feature of the theory, is found to occur almost at the correct value of the field. A remarkable feature of the data is that for sine-wave modulation at low-fields, the second-harmonic spectra are nearly the energy derivative of the first harmonic. This suggests that photon-assisted tunneling in nonuniform fields can be effectively described by an almost rigid lower-energy shift of the edge.

In addition to an accurate determination of the surface field at any value of the applied electrode voltage, Hall-effect and conductivity data have enabled the direct measurement of the effective reduced mobility of electrons in the surface space-charge region. The results indicate that at large band bendings, the random scattering model discussed by Schrieffer[16] is the overwhelming factor controlling the motion of electrons near the surface. Closer to the flat band condition, impurity scattering associated with positively charged surface states may be present. A new effect, which we may term inverted-sign field-effect mobility, has been observed and attributed to a strong mechanism of hole trapping. It is believed that the surface presents a very high density of hole traps created by the particular electropolishing treatment applied to the sample.

ACKNOWLEDGMENTS

The authors are greatly indebted to F. Evangelisti and B. Fornari for assistance during the experiment and to Patricia Giacomoni for drawing the figures.

PHYSICAL REVIEW VOLUME 182, NUMBER 3 15 JUNE 1969

Carrier-Concentration Dependence of Electron-Phonon Scattering in Te-Doped GaSb at Low Temperature

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(Received 16 December 1968)

The thermal conductivity K between 5 and 100°K was measured on Te-doped samples with excess donor concentrations n ranging between $\sim 10^{17}$ and 2×10^{18} cm^{-3}. At temperatures below that of the peak, K was observed to decrease with increasing n, this behavior being associated to electron-phonon scattering. The dependence of K on n was investigated by calculating the additional thermal resistivity $W_{ep}=1/K-1/K_0$ at 6°K, where K and K_0 are, respectively, the experimental and theoretical values of the thermal conductivity. The theoretical conductivity was deduced from the Callaway model, using as parameters the Casimir-boundary mean free path, point-defect scattering calculated from the Klemens relation, and phonon-phonon scattering deduced empirically from the results at higher temperatures. W_{ep} was found to vary approximately as $n_{300°K}^{1.7}$ or $n_{\sim 6°K}^{2.2}$. The excess thermal resistivity is most likely due to scattering of phonons by an electron gas. On the basis of the Ziman model, it is suggested that the observed $W_{ep}(n)$ behavior may arise from a variation of the effective mass m^* with n, due to the nonparabolic (000) band. Tentatively, an alternative argument is considered. The low-temperature thermal conductivity of undoped p-type samples with hole concentrations of about 1.5×10^{17} cm^{-3} was found to be much lower than that of Te-doped samples with comparable electron concentrations. This indicates that the strength of hole-phonon scattering in undoped material is more pronounced than that of electron-phonon scattering in Te-doped material.

INTRODUCTION

In doped semiconductors, the decrease of the thermal conductivity K at temperatures below that of the peak, in comparison to boundary[1] and isotope effects,[2,3] has been shown to arise from electron-phonon scattering Particularly, the carrier-concentration dependence of K was investigated by Carruthers *et al.*[4] in p-type germanium, by Goff and Pearlman[5] and by Albany and Laurence[6] in n-type germanium, and by Challis *et al.*[7] in p-type InSb. In addition, such a scattering mechanism was found by Vook[8] and by Albany and Vandevyver[9,10] to be strongly effective in irradiated germanium. Theoretical works have been carried out by several authors. Ziman's model[11] relates to scattering of phonons by an electron gas, and has been applied to highly doped semiconductors where the impurity levels

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merge with the conduction or valence band. Keyes[12] and Griffin and Carruthers[13] have analyzed the case of isolated impurity states in Sb- and As-doped Ge, where scattering of phonons is due to transitions of bound electrons between singlet ground states and first triplet states arising from valley-orbit splitting. In these two models, the electron-phonon scattering depends on the number of occupied donors. Moreover, Gaur and Verma[14] have discussed the results of Goff and Pearlman on the basis of the models of Ziman and of Griffin and Carruthers. On the other hand, Pyle[15] has analyzed the phonon scattering involved in jumps of carriers from occupied impurity states to unoccupied neighbors. In this mechanism, the scattering depends on the degree of compensation.

