If we make the ansatz

$$\sigma = \sum_{l,m} e^{-i(l+m)\cdot x} X_{1,m}{}^*(\hat{p}) b_m{}^\dagger b_1{}^\dagger - \text{H.c.} \qquad (B3)$$

then σ satisfies (B2) if

$$\hbar\omega[2 - 2\mathbf{q}(t)\cdot(\mathbf{l+m}) + (\mathbf{l+m})^2] X_{1,m}(\mathbf{p}(t))$$

$$+ iF r_0 \frac{\partial}{\partial p_z} X_{1,m}(\mathbf{p}(t))$$

$$= \frac{\hbar\omega}{4l} \left(\frac{4\pi\alpha}{S}\right)^{1/2} [f_m(\mathbf{p}(t) - \hbar\mathbf{l}) - f_m(\mathbf{p}(t))]$$

$$+ \frac{\hbar\omega}{4m} \left(\frac{4\pi\alpha}{S}\right)^{1/2} [f_1(\mathbf{p}(t) - \hbar\mathbf{m}) - f_1(\mathbf{p}(t))] \qquad (B4)$$

and if

$$G^{(2)} = \tfrac{1}{2} \langle 0, \mathbf{k}_0 | [H_1, S] | \mathbf{k}_0, 0 \rangle$$

$$= \text{Re} \left(\frac{4\pi\alpha}{S}\right)^{1/2} \hbar\omega \sum_1 \left(\frac{f_1(\mathbf{p}(t))}{l}\right). \qquad (B5)$$

This verifies that the ansatz (B3) is correct.

In solving (B4) we must choose the integration constant so that, in analogy to (A4), the adiabatic condition

$$\chi_{\mathbf{k}_0}(t_0) - [1 + \lambda S + \lambda^2(\tfrac{1}{2}S^2 + \sigma)] | \mathbf{k}_0, 0 \rangle = O\left(\lambda^3, \frac{\lambda}{\omega t_0}\right)$$

is satisfied.

Infrared Transmission and Fluorescence of Doped Gallium Arsenide

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Experimental measurements of the fundamental optical absorption edge show that with increased doping, n-type GaAs exhibits a shift of the optical absorption edge to higher energy at 77°K, while p-type GaAs at 300°K shows a shift to lower energy. For n-type GaAs at 300°K and p-type GaAs at 77°K, a combination of the two effects is observed. Fluorescence emission for the relatively low doped n-type GaAs occurs at nearly the energy of the band gap, while the highest doped materials emit at higher energies. The p-type fluorescence occurs through the acceptor state at 77°K, but not at 300°K. A deep level, presumably an acceptor level about 0.08 eV above the valence band, was found for Ge-doped GaAs.

I. INTRODUCTION

THE discovery of the efficient emission of infrared light by forward biased GaAs diodes[1] and the subsequent construction of GaAs lasers[2] has created considerable interest in the optical properties of this semiconductor. In order to determine some of the possible effects of material parameters on the performance of these devices, and because of general interest in the properties themselves, the following investigation of transmission and fluorescence of doped GaAs has been carried out.

First, the absorption edge of GaAs is measured for crystals with various types and levels of doping. The shifts in the absorption edge are interpreted as either a "Burstein" type shift,[3] an effect of the impurities

themselves,[4,5] or a combination of the two. It is attempted to fit the first type of shift to the expression of Kaiser and Fan[6] and some difficulties are noted. The fluorescence results are then presented and discussed in relation to absorption data.

II. EXPERIMENTAL TECHNIQUE

The first figure shows the experimental arrangement for both the transmission and fluorescence experiments. A Bausch and Lomb grating monochromator with a grating blazed for 2 μ first order was used in the second order in which case it has a dispersion of 66 Å/mm. A type 7102 photomultiplier was used as a detector in conjunction with a Perkin-Elmer model 107 chopper amplifier.

