# Phase transitions in the superconducting compound $Lu_5Rh_4Si_{10}$ at ambient and high pressure

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The pressure dependence of the superconducting transition temperature $T_c$ up to 68 kbar and resistivity up to 18 kbar were measured on the ternary compound $Lu_5Rh_4Si_{10}$. At ambient pressure, $Lu_5Rh_4Si_{10}$ shows a metalliclike temperature variation of resistivity at temperatures higher than 160 K, exhibits an anomaly at 155 K, and undergoes a superconducting transition at 3.4 K. A tendency to suppress the resistive anomaly and enhance the superconducting transition temperature by the application of pressure was observed. In combination with molar-magnetic-susceptibility data, it is shown that the formation of a charge-density wave opens an energy gap which reduces the density of electronic states at the Fermi surface.

## I. INTRODUCTION

Charge-density waves (CDW's) are now a frequently observed feature of highly anisotropic metals such as one-dimensional conductors[1] and two-dimensional layered compounds.[2] A close relation between charge-density-wave formation and superconductivity is characteristic of the superconductors in the family typified by $NbSe_3$. For example, $NbSe_3$ shows two CDW transitions, but at low temperature, this compound becomes superconducting when pressure is applied[3,4] or when impurities are introduced.[5,6] In our previous work,[7,8] $Lu_5Ir_4Si_{10}$ was shown to exhibit an anomaly in the electrical resistivity and magnetic susceptibility at $T_0=79$ K. Application of pressure at 21 kbar completely suppresses the resistive anomaly and enhances the superconducting transition temperature $T_c$ from 3.8 to 9 K simultaneously. These effects were attributed to the formation (at $T_0$) of a charge-density wave, which was subsequently suppressed by either pressure or crystalline impurities. In this paper, we present similar measurements for $Lu_5Rh_4Si_{10}$, which is isostructural with $Lu_4Ir_4Si_{10}$, to provide additional evidence for CDW formation in this family of compounds.

## II. EXPERIMENTAL DETAILS

All samples were prepared by arc melting stoichiometric mixtures of high-purity elements in a Zr-gettered argon atmosphere. The resulting ingots were turned over and remelted several times to promote homogeneity. The samples were then sealed in quartz ampules with about 160 Torr of argon and annealed at 1250 °C for one day followed by three days at 1050 °C. Mass loss during the synthesis and heat treatment was negligible. Powder x-ray diffraction data confirmed the single phase with the $Sc_5Co_4Si_{10}$-type structure. The dc resistivity at ambient pressure and the ac resistivity at high pressure were measured on samples of rectangular parallelepipeds of approximate dimensions $1\times1\times5$ and $2\times2\times1$ mm$^3$ using a four-probe method. Data on the hydrostatic pressure dependence of $T_c$ and resistivity in $Lu_5Rh_4Si_{10}$ were taken up to 20 kbar using a piston-cylinder self-clamp technique.[9] A 1:1 mixture of *n*-pentane and isoamyl alcohol was used as the pressure-transmitting fluid, with a superconducting Sn manometer to determine the pressure at low temperatures. For measurements above 20 kbar, a diamond anvil cell made of high-purity Cu-Be was used in conjunction with a superconducting quantum interference device (SQUID) detection system. The pressure transmitting fluid was a 4:1 mixture of methanol and ethanol. A Pb manometer was used to determine the pressure at low temperatures. Static-magnetic-susceptibility data were taken in a commercial SQUID magnetometer[10] in a field of 20 kOe.

