

⁶⁹G. D. Whitfield, Phys. Rev. 121, 720 (1961); or Ref. 50.
⁷⁰J. M. Ziman, *Principles of the Theory of Solids* (Cambridge U.P., Cambridge, England, 1965), p. 188, Eq. (7.52).

⁷¹See Ref. 70, Eq. (7.54) and Ref. 51, Eq. (2.10).
⁷²V. Heine, in *The Physics of Metals*, edited by J. Ziman (Cambridge U.P., Cambridge, England, 1969), Vol. 1, Chap. 1, Eq. (1.55).
⁷³L. Nordheim, Ann. Phys. (N.Y.) 9, 607 (1931).

PHYSICAL REVIEW B

VOLUME 5, NUMBER 8

15 APRIL 1972

Transport Properties and the Electronic Structure of $\text{LaSn}_{3-x}\text{In}_x$

Warren D. Grobman

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

(Received 29 July 1971)

Measurements of resistivity, magnetoresistance, and thermopower were made on a series of compounds of the type $\text{LaSn}_{3-x}\text{In}_x$ for temperatures from 1.5 to over 300 K. In addition, Hall effect in LaIn_3 was measured from 10 to 270 K. These experiments are interpreted in the light of previous data on these compounds with the object of obtaining a better understanding of their electronic structure. The main conclusions are that these compounds are transition metals which possess neither local moments nor magnetic phase transitions. They contain small pockets of electrons near the Fermi surface for $x=0$ which disappear as x approaches unity. This result is consistent with measurements of magnetic susceptibilities and superconducting properties. Finally, arguments are presented based on thermopower measurements which indicate that Ce in LaIn_3 is a Kondo system.

I. INTRODUCTION

Intermetallic compounds of the form $\text{LaX}_{3-x}\text{Y}_x$ with X or Y representing Sn, In, Pb, or Tl have been studied extensively in recent years.¹⁻⁸ These papers have been motivated by the possibility of studying superconductivity and magnetism as a function of electron or rare-earth impurity concentration in these compounds. The $\text{LaX}_{3-x}\text{Y}_x$ compounds are also easy to work with experimentally as they possess a simple crystal structure (simple-cubic Bravais lattice), are single phase, congruently melting, and can be grown as large single crystals.¹

As a result of the previous work on these materials, several questions concerning their electronic structure have arisen. The dramatic variation⁸ of T_c with x near $x=0$ in $\text{LaSn}_{3-x}\text{In}_x$, as well as the unusual results for $\chi(T)$ in LaSn_3 has led to speculation that LaSn_3 possesses local moments or strong exchange enhancement.⁵ Here T_c is the superconducting critical temperature and $\chi(T)$ is the magnetic susceptibility as a function of absolute temperature T .

Havinga *et al.*⁶ have done an extensive series of experiments on T_c , $\chi(300\text{ K})$, and $S(300\text{ K})$ vs electron concentration in these compounds, where S is the thermopower. The oscillations in these quantities plotted as a function of electron concentration have been ascribed by them to expansion of a simple-metal Fermi surface through a Brillouin-zone boundary as electron concentration is in-

creased. In addition, they interpret $S_x(300\text{ K})$ vs x by assuming that umklapp processes dominate the thermopower. (In the present paper a subscript x denotes a property measured as a function of the concentration x of In in $\text{LaSn}_{3-x}\text{In}_x$.)

The present measurements of transport properties in $\text{LaSn}_{3-x}\text{In}_x$ vs x were performed in order to test these ideas and to clarify our understanding of the electronic structure of these compounds. We have measured $\rho(H)$, $\rho(T)$, and $S(T)$ vs x for $1.2 \leq T \leq 350\text{ K}$ (ρ is the resistivity), and in LaIn_3 have measured the low-field Hall coefficient R_H for $10 \leq T \leq 270\text{ K}$. The behavior of $\rho(H)$ and $\rho(T)$ has led to the conclusion that these compounds do not possess local moments nor magnetic phase transitions as a function of x or T . Low-temperature measurements of $\rho(T)$ have led us to reject the suggestion⁶ that these materials are simple metals, while S_x for different temperatures suggests that the behavior of S vs x is not explainable on the basis of umklapp processes. Rather, the present measurements, especially $\rho_x(H)$ for $0 < x < 1$, suggest that the unusual behavior of $\text{LaSn}_{3-x}\text{In}_x$ near $x=0$ can be explained by the LaSn_3 Fermi surface possessing small pockets of electrons which disappear as one alloys with In at $x \approx 1$.

Our measurements of $S_x(T)$ indicate that Ce in LaIn_3 (but not in LaSn_3) is a Kondo system, while $\rho(H)$ vs T for LaIn_3 and LaSn_3 , and $R_H(T)$ for LaIn_3 indicate that the number of holes or electrons in these compounds is not temperature dependent. We have also inferred a structural phase trans-

TABLE I. Impurities (ppm) in LaIn_3 samples; ND means not detected ($\leq 3-10$ ppm).

Impurity	Sample I	Sample II
Ti	3000	ND
Sn	10	10
As	10	ND
Y	20	ND
Cu	3	30
Mn	3	3
Ce	100	30
Ho	3	ND
Si	300	ND
Al	100	ND
Mg	20	ND
Na	20	ND
F	3	ND
O	2400	ND
C	800	ND
B	30	ND
Eu	ND	15
Ni	ND	30
Cr	ND	10
Fe	ND	75

formation in $\text{LaSn}_{2.55}\text{In}_{0.45}$ from measurements of $\rho(T)$.

