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THERMOELECTRIC POWER ANOMALY IN n-TYPE InSb AT LOW TEMPERATURES

R. P. Khosla and R. J. Sladek

Department of Physics, Purdue University, Lafayette, Indiana
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In n-type InSb at liquid-helium temperatures the concentration of electrons in the conduction band is independent of temperature, and the electrical resistivity is almost so. This behavior is analogous to that of a normal metal in the residual resistance region, and therefore the simplest theory of electron diffusion predicts that, as for a metal, the thermoelectric power (TEP) of n-type InSb should be proportional to the absolute temperature.[1]

We have found that this prediction is not fulfilled for n-InSb samples measured between 1.6 and 4.9°K. Instead, the measured TEP is much larger and has a different temperature dependence. A description of this anomalous TEP and a possible interpretation of it will be given in this Letter.

The samples measured were of single-crystal material purchased from Cominco Products, Inc. Their high quality was indicated by (1) good agreement between Hall effect and resistivity measurements made on different sets of contacts on a given sample, (2) uniform resistivity measured by traverses at 77°K, (3) high Hall mobilities at 77°K, and (4) low impurity content found by commercial mass spectrographic analyses by Bell and Howell Research Center (made on only two of the samples). The samples were undoped, or doped with Te, and ranged in carrier concentration from 6.9×10^{13} cm^{-3} to 5.4×10^{18} cm^{-3} and were about 2 cm in length and from 0.09 cm^2 to 0.13 cm^2 in cross section. Different cross sections were used to allow detection of possible size effects.

For the TEP measurements the samples were suspended in an evacuated copper can immersed in liquid helium. A heater coil was wound on the bottom end of the sample. Two germanium resistance thermometers were glued to the samples (with GE varnish No. 7031) at two points along the length of the sample, which were away from the ends by several sample widths. The thermometers were calibrated against the vapor pressure of the liquid-helium bath with exchange gas in the can (and no heater current flowing). The correctness of the temperature differences was indicated by the fact that the thermal-conductivity values[2] obtained by using them were apparently correct, since for samples which were comparable in concentration and size to those measured by other investigators,[3] there was good agreement between our and their thermal-conductivity values. The thermoelectric voltages were measured with a Keithley 148 Nanovoltmeter with an accuracy of 2% full scale. The correctness of the voltages were confirmed by TEP measurements we made between 12 and 24°K, since our TEP values are consistent with the results of Puri and Geballe.[4]

The results of our measurements in the liquid-helium range are presented in Fig. 1. From Fig. 1, we see that the TEP of each sample has a nonlinear temperature dependence and exhibits a maximum at some temperature. In order to interpret these results, we first calculated the diffusion TEP for the case of Coulomb scattering by ionized impurities, Q_I, since such scattering seems to account very well for the mobilities[2] of our samples at liquid-helium temperatures. The formulas used were

$$Q_I = -\frac{k_B}{|e|}\left\{\frac{r+\frac{5}{2}}{r+\frac{3}{2}}\frac{F_{r+\frac{3}{2}}(\zeta/k_BT)}{F_{r+\frac{1}{2}}(\zeta/k_BT)}-\frac{\zeta}{k_BT}\right\} \quad (1)$$

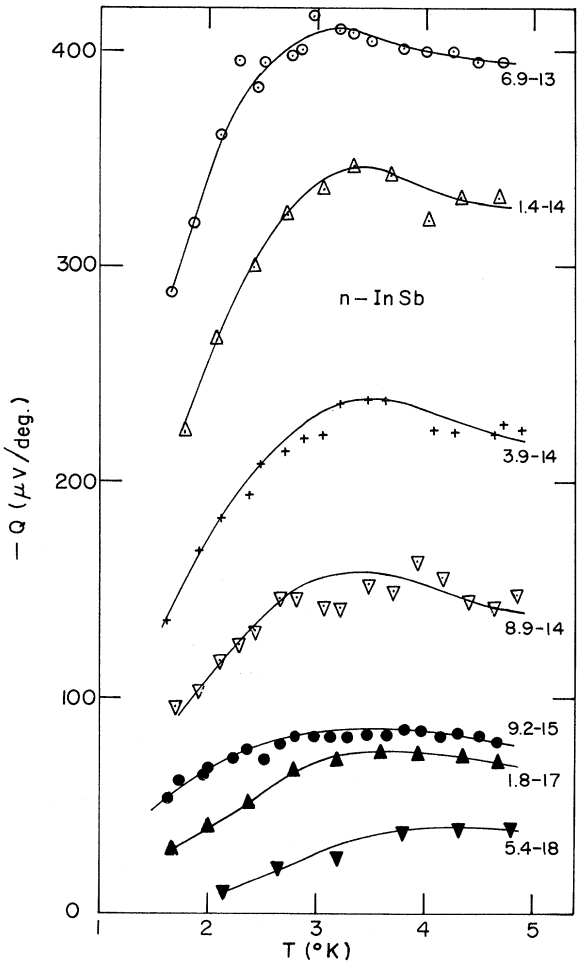

FIG. 1. Experimental thermoelectric power versus temperature for n-type InSb samples having the carrier concentrations indicated (6.9-13 stands for 6.9×10^{13} cm^{-3}, etc.). Smooth curves have been drawn through the experimental points.