Filters for the transmission measurements were necessary to minimize scattered light. These were either the RG-1 or RG-10 filter[7] (red and infrared trans-

[1] R. J. Keyes and T. M. Quist, Proc. I.R.E. **50**, 1822 (1962).
[2] R. N. Hall, G. E. Fenner, J. D. Kingsley, T. J. Soltys, and R. O. Carlsen, Phys. Rev. Letters **9**, 366 (1962); M. I. Nathan, W. P. Dumke, G. Burns, F. H. Dill, Jr., and G. J. Lasher, Appl. Phys. Letters **1**, 62 (1962); T. M. Quist, R. H. Rediker, R. J. Keyes, W. E. Krag, B. Lax, A. L. McWhorter, and H. J. Zeiger, Appl. Phys. Letters **1**, 91 (1962).
[3] E. Burstein, Phys. Rev. **93**, 632 (1954); see also T. S. Moss, Proc. Phys. Soc. (London) **B67**, 775 (1954).

[4] I. Kudman and T. Seidel, J. Appl. Phys. **33**, 771 (1962).
[5] P. Aigrain and J des Cloiseaux, Compt. Rend. **241**, 859 (1955).
[6] W. Kaiser and H. Y. Fan, Phys. Rev. **98**, 966 (1955).
[7] Fish-Schurman Corporation, 70 Portman Road, New Rochelle, N. Y.

mitting) in conjunction with various thicknesses of the KG-3 (infrared absorbing) filter.[7] Values of absorption coefficient were calculated from the transmission measurements using a reflectivity of 0.30.

For the fluorescence measurements, a water filter 1 in. thick with the KG-3 filter 5 mm thick was used for the exciting radiation and a RG-10 filter was used between the sample and the monochromator. When measuring the fluorescence from the same surface on which it is excited, the fluorescence was differentiated from any scattered light which might be present for low emission samples by substituting a Ge sample which will not fluoresce in the region

One feature of this apparatus which may be somewhat unique, and possibly useful in other applications is the fiber optics light guide (American Optical LG-2) used to transmit the light from the source to the sample. This permits the use of a permanently evacuated Dewar flask by eliminating the problem of incident light being multiply reflected between the Dewar walls and adding undesired radiation background to the transmitted or emitted light.

The samples used in these measurements were either single crystal or polycrystalline composed of a few large crystallites. These samples were lapped to a thickness of either 200 or 25 μ and etch polished. Experience with this type of sample in the apparatus described above indicates that they yield a transmission within 5% of that measured for a similar, but optically polished, sample. Measurements in the high transmission regions were not used so that an accuracy of 5% or better could be maintained for absorption coefficient.

Hall measurements were made on a rectangular bar cut from the same slice adjacent to the transmission sample. The relationship between Hall coefficient R and carrier concentration n was taken to be $R = 1/ne$, where e is the charge on the electron. Table I shows the room-temperature electrical properties of the samples used in the transmission measurements.

III. DISCUSSION OF EXPERIMENTAL RESULTS

A. Absorption Edge Measurements

Figure 2 shows the absorption coefficient α as a function of photon energy $h\nu$ for samples of n-type GaAs of various doping levels at 77°K. The sample labeled 5.3×10^{16} electrons/cc essentially duplicates the data of Sturge[8] over the range covered by this measurement.

The most notable feature of these curves is the shift of the absorption edge to higher energy with increased doping. Such behavior is due to filling of the conduction band which shifts the Fermi level ζ to higher energies. This shift can be described for n-type material by the expression of Kaiser and Fan[6] as given below:

$$h\nu(\alpha) = E_g + \{\zeta - kT \ln(\alpha_0 - \alpha)/\alpha\}(1 + m_e/m_h), \quad (1)$$

[8] M. D. Sturge, Phys. Rev. **127**, 768 (1962).

TABLE I. Electrical properties (at 300°K) used in optical absorption measurements.