Section II of this paper describes the experimental details and presents the results of the measurements, while Sec. III will discuss these results.

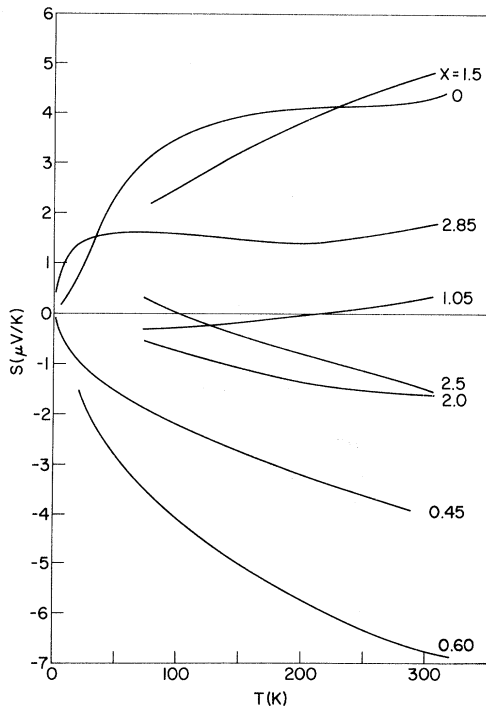


FIG. 1. Thermopower vs temperature in $\text{LaSn}_{3-x}\text{In}_x$ for several values of x .

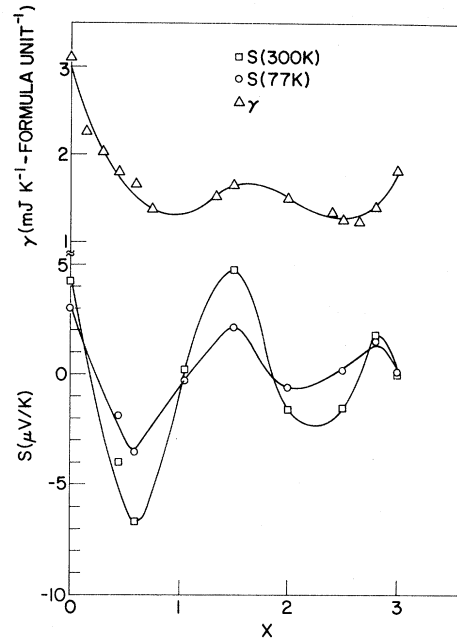


FIG. 2. Thermopower at 300 and 77 K as well as the low-temperature electronic specific heat coefficient γ as a function of x .

II. EXPERIMENTAL DETAILS

Sample Preparation

Most samples were obtained from unannealed ingots pulled from a melt in tantalum crucibles in an argon atmosphere. The Sn and In were 99.999% pure, while an idea of the purity of the La can be obtained from the mass spectrometer analysis of two samples of LaIn_3 shown in Table I. Bars for most measurements were cut from the ingots by a spark-erosion technique. Some bars (usually for thermopower measurements where dimensions and shape are not too important) were obtained from pieces of fractured ingots shaped by lapping on emery paper. The samples are typically quite brittle and corrode badly when unprotected, probably due to hydration of the La by atmospheric H_2O . For this reason samples were stored for long periods in evacuated ampuls and for short periods were preserved under mineral oil.

Thermopower

The thermoelectric power $S_x(T)$, where T is absolute temperature, was measured from 4.2 to 300 K for $x=0, 0.45, 0.6, 2.85, \text{ and } 3.0$, and from 77 to 300 K for $x=1.05, 1.5, 2.0, \text{ and } 2.5$. The apparatus used and the precision and accuracy of the measurements have been previously described.⁹ The random error is small compared to the possible systematic error of about 0.25 $\mu\text{V/K}$. Results of these measurements are shown

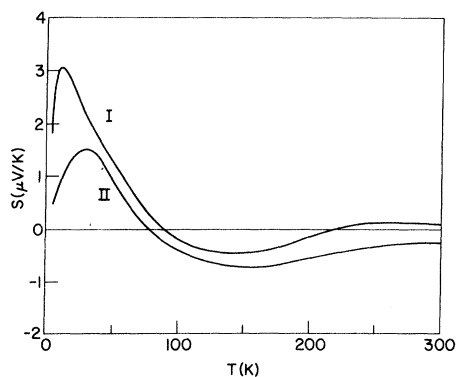


FIG. 3. Thermopower vs temperature for two different samples of LaIn_3 .

in Fig. 1. In Fig. 2 we have plotted S_x (300 K), S_x (77 K), and γ as a function of x , where γ is the previously measured linear coefficient of the low-temperature electronic specific heat.⁷ Figure 3 gives $S(T)$ for two samples of LaIn_3 . Sample I in this figure was cut from a boule pulled from the melt by the Czochralski process, while sample II was melted in a crucible with a conical tip slowly cooled to below the LaIn_3 melting point. Table I gives a mass spectrometer analysis of the magnetic impurities in these two samples. The similarity of the curves at high temperatures indicates the lack of dependence of the properties of these samples on method of preparation—especially since