## III. RESULTS AND DISCUSSION

The normalized resistivity as a function of temperature between 2.6 and 300 K for $Sc_5Ir_4Si_{10}$, $Y_5Ir_4Si_{10}$, and $Lu_5Rh_4Si_{10}$ is shown in Fig. 1. $Sc_5Ir_4Si_{10}$ show normal-metallic behavior through the whole temperature region above $T_c$ with a residual resistivity ratio (RRR) $\rho(300\text{ K})/\rho(4.2\text{ K})\sim20$. The other three compounds exhibit an anomaly at $T_0=250$ K for $Y_5Ir_4Si_{10}$, at 155 K for $Lu_5Rh_4Si_{10}$, and at 83 K for $Lu_5Ir_4Si_{10}$, but with much smaller RRR's. These anomalies are reproducible by independent samples. In the $Lu_5Rh_4Si_{10}$ compound, the resistivity decreases when the temperature is lowered from 300 K, but increases sharply at 155 K, and reaches a maximum at 122 K before resumming a metallic-type temperature variation. The amplitude of the anomaly $\Delta\rho$ is about 13% of the room-temperature resistivity $\rho(300\text{ K})$. No obvious thermal hysteresis is detected when we vary the temperature across the transition temperature. A similar behavior is also seen in the $Lu_5Ir_4Si_{10}$ and $Y_5Ir_4Si_{10}$ compounds but with a sharper and more pro-

 

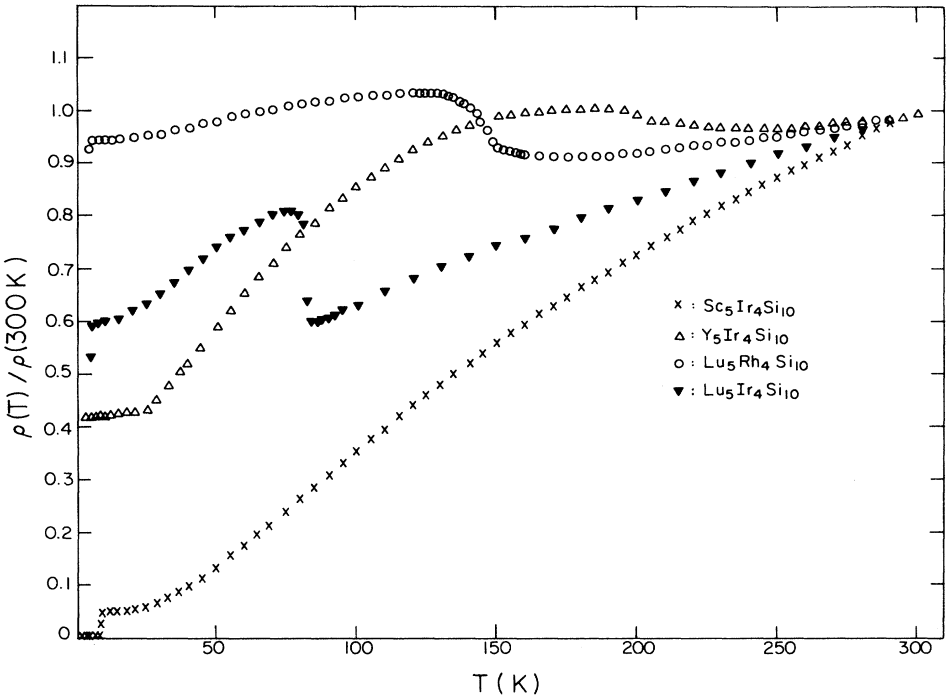


FIG. 1. Normalized resistivity as a function of temperature between 2.6 and 300 K for $Sc_5Ir_4Si_{10}$, $Y_5Ir_4Si_{10}$, $Lu_5Rh_4Si_{10}$, and $Lu_5Ir_4Si_{10}$.

nounced anomaly for the former and a broader and less pronounced anomaly for the latter.

