22})_4\text{Si}_{10}$, which have T_c about 2 K higher than $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, are used to study the enhanced superconducting state with upper-critical-magnetic-field measurements. Changes in the electronic density of states at the Fermi level and the electron-phonon interaction play an important role in these pseudoternary compounds.

I. INTRODUCTION

Since the discovery¹ of a sudden striking enhancement of the superconducting transition temperature T_c from 3.8 to 9 K in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ at a critical pressure of about 20 kbar, several efforts have also been made to try to answer the following questions: What is the nature of this phase transition? What kind of mechanism should be responsible for this dramatic pressure effect on T_c ? For instance, measurements of the volume dependence of T_c in $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ and $\text{Lu}_5(\text{Ir}_{1-x}\text{Rh}_x)_4\text{Si}_{10}$, low-temperature (down to 21 K) powder x-ray diffraction experiments, and isothermal bulk modulus measurement² taken up to 26 kbar at four different temperatures ranging from 293 to 14 K indicate that the transformation responsible for this enormous pressure enhancement of T_c is probably electronic in nature with no major effect on the cohesive energy of the crystal.

Measurements² of the static magnetic susceptibility and the electrical resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ as a function of temperature exhibit a remarkable anomaly at 83 K. This indicates an electronic phase transition involving the development of a charge-density wave (CDW) opens an energy gap over a portion of the Fermi surface. Results of the pressure effects² on the temperature dependence of the electrical resistivity demonstrate that the pressure enhancement of T_c is due to a progressive removal of the CDW in the crystal.

In this paper, we use the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ to study the effect of atomic disorder on CDW formation and the competition between the CDW transition temperature T_0 and the superconducting transition temperature T_c .

II. EXPERIMENTAL METHODS

All samples were prepared from high-purity elements by arc melting in an argon atmosphere. After the melting process, the samples were sealed in quartz ampules under a partial pressure of argon and annealed at 1250 °C for one day followed by three days at 1050 °C. Mass loss during this synthesis and heat treatment was negligible. Powder x-ray diffraction data confirmed the single phase with the $\text{Sc}_5\text{Co}_4\text{Si}_{10}$ -type structure.

Details of the measurement technique for determining the pressure dependence of T_c may be found in Ref. 1. The dc electrical resistivity was measured on a rectangular parallelepiped of approximate dimensions $1 \times 1 \times 5$ mm³ using a four-probe method. The upper critical field as a function of temperature was determined from magnetization-versus-field curves taken in a commercial superconducting quantum interference device (SQUID) magnetometer.

III. RESULTS AND DISCUSSION

The pressure dependence of T_c for the $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ system is shown in Fig. 1. The following observations based on the data can be made.

(1) The compounds can be separated into two groups. With low Sc content ($x = 0.05, 0.1, \text{ and } 0.2$) T_c 's are lower and enhanced by external pressure. With higher Sc content ($x = 0.3, 0.5, 0.95, \text{ and } 1.0$) T_c 's are higher and depressed by external pressure. There is a trend for T_c to approach a certain limiting value of about 7 K at high pressure.

(2) A critical concentration occurs at about $x = 0.3$. Below this concentration, the phases exhibit an electronic

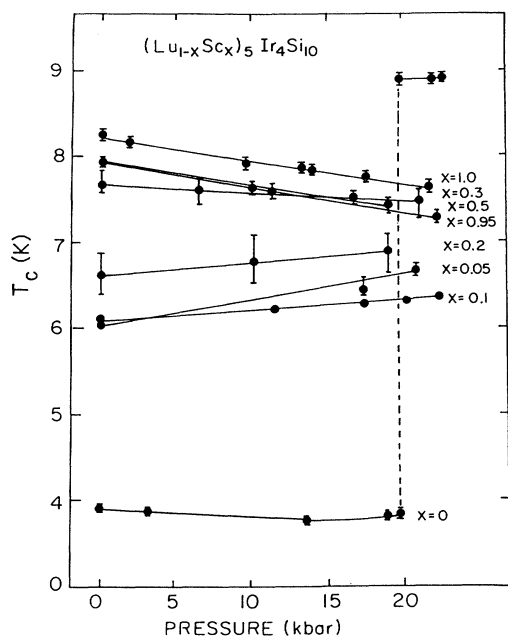


FIG. 1. Pressure dependence of superconducting transition temperature for compounds in the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$. Lines are least-squares fits to the data. Error bars indicate 10%-90% transition widths.

instability and are sensitive to external pressure. This is consistent with previously published data,¹ where the volume of the $x=0.3$ sample at ambient pressure is about the same as that of an $x=0$ sample at 20 kbar.

