Free-Carrier Absorption in *n*-Type GaP

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The possible scattering mechanisms which can cause free-carrier absorption in *n*-type GaP are enumerated, and it is shown that, as a consequence of the many-valleyed nature of the conduction band, the dominant scattering is a combination of intravalley and intervalley scattering by phonons via the deformation-potential interaction. The calculated absorption for these scattering mechanisms has a wavelength dependence of λ^p with $p\approx 1.7$–1.8, which is in accurate agreement with experimental measurements reported here and elsewhere. Several components of the theoretical absorption have identical wavelength dependences, making a unique decomposition impossible. We show, however, that it is possible to obtain an accurate quantitative fit to the data using reasonable values of the adjustable parameters. The values used were obtained from the literature and correspond to a conduction-band deformation potential $E_1^c\approx 10$ eV and energetic-mode coupling parameters $D_i\approx 8\times 10^8$ eV cm^{-1}. By subtracting out the free-carrier absorption, we obtain a value 0.276 ± 0.007 eV for the threshold of the $X_1\rightarrow X_3$ transition. This value is probably more accurate than those previously reported, since the experimental value obtained for this threshold is rather sensitive to the free-carrier-absorption correction.

I. INTRODUCTION

SINCE the first observation of free-carrier absorption (FCA) in *n*-type GaP,[1] there have been several attempts to explain the magnitude and wavelength dependence of this absorption.[2,3] In most of the III–V semiconductors,[2] FCA exhibits a wavelength dependence of λ^p with $p\approx 2.5$ and gives a cross section of $\sigma\approx 2$–5×10^{-17} cm^2 at $\lambda=10\,\mu$. This behavior has been attributed to polar-mode scattering. In the case of *n*-GaP, however, it is found that $p\approx 1.7$–1.8, and $\sigma(10\,\mu)\gtrsim 30\times 10^{-17}$ cm^2. It is easily shown that absorption cross sections calculated for both polar-mode[4] and ionized-impurity[5] scattering have magnitudes which are too small and wavelength dependences which are too steep ($p\approx 2.5$ and $p\approx 3$, respectively) to explain the observed absorption. Acoustic-mode deformation-potential scattering yields the correct wavelength dependence ($p\approx 1.8$), but requires unusually large values of the deformation-potential parameter, E_1, to fit the observed magnitude. Haga and Kimura,[2] for example, obtained $E_1=55$ eV by fitting the data of Spitzer *et al.*[1] with pure acoustic-mode scattering, whereas deformation potentials determined from low-field transport experiments are more typically $\lesssim 10$ eV. This in itself would not necessarily constitute an inconsistency.[6] We show, however, that acoustic-mode scattering is not expected to be dominant in *n*-GaP, and, as a result, excellent fits to the FCA data can be obtained with smaller values of E_1 by properly accounting for additional scattering mechanisms.

The conduction-band minima of GaP, AlP, AlAs, and AlSb are at or very near the X point in the Brillouin zone.[7] Therefore, one has, in these many-valleyed semiconductors, the possibility of intervalley scattering as well as acoustic and nonpolar optical mode (deformation-potential) scattering. In the cases of the zone-center direct-gap materials intervalley scattering can be ruled out entirely and acoustic- and nonpolar optical-mode scattering would be strongly inhibited by symmetry restrictions.[8–11] This leaves polar optical modes as the dominant scatterers in direct-gap materials and, indeed, the cross section for this process is of the right size and has the right wavelength dependence to explain the FCA observed in these materials.[2,4]

The theory of FCA in many-valleyed semiconductors is considerably more complicated and has only been developed for the special case of Ge.[12–14] It is clear from these calculations, however, that the results should be applicable to other many-valleyed semiconductors such as Si and GaP. The salient features of the theory are that acoustic-mode intravalley scattering produces a wavelength dependence with $p\approx 1.8$ and "energetic-mode" scattering (intravalley scattering by zone-center optic modes and intervalley scattering by zone-boundary optic and acoustic modes) produces $p\approx 1.6$–1.8. The p values depend on the relative sizes of the lattice