Sample number	Dopant	Carrier concentration (cm^{-3})	Mobility (cm^2/V-sec)
1	Te	9.0×10^{18}	1480
2	Te	6.8×10^{18}	1860
3	Se	5.4×10^{18}	1580
4	Te	4.6×10^{18}	1870
5	Se	3.2×10^{18}	1790
6	Te	2.0×10^{18}	2350
7	Se	9.4×10^{17}	2800
8	Se	4.1×10^{17}	2830
9	...	5.3×10^{16}	5300
10	Zn	5.7×10^{19}	50
11	Zn	1.7×10^{19}	66
12	Zn	7.0×10^{18}	87
13	Zn	4.1×10^{18}	95
14	Cd	9.7×10^{17}	159
15	Cd	2.9×10^{17}	160
16	Cd	6.1×10^{16}	179

where $h\nu(\alpha) =$ photon energy at which absorption coefficient α occurs, $E_g =$ energy gap, $k =$ Boltzman constant, $T =$ absolute temperature, $\alpha_0 =$ absorption coefficient for "pure" material, $m_e =$ electron effective mass, $m_h =$ hole effective mass.

The influence of the effective mass on photon energy occurs mainly through the Fermi level term. If one uses the data in Fig. 2, and assumes the values $E_g = 1.500$ eV, $\alpha = 300$ cm^{-1}, and $m_e/m_h = 0.144$, an electron effective mass of 0.12 m_0 is required to give the fit shown in Fig. 3. The values of α_0 were taken from the data of Sturge. This electron effective mass value is greater than the largest value obtained by reflectivity[9] or Faraday rotation[10] of 0.089 m_0 for 5×10^{18} electrons/cc. However, a more pronounced effect of the same type has been reported for InSb[6] and InAs[11] which is not really well understood.

We note in Fig. 2 that the high energy portion of the curves is linear within experimental accuracy, but that the slopes of the samples with higher doping are smaller than those with lower doping levels. Samples 5, 6, and 7 have nearly the slope predicted by Eq. (1). For sample 1, however, the observed slope is roughly
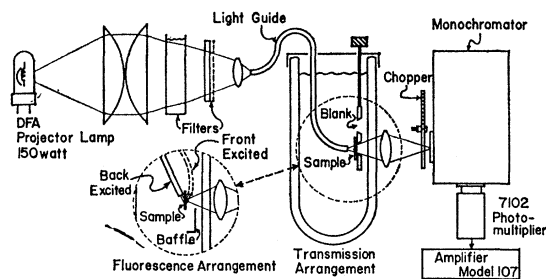

FIG. 1. Schematic drawing of apparatus for measurement of transmission and fluorescence.

[9] W. G. Spitzer and J. M. Whelan, Phys. Rev. **114**, 59 (1959).
[10] M. Cardona, Phys. Rev. **121**, 752 (1961).
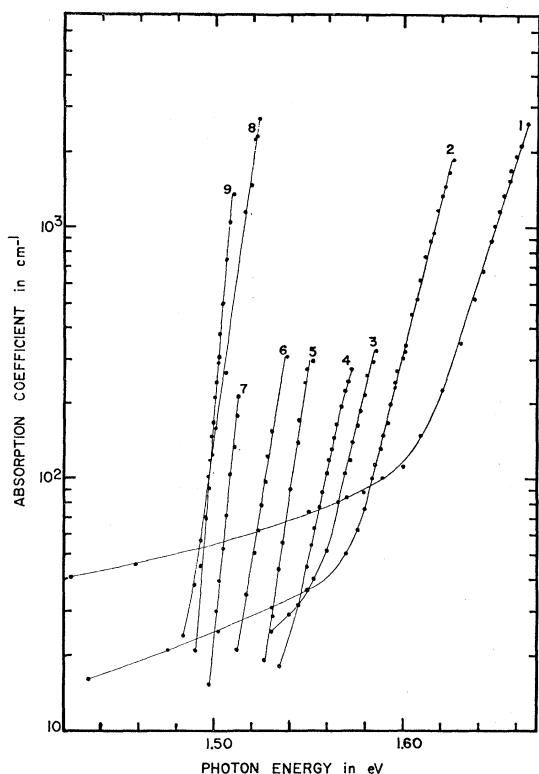[11] F. Stern and R. M. Talley, Phys. Rev. **100**, 1638 (1955).

