XIII-4. Transport Properties and Localized Spins in *n*-Type InSb^{*}

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Resistivity, magnetoresistance, Hall effect, and thermal conductivity measurements have been made on *n*-type InSb samples at liquid helium temperatures. Thermal resistance in excess of that attributable to boundary scattering of phonons and negative magnetoresistance were observed. These results are interpreted as due to electrons which are quasi-localized around some of the impurities scattering conduction electrons and phonons by means of a spin flip process. Curie temperatures and concentrations of localized spins are deduced for some of the samples.

§1. Introduction

Although InSb is probably the most thoroughly investigated of the III-V compound semiconductors, the scattering mechanisms in this material at low temperatures were still, we felt, in need of elucidation. In an attempt to understand these mechanisms in n-type InSb, we have made a detailed experimental study of the thermoelectric power, Q, the thermal conductivity, K, the resistivity, ρ_0 , the magnetoresistance, $\Delta \rho / \rho_0$, and the Hall coefficient, R_H , in samples having electron concentrations from 6.9×10^{13} to 5.4×10^{18} cm^{-3} . The Q data were reported previously in a paper¹⁾ referred to henceforth as I. These data revealed that the Q of each sample had an anomalous temperature dependence and (large) magnitude which was attributed to the mutual drag between conduction electrons of opposite spin due to scattering by electrons localized around some of the donor impurities. Such localized spins were first suggested by Toyozawa²⁾ to explain negative magnetoresistance measured by Sasaki et al.³⁾ on degenerate n-type Ge samples doped with Sb. In such Ge samples Hedgecock and Mathur⁴⁾ have observed anomalies in the thermoelectric power which are presumably of the same origin but which are much smaller in magnitude than those we observed in n-InSb.

Evidence for the effects of localized spins on the other transport properties of n-InSb which we measured will be presented after a brief comment on experimental matters.

The experimental arrangement and the sample holder for measuring the thermal conductivity were the same as described in I, the details of which are given by Khosla.⁵⁾ For measuring ρ_0 , $\Delta \rho / \rho_0$, and R_H a conventional type of sample holder was suspended in a liquid helium Dewar system with a 4" Varian Electromagnet providing transverse magnetic fields up to 6 kG as measured with a Rawson rotating coil fluxmeter. The samples were the same as those used in I. Voltages across the samples and the germanium resistance thermometers were measured potentiometrically.

§ 2. Magnetoresistance

For samples with carrier concentrations $\leq 9 \times 10^{15}$ cm⁻³ $\Delta \rho / \rho_0$ was negative at low fields, passed through a small minimum $\leq 5\%$ below 1 kG, and then became less negative as the field increased further. For higher concentrations $\Delta \rho / \rho_0$ was positive or oscillatory. Analysis of our data revealed that the negative component of the magnetoresistance, $(\Delta \rho / \rho_0)_{neg.}$, was generally smaller the higher the carrier concentration and the higher the temperature (between 1.3°K and 4.2°K). Since this is the behavior predicted by Toyozawa²⁾ for scattering by localized spins, we attribute $(\Delta \rho / \rho_0)_{neg.}$ to such scattering.

From Fig. 1 it can be seen that $(\Delta \rho / \rho_0)_{neg.}$ is a function of $B/(T-\theta)$ as Sasaki⁶⁾ found for *n*-Ge. Our values of θ , listed in Table I, are positive (in contrast to his) and increase gradually as the carrier concentration increases. For our samples having 3.9×10^{14} to 9.2×10^{15} carriers/cm³ the plots of $(\Delta \rho / \rho_0)_{neg.}$ versus $B/(T-\theta)$ tended to saturate. Interpreting this to mean the disappearance of the extra resistance due to localized spins, $\Delta \rho_s$, we can take $\Delta \rho_s = -\rho_0 (\Delta \rho / \rho_0)_{neg.satur.}$. Then we used these values of $\Delta \rho_s$ to deduce the concentration of localized spins, n_s , from a theoretical formula for dilute alloys by Kasuya⁷

$$\Delta \rho_s = 3\pi m^* n_s (g-1)^2 j J_0^2 / 2\hbar \zeta_0 n^2 e^2 , \qquad (1)$$