[1] W. G. Spitzer, M. Gershenzon, C. J. Frosch, and D. F. Gibbs, J. Phys. Chem. Solids **11**, 339 (1959).
[2] E. Haga and H. Kimura, J. Phys. Soc. Japan **19**, 658 (1964).
[3] A. D. Remenyuk, L. G. Zabelina, Yu. I. Ukhanov, and Yu. V. Shmartsev, Soviet Phys.—Semicond. **2**, 557 (1968); **2**, 561 (1968).
[4] S. Visvanathan, Phys. Rev. **120**, 376 (1960). The comments in this paper concerning GaP were presumably made before the author had seen the experimental results of Spitzer *et al.* (Ref. 1).
[5] S. Visvanathan, Phys. Rev. **120**, 379 (1960).
[6] Low-field transport experiments involve electrons which are within a few kT of the conduction-band edge, whereas FCA can excite them well above the band edge when $h\nu\gg kT$. Since the coupling to the phonons may be quite different in these two cases, one should not expect close agreement of the fitted deformation-potential parameters. Differences as large as $E_1=55$ eV as compared with $E_1\lesssim 10$ eV would, nonetheless, be surprising.
[7] H. Ehrenreich, J. Appl. Phys. **32**, 2155 (1961).
[8] For electrons with *s*-like wave functions the matrix elements for acoustic- and nonpolar optical-mode scattering can be shown to vanish (see Refs. 9–11). This selection rule should break down as one moves out into the zone but the small masses of electrons in most direct-gap semiconductors should confine them to the immediate vicinity of $k=0$ even at relatively high energies.
[9] H. Ehrenreich and A. W. Overhauser, Phys. Rev. **104**, 331 (1956).
[10] W. A. Harrison, Phys. Rev. **104**, 1281 (1956).
[11] H. Ehrenreich, Phys. Rev. **120**, 1951 (1960).
[12] H. J. G. Meyer, Phys. Rev. **112**, 298 (1958).
[13] R. Rosenberg and M. Lax, Phys. Rev. **112**, 843 (1958).
[14] H. Riskin and H. J. G. Meyer, Phys. Rev. **123**, 416 (1961).

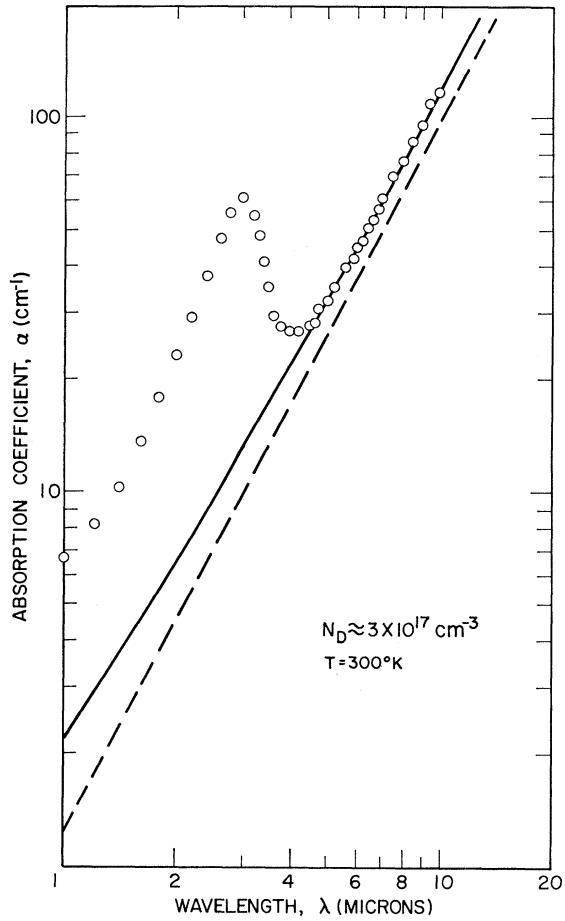

FIG. 1. Experimental (open circles) and calculated (solid line) absorption coefficients for a Te-doped sample of GaP. The absorption peak at 3 μ is due to interband transitions and the background is free-carrier absorption. See the text for a discussion of the calculated absorption. The dotted line shows a power law ($\lambda^{1.83}$) for comparison.

temperature T, the electron temperature after absorption of a photon $\hbar\omega/k$, and the characteristic temperatures θ_i of the relevant phonons. In all cases of interest here $\hbar\omega/k \gg T$, θ_i and the p values are in the range quoted above, which is precisely the range needed to explain the experimental FCA in n-GaP.