(3) It is surprising that no sudden jump in T_c is observed up to 21 kbar even at very low concentration ($x=0.05$). This means that the nature of the pressure-

induced phase transformation in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ is more likely of an electronic type and very easily destroyed by sublattice substitution.

The pressure effects on superconductivity for some alloys related to $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ are summarized in Table I. The alloying of magnetic rare earths on the Lu site suppresses the ambient-pressure T_c , in accord with the pair-breaking theory of Abrikosov and Gor'kov.³ Pressure-induced T_c effects are still observed at around 20–22 kbar and T_c jumps (ΔT_c) are almost the same (about 5 K) even for the low T_c (1 K) in $(\text{Lu}_{0.9}\text{Dy}_{0.1})_5\text{Ir}_4\text{Si}_{10}$. Substituting 10% of Th for Lu, enhances T_c both at ambient and high pressures. This may be due to the different valence between Th (4+) and Lu (3+). In order to study the effects of atomic size and electronic configuration on the anomaly in resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, we have substituted on both the Lu and Ir sites with various elements.

Normalized resistivity as a function of temperature for the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ is shown in Fig. 2. The reason why we choose Sc to substitute for Lu is to chemically “contract” the lattice, since Sc has a smaller metallic radius, resulting in a non-negligible size effect in the alloying process. At $x=0.05$, the resistivity is almost identical to that of $x=0$ in the high-temperature ($T > 250$ K) region; then it starts to deviate as temperature is lowered. No noticeable anomaly appears, and the curve ends with a smaller residual resistivity ratio. With a further increase in x (for instance, with $x=0.1$), the resistivity curve becomes flatter with an even smaller residual resistivity ratio (RRR). For $x=0.3$, the RRR is larger than that of the $x=0$ sample. Combining the results of resistivity under pressure with the value of bulk modulus² 1370 ± 70 kbar for $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, we determine that the unit-cell volume of $x=0.05$ at ambient pressure approximately equals that of $x=0$ at 3.3 kbar. As we have seen in Fig. 2, with 5% Sc substitution,

TABLE I. Pressure effects on pseudoternary superconductors. An asterisk (*) means this was not observed in the measurements.