In GaSb, Holland[16] has observed on a few n- and p-type samples a large depression of the low-temperature thermal conductivity, which was associated with electron-phonon scattering. In the present work, we have investigated the donor-concentration dependence of the low-temperature thermal conductivity in Te-doped GaSb, and compared the results we obtained from undoped p-type crystals with the behavior of n-type samples having comparable electron concentrations.

EXPERIMENTAL

The thermal conductivity K was measured between 5 and 100°K, using the steady-state longitudinal heat-flow technique described elsewhere.[17] K was calculated from the relation $Q=K(s/l)\Delta T$ where Q is the heat power given by an electric heater, s is the cross-sectional area of a rectangular parallelepiped sample, l is the distance between the two thermometers fixed upon the latter, and ΔT is the temperature difference measured along the distance l, using carbon resistors and gold-cobalt thermocouples. The measurement uncertainty was estimated to be $\pm 10\%$ in the temperature range 4–20°K, and less than $\pm 15\%$ at higher temperatures.

The n-type crystals were grown by the Czochralski technique, using stoichiometric melts with sufficient tellurium to produce n-type material. The excess donor concentrations in the Te-doped samples used were in the range $\sim 10^{17}$ to 2×10^{18} cm^{-3}. Residual acceptors, which are believed to be due to lattice defects[18–22] rather than to chemical impurities, are expected to be present in concentrations of about $(1\text{–}2)\times 10^{17}$ cm^{-3}. Undoped single crystals were also prepared from stoichiometric melts, exhibiting hole concentrations of about 1.5×10^{17} cm^{-3}.

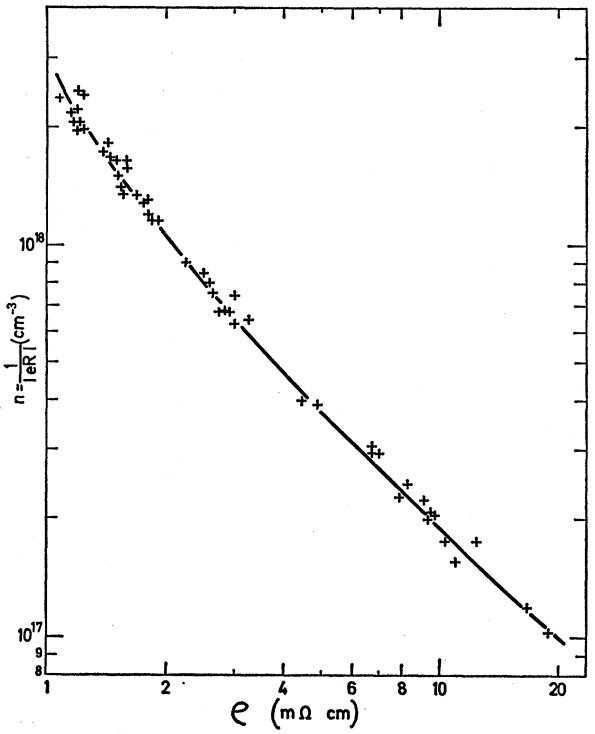

FIG. 1. Variation of the apparent electron concentration $n=1/|eR|$ as a function of electrical resistivity ρ (10^{-3} Ω cm) in n-type Te-doped GaSb at room temperature.