Low-field transport studies confirm the expectation that intervalley scattering is important in n-GaP.[15] Toyama and co-workers find that to explain their Hall mobility data a combination of intervalley and acoustic-mode scattering is required with a considerably less important admixture of polar-mode scattering. Nonpolar intravalley optical-mode scattering is found to be negligible at and below room temperature. This would not necessarily hold true, of course, for hot electrons such as those involved in FCA, since they are sufficiently energetic to *emit* optical phonons.

[15] M. Toyama, M. Naito, and A. Kasami, J. Appl. Phys. Japan **8**, 358 (1969).

II. EXPERIMENTAL

In spite of the fact that infrared-absorption data have already been published[1,3] for n-GaP, we include another measurement here since some of the data in the literature are not self-consistent. For example, the absorption shown for sample 1 in Ref. 3 is anomalously large at short wavelengths, exceeding the absorption for sample 2, even though sample 2 has higher carrier concentration. This behavior can be the result of "excess reflection" caused by surface irregularities introduced during sample preparation. Since the wavelength dependence is of prime importance for the present work, it is important that the sample have specular surfaces free from surface contamination. We also point out that, because most doped GaP samples are rather inhomogeneous, it is difficult to obtain accurate values for the absolute-absorption cross section by combining absorption data with average carrier-concentration data obtained, for example, from Hall measurements.

The room-temperature infrared-absorption coefficient α of n-GaP is shown in Fig. 1 for $1 \leq \lambda \leq 10\,\mu$. For this particular sample the long-wavelength limit to the FCA data was set by the sample thickness. There is little to be gained in going beyond $\lambda = 10$–$12\,\mu$, however, since one runs into structure associated with the strong multiphonon lattice absorption just beyond $12\,\mu$. Absorption coefficients were calculated in the usual way[16] from transmission data taken point by point as a function of λ. The wavelength-dependent reflection coefficient needed in this calculation was obtained from index-of-refraction data measured by Bond.[17] The GaP sample was grown from a Ga solution and was doped with 0.007 mole% Te. This yields a nominal donor concentration[18] of $N_D = 2$–3×10^{17} cm^{-3}. The {111} sample faces were polished with 0.1 μ diamond paste and the final thickness was 0.29 mm.

The absorption cross sections $\sigma = \alpha/N_D$ inferred from Fig. 1 are in essential agreement with those reported elsewhere.[1,3] The absorption band at $3\,\mu$ and 300°K has been associated with $X_1 \rightarrow X_3$ conduction-band transitions[19,20] and the absorption for $\lambda \gtrsim 5\,\mu$ is interpreted to be FCA since the Te levels are almost completely ionized at room temperature.[15] At lower temperatures the absorption in the region $\lambda > 5\,\mu$ is even larger than at 300°K.[1,3] This is due to photoionization of the Te donors.[3,21] In what follows, we are concerned

[16] T. S. Moss, *Optical Properties of Semiconductors* (Butterworth and Co., Ltd., London, 1961).

[17] For $\lambda \leq 4\,\mu$ we used the index of refraction measurements of W. L. Bond, J. Appl. Phys. **36**, 1674 (1955). Beyond 4 μ, n was inferred from the dielectric dispersion oscillator fits of A. S. Barker, Jr., Phys. Rev. **165**, 917 (1968). The index of refraction is ≈ 3 until one nears the lattice bands.

[18] F. A. Trumbore, H. G. White, M. Kowalchik, R. A. Logan, and C. L. Luke, J. Electrochem. Soc. **112**, 782 (1965).

[19] J. W. Allen and J. W. Hodby, Proc. Phys. Soc. (London) **82**, 315 (1963).

[20] R. Zallen and W. Paul, Phys. Rev. **134**, A1628 (1964).

[21] E. Haga, J. Phys. Soc. Japan **19**, 2030 (1964); **20**, 735 (1965).

primarily with the region beyond 5 μ and assume it to be due entirely to FCA.

