Electronic phase transition and partially gapped Fermi surface in superconducting $Lu_5Ir_4Si_{10}$

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The enhancement of the superconducting transition temperature of $Lu_5Ir_4Si_{10}$ from 3.8 to 9.1 K under pressure is caused by the suppression of an electronic phase transition which results in the partial gapping of the Fermi surface. Using electrical resistivity at ambient and high pressure, bulk-modulus, heat-capacity, upper-critical-magnetic-field, and static-magnetic-susceptibility measurements, we estimate that the density of electronic states at the Fermi level is reduced by 36% due to this phase transition. The transition, which occurs at 79 K at ambient pressure, has the experimental characteristics indicative of charge- or spin-density-wave formation.

I. INTRODUCTION

Among the great variety of structural types of ternary rare-earth-metal transition-metal silicides,[1] there exist numerous superconducting compounds.[2,3] The highest reported superconducting transition temperatures (T_c's) of these ternary silicides belong to materials which crystallize in the $Sc_5Co_4Si_{10}$-type structure.[4–8] In a recent paper,[7] we reported a pronounced sensitivity of T_c to pressure for $Lu_5Ir_4Si_{10}$, which resulted in a discontinuous but reversible increase in T_c from 3.8 K to a value in excess of 9.1 K at a critical pressure, p_c, of 21 kbar. There was no measurable volume anomaly associated with this transition, implying an electronic origin to this effect. Phase transitions in which conduction electrons play an important role reflect unusual changes in the electronic properties which can be detected in other physically measurable quantities.

In this paper we present static magnetic susceptibility and electrical resistivity data for $Lu_5Ir_4Si_{10}$ which document the presence of an ambient-pressure phase transition at $T_0=79$ K. High-pressure electrical resistivity experiments show that T_0 decreases with pressure, resulting in the complete suppression of this phase transition at the critical pressure, $p_c=21$ kbar. Thus, the removal of this electronic phase transition by pressure results in the large, discontinuous enhancement of the superconducting critical temperature. Our experimental data indicate this electronic phase transition may involve the development of a charge- or spin-density wave (CDW or SDW) that opens an energy gap over a portion of the Fermi surface. We give a quantitative estimate of the loss in electronic density of states at the Fermi level due to this energy gap. The existence of CDW's and the general tendency of pressure to suppress the formation of CDW's is well documented for two-dimensional compounds such as the transition-metal dichalcogenides[9,10] and anisotropic metals such as the one-dimensional conductors.[11] CDW formation has been observed in the spinel compound CuV_2S_4, where the underlying crystal lattice is certainly three dimensional.[12,13] It has recently been suggested that a CDW or SDW is responsible for the phase transition at 17.5 K in the heavy-fermion compound URu_2Si_2.[14] In addition to these experiments on the electronic phase transition in $Lu_5Ir_4Si_{10}$, we report low-temperature heat-capacity and upper-critical-magnetic-field measurements for this compound. These data show that the superconducting state is described well by the conventional electron-phonon interaction.

II. EXPERIMENTAL METHODS

All samples of $Lu_5Ir_4Si_{10}$ were prepared by arc-melting stoichiometric mixtures of high-purity elements in a Zr-gettered argon atmosphere. The resulting ingots were turned and remelted at least five 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. The lattice parameters of the tetragonal cell (space group $P4/mbm$) were determined from powder x-ray diffraction patterns by the method of least squares using 20 to 24 reflections including an internal silicon standard ($a=5.43083$ Å). No impurity reflections were observed. The sample used for the high-pressure work had lattice parameters $a=12.486(8)$ Å and $c=4.178(4)$ Å, in good agreement with literature values.[5,7] Our experimental density of 9.69 g/cm^3 is consistent with these values. High-pressure measurements of T_c and the electrical resistivity were done using a piston-cylinder-type hydrostatic pressure clamp.[15] The ac electrical resistivity at ambient and high pressure was measured on a rectangular parallelpiped of approximate dimensions $2.2\times2.8\times1.4$ mm^3 using a four-probe method. The technique for determining the isothermal bulk modulus has been detailed elsewhere.[16] Low-temperature heat-capacity measurements were performed using a semiadiabatic heat-pulse-type calorimeter. Details of the measurement technique may be found in Ref. 17. Static magnetic susceptibility data were taken in a commercial superconducting quantum interference device magnetometer[18] in a field of 20.0 kOe. The critical magnetic field as a function of temperature was determined from magnetization versus field curves taken in this same instrument.