The electrical properties of n-type GaSb have been studied by a number of investigators.[23–27] A certain discrepancy can be observed between the published room-temperature values of electrical resistivity ρ and apparent carrier concentration n. For the Te-doped crystals that we used, the relation between $\rho_{300°K}$ and $n_{300°K}$ is shown in Fig. 1. The apparent carrier concentration $n_{300°K}$ corresponds to $n_{300°K}=1/|eR_{300°K}|$, where e is the electron charge and R is the Hall coefficient, and where the Hall factor was assumed equal to 1. Electrical resistivity and Hall coefficient measurements were performed by standard dc technique on four-armed samples cut from the grown crystals. The observed relation $\rho(n)$ is in satisfactory agreement with the data of Long and Hager.[25] The samples cut for thermal-conductivity studies were rectangular parallelepipeds,[17] with their axes approximately in the ⟨111⟩ crystal direction. They were distinguished at room temperature by their electrical resistivities measured

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TABLE I. Room-temperature resistivities $\rho_{300°K}$, apparent carrier concentrations $n_{300°K}=1/|eR_{300°K}|$, and Casimir lengths L [given by $L=2\pi^{-1/2}(l_1l_2)^{1/2}$, where l_1l_2 is the cross section] of the GaSb samples used in thermal-conductivity measurements.

Sample (type, code)	$\rho_{300°K}$ ($10^{-3}\,\Omega$ cm)	$n_{300°K}$ (cm^{-3})	L (cm)
n, 40f	1.3	1.8×10^{18}	0.252
n, 40d	1.4	1.7×10^{18}	0.252
n, 76g	1.45	1.6×10^{18}	0.306
n, 76f	1.5	1.5×10^{18}	0.305
n, $47f_2$	1.8	1.2×10^{18}	0.307
n, 74a	2.4	8.6×10^{17}	0.45
n, 57f	6.9	2.7×10^{17}	0.307
n, 34a	8.6	2.1×10^{17}	0.337
n, 59Md	1.5	1.5×10^{18}	0.314
n, 40c	1.2	2×10^{18}	0.466
n, $47e_1$	1.6	1.4×10^{18}	0.539
n, 17b	2.2	10^{18}	0.331
n, $47e_2$	2.0	1.1×10^{18}	0.312
n, $58Gb_1$	2.4	8.6×10^{17}	0.305
n, 57e	6.5	3×10^{17}	0.309
n, 34c	8.2	2.2×10^{17}	0.391
n, 34d	11.6	1.6×10^{17}	0.425
p, 29f	58	1.5×10^{17}	0.552
p, 29b	59	1.4×10^{17}	0.396
p, 29c	59	1.4×10^{17}	0.418

by a four-probe technique. The corresponding apparent carrier concentrations at room temperature were deduced from the relation $\rho(n)$ shown in Fig. 1. The parameters of the samples used for thermal-conductivity measurements are listed in Table I. The latter contains the equivalent sample size L (Casimir length) which is given by $L=2\pi^{-1/2}(l_1l_2)^{1/2}$, where l_1l_2 is the cross section of the sample. This parameter will be used as explained later.

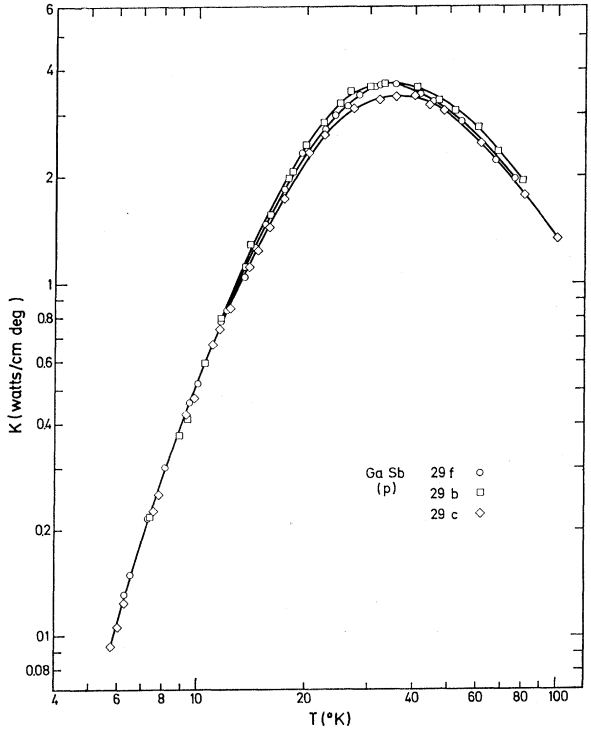

FIG. 2. Variation of the thermal conductivity K as a function of temperature for three samples of undoped GaSb.