III. DISCUSSION

As a first step in attempting to explain the observed absorption we can eliminate several possible scattering mechanisms. Calculations based on Visvanathan's[4] expression for absorption due to polar-mode scattering indicate that $\alpha(10\,\mu)\lesssim 5$ cm^{-1} and $p\approx 2.5$. Thus, polar-mode scattering can be neglected for all wavelengths of interest here. Although theoretical expressions for the absorption due to ionized-impurity scattering[2,5,12,13] are considered to be less reliable,[13] the intravalley contribution can be estimated from Visvanathan's work[5] and is found to give $\alpha(10\,\mu)<10$ cm^{-1} and $p\gtrsim 3$. Both polar-mode and ionized-impurity interactions give rise to small-angle scattering and, as a result, do not contribute significantly to intervalley scattering. Other scattering mechanisms such as piezoelectric, neutral-impurity, and space-charge scattering are sometimes found to be important at low temperatures but should not be important in the present work and will not be considered further.

The only remaining scattering mechanisms are those involving the scattering of electrons by optical and acoustic phonons via deformation-potential interactions. This mechanism can cause intravalley scattering as well as equivalent ($X_1\rightarrow X_1$) and nonequivalent ($X_1\rightarrow L_1$ and $X_1\rightarrow\Gamma_1$) intervalley scattering.[22] Nonequivalent intervalley scattering has been treated by Risken and Meyer[14] but need not be considered here since it is only energetically possible when $h\nu\gtrsim 0.5$ eV ($\lambda\lesssim 2.5\,\mu$). Hence, we restrict ourselves to scattering processes involving only the three equivalent valleys at the X points.

Rosenberg and Lax[13] have shown that under these conditions the FCA has only two distinguishable contributions: (1) absorption due to intravalley scattering by TA and LA phonons, and (2) absorption due to energetic-mode scattering. In the notation of Rosenberg and Lax,[13] the absorption coefficients are given (in cgs units) by

$$\frac{\alpha_{\mathrm{TA}}}{N}=\frac{4e^2}{3\hbar^2 c}\left(\frac{2}{\pi k}\right)^{1/2}\frac{(\det\mathbf{m})^{1/2}\langle m^{-1}\rangle f_1(\beta)\Xi_u^{\,2}}{n\rho v_T^{\,2}T^{1/2}}\times\frac{\sinh(x)K_2(x)}{x},\quad(1)$$

$$\alpha_{\mathrm{LA}}/N=(v_T/v_L)^2 f_1^{-1}(\beta)[(\Xi_d/\Xi_u)^2+(\Xi_d/\Xi_u)f_2(\beta)+\tfrac{1}{2}f_2(\beta)-f_1(\beta)](\alpha_{\mathrm{TA}}/N)\quad(2)$$

$$=(\Gamma_2/\Gamma_1)(\alpha_{\mathrm{TA}}/N),$$

[22] C. Herring and E. Vogt, Phys. Rev. **101**, 944 (1956).

TABLE I. Numerical values of parameters used in FCA calculations.

Parameter	Value	Reference
n	≈ 3.0	a
ρ	4.13 g cm^{-3}	b
v_L	6.28×10^5 cm sec^{-1}	b
v_T	3.8×10^5 cm sec^{-1}	b
m_1^*/m_0	1.76 ± 0.2	c
m_2^*/m_0	0.191 ± 0.005	c
$\Xi_u{}^c$	23 eV	
$\Xi_d{}^c/\Xi_u{}^c$	$+0.1$	d
D_i	8×10^8 eV cm^{-1}	e
ϵ_0	11.1	f
ϵ_∞	9.1	f
$(\det\mathbf{m})^{1/2}\langle m^{-1}\rangle$	$0.917m_0{}^{1/2}$	g
$f_1(\beta)$	0.144	g
$f_2(\beta)$	1.06	g
Γ_2/Γ_1	1.28	g
$\theta_{\mathrm{LO}}{}^\Gamma$	570°K	h
$\theta_{\mathrm{LO,TO}}{}^x$	500°K	h
$\theta_{\mathrm{LA}}{}^x$	340°K	h
$\theta_{\mathrm{TA}}{}^x$	150°K	h

[a] See Ref. 17.
[b] R. Weil and W. O. Groves, J. Appl. Phys. **39**, 4049 (1968).
[c] A. Onton, Phys. Rev. **186**, 786 (1969).
[d] This value was inferred from a discussion given by Long (Ref. 24) for electrons in Si. The superscript "c" refers to the conduction band.
[e] This value was used for all energetic phonons and is discussed in the text.
[f] A. S. Barker, Jr., Phys. Rev. **165**, 917 (1968).
[g] This quantity was calculated from expressions given in Ref. 13.
[h] See Ref. 25.

