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Electrical and Optical Properties of Te doped GaSb

By

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Abstracts

To examine the property of impurity states in n- and p-type GaSb, the crystals were prepared by doping it with Te. The Hall and resistivity measurements were made and, simultaneously, the free carrier absorption was measured. The undoped crystals were always p-type. The perfect compensation was found to occur at the 0.003 at% amount of Te added to the melt. In the p-type GaSb, it was found that the acceptor level, which results from vacancies in Sb sub-lattice site, is located at 0.021 eV. In the n-type GaSb, the donor concentration changed from 0.5 to $10 \times 10^{18} \text{cm}^{-3}$ by increasing the Te amount. It was suggested that the acceptor introduced into the n-type crystal corresponds to the deeper acceptor level, which was observed in the highly compensated p-type sample. The concentration of its acceptor was found to be fairly larger than that of the native acceptor.

I Introduction

Gallium antimonide has been prepared from high purity elements by various laboratories and the resultant material has been found to contain an almost constant concentration of acceptors of between 1 and $2 \times 10^{17} \text{cm}^{-3}$ ⁽¹⁾⁻⁽⁵⁾. Effer and Etter⁽⁶⁾ suggested that the p-type nature of GaSb is due to lattice defects rather than to impurities. Their suggestion is based on the fact that the crystal of a reduced hole concentration had been grown from antimony rich melts. They reported that the best p-type sample had a hole concentration of $9.2 \times 10^{15} \text{cm}^{-3}$, and a Hall mobility of $5230 \text{ cm}^2 \text{V}^{-1} \text{ sec}^{-1}$ at 77°K.

The n-type GaSb is usually prepared by doping with tellurium⁽⁷⁾ or selenium⁽⁸⁾. Then, even though the n-type GaSb is theoretically favoured to have a high electron mobility of $4.3 \times 10^4 \text{ cm}^2 \text{V}^{-1} \text{ sec}^{-1}$ ⁽⁸⁾, the observed value has been found to be $5 \times 10^3 \text{ cm}^2 \text{V}^{-1} \text{ sec}^{-1}$ or less. The reason seems to arise from the fact that the n-type crystal is prepared by doping with n-type impurities and, accordingly,

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relatively large impurity states are produced in the crystal. Another cause is related to its conduction band structure. As the conduction band minimum is located at L point, and the energy separation from the second minima at L point is $0.074\text{eV}^{(9)}$, the conduction electron is easily excited to higher bands with a low electron mobility by increasing the carrier concentration and/or a temperature rise.

To examine the conditions to obtain a high purity n-type GaSb with a high electron mobility, we prepared GaSb crystals by doping with tellurium, and investigated these electrical and optical properties.

II Experimental Procedures

The compound was prepared from six-nine purity Ga and Sb by using the horizontal Bridgman technique. The undoped crystals were always p-type and had a hole concentration of 1 to $3 \times 10^{17}\text{cm}^{-3}$ at room temperature. A sufficient amount of Te was added to the melt to obtain n-type crystals. In order to examine the effect of the dopant, its amount was changed in the range of 0.0007 to 0.7 at%. The samples were rectangular parallelepipeds 1 - 2 mm thick, 2 - 4 mm wide and 10 - 12 mm long. The Hall and conductivity measurements could be made by the conventional dc method, where the magnetic field was about 9800 gauss. The samples for the optical measurement were cut from the vicinity of the former one and optically polished to 0.2 mm thick. The infrared transmission measurement was performed in the spectral region of 400 to 4000cm^{-1} by using a double beam spectrometer at room temperature. The absorption coefficient α was derived from the equation,

$$T = \frac{(1-R)\exp(-\alpha d)}{1-R^2\exp(-2\alpha d)} \quad (1)$$

where T and R are the bulk transmittance and reflectance, respectively, and d is the sample thickness. The reflectance was taken from the reference⁽¹⁰⁾.