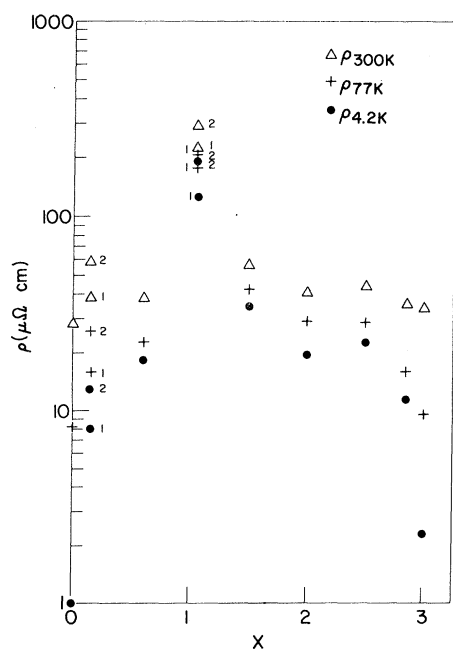


FIG. 4. Resistivity of $\text{LaSn}_{3-x}\text{In}_x$ at $T=4.2$, 77, and 300 K. (1 and 2 refer to two different samples of a given composition.)

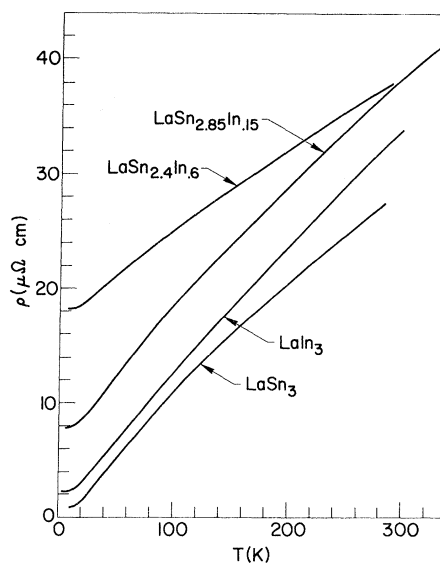


FIG. 5. Resistivity of $\text{LaSn}_{3-x}\text{In}_x$ for $x=0$, 0.15, 0.6, and 3.0.

$S(T)$ is so sensitive to the detailed electronic structure. (A similar independence of preparation method was seen in the magnetic susceptibility of LaSn_3 which was identical over a wide temperature range for a pulled boule and an unannealed arc-melted sample.¹⁰) The difference between $S(T)$ for samples I and II for $T < 50$ K is probably due to the Kondo behavior of Ce impurity in the La starting material. This will be discussed in Sec. III.

Resistivity and Magnetoresistance

These measurements were made by a standard four-probe technique on samples of approximate dimensions $0.1 \times 0.1 \times 1 \text{ cm}^3$ cut from ingots by spark erosion. The samples were glued to a 0.001-in.-thick plastic film fastened to a copper block with GE 7031 varnish. The copper block contained heaters and a GaAs thermometer and was placed in an evacuated can which fit inside the 1-in. bore of a 45-kG superconducting magnet. Leads were 0.005- or 0.010-in.-diam Pt wire spot welded or In soldered to the samples.

The resistivity of $\text{LaSn}_{3-x}\text{In}_x$ was measured at 4.2, 77, and 300 K for $x=0.0$, 15.0, 0.6, 1.05, 1.5, 2.0, 2.5, 2.85, and 3.0. In addition, $\rho(T)$ was measured from 4.2 K to ≥ 300 K for $x=0.0$, 0.15, 0.6, and 3.0. These data are shown in Figs. 4 and 5. Transverse and longitudinal magnetoresistance were measured¹¹ from 0 to 45 kG for $x=0.15$, 0.3, and 0.6. These measurements will be discussed in Sec. III.

Kohler plots ($\Delta\rho/\rho_0$ vs H/ρ_0) of $\rho_{\text{trans}}(H)$ for LaIn_3 and $\rho_{\text{long}}(H)$ for LaSn_3 are shown in Fig. 6 for temperatures of 5 to 45 K and fields of 0 to 45 kG.