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Fig. 1. The negative component of the magnetoresistance as a function of the magnetic induction divided by the difference in the absolute temperature and a characteristic temperature, θ , chosen so data for different temperatures (closed or open points) fall on the same curve. The values of θ are listed in Table I. The numbers indicate carrier concentration. Thus 9.2-15 indicates 9.2×10^{15} cm⁻³.

$n (10^{14} \mathrm{cm}^{-3})$	m^*/m_0	(10^{ζ_0}eV)	J_0/A_0^\dagger	$ \begin{array}{c} J_0 \\ (10^{-5} \mathrm{eV}) \end{array} $	$-g^{\dagger}$	$(10^{-3}\Omega \text{cm})$	n_s (10 ¹⁴)	n_s/n (%)	θ (°K)
0.69	.0130	0.47	.029	1.82	51.7	No sat.			0.10
1.4	.0131	0.76	.028	1.76	51.2	No sat.	_		0.14
3.9	.0132	1.5	.028	1.76	50.8	10.7	.17	4.5	0.20
8.9	.0133	2.6	.024	1.51	50.3	1.77	.20	2.3	0.25

44.2

1.70

Table I. Various parameters for samples exhibiting negative magnetoresistance.

[†] The signs of these quantities were given incorrectly in ref. 1).

.027

where g is the electron g factor and is very large in magnitude in n-InSb; j is the quantum number which, as in I, we shall take=1/2; J_0 is the exchange integral between a conduction and a localized electron; ζ_0 is the Fermi energy; n is the electron concentration; and the other symbols have their usual meanings. Values for J_0 are obtained by multiplying the J_0/A_0 values found in I by assuming, following Toyozawa,²⁾ that the Coulomb integral A_0 is about 0.9 the donor ionization energy for which we take the effective mass theory value of .00069 eV. The values of n_s obtained using eq. (1) are given in Table I. Note that n_s increases with carrier concentration, n, but n_s/n decreases with increasing *n* in accord with Toyozawa's theory.²⁾ Other theoretical expressions for the anomalous resistivity in dilute alloys^{8,9)} do not contain the $(g-1)^2$ factor in eq. (1) and thus require a very large "quantum number" to give reasonable values of n_s . Our objection to using such a viewpoint is

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that we found it impossible to fit our thermoelectric data using it with reasonable values for the other parameters.

.28

0.30

0.35

§3. Hall Coefficient

0.025

For each sample the value of R_H at liquid helium temperature was about equal to that at 77°K (except in the lowest concentration samples which exhibit some magnetic freezeout¹⁰ at liquid helium temperatures at fields above about 1.5 kG) indicating either that the fraction of carriers in localized spin states, n_s/n , is small or that these states are scattering rather than more conventional bound states. We did deduce small values of n_s/n from the magnetoresistance data (see Table I). However, if these values are reasonably accurate, an observable effect on R_H should have occurred if the localized spins were "bound". Since none was seen, we tentatively infer that the localized spin states are scattering states rather than conventional bound states.

92

.0150

§4. Thermal Conductivity

Our measured K values indicate that heat conduction is by phonons and that, for the lowest concentration samples below 3°K all the scattering is due to the crystal boundaries. We deduce this from the fact that use of the measured sample dimensions (including the correction for finite sample length) in the theoretical formula for diffuse scattering at the boundaries yielded values of K which are in quantitative argeement with our measured values. (Such agreement has been found for a number of very pure semiconductors by Holland.¹¹⁾) However, additional thermal resistance appears at higher temperatures in our lowest concentration samples and is important at all measurement temperatures in our higher concentration samples. Challis et al.¹²⁾ have also measured the thermal conductivity of InSb samples having a large range of carrier concentrations, but they interpreted their results to mean that 50% of the phonons are specularly reflected and that the extra thermal resistance which cannot be accounted for by boundary scattering is due to scattering of phonons by band electrons as worked out for semiconductors by Ziman¹³⁾.