III Experimental Results

a. Compensation Effect

Fig. 1 shows the carrier concentration at 77°K as a function of the Te amount. It was found that the hole concentration for non-doped samples agree well with those for high purity crystals, carefully prepared by various authors⁽¹⁾⁻⁽⁴⁾. The hole concentration decreases rapidly with the addition of small amount of Te until the perfect compensation occurs around 0.003 at% Te. By further Te doping, the samples change into n-type crystals and the electron concentration increases by increasing the impurity. Above 0.073 at% Te or more, the electron concentration

was found to saturate at a value of about $3 \times 10^{18} \text{cm}^{-3}$.

As the Te amount in the figure means the total amount added to the melt, the Te concentration introduced into the solid should correspond to the amount in the liquid multiplied by the distribution coefficient. When the hole concentration $N_A - N_D$ for non-doped GaSb is assumed to be compensated by the 0.003 at% Te added in the melt, the distribution coefficient is 0.094. The present value agrees with the reported values of 0.01 to 0.4⁽²⁷⁾⁻⁽²⁹⁾.

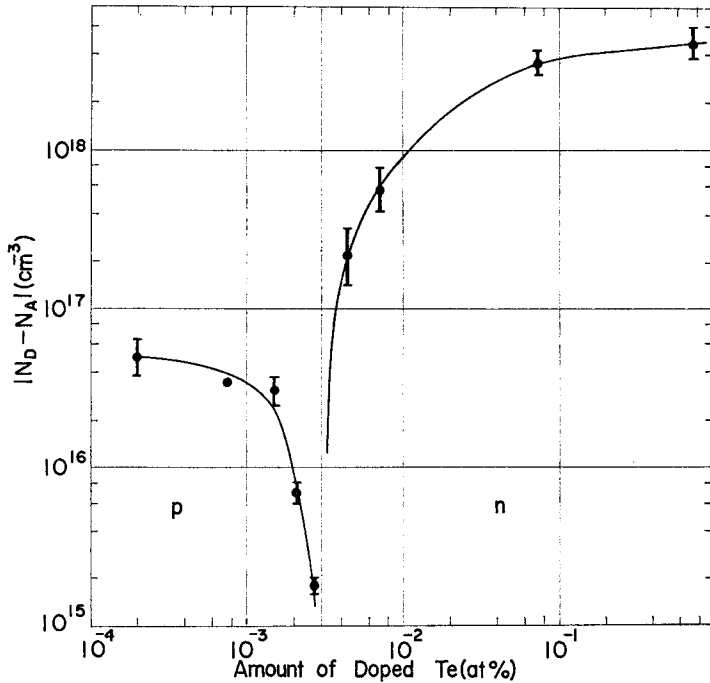


Fig. 1 The change of carrier concentration as a function of doped Te. It is noted that by being doped with Te, the crystal varies from p- to n-type, and the perfect compensation occurs at 0.003 at% Te.

b. Electrical Properties

Electrical resistivity and Hall coefficient data on several p-type samples are, respectively, shown in Figs. 2 and 3. The temperature dependences for all samples, except Sample P73, are similar to each other. Sample P73 is highly compensated by Te. It is noted that the resistivity increases with increasing the Te concentration.

The value of the free hole concentration N_p is obtained from the mass action law⁽¹¹⁾ as follows,

$$\frac{N_v}{g} \frac{N_A - N_D - N_p}{N_p(N_D + N_p)} = \exp(E_A/kT), \quad (2)$$

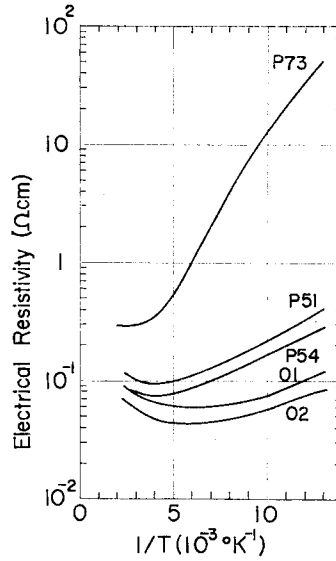


Fig. 2 Electrical resistivity as a function of temperature in p-type GaSb.