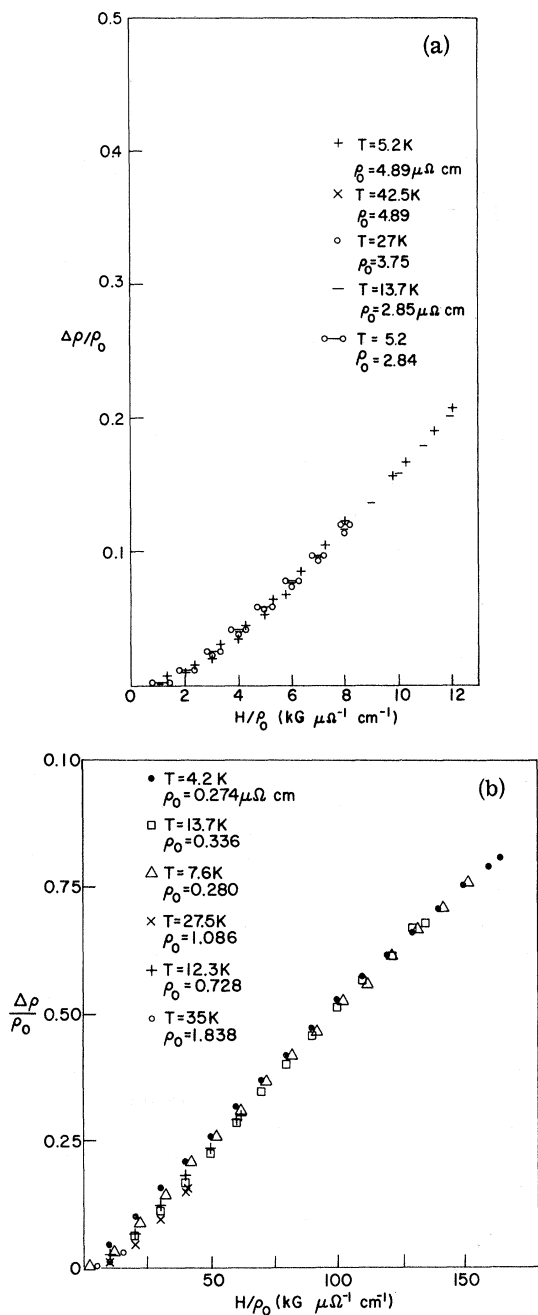


FIG. 6. (a) Kohler plot of the transverse magnetoresistance in LaIn₃, and (b) Kohler plot of the longitudinal magnetoresistance of LaSn₃.

Here $\rho_0 \equiv \rho(H=0)$ and $\Delta\rho \equiv \rho(H) - \rho_0$. The quantity $\rho_{\text{trans}}(H)$ for LaSn₃ also follows Kohler's law and the $T=4.2$ K curves for this magnetoresistance is shown in Fig. 7. The most significant error in these measurements would be in the temperature calibration. This error is at most the lesser of 1% or 2 K.

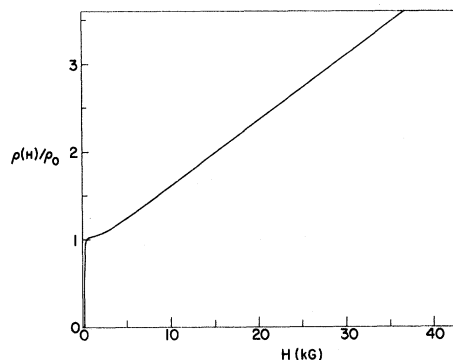


FIG. 7. Transverse magnetoresistance of LaSn₃ at $T=4.2$ K.

Hall Effect

The Hall coefficient R_H was measured in LaIn₃ from $T=10$ to 292 K by a dc technique.¹² Figure 8 shows the circuit used as well as the final results of the measurement. The experimental procedure for each point was to measure the Hall voltage for a magnetic field (~ 20 kG) of both polarities, and for a sample current of both polarities, choosing every combination of polarities and repeating the measurement several times for each combination. The Hall coefficient is then found from

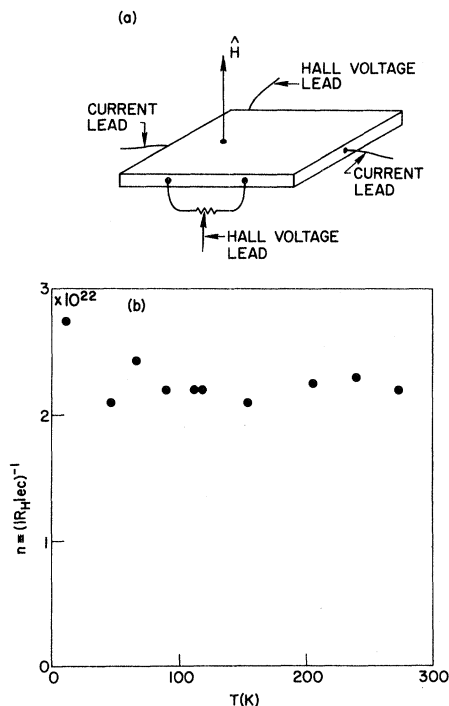


FIG. 8. (a) Circuit used for measurements of the low-field Hall coefficient, R_H in LaIn₃, and (b) R_H vs T for LaIn₃.

$$R = \frac{\bar{V}t}{IH}, \quad 4\bar{V} = V_{++} + V_{--} - V_{+-} - V_{-+},$$

where t is sample thickness, H is the magnetic field, and I is the current. V_{ij} is the voltage on the Hall leads where $i=+$ or $-$ refers to the magnetic field polarity and $j=+$ or $-$ refers to the polarity of the current. Measurement of R by this technique can cancel the effects of most spurious voltages arising from thermomagnetic or thermoelectric effects, or from an Ohmic potential drop along the sample.¹² One standard deviation is about 1.5×10^{21} for the points in Fig. 8.

III. DISCUSSION

In this section we will discuss conclusions about the electronic structure of $\text{LaSn}_{3-x}\text{In}_x$ that can be drawn from the data presented in Sec. II. Some of the major conclusions to be obtained were summarized in Sec. I.