and

$$\frac{\alpha_E{}^i}{N}=\left(\frac{\hbar^2}{4k^2T^2}\right)\left(\frac{v_T{}^2D_i{}^2}{f_1(\beta)\Xi_u{}^2}\right)\times\left[\frac{(x+z_i)^2K_2(x+z_i)+(x-z_i)^2K_2(x-z_i)}{x^2z_iK_2(x)\sinh(z_i)}\right]\frac{\alpha_{\mathrm{TA}}}{N},\quad(3)$$

where N is the density of free carriers, n is the refractive index, k is Boltzmann's constant, $x=\hbar\omega/2kT$, $z_i=\theta_i/2T$, θ_i is the characteristic temperature of the ith energetic-mode phonon, $\beta=m_2^*/m_1^*$, Ξ_u, Ξ_d, and D_i are deformation-potential parameters,[23] and $K_2(x)$ is the modified Bessel function of order 2. Numerical values for the functions $f_1(\beta)$, $f_2(\beta)$, $\det\mathbf{m}$, and $\langle m^{-1}\rangle$ have been calculated from expressions given in Ref. 13 and are listed in Table I along with other physical parameters for n-GaP which are needed in these calculations. As can be seen from Eqs. (1) and (2), α_{TA} and α_{LA} have identical wavelength dependences, Furthermore, the expression in brackets in Eq. (3) has a very weak wavelength dependence when $x\gtrsim z$. This is shown in Fig. 2 where we have plotted the wavelength-dependent parts of these expressions at 300°K for various values of z. The TA and LA intravalley contributions have a perceptible curvature but are nearly proportional to λ^p with $p\approx 1.8$. The energetic-mode contribution is more

[23] The D_i parameters used here are the same as the parameters B_τ in Ref. 13. We have changed notation to facilitate comparison with other work. Note that the units of these coupling parameters are erg cm^{-1}, whereas Ξ_u and Ξ_d have units of ergs.

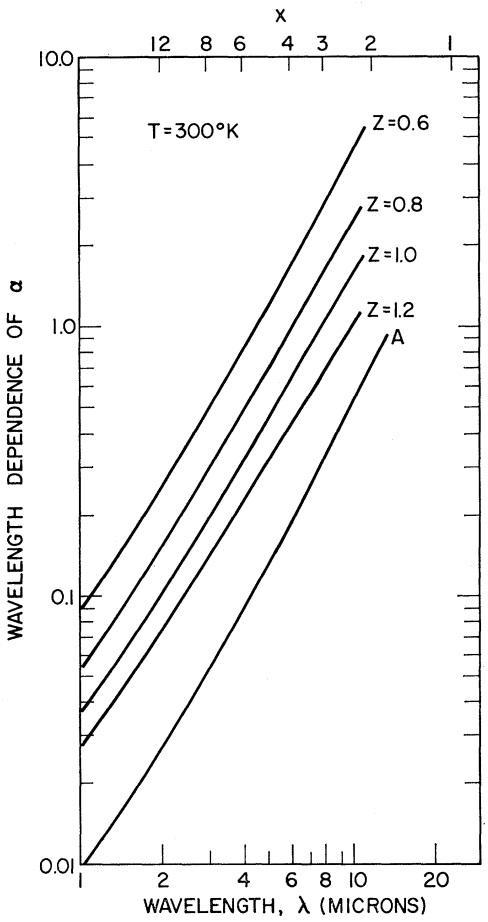

FIG. 2. Wavelength-dependent parts of Eqs. (1)–(3). The curve labeled "A" shows the wavelength dependence of Eqs. (1) and (2) (acoustic-mode intravalley scattering) and the other curves give the wavelength dependence of Eq. (3) (energetic-mode intervalley and intravalley scattering) for various choices of the reduced phonon temperature $z_i=\theta_i/2T$. At 300°K, x is related to λ by $x=24/\lambda(\mu)$.

nearly of the form λ^p and has p values between 1.6 and 1.8.