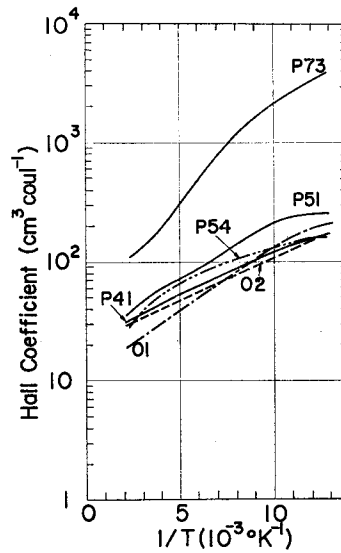


Fig. 3 Hall coefficient as a function of temperature in p-type GaSb.

where N_A and N_D are the acceptor and donor concentrations, E_A is the acceptor energy level, N_V is the equivalent density of state in the valence band, and g is the acceptor state spin degeneracy. For non-doped GaSb and the samples with low

Table 1 Impurity concentration and acceptor energy level in *p*-type GaSb

Sample	N_A (cm ⁻³)	N_D (cm ⁻³)	E_A (eV)
O1	1.2×10^{17}	0.24×10^{17}	0.021
O2	1.9	0.16	0.019
P54	1.6	0.14	0.021
P51	0.87	0.15	0.020
P3	0.68	0.43	—
P73	0.54	0.47	0.055

compensation degrees, by assuming the condition of $N_D < N_p < N_A$, we obtain,

$$R_H T^{3/4} = 3.02 \times 10^{11} N_A^{-0.5} \exp(E_A/2kT). \tag{3}$$

By using the equation, an impurity activation energy was estimated to be 0.021 eV for all samples, except Sample P73. The concentrations of acceptor and donor are shown in Table 1. The present activation energy is in good agreement with the data reported by some authors^{(1),(2)}. On the other hand, for the highly compensated sample such as Sample P73, by assuming the conditions of $N_p < N_D$ and $N_p < N_A - N_D$, we obtain,

$$R_H T^{1.5} = 1.46 \times 10^4 K^* / (1 - K^*) \exp(E_A/kT), \tag{4}$$

where K^* is the compensation ratio. The equation was well fitted for the data of Sample P73. Then, an acceptor energy level was determined to be 0.055 eV, and the compensation ratio to be 0.87. This deeper acceptor level has been previously reported by other authors⁽¹²⁾ to be a result of the compensation with Te.

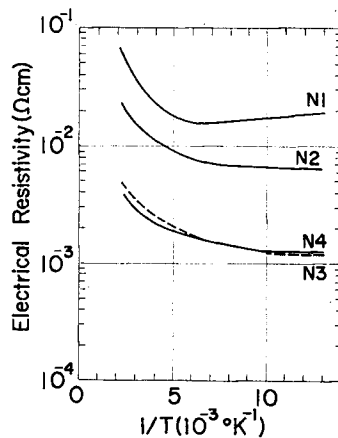


Fig. 4 Electrical resistivity as a function of temperature in *n*-type GaSb.

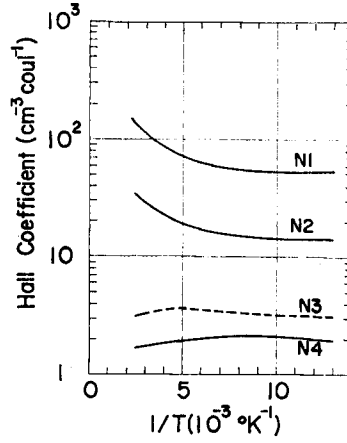


Fig. 5 Hall coefficient as a function of temperature in n-type GaSb.