Normalized resistivity as a function of temperature for $Lu_5Rh_4Si_{10}$ at five different pressures is presented in Fig. 2. At ambient pressure, the resistivity shows an anomaly at $T_0 = 155$ K. Application of pressure at 8.6 and 14.4 kbar depresses $T_0$ severely down to about 125 and 110 K, respectively. The anomaly is much broader and the RRR is smaller than at ambient pressure. As the pressure increases to 17.9 kbar, the transition becomes very sluggish, and more than one relative maximum seems to exist in the curve. Further application of pressure at 18.3 kbar yields only one clear maximum in resistivity at 64 K, but with a different shaped anomaly from that at ambient

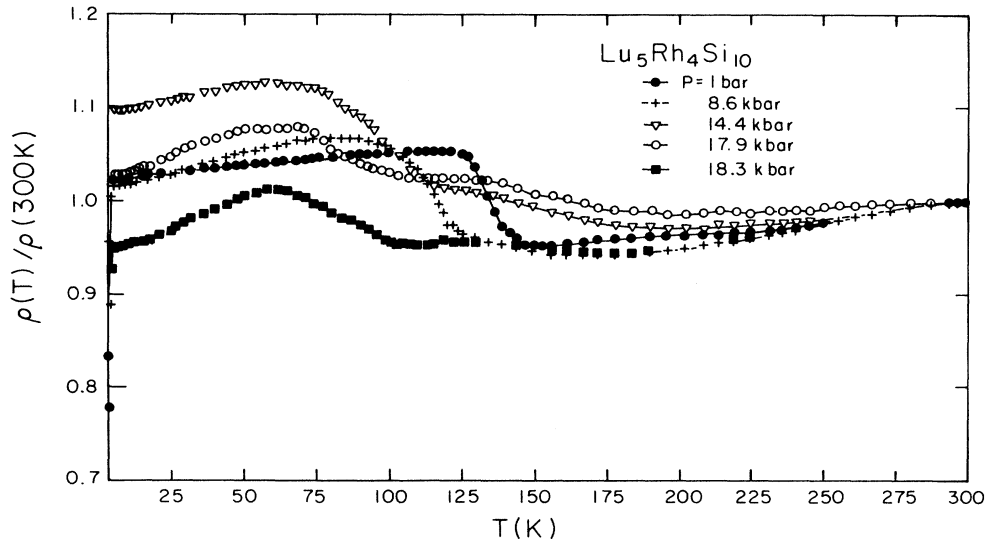


FIG. 2. Normalized resistivity as a function of temperature for $Lu_5Rh_4Si_{10}$ at five distinct pressures.

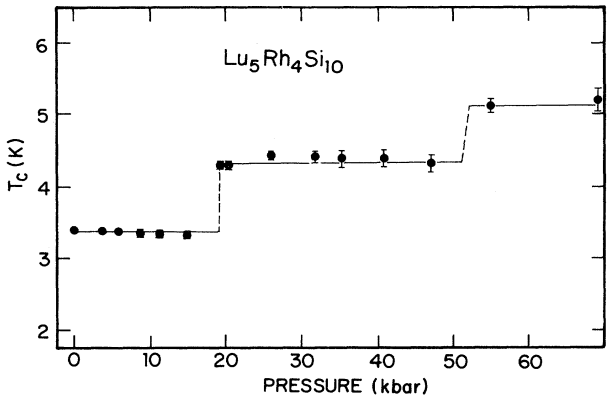


FIG. 3. Pressure dependence of the superconducting transition temperature for $Lu_5Rh_4Si_{10}$. Error bars indicate 10%-90% transition widths.

pressure. At this point, we note that the superconducting transition temperature $T_c$ is 4.3 K at pressures of 17.9 and 18.3 kbar, compared with a $T_c$ of 3.4 K at pressures of 1 bar, 8.6 kbar, and 14.4 kbar. This indicates specific differences between $Lu_5Rh_4Si_{10}$ and $Lu_5Ir_4Si_{10}$. In the latter case, the anomaly in resistivity is totally suppressed at 21.42 kbar, and $T_c$ jumps from 3.8 to 9.1 K at the same time, while in the former case, the $T_c$ jumps from 3.4 to 4.3 K at 17.9 kbar, but the resistivity anomaly is still partially retained. Therefore, we postulate that a more complicated CDW structure forms in $Lu_5Rh_4Si_{10}$ than in $Lu_5Ir_4Si_{10}$. The changing shape of the resistive anomaly with increasing pressure may reflect that the type of CDW is pressure dependent or that the coupled CDW's are separated by the application of pressure. One could anticipate that at higher pressures the anomaly could be suppressed completely for $Lu_5Rh_4Si_{10}$, resulting in another jump in $T_c$ to a higher value.