Compound	$T_c(0)$ (K)	P_c (kbar)	ΔT_c (K)	dT_c/dp (10^{-5} K/bar)	T_0 (K)
$\text{Lu}_5\text{Ir}_4\text{Si}_{10}$	3.72	20.5	5.4	-1.17	81
$\text{Lu}_5\text{Ir}_4(\text{Si}_{0.9}\text{C}_{0.1})_{10}$	3.74	20.8	5.4	-1.34	82
$(\text{Lu}_{0.95}\text{Sc}_{0.05})_5\text{Ir}_4\text{Si}_{10}$	6.06	*	*	3.14	*
$(\text{Lu}_{0.9}\text{Sc}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	6.09	*	*	1.23	*
$(\text{Lu}_{0.8}\text{Sc}_{0.2})_5\text{Ir}_4\text{Si}_{10}$	6.54	*	*	1.76	*
$(\text{Lu}_{0.7}\text{Sc}_{0.3})_5\text{Ir}_4\text{Si}_{10}$	7.64	*	*	-0.59	*
$(\text{Lu}_{0.5}\text{Sc}_{0.5})_5\text{Ir}_4\text{Si}_{10}$	7.90	*	*	-2.60	*
$(\text{Lu}_{0.2}\text{Sc}_{0.8})_5\text{Ir}_4\text{Si}_{10}$	7.68	*	*	-3.11	*
$(\text{Lu}_{0.05}\text{Sc}_{0.95})_5\text{Ir}_4\text{Si}_{10}$	7.95	*	*	-2.91	*
$(\text{Lu}_{0.9}\text{Y}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	3.90	20.2	5.0	0.22	73 and 270
$(\text{Lu}_{0.9}\text{Th}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	4.88	*	*	5.10	105
$(\text{Lu}_{0.9}\text{Tm}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	2.84	23.4	5.0	-1.93	86
$(\text{Lu}_{0.9}\text{Er}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	2.74	23.1	5.4	-1.34	82
$(\text{Lu}_{0.9}\text{Ho}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	1.89	20.8	5.4	-1.34	82
$(\text{Lu}_{0.9}\text{Dy}_{0.1})_5\text{Ir}_4\text{Si}_{10}$	1.00	20.5	5.3	-0.52	78
$\text{Lu}_5(\text{Ir}_{0.86}\text{Rh}_{0.14})_4\text{Si}_{10}$	5.72	*	*	3.04	*
$\text{Lu}_5(\text{Ir}_{0.78}\text{Rh}_{0.22})_4\text{Si}_{10}$	5.68	*	*	1.10	*
$\text{Lu}_5(\text{Ir}_{0.95}\text{Co}_{0.05})_4\text{Si}_{10}$	5.52	*	*	3.38	*

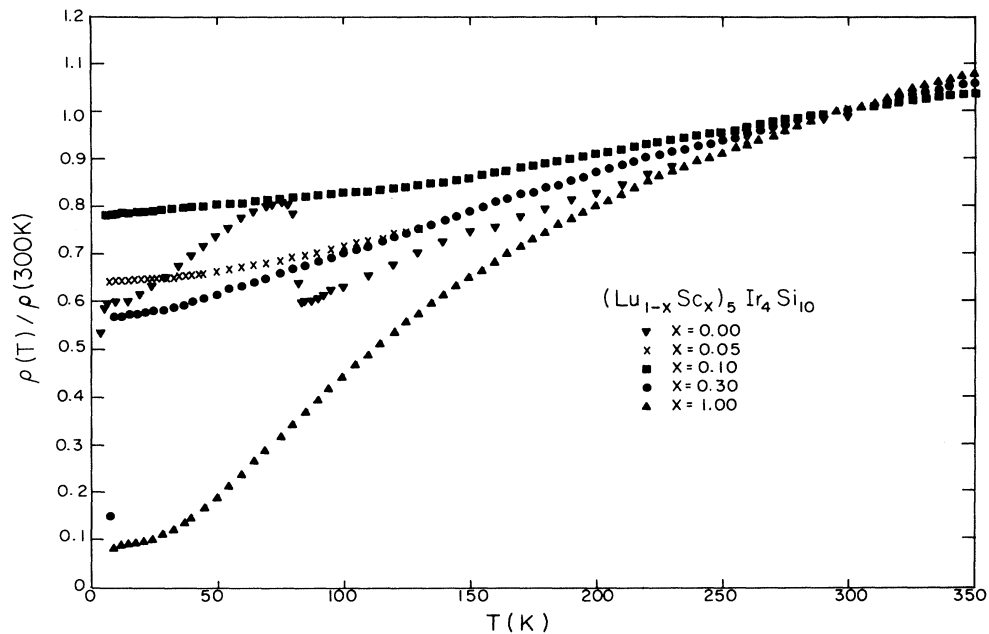


FIG. 2. Normalized resistivity as a function of temperature for the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ ($x=0, 0.05, 0.1, 0.3,$ and 1.0).

the anomaly in the resistivity is totally suppressed. Contrasting this with the pressure data of pure $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ shown in Ref. 2, we conclude that the substitution of Sc not only gives rise to a size effect but induces an atomic disorder to suppress the anomaly in resistivity for $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$. This observation is consistent with the presence of a CDW in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$.