The variation of K as a function of temperature for a few undoped p-type samples is shown in Fig. 2. For clarification, we have plotted in Figs. 3(a) and 3(b) the results for the Te-doped samples used. In the latter, the low-temperature thermal conductivity was observed to decrease with increasing donor concentration. In order to investigate this dependence, we have compared the experimental results at temperatures below that of the peak to the theoretical data calculated on the basis of the Callaway model,[28] which has been successfully used.[29] From this procedure, one may estimate the thermal resistance due to scattering mechanisms included in the model, and thus deduce the contribution of electron-phonon scattering to the experimental thermal resistance measured in the materials studied. Holland's analysis,[30] which considers the heat conduction by transverse and longitudinal phonons, was found to be more adequate in the high-temperature region above the maximum. This formulation, which gives, however, the same results as that of Callaway in the boundary and mass-difference scattering region, was not applied here.

ANALYSIS OF RESULTS AND DISCUSSION

In the absence of electron-phonon scattering, boundary and mass-difference scatterings are dominant in the temperature range below the peak. However, as the low-temperature measurements were limited to $\sim5°K$ in the present work, it was felt more appropriate to evaluate the contribution of phonon-phonon interaction. The Callaway expression[28] of K includes the relaxation times of three scattering mechanisms: boundary effect ($\tau_b^{-1}=v/L$), mass-difference scattering ($\tau_d^{-1}=Aw^4$), and three-phonon interaction, where the relaxation time for umklapp (τ_U) and normal (τ_N) processes are assumed to be of the form $\tau_U^{-1}=B_1w^2T^3$ and $\tau_N^{-1}=B_2w^2T^3$. The total relaxation time τ_c ($\tau_c^{-1}=\sum_i\tau_i^{-1}$) is given by

$$\tau_c^{-1}=v/L+Aw^4+(B_1+B_2)w^2T^3.$$

In these relations, v represents the average phonon velocity, taken equal to 3.09×10^5 cm/sec in GaSb.[16] The Casimir length L [$=2\pi^{-1/2}(l_1l_2)^{1/2}$] was deduced for each sample from its cross section l_1l_2. The values of L for all the samples used ranged between 0.25 and 0.55 cm. The mass-difference parameter $A=0.813\times10^{-44}$ sec^3 was deduced[16] from Klemens relation[2] applied to a compound. The parameter B_1+B_2 was estimated from high-temperature results above the peak, where phonon-phonon scattering predominates. The simpler Callaway expression for K was found to