One of the most interesting problems about the compositions near LaSn_3 is the question of whether they possess local moments or strong magnetic interactions.⁵ The magnetic susceptibility χ for LaSn_3 is of the Curie-Weiss form⁵:

$$\chi = \frac{C}{T - \Theta}$$

for $100 \leq T \leq 700$ K. Below ~ 100 K, χ deviates from this form. It has been questioned at various times whether this susceptibility implies local moments or a strong exchange-enhanced Pauli susceptibility for these compounds.⁸ It has even been conjectured⁹ that some sort of unusual magnetic interaction leads to the large decrease of T_c with x in $\text{LaSn}_{3-x}\text{In}_x$ near $x=0$, where T_c is the superconducting transition temperature. Our results for $\rho(T)$ and $\rho(H)$ suggest that there are no striking magnetic transitions or local moments in LaSn_3 . In the case of magnetoresistance, one would expect at least a small negative transverse magnetoresistance at low fields which was temperature dependent if there were local moments. We have seen no such negative magnetoresistance of more than 1 part in 10^3 (the noise limit of our apparatus). Also, the validity of Kohler's law for LaSn_3 over a wide range of temperatures and fields indicates both the absence of local moments and of magnetic ordering. The resistivity up to room temperature for LaSn_3 (Fig. 5) shows no anomalies which one should observe if there were magnetic ordering.

There appears to be a Kondo effect in LaIn_3 as evidenced by the thermopower, $S(T)$, shown in Fig. 3. Ce was the dominant magnetic impurity common to both samples (see Table I). The $3\text{-}\mu\text{V/K}$ value for S at $T=10$ K is an anomalously large value for any non-Kondo system at 10 K including the other members of the $\text{LaSn}_{3-x}\text{In}_x$ system¹³ (see Fig. 1).

Consider an impure system which has a resistivity ρ_m due to magnetic scatterers, ρ due to other scattering centers, and thermopowers S_m and S due to magnetic or other scatterers, respectively, when they act alone. In this case, when magnetic and nonmagnetic scatterers are both present, the total thermopower is¹⁴

$$S_{\text{total}} = (\rho_m S_m + \rho S) / (\rho_m + \rho). \quad (1)$$

At the intermediate compositions, ρ_m for 100 ppm Ce would be dominated by ρ due to disorder (see the residual resistivities in Fig. 4) and the large denominator in Eq. (1) would destroy the anomalous thermopower. Thus it is only in almost pure LaIn_3 or LaSn_3 that one would see a Kondo peak in $S(T)$ at small Ce concentrations. Further, the resistivity in LaIn_3 or LaSn_3 at low temperatures still is significant compared with that due to a few hundred ppm of Ce so any Kondo peak in $S(T)$ should be dependent on Ce concentration. This is probably the explanation for the larger peak in Fig. 3 for sample I which, as we see from Table I has more Ce than sample II. There is probably 30 to a few hundred ppm of Ce in our LaSn_3 samples as well, but no Kondo peak is observed there. We attribute this to the different conduction wave functions at a Ce site surrounded by Sn atoms rather than In atoms.

The electronic structure for the compositions near LaSn_3 is probably unusual for a transition metal. This is evidenced by the susceptibility and Knight-shift measurements in the literature,¹⁵ as well as by our results for $S(T)$ vs x in $\text{LaSn}_{3-x}\text{In}_x$. Furthermore, our measurements of $\rho_x(T)$, which show a large increase in resistivity at all temperatures near $x=1$ (see Fig. 4), as well as the results obtained by others⁶ for T_c and γ , where γ is the low-temperature electronic specific heat, seem to show a minimum in the density of states near LaSn_2In_1 . It is just at this point that the interpretation of our results for $S(T)$ shows a changing character for the conduction states. Consider $S_x(300\text{ K})$ or $S_x(77\text{ K})$ plotted along with γ_x . The simplest rigid-band model of S_x would give the relation¹⁶

$$S_x \propto \frac{1}{N} \frac{\partial N}{\partial E_F} = \frac{1}{N} \frac{\partial N}{\partial x} \propto \frac{\partial N}{\partial x}, \quad (2)$$

where N is the electronic density of states at the Fermi surface and E_F is the Fermi energy. We see in Fig. 2 that smooth curves through our data points for $S_x(300\text{ K})$ and $S_x(77\text{ K})$ are approximately proportional¹⁶ to $\partial\gamma/\partial x$ for $x > 1$. However, Eq. (2) cannot hold even approximately for $x < 1$. The implication then is that the states at the Fermi surface for compositions near LaSn_3 ($x < 1$) are more complicated than states for $x > 1$. Alternatively, the failure of Eq. (2) for $x < 1$ may be due to the breakdown of the rigid-band approximation which was

used to derive (2). Levin *et al.*¹⁷ have shown using the coherent-potential approximation (CPA) that $S_x(T)$ may not follow the rigid-band model predictions for certain potential or composition values.¹⁶ However, since Eq. (2) does hold approximately over two-thirds of the composition range, we chose to attribute the change of behavior near $x=1$ to a change in the character of the states at E_F . We note that the rapid fall of γ and T_c with increasing x for $x < 1$ might indicate the presence of small pockets of electrons in LaSn_3 which rapidly disappear as one lowers the electron concentration by replacing Sn atoms by In. Such a picture is consistent with the anomalous susceptibility in LaSn_3 as well as the unusual behavior of $S(T)$ vs x near LaSn_3 . This interpretation is also substantiated by $\rho(H)$ for $x=0.15, 0.3$, and 0.6 as discussed later.