Figure 3 shows the normalized resistivity as a function of temperature for the pseudoternary systems $(\text{Lu}_{0.9}\text{R}_{0.1})_5\text{Ir}_4\text{Si}_{10}$ ($\text{R}=\text{Dy}, \text{Ho}, \text{Er}, \text{Tm}, \text{Sc}, \text{Y},$ and Th). An anomaly is still seen for $\text{R}=\text{Dy}-\text{Tm}$ and Y , is smeared out for $\text{R}=\text{Th}$, and disappears completely for $\text{R}=\text{Sc}$. These results are consistent with the data presented in Table I, namely, that an anomaly in resistivi-

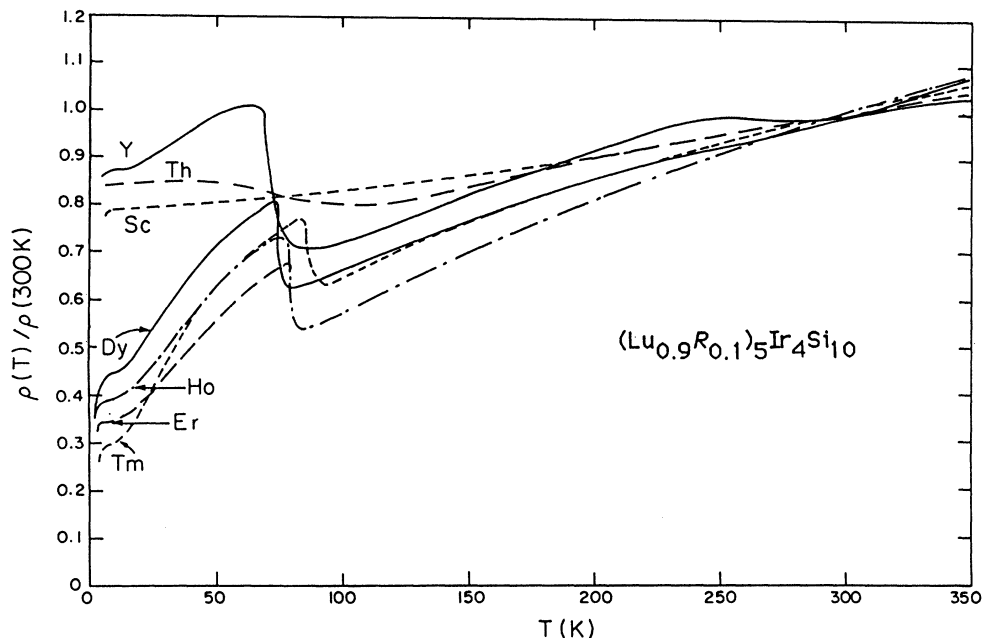


FIG. 3. Normalized resistivity as a function of temperature for the pseudoternary system $(\text{Lu}_{0.9}\text{R}_{0.1})_5\text{Ir}_4\text{Si}_{10}$ ($\text{R}=\text{Dy}, \text{Ho}, \text{Er}, \text{Tm}, \text{Sc}, \text{Y},$ and Th).