Analysis shows that the additional thermal



Fig. 2. The mean free path deduced from the additional thermal resistance as a function of temperature. The mean free paths for boundary scattering, calculated from the dimensions¹¹) were between 0.34 cm and 0.43 cm for all samples so that values of $L_{add, W} < 0.3$ cm are most significant.

resistance, add. W, of our *n*-InSb samples can be characterized by a mean free path, $L_{add.W}$, which decreases strongly with increasing temperature for all samples at the higher temperatures and has a more complicated dependence at lower temperatures as shown in Fig. 2.

From Fig. 2 it can be seen that above 2.5 or 3.0°K exponential dependences occur. It is not clear how significant these exponentials are, since in this temperature range, $L_{add, W}$, can be represented almost as well by functions of the form T^{-x} where 2.5 < x < 3.9 for the various samples. In any case, neither the exponentials nor the power laws agree with the theoretical temperature dependence for the mean free path of phonons scattered by ordinary conduction electrons.¹³⁾ In addition, the magnitudes of our observed Ladd. W values are very much smaller than those predicted by Ziman's theory. Thus we do not believe ordinary electron scattering of phonons can account for our results. Estimates of the thermal resistance due to isotope and other point impurity scattering¹¹⁾ based on theoretical formulas indicated that such mechanisms are apparently not important enough to explain our add. W either.

In order to explain our add. W we note that the temperature dependences we observed for $L_{\text{add. }W}$ are similar to those of spin relaxation times.^{14–16)} Thus we attribute our add. W to scattering of phonons by localized spins. Another argument in favor of this is that, at least at the highest temperatures as can be seen from Fig. 2, $L_{add, W}$ depends much less on carrier concentration, n, at low n than it does at high n, and the concentration of localized spins, n_s , has the same general type of behavior. (See Table I) Quantitative interpretation of out $L_{\text{add.}W}$ awaits a theory of scattering of phonons by localized spins. It is interesting to point out that perhaps phonon-localized spin scattering may be responsible for the unexplained additional thermal resistance observed in a number of other semiconductors also.¹¹⁾

From this investigation it can be concluded that localized spins affect the magnetoresistance and the thermal conductivity of *n*-InSb at low temperatures as well as producing a thermoelectric power anomaly.¹⁾ From $\Delta \rho / \rho_0$ apparently reasonable values of the concentration of localized spins and a Curie temperature, θ , can be deduced for each of a number of samples. Since the θ values are positive, it appears that the localized spins tend toward ferromagnetic alignment at lowest temperatures as is also suggested by the thermoelectric power data in I.

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DISCUSSION

Tanaka, S.: We have measured the electrical resistivity and the magnetoresistance of *n*-InSb whose carrier concentration are more than 1.8×10^{14} /cc from 4.2 to 0.1° K, and we have found resistance anomaly and negative magnetoresistance. The temperature dependence of the resistivity was analysed by Kondo's theory. The exchange integral J is negative and is estimated to be -1 meV, which is quite different from your results. The magnetoresistance at low magnetic field is negative and does not depend on the direction of the field, which indicates the existence of the antiferromagnetic interaction between localized spins.

Sladek, R. J.: The discrepancy in the sign and magnitude of J may be due to the fact that we obtained our value in a different way, using thermoelectric power data, although both methods should presumably yield the same results. For a positive J, Kondo's theory predicts different temperature dependence of resistivity from that observed by us down to 1.3° K, which is in qualitative agreement with your results. We suggest that the temperature dependence of resistivity may not be due to the Kondo effect.