The question of the presence of d bands in LaSn_3 is an important one. Indeed, Havinga has suggested the absence of d bands near the Fermi energy in $\text{LaSn}_{3-x}\text{In}_x$ and similar compounds.⁶ This model of the composition dependence of the properties of $\text{LaSn}_{3-x}\text{In}_x$ being due to a nearly spherical Fermi surface crossing Brillouin-zone boundaries can be shown to be untenable.¹⁸ Furthermore, Havinga can explain the variation of $S(300\text{ K})$ with x in his model only by assuming umklapp processes to dominate the scattering. However, our data for S_x at 77 K are proportional to $S_x(300\text{ K})$ (see Fig. 2) while if umklapp processes dominated, $S_x(300\text{ K})$ and $S_x(77\text{ K})$ should be very different since umklapp processes are so temperature dependent. Also, a nearly-free-electron picture predicts¹⁹ that $\rho_{\text{long}}(H)$ and $\rho_{\text{trans}}(H)$ are approximately the same size both in LaSn_3 and in LaIn_3 . Finally, plots of $\log[\rho(T) - \rho(4.2\text{ K})]$ vs $\log T$ show a low-temperature resistivity

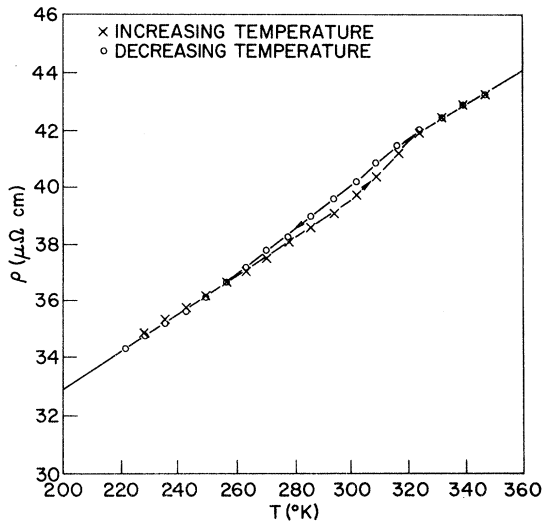


FIG. 9. $\rho(T)$ for $\text{LaSn}_{2.4}\text{In}_{0.6}$ for $200 \leq T \leq 350\text{ K}$.

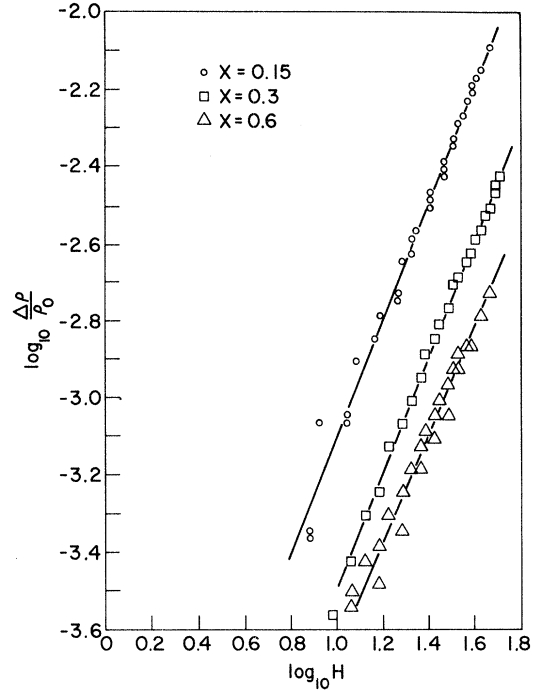


FIG. 10. Log-log plot of $\Delta\rho/\rho_0$ vs H for $\text{LaSn}_{3-x}\text{In}_x$ for $x=0.15, 0.3$, and 0.6 .

tivity proportional to T^n where n is 3 for $x=3$ and 0.6 , and $n=4$ for LaSn_3 . These values of the exponents are typical of transition metals²⁰ and are an important indicator that in $\text{LaSn}_{3-x}\text{In}_x$ there are d states at the Fermi energy.

Our measurements of $\rho(H)$ in LaSn_3 and LaIn_3 indicate that the density of holes and of electrons (n_h and n_e) are not temperature dependent below 50 K . This observation is substantiated for LaIn_3 by the fact that, within experimental error, R_H is temperature independent from liquid-helium to room temperature. In both materials the fact that Kohler's law holds up to 50 K (and perhaps higher) is evidence that n_h and n_e are temperature independent in this temperature range.

A phase transition was seen in $\text{LaSn}_{2.55}\text{In}_{0.45}$ near room temperature during measurements of $\rho(T)$ for this sample. Figure 9 shows the resistivity of this sample near room temperature both while heating and cooling through the transition. This behavior was reproducible and the hysteresis in this curve is evidence that we are observing a structural rather than an electronic phase transition.