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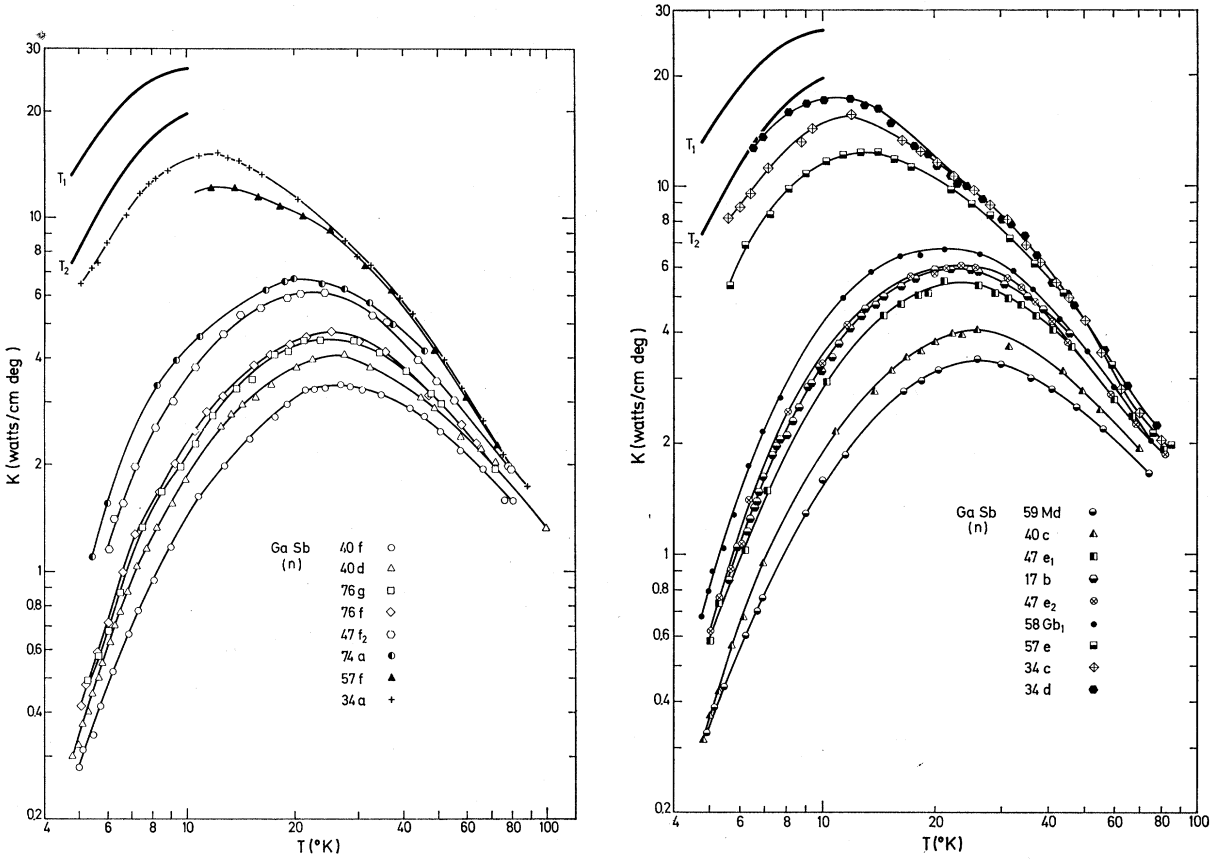

(a) (b)

FIG. 3. Variation of the thermal conductivity K as a function of temperature in n-type GaSb. Experimental curves correspond to Te-doped samples with excess donor concentrations ranging between $\sim 1.5\times10^{17}$ and 2×10^{18} cm^{-3}. Theoretical curves T_1 and T_2 were calculated on the basis of Callaway equation, using for the mass-difference scattering parameter the calculated value $A=0.813\times10^{-44}$ sec^3; for the phonon-phonon interaction parameter, the fitted value $B_1+B_2=1.1\times10^{-22}$ sec deg^{-3}; and for size effect, the Casimir length $L_1=0.55$ cm (for T_1) and $L_2=0.25$ cm (for T_2), which correspond to the highest and lowest values of L for the samples used.

be inadequate to fit the experimental data in this temperature range. Therefore we used, as previously described,[31–33] the general Callaway expression[28]

$$K=(k/2\pi^2v)(I_1+\beta I_2).$$

The term $(k/2\pi^2v)\,I_1$ is the simpler Callaway expression, generally used from the lower temperatures to just above the peak. The second term, which is due to the conservative nature of normal processes, is negligible at low temperatures below the peak, but becomes important above the latter,

$$I_1=\left(\frac{kT}{\hbar}\right)^3\int_0^{\theta/T}\tau_c\frac{x^4e^x}{(e^x-1)^2}dx,$$

$$I_2=\left(\frac{kT}{\hbar}\right)^3\int_0^{\theta/T}\frac{\tau_c}{\tau_N}\frac{x^4e^x}{(e^x-1)^2}dx,$$