III. RESULTS AND DISCUSSION

The previously reported[7] effect of hydrostatic pressure on the T_c of $Lu_5Ir_4Si_{10}$ is reproduced in Fig. 1. The reversibility of the phase transformation from the low-T_c to high-T_c state is illustrated by the order in which the data were taken. We have observed this effect in other samples of $Lu_5Ir_4Si_{10}$, always at a critical pressure of $p_c = 21 \pm 1$ kbar. Data for the compressions of an indium-jacketed $Lu_5Ir_4Si_{10}$ sample at two temperatures are shown in Fig. 2. The total length of the jacketed sample at $p = 1$ bar was 6.35 mm, while the equivalent length of the $Lu_5Ir_4Si_{10}$ sample was 2.44 mm for the 6.35-mm-diameter sample holder. The change in sample length between 293 and 14.3 K in Fig. 2 is due primarily to the thermal expansion of the indium jacket. The size of the data points which are shown corresponds to $(\pm 5 \times 10^{-4})V_0$, where V_0 is the volume of the $Lu_5Ir_4Si_{10}$ sample. The smoothness of the data suggests that if a transition occurs in this pressure region, the relative volume change, $\Delta V/V_0$, associated with it must be of the order of or smaller than 10^{-3} (0.1%). The isothermal bulk modulus determined in these measurements is 1370±70 kbar, independent of temperature. Two additional sets of data taken at 201 and 100 K are similarly featureless. We note that the lowest temperature of 14.3 K for these measurements is close to the high T_c of 9.1 K and that the maximum pressure of 26 kbar considerably exceeds p_c. Since the high sensitivity of this experiment would reveal a relative volume change of $\Delta V/V \sim 0.1\%$, these data provide strong evidence that the effect on T_c is electronically driven with no major change in the cohesive energy of the crystal.

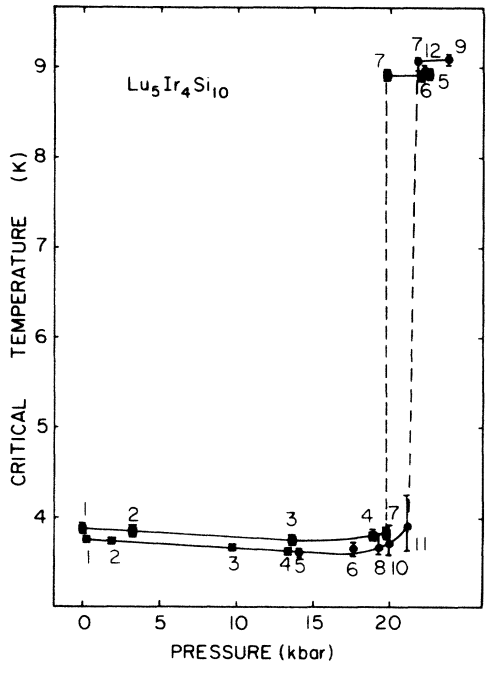

FIG. 1. Pressure dependence of the superconducting transition temperature for two samples of $Lu_5Ir_4Si_{10}$. Numbers next to the data points represent the order in which the data were taken for each sample.

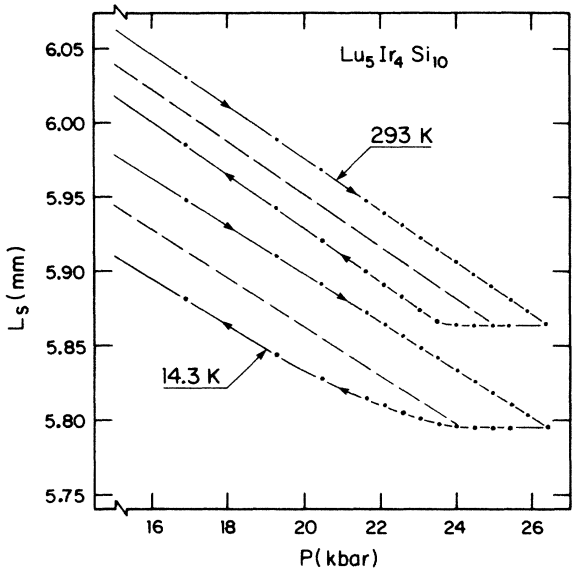

FIG. 2. Actual experimental data for the compressions of an indium-jacketed $Lu_5Ir_4Si_{10}$ sample at two temperatures in a piston-displacement experiment; the hysteresis loop is due primarily to friction effects in the apparatus.