The electrical resistivity of $Lu_5Ir_4Si_{10}$ and $Lu_5Rh_4Si_{10}$ can be consistently explained by assuming the formation of charge-density waves. When a CDW forms, gaps open at the Fermi surface at those portions that satisfy the nesting condition. The increase in resistivity at $T_0$ for these two compounds can be attributed to the decrease in area of the Fermi surface resulting from the opening of the gaps. The formation of a CDW is determined by the competition between two terms in the free energy of the system: the strain energy, which increases with the formation of superlattice disorder, and the gain in electronic energy resulting from the opening of the gaps. The gain in electronic energy increases with decreasing temperature because the Fermi surface is sharper at low temperature. By applying pressure, we expect a stiffening of the lattice, which increases the strain energy. To offset this increase in energy, the electronic-energy gain must be larger to stabilize the CDW state. Consequently, the CDW transition temperature $T_0$ is lowered. This is similar to the pressure dependence of the CDW transition temperature in the layered compound $2H$-$NbSe_2$,[11] quasi-one-dimensional transition-metal trichalcogenide $NbSe_3$,[12] and even the three-dimensional cubic spinel compound $CuV_2S_4$.[13]

The pressure dependence of the superconducting transition temperature $T_c$ for $Lu_5Rh_4Si_{10}$ measured inductively is given in Fig. 3. $T_c$ is slightly depressed by pressure at $p < 18$ kbar, while it jumps suddenly from 3.4 to 4.3 K as the pressure increases above 18 kbar. $T_c$ is basically pressure independent between 18 and 50 kbar; however,

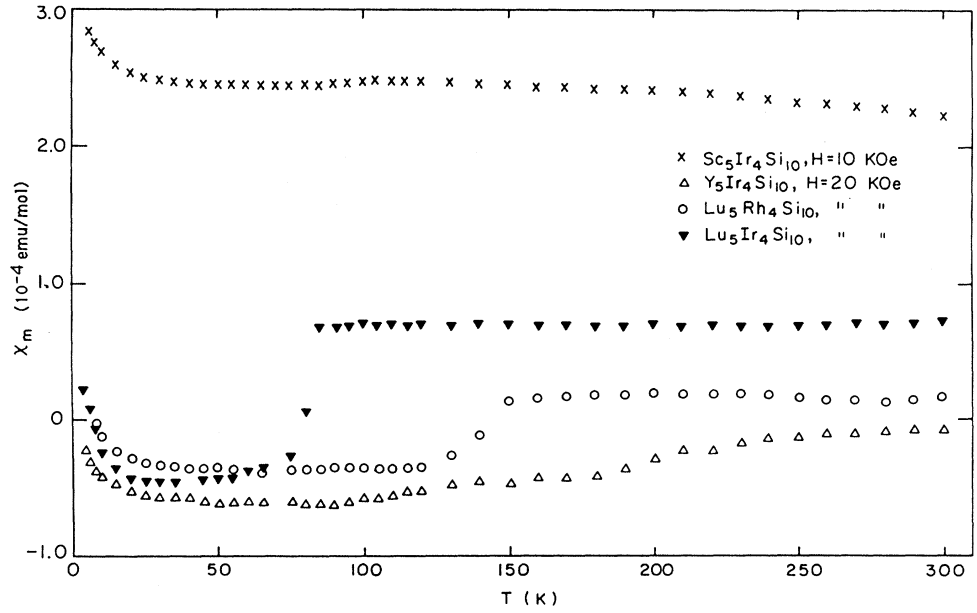


FIG. 4. Molar magnetic susceptibility as a function of temperature between 4 and 300 K for $Sc_5Ir_4Si_{10}$, $Y_5Ir_4Si_{10}$, $Lu_5Rh_4Si_{10}$, and $Lu_5Ir_4Si_{10}$.