Figs. 4 and 5 show the temperature dependence of the Hall coefficient and electrical resistivity for n-type samples, respectively. Hall data are separated in two groups. For samples with a relatively higher R_H value, the Hall coefficient increases monotonically with increasing temperature. On the other hand, for samples with a lower R_H value, the coefficient reveals a maximum in the temperature dependence. These dependences could be explained quantitatively by assuming the existence of two conduction bands, where the electron transition occurs between Γ and L bands. According to Sagar⁽⁷⁾, the two band expressions for the electrical conductivity and the Hall coefficient are given by

$$\sigma = e(n_0\mu_0 + n_1\mu_1) \quad , \quad (5)$$

$$R_H = \frac{1}{ec} \frac{(n_0\mu_0^2 + n_1\mu_1^2)}{(n_0\mu_0 + n_1\mu_1)^2} \quad , \quad (6)$$

where the subscripts, 0 and 1, are used to denote the properties of Γ and L bands, respectively. μ represents the mobility, n the carrier concentration, and e and c denote the electric charge and the light velocity, respectively. Here, the total number of the carrier, N_s is expressed by

$$N_s = n_0 + n_1 = 4\pi(2kT)^{1.5}/h^3 [m_0^{1.5} F_{1/2}(\eta) + 4m_1c^{1.5}K^{0.5}F_{1/2}(\eta - \Delta)] \quad , \quad (7)$$

where

$$F_{1/2} = \int_0^\infty \frac{x^{1/2}}{e^{x-\eta} + 1} dx \quad , \quad (8)$$

and $\eta = E_F/kT$ and $\Delta = \Delta E/kT$, and E_F is the Fermi energy measured from the edge of the Γ band. ΔE is the energy separation between the Γ and L bands. When both relaxation times for carriers existing in the two bands are independent of

energy, and the mobility ratio μ_1/μ_0 is 0.178, the above equation is numerically solved by applying appropriate values of the parameters as follows,

$$N_e = 1.26 \times 10^{18} T^{1.5} [F_{1/2}(\eta) + \epsilon F_{1/2}(\eta - \Delta)] \quad , \quad (9)$$

if the ratio of the effective density of states ϵ is given as 56.7. Then, the total number of carriers could be calculated by using the observed value of R_H from Eqs. (6) and (9). The results are listed in Table 2.

Table 2 Summary of electrical properties and carrier concentration in n-type GaSb at 77°K

Sample	$R_H(\text{cm}^3\text{coul}^{-1})$	$\rho(\Omega\text{cm})$	$\mu(\text{cm}^2\text{V}^{-1}\text{sec}^{-1})$	$N_e(\text{cm}^{-3})$
N4	2.0	1.3×10^{-3}	1500	4.8×10^{18}
N3	3.4	1.2	2800	4.0
N2	16	6.6	2400	0.54
N1	55	19	2900	0.11

c. Infrared Absorption

The absorption coefficients as a function of the wave number for p-type samples are shown in Fig. 6. The curves are similar to those observed for p-type samples of Ge⁽¹⁴⁾, InAs⁽¹⁵⁾ and GaAs⁽¹⁶⁾, which indicate that the absorption is produced by interband transitions within the valence band. The effective mass

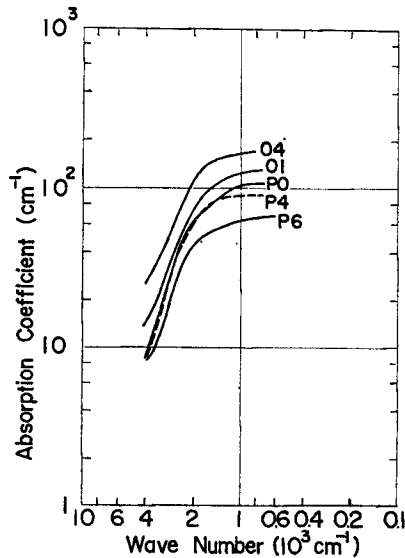


Fig. 6 Absorption coefficient as a function of wave number in p-type GaSb.

ratio of the two types of holes can be estimated from the analysis of the interband transitions, if the energy bands have spherical surfaces of constant energy. Then, the absorption coefficient is given by the equation,

$$\alpha = A\nu^{0.5} \left[\exp\left(-\frac{m_L}{m_L - m_H} \frac{h\nu}{kT}\right) - \exp\left(-\frac{m_H}{m_L - m_H} \frac{h\nu}{kT}\right) \right], \quad (10)$$