for the noncompletely degenerate samples, and

$$Q_I = \frac{-\pi^2 k_B{}^2 T}{3|e|\zeta}\left\{\frac{d\ln n(E)}{d\ln E}+\frac{d\ln k^2}{d\ln E}+\frac{d\ln\tau}{d\ln E}\right\}_\zeta \qquad (2)$$

for the degenerate samples,[5] where r is the exponent in the scattering time, $\tau \sim E^r$, the $F_j(x)$'s are Fermi-Dirac integrals, ζ is the Fermi energy, $n(E)$ is the density of electronic energy states, k is the electron wave vector, and the other symbols have their usual meanings. We used $r=\frac{3}{2}$, and for calculations employing Eq. (2), used the nonparabolic conduction band of Kane.[6] Then subtraction of the calculated values of Q_I from the measured values of Q yielded values of the excess TEP, $-\Delta Q$.

The values of $-\Delta Q$ for each of our samples are plotted as a function of temperature in Fig. 2. Note that for each sample $-\Delta Q$ goes through a maximum, and at a given temperature $-\Delta Q$ is smaller, the higher the carrier concentration. This behavior is reminiscent of phonon-drag TEP.[7] However, we do not think that the excess TEP is due to the phonon-drag effect for the following reasons: First, we found that $-\Delta Q$ did not depend on the cross-sectional area of the samples, whereas the TEP due to phonon drag would be expected to be size dependent in our temperature range, since boundary scattering of phonons is important, as indicated by the fact that the ther-

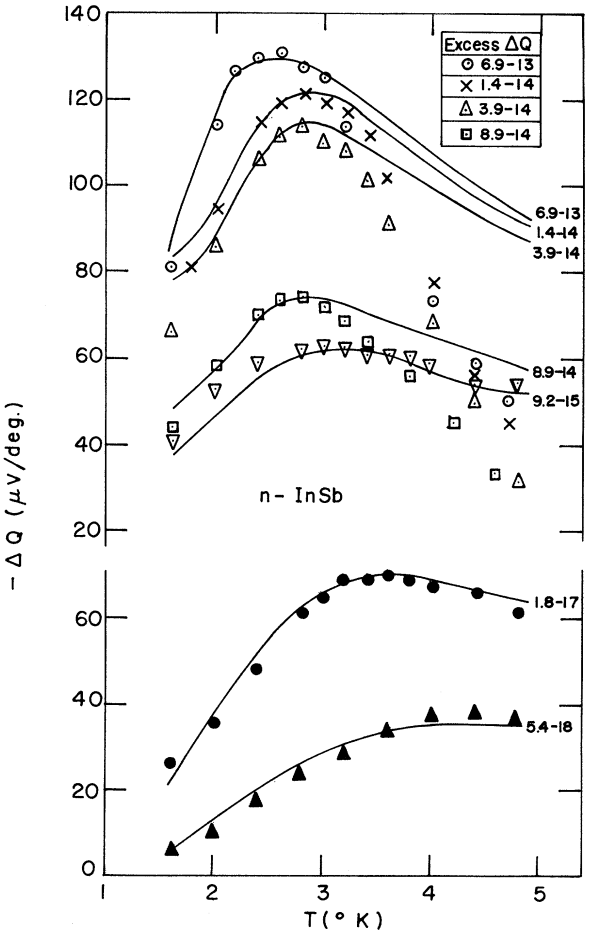

FIG. 2. Fit of Kasuya's exchange scattering term, shown by the curves, to the anomalous excess TEP, shown by the points.

mal conductivities of our samples did depend on their cross-sectional sizes. Second, Puri and Geballe[4] have found that even in n-InSb of highest purity for which phonon drag would be largest, the phonon-drag TEP was only about 50 μV/deg in the temperature range 10 to 30°K and was negligible outside this temperature range (in the absence of a strong magnetic field).

We suggest that the anomalous TEP is due to a highly energy- and temperature-dependent type of scattering. This scattering is most likely due to electrons localized in magnetic states around donor impurities. Toyozawa[8] has shown theoretically how a small fraction of the donors have such states which are occupied by electrons even though most of the donors are ionized.

Since previous experimental evidence for the magnetic states consists mainly of negative magnetoresistance effects at liquid-helium temperatures,[9] we have made magnetoresistance measurements on our samples also in order to detect them. We did indeed find negative magnetoresistance[2] which depended on carrier concentration and temperature as predicted by Toyozawa's theory indicating that localized magnetic states are present.