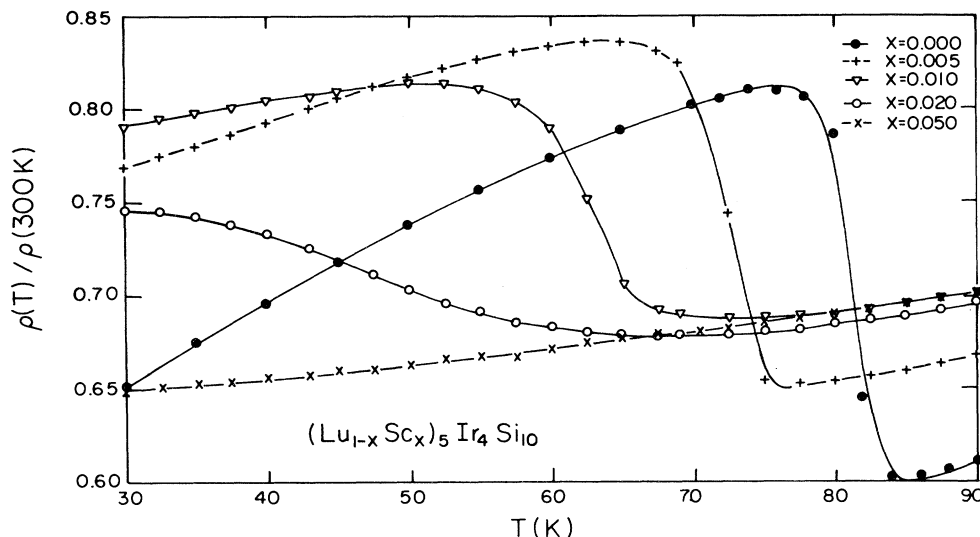


FIG. 4. Normalized resistivity as a function of temperature between 30 and 90 K for the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ ($x = 0, 0.005, 0.01, 0.02, \text{ and } 0.05$).

ty is always associated with a jump in T_c at high pressure. Because the atomic volume of these atoms (except Sc) is larger than that of Lu, no alloying compression comes into the system to suppress the anomaly. On the other hand, all the pure ternary compounds $R_5\text{Ir}_4\text{Si}_{10}$ ($R = \text{Dy} - \text{Tm}$ and Y) have an anomaly in the resistivity.⁴ Thus, the presence of the anomaly in the $R_5\text{Ir}_4\text{Si}_{10}$ compounds results in a reduced effect in the $(\text{Lu}_{0.9}\text{R}_{0.1})_5\text{Ir}_4\text{Si}_{10}$ system. The $(\text{Lu}_{0.9}\text{Th}_{0.1})_5\text{Ir}_4\text{Si}_{10}$ compound shows a slightly different feature. The anomaly in the resistivity is much less pronounced than that of the others. This result correlates with the pressure data where the T_c jump is absent, but with a bigger positive value of dT_c/dp . It is probably due to the tetravalent nature of Th as distinct from the trivalent rare earths.

It has been suggested⁵ that CDW formation and superconductivity are antagonistic, both competing for states near the Fermi surface. In our previous report,² we have shown a suppression of the CDW and an increase of superconducting transition temperature by the application of pressure. At present, we use a quite separate way to study the interplay between superconductivity and CDW in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ by doping with Sc impurities on Lu sites. The presence of an impurity in a CDW material may lead to a change of the CDW transition temperature T_0 and also to a possible smearing of the CDW transition itself. The mathematical analogy between the theories describing a (BCS) superconductor and CDW state suggests⁶ that magnetic impurities in a superconductor play a role equivalent to that of nonmagnetic impurities in a CDW system. In analogy to superconductors, the critical mean free path $\lambda_{cr} = v_F b_{cr}$ for the electrons in a CDW system^{7,8} is $\lambda_{cr} \propto 1.14 v_F / T_0$. Here v_F is the Fermi velocity, $1/b_{cr}$ is the critical pair-breaking parameter for a BCS superconductor, and T_0 is the CDW transition temperature in the absence of impurities. The quantity λ_{cr} may be associated with an effective localization length applicable to

any disorder. Consequently, in the dilute impurity limit, T_0 decreases linearly with increasing impurity concentration. In addition to scattering electrons and inducing a finite electron lifetime, an impurity may also induce Friedel oscillations in the electron-charge distribution, leading to a static lattice distortion. A direct result of this effect is a smearing of the CDW transition.