and

$$\beta=\int_0^{\theta/T}\frac{\tau_c}{\tau_N}\frac{x^4e^x}{(e^x-1)^2}dx\Big/\int_0^{\theta/T}\frac{1}{\tau_N}\left(1-\frac{\tau_c}{\tau_N}\right)\frac{x^4e^x}{(e^x-1)^2}dx,$$

with $x=\hbar w/kT$. The factors k, $\hbar$, and Θ are, respectively, Boltzmann constant, Planck constant, and Debye temperature ($\Theta=270$°K).

A satisfactory fit was computed for high-temperature results (30–100°K) obtained on highly compensated samples which exhibited the highest K in this temperature range. Using the calculated value for A and the values L for the samples considered (the effect of the latter being almost negligible), the fit was performed, as shown in Fig. 4, for $B_1+B_2=1.1\times10^{-22}$ sec deg^{-3} and $B_1/(B_1+B_2)=0.25$. Therefore, the above values of A and B_1+B_2, and the Casimir length, corresponding to each sample, were used to compute the thermal conductivity in the low-temperature side of the peak (<10°K). This is shown in Figs. 3(a) and 3(b) for the two samples with the highest and lowest

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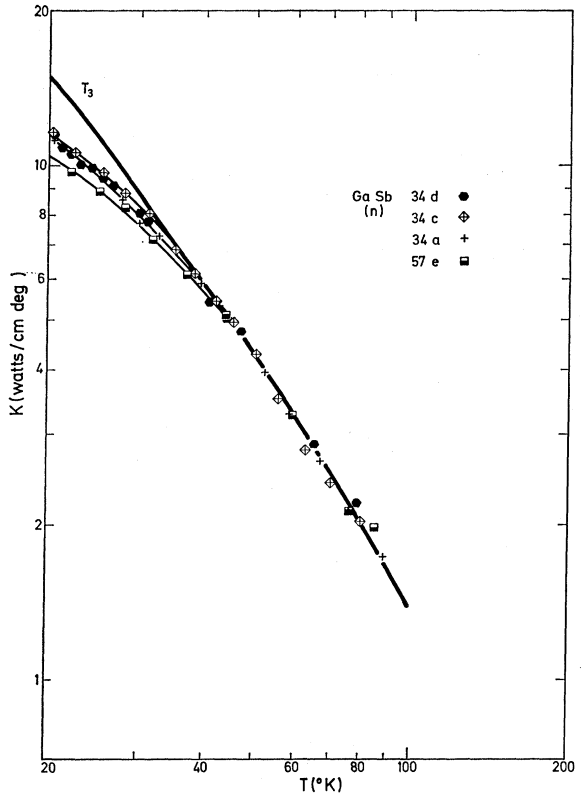

FIG. 4. Fit of the high-temperature thermal conductivity of n-type samples with lowest excess electron concentrations [$(1.6–3)\times10^{17}$ cm^{-3}], using the general Callaway equation. The fitted curve T_3 was obtained for the calculated value $A=0.813\times10^{-44}$ sec^3 and the adjusted values $B_1+B_2=1.1\times10^{-22}$ sec deg^{-3} and $B_1/(B_1+B_2)=0.25$.