Although there is no theory specifically for the thermoelectric power associated with the presence of the magnetic states in semiconductors, there is a theory by Kasuya[10] for transition-metal alloys, which predicts a contribution to the TEP due to s-d exchange interactions which is much larger than the ordinary diffusion contribution and which goes through a maximum at low temperatures. Since, as shown in Fig. 2, $-\Delta Q$ for our n-InSb samples also exhibits this general type of behavior, we infer that the localized spin states influence the TEP of n-InSb in a manner which is similar to the effect of d states on the TEP of transition-metal alloys. Therefore, to our $-\Delta Q$ values we have fitted curves obtained from Kasuya's expression[10]

$$Q_{sd}=\frac{2k_B}{|e|}(g-1)^3\left(\frac{J_0}{A_0}\right)^3\langle j_Z\rangle \times(j^2+j-\langle j_Z^2\rangle-\langle j_Z\rangle)\frac{x^2}{1-e^{-x}}, \qquad (3)$$

where g is the Lande g factor, J_0 is the s-d exchange integral, A_0 is the s-s Coulomb-scattering integral, j_Z is the value of the Z component of total angular momentum j, and $x=H_0/k_BT$, where H_0 is the molecular field. For applying Eq. (3) to n-InSb we have calculated values for g (listed in Table I) from a formula by Roth, Lax, and Zwerdling[11] using material constants for InSb; computed the various functions of j using Kasuya's expressions and $j=\frac{1}{2}$; and regarded J_0/A_0 and H_0 as parameters to be determined by our fits. The curves calculated from Eq. (3) are shown in Fig. 2, and it can be seen that good fits are obtainable for our more degenerate samples and even for our less degenerate samples up to 3°K. The values we found for H_0 and J_0/A_0 (given in Table I) are approximately independent of carrier concentration at low concentrations, but increase with carrier concentration at large concentrations. The magnitude of the J_0/A_0 values is quite reasonable in view of the value of 0.028 obtained for n-type germanium (doped with Sb) by Hedgecock and Mathur[12] from magnetoresistance data. With this J_0/A_0 value and Kasuya's theory, these authors were also able to account for excess TEP which they observed in their Ge samples at liquid-helium temperatures and which they attributed to Toyozawa's localized spin states.

Thus, we conclude that the excess TEP which we have discovered in n-InSb is due to scattering by electrons in localized spin states and, for most samples, can be accounted for quantitatively by employing a theory developed for the TEP due to s-d exchange scattering in alloys.

Some features of the excess TEP which we cannot account for at present, e.g., the discrepancy between the excess TEP and that

Table I. Parameters used to fit Kasuya's exchange term to the anomalous TEP in n-InSb at liquid-helium temperatures.

Sample[a]	g	H_0 (10^{-4} eV)	$-\frac{J_0}{A_0}$
6.9-13	51.7	5.3	0.030
1.4-14	51.2	5.9	0.029
3.9-14	50.8	5.9	0.029
8.9-14	50.3	5.9	0.025
9.2-15	47.4	6.4	0.026
1.8-17	30.1	6.9	0.042
5.4-18	14.2	8.7	0.073

[a]The sample numbers indicate the exhaustion carrier concentration; e.g., 6.9-13 means 6.9×10^{13} cm^{-3}, etc.

calculated from Kasuya's formula for the most pure samples above 3.0°K and the dependence of H_0 and J_0/A_0 on carrier concentration, are under investigation.

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NEW MECHANISM FOR SUPERCONDUCTIVITY*

W. Kohn

University of California, San Diego, La Jolla, California

and

J. M. Luttinger

Columbia University, New York, New York

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It is the purpose of this note to point out a new mechanism which provides an instability against Cooper-pair formation. We find that a weakly interacting system of fermions cannot remain normal down to the absolute zero of temperature, no matter what the form of the interaction. This mechanism has nothing to do with the conventional electron-phonon attractive interaction in metals, or the long-range attractive van der Waals forces in He^3. It is present even in the case of purely repulsive forces between the particles, and is due to the sharpness of the Fermi surface for the normal system.

To understand what is involved, we first take an over-simplified view of the effect. It has long been known[1] that if a charge is placed in a metal, the screening is such that there remains a long-range oscillatory potential of the form $\cos(2k_F r+\varphi)/r^3$ (k_F is the Fermi momentum). This leads to a long-range interaction between charges. Formally, the source of this long-range force is the singularity of the dielectric constant as a function of the momentum transfer $\vec{q}$, when $q=2k_F$.[1] This singularity in the Fourier transform of the interaction gives rise to a long-ranged oscillatory force in ordinary space. All that is necessary for this effect is a sharp Fermi surface; a rounding of the Fermi surface due to (say) finite temperature or impurities will give rise to an interaction which drops off exponentially at very large distances.

It is plausible to suppose that, similarly, the effective interaction between the fermions themselves will have a long-range oscillatory part. By taking advantage of the attractive regions, Cooper pairs can form thus giving rise to superconductivity.

To investigate this possibility more systematically we consider the following model: an isotropic system of spin-$\frac{1}{2}$ fermions with weak short-range forces between them. The criterion we use for the onset of superconductivity is that the scattering amplitude for pairs of quasiparticles of equal and opposite momenta