Normalized resistivity as a function of temperature be-

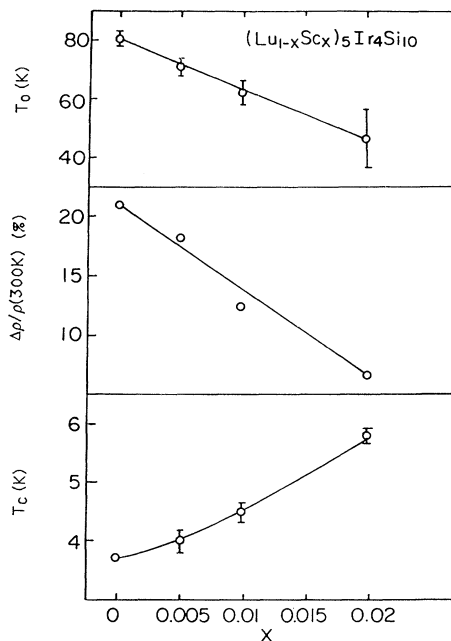


FIG. 5. Alloy concentration dependence of CDW transition temperature T_0 , amplitude of anomaly $\Delta\rho/\rho(300\text{ K})$, and superconducting transition temperature T_c for the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ ($x = 0, 0.005, 0.01, \text{ and } 0.02$).

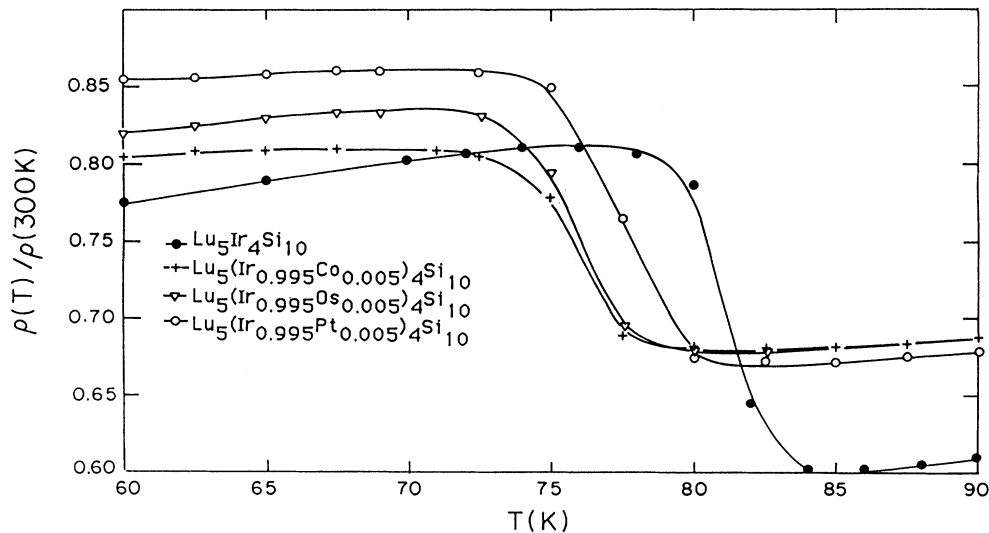


FIG. 6. Normalized resistivity as a function of temperature between 60 and 90 K for pure and $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ doped with 5 at. %, Co, Os, and Pt.

tween 30 and 90 K for pure and doped samples is shown in Fig. 4. It shows that replacing 0.5% of the Lu atoms by Sc atoms results in a decrease of CDW transition temperature by approximately 9 K. At 5 at. % Sc doping, the transition is so depressed that it is no longer detectable. The concentration dependence of the CDW temperature T_0 , the amplitude of anomaly in resistivity $\Delta\rho/\rho(300\text{ K})$, and the superconducting transition temperature T_c for the pseudoternary system $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ ($x=0.0, 0.005, 0.01$, and 0.02) are presented in Fig. 5. It is shown that the impurities lower the T_0 , broaden and smear the CDW transition, while increasing T_c just as one would expect in this system. We calculate the initial concentration dependence of T_0 and T_c to be $(dT_0/dx)_{x=0} = -18.5\text{ K/at. \%}$ and $(dT_c/dx)_{x=0} = 0.5\text{ K/at. \%}$. These values are consistent with the lack of observation of an anomaly and the enhancement of T_c to 5.8 K for 5 at. % Sc substitution for Lu. Thus, from the view of volume change, the effect of impurity is at least six times more sensitive than that of pressure for suppressing the CDW transition in this system. This also provides information that the impurity may induce a change in the band structure or density of states near the Fermi surface.