L values ($L=0.25$ and 0.55 cm). The general Callaway expression gives below ~8°K the same results as the simpler expression, and the values at 10°K are ~4% higher than the data deduced from the latter. It must be pointed out that, if one considers only boundary and mass-difference scatterings, the corresponding values of K at about 5–6°K are nearly 10% higher than the above calculated data which include phonon processes. However, this difference increases rapidly with increasing temperature, reaching about 40% at 10°K. It can be seen that the experimental curves are lower than the calculated data. This depression rises with increasing donor concentration n, representing nearly a factor 25 for n about 10^{18} cm^{-3}. We deduced at 6°K for each sample the additional thermal resistivity $W_{ep}=1/K-1/K_0$, which is considered to represent approximately the contribution of electron-phonon scattering: K and K_0 are, respectively, the measured and calculated values of the thermal conductivity at 6°K. Because of the above considerations, W_{ep} was estimated at the lower temperature available, where the contribution of phonon-phonon scattering can be neglected. In Fig. 5, we have plotted the variation of W_{ep} as a function of the apparent electron concentration $n_{300°K}$ measured at room temperature. This variation can be represented approximately by $W_{ep}\approx n_{300°K}^{1.7}$. In GaSb, the second (111) conduction-band edge is separated by an energy of about 0.08 eV from the principal (000) conduction-band edge.[23–27] At temperatures low enough, the population of the second conduction band is expected to occur when electron concentrations exceed $\sim10^{18}$ cm^{-3}. Thus, it is more appropriate to consider the electron concentrations at about liquid helium temperature. From the data of Long and Hager,[25] we estimate the values of n at ~6°K for the Te-doped samples measured. $n_{\sim6°K}$ is $\sim1.3n_{300°K}$ at low concentrations ($n_{300°K}\simeq1.5\times10^{17}$ cm^{-3}), and represents nearly $2.2n_{300°K}$ at about $n_{300°K}=10^{18}$ cm^{-3}. Thus, the plot of W_{ep} versus $n_{\sim6°K}$ leads to a steeper increase of the additional thermal resistivity with increasing electron concentration, approaching the law $W_{ep}\approx n_{\sim6°K}^{2.2}$. However, a little less steep increase of W_{ep} with n could not be ruled out. The limit of such behavior would correspond approximately to a negligible contribution of $1/K_0$ in the expression of W_{ep}. In this case, W_{ep} would vary nearly as $n_{300°K}^{1.4}$ or $n_{\sim6°K}^{1.9}$. Therefore, the possibility for W_{ep} to increase

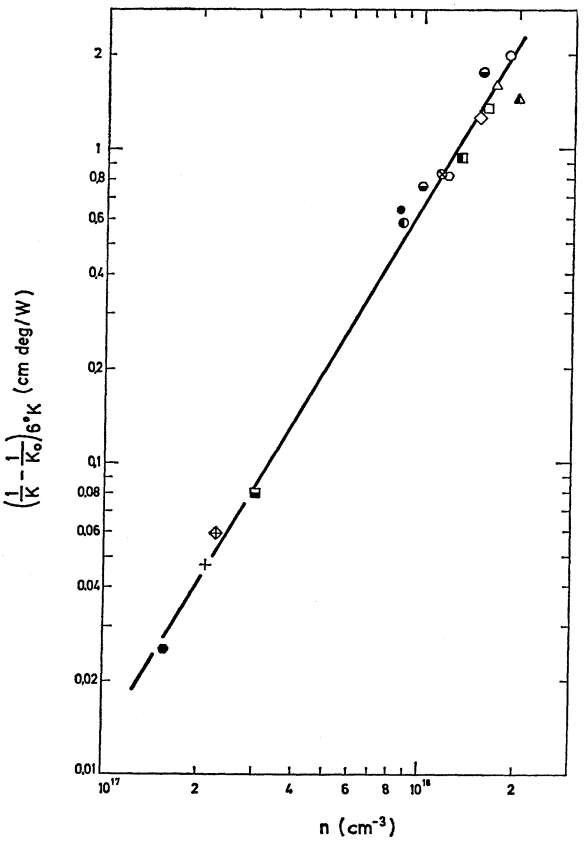

FIG. 5. Variation of the additional thermal resistivity ($1/K-1/K_0$) at 6°K as a function of the apparent carrier concentration ($n=1/|eR|$) at 300°K; K and K_0 are the experimental and theoretical values of the thermal conductivity at 6°K, the latter value being calculated on the basis of the Callaway equation.

a little less rapidly with n than appears in the preceding analysis might perhaps arise from specular reflection, which would give rise to a higher value of L than expected from size effect. Nevertheless, these considerations do not affect the striking feature of the present results.