FIG. 2. Optical absorption of n-type GaAs at 77°K.

half this value, even when the low-energy tail of this curve is subtracted from the higher energy data. Thus, the slope of the curves in Fig. 2 for the most heavily doped samples does not agree with Eq. (1). This effect shows up in a different way if one tries to fit Eq. (1) for various values of absorption coefficient. In general a different effective mass is required to get a good fit of the data for different values of absorption coefficient.

It is not surprising that one has difficulties fitting a theory which assumes among other things that the conduction band is parabolic and that it does not change shape or shift with doping. We can see from the above results, however, that the nonparabolic nature of the conduction band of GaAs alone cannot explain the difficulties, since the highest effective mass obtained from reflectivity and Faraday rotation are considerably less than those obtained above.

The low-energy end of the curves in Fig. 2, particularly for the two most heavily doped samples, show a tendency to level out. The absorption in this region would be expected to be at least partly due to transitions between conduction band minima as observed by Spitzer and Whelan.[9] There seems to be additional absorption present, however, which increases more rapidly than the carrier concentration in Te-doped samples 1 and 2. This may be due to a mechanism similar to that proposed by Vieland and Kudman[12] for

[12] L. J. Vieland and I. Kudman, Phys. Chem. Solids 24, 437 (1963).

GaAs heavily doped with Se. In this case, such additional absorption is postulated as being caused by a Ga vacancy-Se_3 complex.

Note in Fig. 3 that the data for Te- and Se-doped GaAs fall on essentially the same line, while that for Ge and S fall definitely below. The data for Sn and Si seem to agree with Te and Se but are not really extensive enough to be conclusive.

The Ge-doped GaAs can be understood on the basis of self-compensation since Ge can substitute for either a Ga or As atom in the GaAs lattice. Analysis of the Ge content and examination of the Hall mobility show that self compensation is present. The difference between the solid curve and the points for Ge-doped samples is roughly half that predicted for compensated samples on the basis of the calculations of Stern and Dixon.[13] The differences in the case of S-doped GaAs are not understood at the present, and measurements of lattice dimension indicate that changes of this parameter do not explain the difference.

The absorption edge for n-type GaAs at 300°K is shown in Fig. 4. Again we note the shift of the absorption edge to higher energy with increased doping. A fit of this data at an absorption coefficient of 300 cm^{-1} using $E_g = 1.405$, and $m_e/m_h = 0.144$ yields an electron effective mass of 0.095 m_0. Also, the slope of the highly doped samples is less than that expected according to Eq. (1), showing that the same difficulties encountered with the 77°K data are present with the 300°K data, but to a lesser degree.

There is, however, an additional complication evident at low doping levels. Note that the absorption edge for sample 9 occurs at higher energy than that for samples 7 and 8, even though the latter two are more heavily doped. This difference is greater than the experimental uncertainties and may be related to an impurity interaction effect similar to that observed
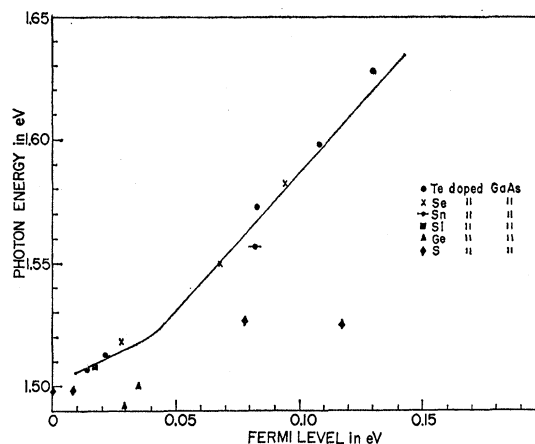

FIG. 3. Energy at which an absorption coefficient of 300 cm^{-1} occurs as a function of Fermi energy.

[13] F. Stern and R. Dixon, J. Appl. Phys. 30, 268 (1959).

by Kudman and Seidel in p-type GaAs.[4] A shift in this direction would decrease the observed Burstein shift and result in effective masses which are too large. An effect of this type has been suggested by Aigrain and Cloizeaux[5] to explain similar difficulties observed for InSb.