To investigate the electronic density of states near the Fermi level in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, we vary the Fermi level slightly by the impurity doping and measure the temperature dependence of the resistivity. Os and Pt, which are different from Ir by only one in the atomic number, are chosen as dopants in order to minimize the lattice distortion due to the substitutional disorder. We also use Co as a dopant that is isoelectronic to Ir. Normalized-resistivity data as a function of temperature between 60 and 90 K for pure $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ and $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ doped with 0.5 at. % Co, Os, and Pt are shown in Fig. 6. It is found that Os doping is more effective than Pt doping in suppressing the anomaly as measured by the jump in the

transition temperature and the resistivity. However, the superconducting transition does not change in a complementary manner. For both the Os- and Pt-doped samples, T_c increases as T_0 decreases, as expected; however, T_c increases more for the Pt-containing compound, which is not consistent with what we observe in the $(\text{Lu}_{1-x}\text{Sc}_x)_5\text{Ir}_4\text{Si}_{10}$ system. This inconsistency indicates that size effects, which also occur during doping, may be as important as the electronic effects. The reduction of T_0 when isoelectronic Co is substituted for Ir is larger than with Os or Pt substituting for Ir, confirming the importance of size effects.

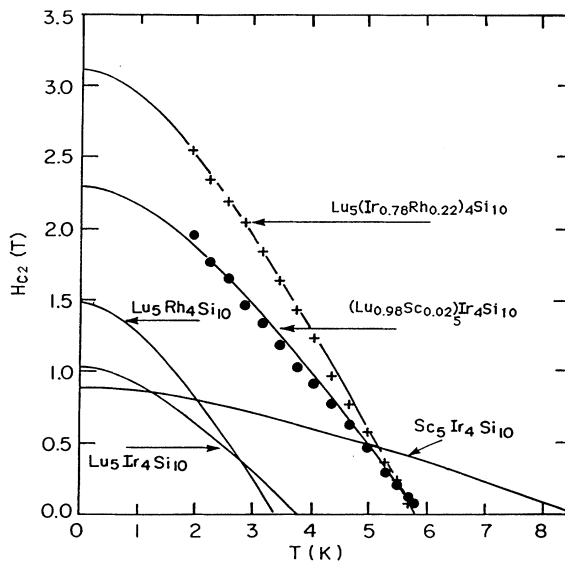


FIG. 7. Upper critical field for $\text{Lu}_5(\text{Ir}_{0.78}\text{Rh}_{0.22})_4\text{Si}_{10}$, $(\text{Lu}_{0.98}\text{Sc}_{0.02})_5\text{Ir}_4\text{Si}_{10}$, $\text{Sc}_5\text{Ir}_4\text{Si}_{10}$, $\text{Lu}_5\text{Rh}_4\text{Si}_{10}$, and $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$. The last three data are taken from Ref. 8. Solid lines are fit to the WHHM theory.

TABLE II. Upper-critical-magnetic-field parameters for some $R_5T_4Si_{10}$ compounds.

Compound	T_c (K)	ρ_{res} ($\mu\Omega$ cm)	$H_{c2}(0)$ (T)	α (Oe/K)	$-(dH_{c2}/dT)_{T=T_c}$ ^a (T/K)	γ_{calc} (erg/cm ³ K ²)	γ_{cal} (mJ/mol K ²)	ξ_{GL} (Å)	λ_{GL} (Å)	κ_{GL}
$Sc_5Ir_4Si_{10}$ ^b	8.23	75	0.90	0.08	0.15	446.4	8.4	150	1920	11.9
$Lu_5Rh_4Si_{10}$ ^b	3.91	160	1.48	0.34	0.64	892.9	17.4	115	4110	35.8
$Lu_5Ir_4Si_{10}$ ^b	3.88	195	1.04	0.21	0.39	446.4	8.8	160	4550	30.9
$(Lu_{0.98}Sc_{0.02})_5Ir_4Si_{10}$	5.90	193	2.30	0.30	0.57	659.0	12.9	100	3670	37.1
$Lu_5(Ir_{0.78}Rh_{0.22})_4Si_{10}$	5.80	325	3.12	0.41	0.78	535.7	10.5	85	4810	56.3

^aThese values were obtained from a fit of the data to the WHHM theory.