The sharp increase of W_{ep} with increasing n is a rise which is much more steep than that observed in germanium[4–6] or InSb.[7] The above additional resistivity is most likely to be related to scattering of phonons by an electron gas, i.e., electrons in the (000) conduction band. Ziman's model indicates that, in the case of degenerate electron gas, the relaxation time τ for electron-phonon scattering becomes independent of the density of impurities, but varies, however, as $(m^*)^{-2}$ at a given temperature. m^* represents the density-of-states effective mass. A qualitative explanation which may tentatively involve a dependence of K on n is related to the variation of m^* with increasing electron concentration. This is the case in a nonparabolic band model (Kane, Cohen). In the Kane model, which has been used for GaSb,[26,27] m^* in the (000) band is proportional to the low-temperature Fermi level E_f, and the electron concentration[26] can be considered to increase approximately as E_f. Therefore, if one assumes that the $\tau^{-1}\sim m^{*2}$ law is still valid for a Kane model, the electron-phonon scattering would give rise to a thermal resistivity which varies as n^2, a variation which is close to the observed one. However, this problem requires further analysis.

An alternative argument might be considered. This is related to the possibility for impurity states to exhibit localized level properties even when merged with the conduction or valence band. The existence of electrons localized around some of the donor impurities was suggested by Toyozama[34] in order to explain the results of magnetoresistance measurements of Sasaki *et al.*[35] on degenerate Sb-doped germanium. Thermal-conductivity results of highly doped GaAs[36] were believed to be due to such properties. Evidence for the presence of localized spins in highly degenerate samples of Te-doped InSb was obtained by Khosla and Sladek,[37] who showed that the additional thermal resistance of the latter at low temperature arose most likely from a phonon-localized spin scattering mechanism. Therefore the above argument, which provides a dependence of K on concentration and type of impurity, cannot be ruled out. Further investigations on n-type GaSb with a different type of doping would be very fruitful. On the other hand, the effects of compensation, due to residual acceptors, on electrical properties of n-type GaSb[27,38,39] have been emphasized. For thermal conductivity, and on the basis of electron-phonon scattering models, compensation effects might be correlated, particularly in the case of isolated or semi-isolated impurity states, to impurity conduction and variation of impurity levels.[40,41] In n-type GaSb, however, excess electrons are found to be in the (000) band at lower doping levels. Nevertheless, such effects might be expected to be important in the case where the preceding argument is found to be effective.

A final remark concerns data on undoped p-type samples. It can be seen that the low-temperature thermal conductivity is lower than the calculated curves by a factor of about 100, whereas in Te-doped samples with comparable electron concentrations, this factor is about 10. The additional thermal resistivity W_{ep} at 6°K deduced for the undoped samples is nearly 5 $(\mathrm{W/cm\ deg})^{-1}$, whereas Te-doped samples with electron concentrations of about 1.5×10^{17} cm^{-3} exhibit a value of about 0.025 $(\mathrm{W/cm\ deg})^{-1}$. This large difference indicates that hole-phonon scattering in undoped GaSb is much more strong than electron-phonon scattering in Te-doped GaSb. The former mechanism arises from the scattering of phonons by residual acceptor levels which are at approximately 0.03 eV above the valence band,[20,21,42] and which are suggested to be doubly charged centers.[21,22,27] Preliminary results on lightly Te-doped GaSb, which was maintained p-type, showed that the low-temperature thermal conductivity increases with increasing compensation,[43] reaching values of the same order as the above data on slightly n-type samples. This behavior is in qualitative agreement with Keyes model,[12] which indicates that scattering strength depends on the concentration of neutral donors or acceptors.

ACKNOWLEDGMENTS

The authors are indebted to Dr. Gouskov of Centre d'Etudes d'Electronique des Solides at Montpellier for preparing the single crystals used. They wish to thank Blanjot for his technical assistance.

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