The electrical resistivity measured as a function of temperature at ambient and high pressures is shown in Fig. 3. In every instance, the pressure was fixed at room temperature and the data were taken as the sample cooled slowly to below the superconducting transition. Each isobaric sequence was followed by an increase in pressure until the highest pressure sequence at 21.42 kbar was completed. At this point, the pressure was released and the ambient-pressure resistivity remeasured. This second set of data at ambient pressure was identical to the initial one, confirming the complete reversibility of the phase transition. Absolute values for $\rho(300\text{ K})$ and $\rho(7\text{ K})$ at ambient pressure are 85.6 and 56.2 $\mu\Omega$ cm, respectively, while the corresponding numbers at 21.42 kbar are $\rho(300\text{ K}) = 94.4$ $\mu\Omega$ cm and $\rho(10\text{ K}) = 3.52$ $\mu\Omega$ cm. At ambient pressure the resistivity shows a clear, sharp jump at $T_0 = 79$ K.

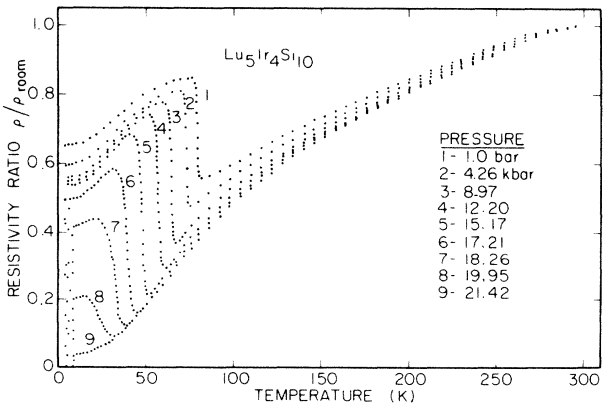

FIG. 3. Electrical resistivity normalized to the value at 300 K versus temperature for $Lu_5Ir_4Si_{10}$ at nine distinct pressures. Some data points are omitted for clarity.

This feature is characteristic of a phase transition associated with an increase in conduction-electron scattering, or the loss of a portion of the Fermi surface. It resembles the type of anomaly one expects from the formation of a CDW or SDW or perhaps a crystallographic transformation. The last possibility is remote considering the lack of volume discontinuity in the bulk-modulus measurements. However, as a further check, we performed powder x-ray diffraction experiments down to 21 K which revealed no detectable deviation from the primitive tetragonal symmetry observed at room temperature. In addition, this anomaly in the resistivity is insensitive to applied magnetic fields, showing no measurable shift in a field of 20 kOe.

In contrast to the lack of magnetic field sensitivity, pressure has a major effect on T_0, the temperature at which the anomaly occurs. From the data of Fig. 3, T_0 is depressed monotonically by the application of pressure. We compute the initial pressure dependence of T_0 to be $dT_0/dp)_{p=0} = -1.4$ K/kbar. This slope increases in magnitude with pressure, resulting in a phase boundary with negative curvature in a T_0 versus pressure graph. In addition to lowering T_0, the size of the anomaly is also suppressed as p_c is approached (e.g., see curve 8). When the pressure exceeds p_c, this resistive anomaly is destroyed completely. These data demonstrate conclusively that the complete suppression of this phase transition correlates with the stabilization of the high-T_c superconducting state.

The effect of this phase transition is also evident in the molar magnetic susceptibility shown in Fig. 4. The data are essentially temperature-independent from 380 K to $T_0 = 79$ K, with a value of $\chi(85\text{ K}) = 0.732 \times 10^{-4}$ emu/mol. At T_0, χ decreases sufficiently over a narrow temperature range to become diamagnetic. The magnetic susceptibility maintains a relatively constant value of $\chi(35\text{ K}) = -0.468 \times 10^{-4}$ emu/mol until an increase at the lowest temperatures causes χ to become positive again. This low-temperature upturn is probably due to the presence of a few ppm of paramagnetic impurity in the sample which is undetectable by our other experiments.

We analyze this susceptibility data by writing the total temperature-independent part of the susceptibility as,

$$\chi_0 = \chi^{\text{core}} + \chi^{\text{Pauli}} + \chi^{\text{Landau}} , \tag{1}$$

where χ^{core} is the core diamagnetism term, χ^{Pauli} is the Pauli paramagnetism due to the conduction electrons, and χ^{Landau} is the diamagnetic orbital contribution due to the conduction electrons. The core diamagnetism may be estimated from tabulated values[19] using Ir^{3+} which yields a value of $\chi^{\text{core}} = -2.35 \times 10^{-4}$ emu/mol. Representing the conduction-electron band effects by an effective mass, m^*, permits one to relate χ^{Pauli} to χ^{Landau} (Ref. 20)