^bThese data were derived from Ref. 8.

It will be interesting to understand the superconducting properties of $Lu_5Ir_4Si_{10}$ under higher pressure compared with those at ambient pressure. Instead of measuring the heat capacity and static magnetic susceptibility under high pressure, we used two pseudoternary samples ($Lu_{0.98}Sc_{0.02})_5Ir_4Si_{10}$ and $Lu_5(Ir_{0.78}Rh_{0.22})_4Si_{10}$ to study the upper critical field. The former is strongly enhanced in T_c ($\Delta T_c \sim 2$ K) at a very low Sc concentration, while the latter has the highest T_c in its pseudoternary system. The upper critical fields for ($Lu_{0.98}Sc_{0.02})_5Ir_4Si_{10}$ and $Lu_5(Ir_{0.78}Rh_{0.22})_4Si_{10}$, as well as those for the end members $Sc_5Ir_4Si_{10}$, $Lu_5Ir_4Si_{10}$, and $Lu_5Rh_4Si_{10}$,⁹ are shown in Fig. 7. The data were analyzed with use of the Werthamer-Helfand-Hohenberg-Maki (WHHM) theory¹⁰⁻¹² in the dirty limit¹³ (short mean free path) so that

$$\alpha = 5.28 \times 10^{-5} \left[-\frac{dH_{c2}}{dT} \right]_{T=T_c}, \quad (1)$$

$$\left[-\frac{dH_{c2}}{dT} \right]_{T=T_c} = 4.48 \times 10^4 \gamma_{calc} \rho_{res}, \quad (2)$$

where the Maki parameter α is in units of Oe/K, γ_{calc} is the calculated electronic contribution to the normal-state heat capacity in units of erg/cm³ K², and ρ_{res} is the residual resistivity in units of Ω cm. The values at $T=0$ K of the Ginzburg-Landau (GL) coherence length ξ_{GL} , the GL penetration depth λ_{GL} , and GL parameter κ were calculated (in units of cm) by the following relations:¹¹

$$\xi_{GL} = 8.57 \times 10^{-7} (\gamma_{calc} \rho_{res} T_c)^{-1/2} (1-t)^{-1/2}, \quad (3)$$

$$\lambda_{GL} = 6.42 \times 10^{-3} \left[\frac{\rho_{res}}{T_c} \right]^{-1/2} (1-t)^{-1/2}, \quad (4)$$

$$\kappa_{GL} = 7.49 \times 10^3 \gamma_{calc}^{1/2} \rho_{res}, \quad (5)$$

where $t = T/T_c$. The results are listed in Table II.

For $(Lu_{0.98}Sc_{0.02})_5Ir_4Si_{10}$, the $H_{c2}(0)$, $-(dH_{c2}/dT)_{T=T_c}$, and γ_{calc} are larger than those of the two end members. So, the enhancement of T_c is mainly due to the enhancement in the electronic density states at Fermi level. For $Lu_5(Ir_{0.78}Rh_{0.22})_4Si_{10}$, even though T_c , $H_{c2}(0)$, and $-(dH_{c2}/dT)_{T=T_c}$ are larger than those of two end members, γ_{calc} and ξ_{GL} are smaller than those of $Lu_5Ir_4Si_{10}$ due to the bigger ρ_{res} caused by the comparable atomic-disorder effects. Thus the enhancement of T_c may be caused by an enhancement of the electron-phonon interaction.

IV. CONCLUSION

Our experiments reveal that the CDW formation in these compounds is highly sensitive to atomic disorder. The doping effects maintain the correlation between T_c and T_0 . These results for the pseudoternary systems $(Lu_{1-x}Sc_x)_5Ir_4Si_{10}$ and $Lu_5(Ir_{1-x}Rh_x)_4Si_{10}$ are very similar to those of other CDW systems such as $1T-TaS_2$,¹⁴ $(Ti_{1-x}V_x)Se_2$,¹⁵ $(Ta_{1-x}Nb_x)S_3$,¹⁶ and $2H-TaS_2$.¹⁷

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