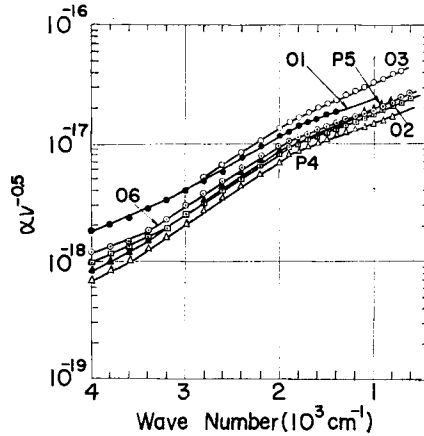


Fig. 7 The change of $\alpha\nu^{0.5}$ as a function of wave number in p-type GaSb.

where A is the constant, ν is the wave number, and m_L and m_H are the effective masses of the heavy and light hole, respectively. The logarithmic values of $\alpha\nu^{0.5}$ are shown in Fig. 7 as a function of the wave number. The shapes for all specimens are similar to each other, where the slope changes slightly around $\nu=1900\text{ cm}^{-1}$. The change occurs from the fact that one of the hole bands is nonspherical. As the second term in Eq. (10) is neglected for $h\nu \ll kT$, the ratio of m_H/m_L was estimated to be 5.0 from the slope of the straight line in the region of 2000 to 3000 cm^{-1} . The present result agrees with the reported one⁽⁹⁾.

For n-type crystals, the absorption coefficients are shown in Fig. 8, where the dotted lines are the theoretical absorption coefficients, which will be discussed later. In the wave number region measured here, the spectra can be explained as a result of the superposition of some optical transitions, i.e., the free carrier absorption, the absorption due to the interconduction band transition and the absorption due to the transition from the impurity level to the conduction band. For wave numbers, $\nu < 2000\text{ cm}^{-1}$, the absorption curves rise smoothly with decreasing wave number. This absorption could be understood to arise mainly from the free carrier absorption as discussed in the previous report⁽¹⁷⁾. The infrared absorption due to the optical transition of free carriers is closely related to the following scattering mechanisms in the conduction phenomena.

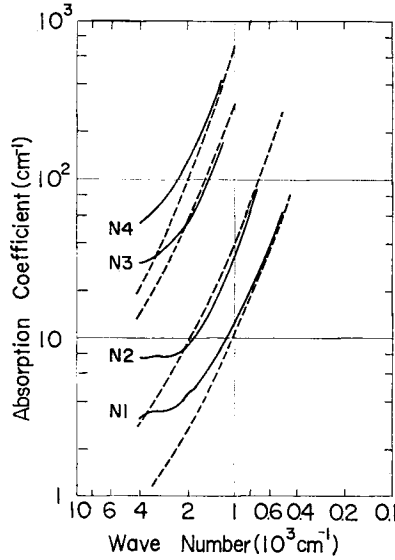


Fig. 8 Absorption coefficient as a function of wave number in n-type GaSb.

Perfectly free carriers do not produce absorption. Absorption arises from the fact that there is a scattering of carriers in motion. The treatment of the effect of acoustic mode scattering by the method of second order perturbation was reported by Fan^{(18), (19)}. For spherical energy bands, the photon capture cross section can be written by

$$\frac{\alpha_{ac}}{N_e} = \frac{4\pi}{cn} \frac{1}{9\pi^{3/2}} \frac{e^3}{m^{*2}} \frac{1}{\nu^2} \left(\frac{h\nu}{kT} \right)^{1/2} \frac{1}{\mu_{ac}}, \quad (11)$$

where n is the refractive index, and μ_{ac} is the mobility corresponding to the acoustic mode scattering reported by Bardeen and Shockley⁽²⁰⁾,

$$\mu_{ac} = 3.2 \times 10^{-5} \rho s^2 \left(\frac{m^*}{m} \right)^{5/2} / T^{3/2} E_1^2, \quad (12)$$

where ρ is the density of the material, s is the sound velocity, and E_1 is the deformation potential constant of the band edge.