Before attempting to estimate the over-all magnitude of the absorption, it is necessary to determine which phonons are involved in the various scattering processes. Using geometrical arguments analogous to those given by Long,[24] it can be shown that scattering between adjacent valleys (commonly referred to as f scattering) is an umklapp process involving $\langle 111\rangle$ reciprocal-lattice vectors and X-point phonons. These phonons and their characteristic temperatures[25] are listed in Table I. In GaP the conduction-band minima are at the X points[25,26] and so scattering directly across the zone to second-nearest-neighbor valleys (g scattering) is equivalent to intravalley scattering. The phonons involved in intravalley scattering are the long-wavelength TA and LA phonons and the Γ-point LO phonon. In transport experiments, where the electrons are confined very near the X points, the selection rules for intervalley scattering[27] limit the number of phonons which can scatter electrons from one valley to another. In the case of GaP these rules would permit only the LA phonon to participate in $X_1\rightarrow X_1$ intervalley scattering.[28] Because of the relatively large masses at the X point, however, photoexcited electrons with energies $\gtrsim 0.1$ eV are spread over an appreciable fraction of the Brillouin zone and can no longer be thought to be located near the X point. The strict X point selection rules are therefore not applicable to scattering among these "high-energy" ellipsoids. Birman *et al.*[27] give a relaxed set of selection rules for scattering involving electrons near the X points (along the line Δ). These rules allow X-point phonons from all branches to participate in intervalley scattering. In all cases the allowed phonons for intravalley scattering are zone center LO, LA, and TA phonons. Thus the total absorption coefficient can be decomposed as follows:

$$\alpha=\alpha_{\mathrm{TA}\Gamma}+\alpha_{\mathrm{LA}\Gamma}+\alpha_{\mathrm{E}}, \tag{4}$$

with

$$\alpha_{\mathrm{E}}=\alpha_{\mathrm{LO}\Gamma}+\alpha_{\mathrm{IV}},$$

where E refers to energetic-mode scattering, IV refers to intervalley scattering (X-point phonons), and we assume that the relaxed selection rules apply.

Since there is no way to separate the individual contributions on the basis of wavelength dependence, one must look to other experiments for determinations of the individual coupling constants Ξ_u, Ξ_d, and D_i. The values used in calculating the curve shown in Fig. 1 are listed in Table I. These values were obtained in the following way. Since there are no materials for which a complete set of parameters is available, one is forced to use values from various materials. The band structures of GaP and Si are extremely similar, leading us to choose a value of Ξ_d/Ξ_u which has been inferred for Si.[24] Having made this choice, one can estimate Ξ_u by requiring the conduction-band deformation potential to be $E_1\approx 10$ eV [15] and using the relation[22] $E_1=\Xi_d+\frac{1}{3}\Xi_u$. This gives $\Xi_u\approx 23$ eV ($\Xi_u\approx 19$ eV for Ge so our value is not unreasonable). It should be pointed out that Γ_2/Γ_1 (and hence $\alpha_{\mathrm{LA}}/\alpha_{\mathrm{TA}}$) is not very sensitive to the precise value of Ξ_d/Ξ_u. For example, if $-0.2\leq\Xi_d/\Xi_u\leq+0.2$ then $0.5\leq\Gamma_2/\Gamma_1\leq 1.5$. Thus the TA and LA modes will make roughly equal contributions to the intravalley scattering. The choices of the D parameters are neces-

[24] D. Long, Phys. Rev. 120, 2024 (1960).
[25] J. L. Yarnell, J. L. Warren, R. G. Wenzel, and P. J. Dean, *Neutron Inelastic Scattering* (International Atomic Energy Agency, Vienna, 1968), Vol. 1, p. 301.
[26] R. Faulkner (private communication).
[27] J. L. Birman, M. Lax, and R. Loudon, Phys. Rev. 145, 620 (1966).
[28] It is interesting to note that Toyama *et al.* (Ref. 15) used a six-ellipsoid model for which a more relaxed set of selection rules would apply. They concluded, however, that the LA phonon was dominant in intervalley scattering and that the coupling constants for other phonons were much smaller. This probably indicates the correctness of the three-ellipsoid model.