$$\chi^{\text{Landau}} = -\frac{1}{3}\left[\frac{m}{m^*}\right]^2 \chi^{\text{Pauli}} , \tag{2}$$

$$\chi^{\text{Pauli}} = 2\mu_B^2 N_b(0) , \tag{3}$$

where μ_B is the Bohr magneton and $N_b(0)$ is the bare density of states at the Fermi level per spin direction. Combining Eqs. (1), (2), and (3) yields an expression involving the effective mass and density of states; namely,

$$\chi_0 - \chi^{\text{core}} = \left[\frac{r}{1.545 \times 10^4}\right]\left[1 - \frac{1}{3}\left[\frac{m}{m^*}\right]^2\right] N_b(0) . \tag{4}$$

In this equation, r is the number of atoms per molecule ($r = 19$), susceptibilities are expressed in units of emu/mol and the density of states, $N_b(0)$, is given in terms of states/eV atom spin. The left-hand side of the equation is known from our experimental data and tabulated values of χ^{core}, while the right-hand side of the equation involves two unknowns that characterize the electronic state of the compound; namely, m^*/m and $N_b(0)$. Below the phase transformation temperature, $T_0 = 79$ K, we obtain an independent experimental determination of the enhanced density of electronic states at the Fermi level $N^*(0) = N_b(0)(1+\lambda) = N_b(0)(m^*/m)$ from our low-temperature heat-capacity measurements.

The low-temperature heat capacity of $Lu_5Ir_4Si_{10}$ is shown in Fig. 5, where C/T is plotted against T^2 in a standard fashion. The superconducting transition at $T_c = 3.77$ K is evident in the inset. All data above T_c were fit to an equation of the form $C = \gamma T + \beta T^3 + \alpha T^5$, where γT is the usual electronic contribution, β is the lattice specific-heat coefficient and the αT^5 term accounts for any anharmonicity of the lattice. Utilizing the coefficients γ and β, we calculate $N^*(0)$, the enhanced density of electronic states at the Fermi level, $N^*(0) = 3\gamma/2\pi^2 N r k_B^2$, as well as the Debye temperature, $\Theta_D = (12\pi^4 N r k_B/5\beta)^{1/3}$, where N is Avogadro's number and k_B is Boltzmann's constant. The Debye temperature

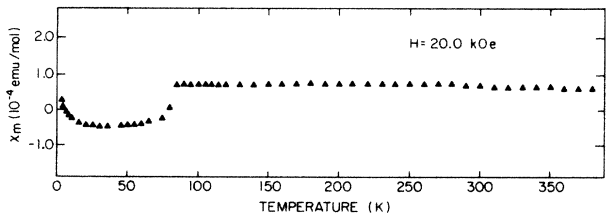

FIG. 4. Static molar magnetic susceptibility as a function of temperature for $Lu_5Ir_4Si_{10}$ measured in a field of 20.0 kOe.

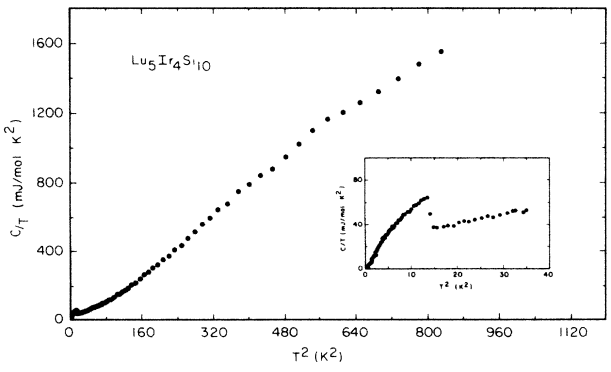

FIG. 5. Specific heat divided by temperature T vs T^2 for $Lu_5Ir_4Si_{10}$ between 0.5 and 830 K^2. The inset shows clearly the superconducting transition at $T_c = 3.77$ K.