The effect of polar mode scattering has been treated by Visvanathan⁽²¹⁾. For a classical distribution of carriers, the photon capture cross section obtained in the form of an integral can be written by

$$\frac{\alpha_{op}}{N_e} = \frac{4\pi}{cn} \frac{\sqrt{2}}{3} \frac{e^4(\epsilon_\infty^{-1} - \epsilon_0^{-1})}{m^*} \frac{\hbar\omega_e}{(h\nu)^{2.5}} \frac{e^{2Z} + 1}{e^{2Z} - 1} \left(1 + \frac{11}{8X} - \frac{4}{\sqrt{2\pi X}} \right) \quad (13)$$

where ϵ_∞ and ϵ_0 are the high and low frequency dielectric constants, respectively, ω_1 is the longitudinal optical frequency, $Z = \hbar\omega_1/2kT$, and $X = h\nu/2kT$.

The effect of ionized impurity scattering can also be treated by the second order perturbation method, using the Born approximation for the electron interaction with a Coulomb or screened Coulomb center⁽²²⁾. A preferable approach is to treat the absorption as the inverse process of Bremsstrahlung, giving the following result^{(18), (23)},

$$\frac{\alpha_{imp}}{N_e} = \frac{4\pi}{cn} N_{imp} \frac{8\pi}{3} \left(\frac{Ze^2}{\epsilon}\right)^2 \frac{e^2 \hbar^2}{m^*} \frac{1}{(2\pi m^* kT)^{1/2}} \frac{1}{(\hbar\nu)^3} \times (1 - e^{-2x}) e^x K_0(X) \quad , \quad (14)$$

in the case of a classical distribution of carriers, where K_0 is the modified Bessel function of the order zero, N_{imp} is the ionized impurity concentration, and Ze is the charge of an impurity center.

Fig. 9 shows the theoretical cross section as a function of the wave number at room temperature, where the impurity concentration and the deformation potential

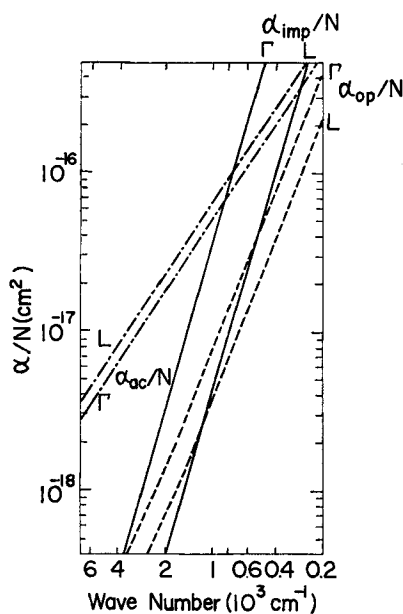


Fig. 9 Dependence of the theoretical cross section on wave number.

Table 3 Material parameters

ΔE	0.074 eV ⁽⁷⁾	ρ	5.62 gcm ⁻³ ⁽²⁴⁾
m_0^*/m	0.047 ⁽¹⁸⁾	s	4.36×10^5 cm sec ⁻¹ ⁽²⁵⁾
m_{1t}/m	0.143 ⁽³⁰⁾	ϵ_∞	14.44 ⁽³¹⁾
K	8.6 ⁽³⁰⁾	ϵ_0	15.69 ⁽³¹⁾
m_L/m	0.065 ⁽⁹⁾	$\hbar\omega_e$	340 °K ⁽⁶⁾
m_H/m	0.23 ⁽⁹⁾		

were assumed to be $1 \times 10^{18} \text{cm}^{-1}$ and 24.9 eV, respectively. The other values for parameters were taken from Table 3. The cross section for the absorption due to the ionized impurity scattering has the largest wave number dependence, and greatly differs between the I' and L bands. If the donor states ionize perfectly, the ionized impurity concentration (N_{imp}) and the carrier concentration (N_c) are given by $N_D + N_A$ and $N_D - N_A$, respectively. The experimental absorption coefficient was fitted to the theoretical equations to obtain the correct values of E_1 and N_{imp} as follows. The total absorption coefficient α , which equals to $\alpha_{ac} + \alpha_{op} + \alpha_{imp}$, is written into the form,

$$\frac{\alpha - \alpha_{op}}{\alpha_{ac}} = \left(\frac{E_1}{24.9} \right)^2 + \left(\frac{N_{imp}}{N_c} \right) \frac{\alpha_{imp}}{\alpha_{ac}} \quad (15)$$