sarily somewhat arbitrary. Typical coupling constants for intervalley scattering are as follows: $D=7\times10^8$ eV cm^{-1} for the LA phonon in GaP,[15] $D=1\times10^9$ eV cm^{-1} for GaAs,[29] and $D\approx4\times10^8$–2×10^9 eV cm^{-1} for Ge.[10–12] It is possible to make physical arguments concerning the expected relative sizes of the D_i parameters but, in the absence of independent determinations, any speculation along these lines is not justified. In our calculations we therefore assume that all the energetic phonons have the same coupling constant and find that a value of $D\approx8\times10^8$ eV cm^{-1} gives good agreement between experimental and theoretical curves as shown in Fig. 1. These choices of coupling constants are certainly not unique but they serve to illustrate that it is possible to fit both the wavelength dependence and the magnitude of the free-carrier absorption using physically reasonable scattering mechanisms and parameters. The present agreement between experimental and theoretical FCA is sufficiently improved over previous work that we can obtain a more reliable value for the X_1–X_3 splitting. By subtracting the theoretical curve from the experimental points in Fig. 1 we obtain a threshold for the $X_1\rightarrow X_3$ transition of 0.276 ± 0.007 eV. This is to be compared with the value of 0.29 ± 0.01 eV obtained by Dean and co-workers[30] and values of 0.29–0.30 eV obtained by others.[1,3,19,20] As can be seen from Fig. 3 the threshold is quite sharp and not subject to the low-energy tail reported by Allen and Hodby.[19] Such a tail would be obtained if the FCA correction were made on the basis of a simple λ^p dependence (linear extrapolation on a log-log plot) instead of the more complicated λ dependence shown in Fig. 1.

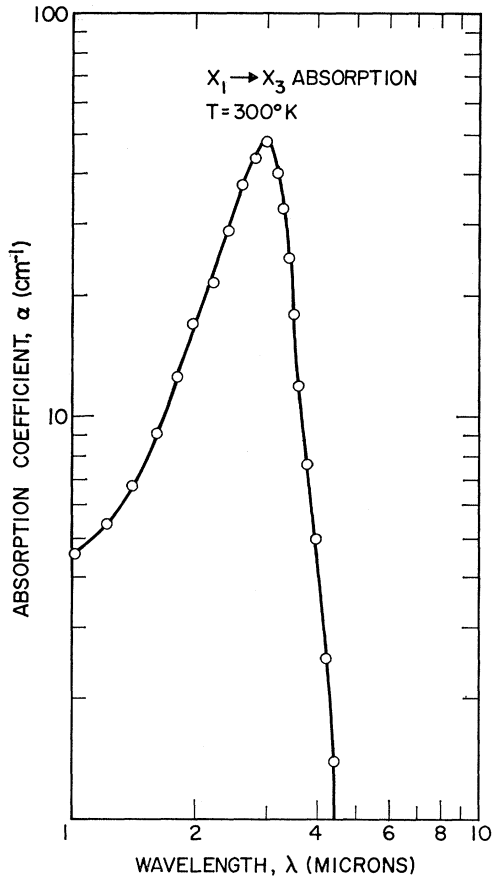

FIG. 3. 3-μ absorption band corrected for FCA by subtracting the theoretical curve from the experimental points in Fig. 1. The sharp threshold at 4.5 μ is caused by the onset of $X_1\rightarrow X_3$ transitions.

IV. CONCLUSIONS

The scattering mechanisms which are important in free-carrier absorption in n-GaP have been identified and it has been shown that one can obtain quantitatively accurate fits to the data using theoretical expressions given by Rosenberg and Lax.[13] The agreement between experimental and theoretical wavelength dependences is excellent, but there is no way, on the basis of wavelength dependence alone, to determine the relative contributions of the various partial-absorption coefficients. Using values of Ξ_u, Ξ_d, and D_i which have been determined for materials similar to GaP, we obtain a total-absorption coefficient which is in agreement with experiment. It must be emphasized that equally good fits could be obtained with quite different sets of coupling parameters so that the final comparison of theory and experiment will have to await independent determinations of the electron-phonon coupling parameters for GaP.

It is of interest to note that FCA has been observed in n-AlSb,[31] another of the many-valleyed III–V semiconductors, and that it exhibits a wavelength dependence of λ^p with $p\approx2$ and a cross section of $\approx15\times10^{-17}$ cm^2 at $\lambda\approx9\,\mu$. Thus it would appear, as expected, that the scattering mechanisms discussed here for GaP are also important in AlSb.

ACKNOWLEDGMENTS

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[29] E. M. Conwell and M. O. Vassell, Phys. Rev. 166, 797 (1968).
[30] P. J. Dean, G. Kaminsky, and R. B. Zetterstrom, J. Appl. Phys. 38, 3551 (1967).
[31] W. J. Turner and W. E. Reese, Phys. Rev. 117, 1003 (1960).