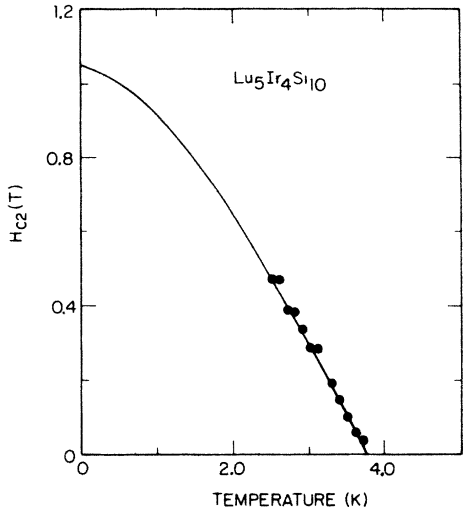

FIG. 6. Upper critical field versus temperature for $Lu_5Ir_4Si_{10}$. The curve represents the best fit of the data to the WHHM theory (see text for details).

of $\Theta_D = 366$ K is somewhat lower than those reported for the isostructural scandium transition-metal silicides.[21] The experimental value of $N^*(0) = 0.26$ states/eV atom spin is used in the analysis below. The jump in the specific heat at T_c is sharp, with a value of $\Delta C = 124$ mJ/mol K. In combination with our experimental values of $\gamma = 23.42$ mJ/mol K^2 and T_c, we compute the ratio $\Delta C/\gamma T_c = 1.41$, in excellent agreement with the BCS value of 1.43.

The combination of magnetic susceptibility and heat-capacity data provides an experimental determination of $N_b(0)$ and $m^*/m = 1+\lambda$ at temperatures below the phase transition. Specifically, we use Eq. (4) with $\chi_0 = \chi(35\ \text{K})$ and our heat-capacity data that give $N_b(0)(m^*/m) = 0.26$ states/eV atom spin to obtain $m^*/m = 1.43$ and $N_b(0) = 0.18$ states/eV atom spin for $T < T_0$. This results in a value for the electron-phonon coupling parameter, λ, of 0.43. As a check on this result, we use the formalism of McMillan[22] with $\mu^* = 0.10$ and calculate $\lambda = 0.50$ based on our calorimetric T_c of 3.77 K. These two values are in reasonable agreement.

As the system goes through the phase transition at $T_0 = 79$ K, we attribute the change in susceptibility at T_0 to an increase in $N_b(0)$ and thus an enhanced Pauli susceptibility. We can estimate the new effective electron mass by noting that the superconducting transition temperature is $T_c = 9.1$ K when this phase transition is prevented from occurring. Using this value for T_c in the McMillan equation yields $\lambda = 0.66$, or a mass enhancement of $m^*/m = 1.66$ for $Lu_5Ir_4Si_{10}$ when no phase transition occurs. Taking this ratio of m^*/m and using $\chi_0 = \chi(85\ \text{K})$ as the experimental value on the left-hand side of Eq. (4) yields a value of $N_b(0) = 0.28$ states/eV atom spin. Therefore, the electronic phase transition which occurs at 79 K is responsible for a 36% reduction in $N_b(0)$ as the sample cools through the transition. This is consistent with the occurrence of a CDW or SDW transition at T_0 which opens an energy gap over about 36% of the Fermi surface.

Experimental results for H_{c2} versus temperature are shown in Fig. 6. The curve is the calculated $H_{c2}(T)$ based on the Werthamer-Helfand-Hohenberg-Maki (WHHM) theory[23–25] and gives a best fit with the spin-orbit scattering parameter $\lambda_{SO} = 9.0$ and a Maki parameter $\alpha = 0.21$. This fit projects a value for $H_{c2}(0)$ of 1.04 T. The experimental value of the initial slope $(-dH_{c2}/dT)_{T_c}$ is 0.39 T/K. In the dirty limit, this theory yields the relationship $(-dH_{c2}/dT)_{T_c} = 4.48\gamma\rho$, where H_{c2} is in Teslas, γ is in ergs/cm$^2K^2$ and the residual resistivity, ρ, is in Ω cm.[26] Using our experimental values for γ(23.42 mJ/mol K^2) and ρ(10 K) measured at ambient pressure (56.2 $\mu\Omega$ cm), we calculate $(-dH_{c2}/dT)_{T_c} = 0.30$ T/K, in good agreement with the measured value.

IV. CONCLUSIONS

An electronically driven phase transition which occurs at 79 K opens an energy gap over a portion of the Fermi surface of $Lu_5Ir_4Si_{10}$. This gap is responsible for a 36% reduction in the density of electronic states at the Fermi level. Experimentally, this loss of states is evident in the resistivity and magnetic susceptibility. The superconducting properties also reflect this loss, since complete suppression of this phase transition by the application of hydrostatic pressure corresponds to a dramatic, sharp increase in T_c from 3.8 to 9.1 K. The identification of this transition with either a CDW or SDW requires further investigation. Low-temperature single-crystal x-ray studies are in progress. Previous high-pressure work[7] indicates that an electronic phase transition of this nature may be present in other isostructural ternary silicides.

ACKNOWLEDGMENTS

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