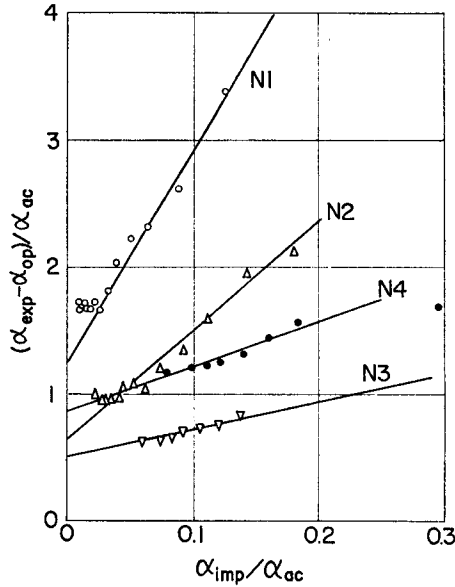


Fig. 10 The relation of $(\alpha_{exp} - \alpha_{op})/\alpha_{ac}$ to α_{imp}/α_{ac} .

By putting the experimental absorption coefficient into the equation, two values for $(\alpha - \alpha_{op})/\alpha_{ac}$ and α_{imp}/α_{ac} could be numerically evaluated. This relation is shown for several samples in Fig. 10. Here, the theoretical absorption coefficient was expressed as $(\alpha_{0,t} + f\alpha_{1,t})/(1+f)$, where f is the carrier concentration ratio between two bands. From the slope, we determined the value of N_{imp}/N_c for each sample. From the intercepts, we estimated the correct value of the deformation potential constant to be 22.1 ± 1.9 eV. As the carrier concentration was obtained from the electrical measurement as listed in Table 2, the total impurity concentration could be obtained. The result is listed in Table 4.

Table 4 Impurity concentration in *n*-type GaSb

Sample	N_{imp}/N_e	$N_{imp}(\text{cm}^{-3})$	$N_D(\text{cm}^{-3})$	$N_A(\text{cm}^{-3})$
N4	3.5	17×10^{18}	11×10^{18}	6.1×10^{18}
N3	2.2	7.8	5.7	2.1
N2	2.9	1.1	0.76	0.37
N1	6.6	0.80	0.46	0.34

IV Discussion

From the electrical and optical data, we have deduced the state of impurity in crystals as summarized in Fig. 11. As the acceptor concentration in non-doped GaSb has been found to be an almost constant value of 1 to $2 \times 10^{17} \text{cm}^{-3}$ for all the samples prepared by various growth methods, the acceptor is believed to be related to a native defect in the crystal. We assume that this native acceptor level results from vacancies in the Sb sub-lattice site. The assumption seems to be reasonable from the fact that the sample grown from Sb rich melt has a reduced hole concentration. By being doped with Te, the acceptor concentration is found to decrease gradually. The substitution of the Te impurity for the Sb site vacancy is an easy process, because the atomic radius of Te is very similar to that of Sb, as compared with that of Ga. In the p-type region, the donor concentration, which arises from the Te impurity, increases abruptly around the compensation point. Here, the total concentration of Te introduced to the solid is given by the sum of the donor concentration and the number dropped into the Sb vacancies. This is shown by the dotted line in Fig. 11.