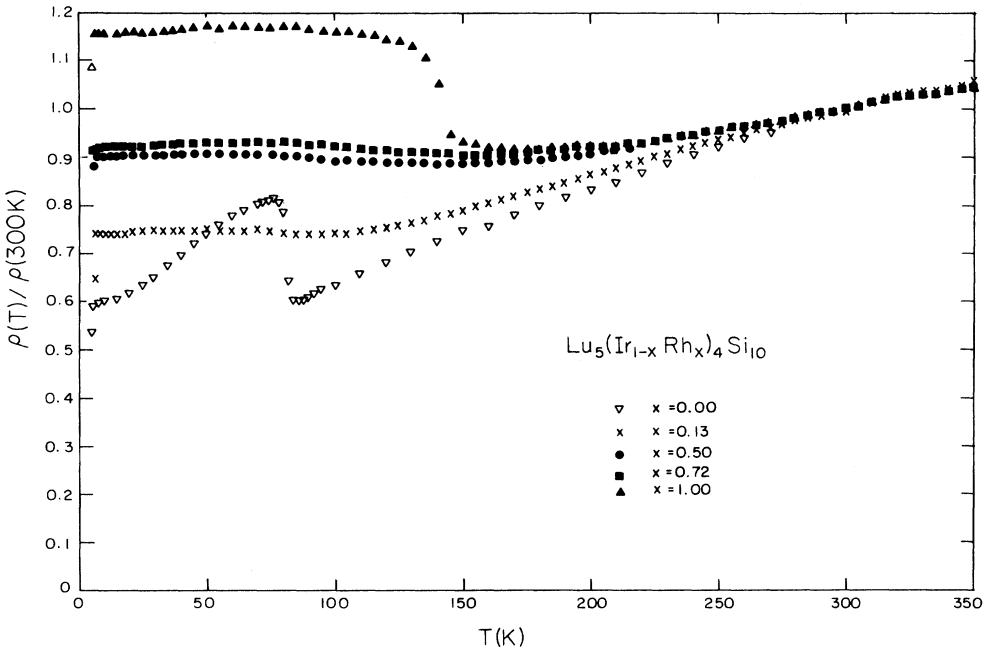


FIG. 5. Normalized resistivity as a function of temperature for the pseudoternary system $Lu_5(Ir_{1-x}Rh_x)_4Si_{10}$ ($x=0$, 0.13, 0.5, 0.72, and 1.0).

another jump in $T_c$ from 4.3 to 5.1 K is observed as the pressure exceeds 50 kbar. This is consistent with the resistivity measurements. Now, we briefly discuss the correlation between $T_0$ and $T_c$ with the parameter of pressure. A supplementary contribution to the decrease of $T_0$ with pressure can be expected from the stiffening of the lattice, since the distortion associated with a CDW will then cost more strain energy. This stiffening should also cause a decrease of $T_c$ (Ref. 14) at the same time. The onset of the CDW will open a gap at the Fermi surface, reduce the density of states at the Fermi level $N(E_F)$, and then decrease the $T_c$. When pressure is applied, both the area where the gap opens and the amplitude of the gap decrease; therefore the reduction of $N(E_F)$ will be smaller and $T_c$ will increase.[15] However, the soft-mode model, which was proposed by Testardi[16] for the explanation of the variation of $T_c$ with pressure in $A15$ superconductors cannot be ruled out. The soft-mode associated with the structure distortion may decrease $\langle\omega^2\rangle$ as long as $T_0 > T_c$ and should be more efficient when $T_0 \sim T_c$. From this point of view, the absence of a jump[7] in $T_c$ at 21 kbar for $Y_5Ir_4Si_{10}$ can be explained by assuming that the applied pressure is not high enough to suppress the anomaly that appears in resistivity at about 250 K.