Finally, we discuss our results for the liquid-helium temperature magnetoresistance in the intermediate composition compounds. Our results for $\Delta\rho/\rho_0$ for $x=0.15, 0.3$, and 0.6 at 4.2 K are shown in Fig. 10, where $\log_{10} \Delta\rho/\rho_0$ is plotted vs $\log_{10} H$. In the range $3 \times 10^{-4} < \Delta\rho/\rho_0 < 10^{-2}$, $\Delta\rho/\rho_0$

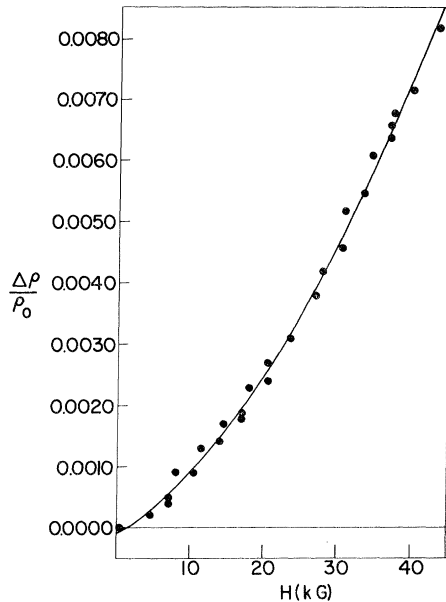


FIG. 11. Dots are experimental values of $\Delta\rho/\rho_0$ for $\text{LaSn}_{2.85}\text{In}_{0.15}$ as a function of H , while the line is the function $\Delta\rho/\rho_0 = -0.0001558 + 8.067 \times 10^{-5}H + 2.680 \times 10^{-6}H^2$.

cannot be fit by a curve which is proportional to H^2 . Rather, the slope of the lines in Fig. 10 is close to 1.5. These data can be fit well by

$$\Delta\rho/\rho_0 = AH + BH^2. \quad (3)$$

This is shown in Fig. 11 where we see the excellent fit to our data on $\text{LaSn}_{2.85}\text{In}_{0.15}$ for $A = 8.07 \times 10^{-5}/\text{kG}$ and $B = 2.68 \times 10^{-6}/\text{kG}^2$. (The slight displacement of the zero of the ordinate in Fig. 2 corresponds to a small error in extrapolating $\rho(H)$ to $H=0$ to determine ρ_0 and is not significant in this discussion.) The linear and quadratic terms become equal at 10–20 kG.

This data is difficult to fit on a one-band model. If the usual replacement of the collision term in the Boltzmann equation by $(f - f^0)/\tau$ is valid, which it is in this low-temperature impurity-scattering dominated regime, then the magnetoconductance $\sigma(H)$ is given by²¹

$$\sigma(H) = \sum \int d\epsilon \int \frac{dA}{|\nabla|} \vec{v} \cdot \vec{v} \tau \frac{\partial f^0}{\partial \epsilon} \frac{1}{1 + (\omega_c \tau)^2}. \quad (4)$$

In Eq. (4), the sum is over Fermi surface sheets, f^0 is the Fermi function, and A , v , τ , and ω_c are the area, velocity, collision time, and cyclotron frequency near the Fermi surface, respectively, while ϵ is the electron energy. If only one term in the sum in (4) contributed to $\sigma(H)$, then Eq. (4) would imply that $\sigma(H)$ should be proportional to H^2 in Figs. 10 and 11 since there $[\sigma(H) - \sigma_0]/\sigma_0 \ll 1$, where $\sigma_0 \equiv \sigma(H=0)$. Another more general way to see this is to note that $\sigma(H)$ is an even function for a crystal with inversion symmetry. Then only even powers of H can appear in the Jones-Zener expansion^{19,20} of $\sigma(H)$ so that $(\sigma - \sigma_0)/\sigma_0$ should vary as H^2 at low enough fields.

One way out of this dilemma is to invoke a scattering center that is not invariant under spatial inversion (for example, magnetic scattering centers). However, since there are many electrons per unit cell in these compounds, and the Fermi surface certainly consists of many sheets, it is simpler and more realistic to assume that there is a small number of carriers which are in the regime $\omega_c \tau \gg 1$ at even low fields in at least one of the terms in Eq. (4). It is easy to show that such a term in Eq. (4) will lead to a small linear magnetoresistance at low fields—small because there are few carriers involved, but linear because these carriers are in the large $\omega_c \tau$ regime at a few kilogauss owing to their possessing a fairly small effective mass. [Large $\omega_c \tau$ could imply $\rho(H)$ linear since Fig. 7 shows that LaSn_3 has $\rho(H)$ linear at large fields.] The joint requirements of small effective mass and small number of carriers is easily met if E_F is near a small zone boundary gap in an s - p band. Such small pockets have already been suggested earlier in this paper on the basis of $\rho_x(T)$ and $S_x(T)$ measurements in these compounds.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge many informative conversations with D. E. Eastman, R. J. Gambino, J. R. Schrieffer, P. E. Seiden, A. M. Toxen, and L. B. Welsh. I am also grateful for the fine technical assistance and data reduction provided by G. J. Padilla, and to R. J. Gambino and R. R. Ruf for providing the samples used in this study. The mass spectrometer analysis was provided by R. W. Johnson of the Analytical Chemistry Group at the IBM Research Center.

¹R. J. Gambino, N. R. Stemple, and A. M. Toxen, *J. Phys. Chem. Solids* **29**, 295 (1968).

²E. E. Havinga, *Phys. Letters* **28A**, 350 (1968).

³E. Bucher *et al.*, *Helv. Phys. Acta* **41**, 723 (1968).

⁴L. B. Welsh, R. J. Gambino, and A. M. Toxen, *J. Appl. Phys.* **42**, 1545 (1971).