The perfect compensation occurs around 0.003 at% Te. By being doped with Te above its amount, we obtain n-type crystals. As expected, the donor concentration increases with the increase of doped Te and reaches to about $1 \times 10^{19} \text{cm}^{-3}$. On the other hand, the acceptor concentration is relatively high, as compared with the native acceptors. This means the occurrence of a new acceptor level, by being doped with Te. As mentioned in the previous section, the acceptor level was observed to be 0.055 eV in the highly compensated p-type sample. This 0.055 eV acceptor level might be suggested to correspond to the new one in the n-type GaSb. A possible acceptor state is thought to exist in one of the various crystal imperfections, that is, the further ionization of Te substituted for the Sb sub-lattice site, Te at an interstitial site and Te at the Ga lattice site combined by a vacancy. This crystal imperfection which is related to the new acceptor level should be specified by the further experiments.

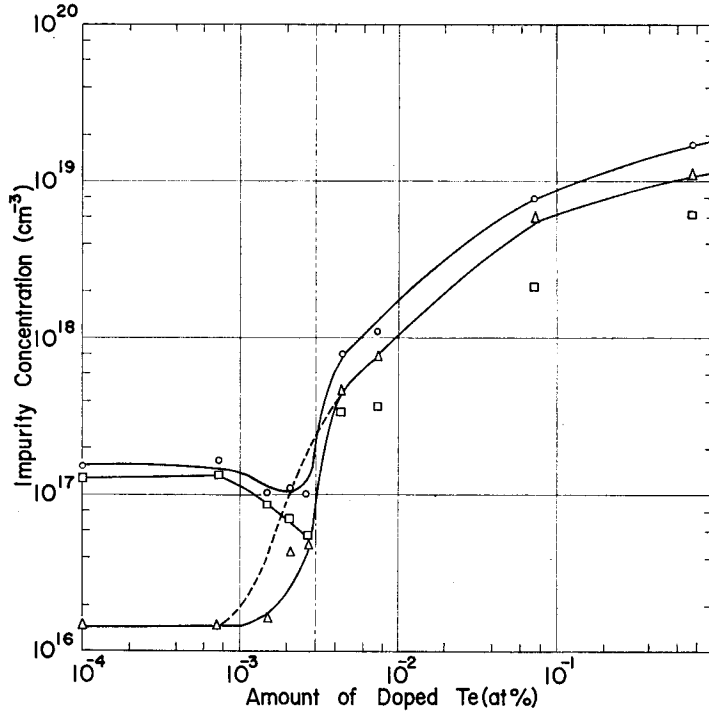


Fig. 11 Impurity concentration as a function of amount of doped Te. \circ , \triangle and \square show the total impurity, donor and acceptor concentration, respectively.

As shown in Fig. 11, the total impurity concentration in the n-type GaSb is found to be $5 \times 10^{17} \text{ cm}^{-3}$ even in the purest sample. According to Brooks⁽²⁶⁾, the mobility due to the ionized impurity scattering is given as

$$\mu_{imp} = 3.2 \times 10^{15} \left(\frac{m}{m^*} \right)^{1/2} \frac{\epsilon^2 T^{3/2}}{N_A + N_D} / \log \left[1 + 3 \times 10^{14} T^2 \epsilon \left(\frac{m^*}{m} \right) / n \right], \quad (16)$$

in the dimension of $\text{cm}^2 \text{ V}^{-1} \text{ sec}^{-1}$. From the equation, we obtain $2570 \text{ cm}^2 \text{ V}^{-1} \text{ sec}^{-1}$ at 77°K for Sample N1, which is in good agreement with the experimental value. Therefore, the electron mobility in the present samples is thought to be dominated by the impurity scattering, and could not be expected to have a theoretical value.

V Conclusion

To examine the property of impurity states in n- and p-type GaSb, the crystal was prepared by doping with tellurium. The perfect compensation was found to

occur at the amount of Te of 0.003 at% added to the melt. In the p-type GaSb, it was found that the acceptor level is located at 0.021 eV from the top of valence band, and the other acceptor level is formed at 0.055 eV in the highly compensated sample. The former acceptor results from vacancies in the Sb sub-lattice site. It was suggested that the acceptor introduced into the n-type crystal corresponding to the deeper acceptor level observed in the highly compensated p-type sample. Its concentration is fairly larger than that of the native acceptor. Therefore, it was concluded that the total impurity concentration in the n-type GaSb can not be lowered by being doped with tellurium.

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