The molar magnetic susceptibility as a function of temperature between 4 and 300 K for $Sc_5Ir_4Si_{10}$, $Y_5Ir_4Si_{10}$, $Lu_5Rh_4Si_{10}$, and $Lu_5Ir_4Si_{10}$ is shown in Fig. 4. The size, width, and transition temperature of the anomaly for these four compounds are mutually consistent for both the magnetic-susceptibility and electrical-resistivity measurements. The sharp decrease in the magnetic susceptibility, which coincides with the increase in the resistivity, is attributed to a partial opening of energy gaps and a loss of electronic density of states at the Fermi surface due to the CDW transition.

Figure 5 shows the normalized resistivity as a function of temperature for the pseudoternary system $Lu_5(Ir_{1-x}Rh_x)_4Si_{10}$. In this case, there exists an anomaly in resistivity for both end members, but no feature is seen for the intermediate-composition ($0.13 \leq x \leq 0.72$) compounds. Because the unit-cell volumes of $Lu_5Ir_4Si_{10}$ and $Lu_5Rh_4Si_{10}$ are about the same (with only a 0.7% difference), the atomic disorder or chemical effects in the pseudoternary system are the dominant factors in suppressing the anomaly in the resistivity. This is consistent with the fact that impurity effects and lattice disorder have a profound influence on the CDW transition.[17] This observation can also explain the existence of a nonlinear concentration dependence of the superconducting transition temperature $T_c$ in this system.[7]

## IV. CONCLUSION

We have presented electrical-resistivity and static-magnetic-susceptibility data for the isostructural compounds $Lu_5Ir_4Si_{10}$, $Lu_5Rh_4Si_{10}$, $Y_5Ir_4Si_{10}$, and $Sc_5Ir_4Si_{10}$. These experiments document the presence of an ambient pressure phase transition at $T_0 = 83$, 155, and 250 K for the first three compounds, respectively, while $Sc_5Ir_4Si_{10}$ show normal-metallic behavior over the temperature range above $T_c$.

High-pressure electrical-resistivity experiments for $Lu_5Rh_4Si_{10}$ show that $T_0$ decreases monotonically with

pressure, the anomaly is partially suppressed, and $T_c$ is enhanced discontinuously from 3.4 to 4.3 K by application of pressure above 18 kbar. A complete suppression of the CDW transition is expected as the pressure is increased up to 50 kbar, where another jump in $T_c$ is observed. Furthermore, the $Lu_5(Ir_{1-x}Rh_x)_4Si_{10}$ system demonstrates clearly the suppression of the CDW and the simultaneous enhancement of $T_c$.

All of these experimental data indicate this electronic phase transition involves the development of a charge-density wave (CDW) that opens an energy gap over a portion of the Fermi surface. The formation of a CDW has been observed and is well documented for two-dimensional compounds such as the transition-metal dichalcogenides,[2,18] anisotropic metals such as one-dimensional conductors,[19] and the cubic spinel compound $CuV_2S_4$ where the underlying crystal lattice is certainly three dimensional.[13,20]

A crucial experiment to be performed in the future is a low-temperature single-crystal x-ray or electron diffraction measurement to look for specific evidence of a CDW superlattice in $Lu_5Ir_4Si_{10}$. In addition, band-structure calculation would provide great insight to understand why the CDW is observed in $Lu_5Ir_4Si_{10}$, $Lu_5Rh_4Si_{10}$, and $Y_5Ir_4Si_{10}$, but not in $Sc_5Ir_4Si_{10}$ and the other isostructural superconductors. These conclusions might also clarify the effective dimensionality (or anisotropy) of $Lu_5Ir_4Si_{10}$.

## ACKNOWLEDGMENTS

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