⁵A. M. Toxen and R. J. Gambino, *Phys. Letters* **28A**, 214 (1968).

⁶E. E. Havinga, H. Damsma, and M. H. Van Maaren, *J. Phys. Chem. Solids* **31**, 2653 (1970); for γ vs x see Ref. 3.

⁷R. J. Gambino, A. M. Toxen, and B. J. van der Hoeven, in *Proceedings of the Twelfth International Low Temperature Conference, Kyoto, 1970*, edited by E. Kanda (Academic Press of Japan, Kyoto, 1971).

⁸A. M. Toxen, *Proceedings of the Conference on the*

Science of Superconductivity, Stanford, 1969 (unpublished).

⁹W. D. Grobman, J. Appl. Phys. **42**, 1456 (1971).

¹⁰A. M. Toxen and R. D. Parks (private communication).

¹¹See Figs. 10 and 11.

¹²O. Lindberg, Proc. IRE **40**, 1414 (1952).

¹³In addition, R. J. Gambino, W. D. Grobman, and A. M. Toxen (unpublished) will report on the Kondo behavior of CeIn₃.

¹⁴D. K. MacDonald, *Thermoelectricity: An Introduction to the Principles* (Wiley, New York, 1962), Sec. 4.1.1.

¹⁵L. B. Welsh, A. M. Toxen, and R. J. Gambino, Phys. Rev. (to be published).

¹⁶The use of γ rather than the bare density of states does not affect the validity of the argument since we are only

making qualitative arguments about the curves in Fig. 2. Equation (2) assumes the rigid-band model.

¹⁷K. Levin, B. Velicky, and E. Ehrenreich, Phys. Rev. B **2**, 1771 (1970).

¹⁸W. D. Grobman, J. Phys. Chem. Solids (to be published).

¹⁹J. M. Ziman, *Electrons and Phonons* (Oxford U. P., London, 1960).

²⁰G. Gladstone, M. A. Jensen, and J. R. Schrieffer, *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), Chap. 13.

²¹Reference 19, p. 487.

²²A. C. Smith, J. F. Janak, and R. B. Adler, *Electronic Conduction in Solids* (McGraw-Hill, New York, 1966), Sec. 9.1.

Existence of Mobility Edges in Anderson's Model for Random Lattices*

E. N. Economou[†] and Morrel H. Cohen

The James Franck Institute, The University of Chicago, Chicago, Illinois 60637

(Received 15 April 1971)

Anderson's theory of localization is critically reviewed and extended with particular emphasis on some controversial aspects. It is shown that when the randomness exceeds a certain critical value, all the eigenstates become localized in agreement with Anderson's original result. When the randomness is less than this critical value, the tails of a band consist of localized states. The character of the states changes sharply from localized to extended at mobility edges, in agreement with the Mott-CFO (Cohen-Fritzsche-Ovshinsky) model. As the randomness increases, the mobility edges move inwards into the band and they coincide at Anderson's critical value of the randomness. A criterion is developed which, under certain conditions, imposes upper limits on the extent of the portions of the energy spectrum consisting of extended states. These conditions are fulfilled exactly in the case of a Lorentzian distribution of single-site energies and approximately within the framework of any single-site approximation. Thus in the Lorentzian case upper bounds are obtained for the positions of the mobility edges and the critical value of the randomness for which Anderson's transition takes place. These results are in agreement with the Mott-CFO model.

I. INTRODUCTION

Considerable attention has been given recently to the problem of the electronic structure of disordered materials. Perhaps the most significant contribution to the field is Mott's notion¹ that there exist energies of sharp transition from localized states in the band tails to extended states in the interior of the band.¹⁻³ This notion is based on results obtained by Anderson in a remarkable paper entitled "The Absence of Diffusion in Certain Random Lattices"⁴ as well as some other results suggesting the existence of localized states in the extreme tails of the bands in disordered materials.⁵⁻¹²

Anderson⁴ considers what is essentially a tight-binding model, in which a single band is formed from *s*-like atomic orbitals with energies $\epsilon_{\vec{l}}$, corresponding to the site \vec{l} . The bandwidth is $B = 2VZ$, where V is an overlap energy integral and Z is the coordination number. Anderson introduces random-

ness into the system by assuming that the quantities $\epsilon_{\vec{l}}$ are random variables possessing a common distribution function with a width Γ around the mean value. He finds that there is a critical value $\Gamma_c \sim B \ln Z$ of Γ such that for $\Gamma \geq \Gamma_c$ the states at the middle of the band (and by inference all the states) are localized. On the other hand, for $\Gamma < \Gamma_c$ the states in the middle of the band are extended. Anderson has demonstrated that his results hold only if the off-diagonal matrix elements $V_{\vec{l}\vec{m}}$ are of range sufficiently short to give rise to bands of finite width. His main line of argument is as follows: The localizability of an eigenstate belonging to an eigenenergy $E = 0$ is related to the convergence of a renormalized perturbation expansion (RPE) for the self-energy $\Delta_{\vec{l}}(0)$. Since one is dealing with random quantities, the convergence is a matter of probability. We show here by comparison with a geometrical series that the series for $\Delta_{\vec{l}}(0)$ ($E = 0$ corresponds to the middle of the band) converges