The absorption edge measurements for p-type GaAs at 77°K are shown in Fig. 5. We note that a definite Burstein type shift is observed for the two highest doped samples while the samples with lower doping are clustered together. The data for the former two samples can be analyzed according to an equation similar to Eq. (1), but with the term $[1+(m_e/m_h)]$ replaced by $[1+(m_h/m_e)]$. Such analysis leads to effective mass values of roughly $2m_0$. However, the presence
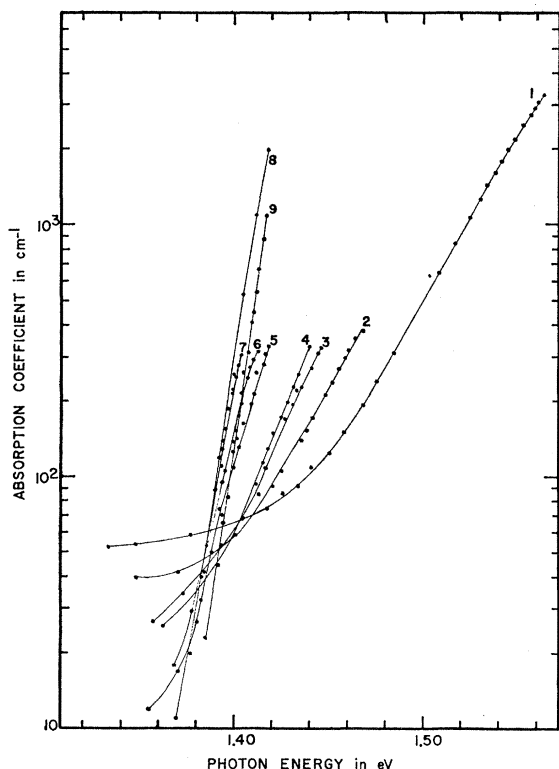

FIG. 4. Optical absorption of n-type GaAs at 300°K.

of light and heavy hole bands make such an analysis even less valid for p-type GaAs than the corresponding analysis is for n-type material.

Effects of the light and heavy hole bands have been considered by Gobeli and Fan in the optical absorption measurements on p-type InSb,[14] and they alter the predictions of the simple theory considerably. The present data, however, is not extensive enough to make a more complete analysis possible. The room temperature absorption measurements for p-type GaAs are shown in Fig. 6. In this case we see that the increased

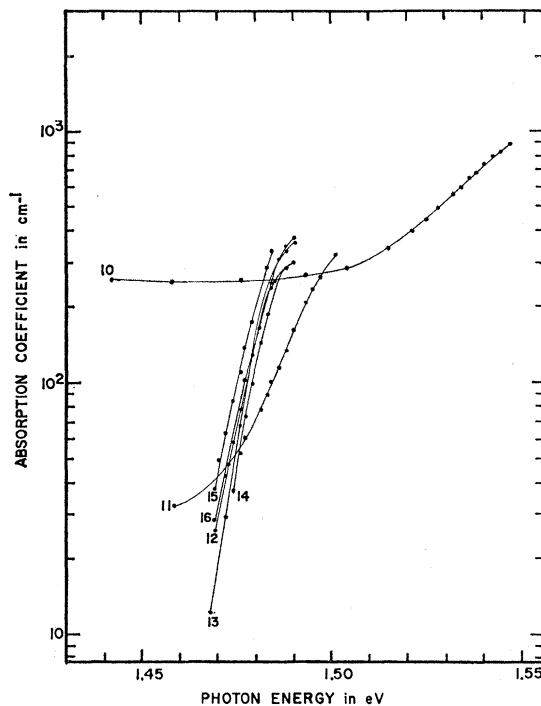[14] G. W. Gobeli and H. Y. Fan, Phys. Rev. **119**, 613 (1960).

FIG. 5. Optical absorption of p-type GaAs at 77°K.

doping causes shifts to lower energy as previously observed.[4]

B. Fluorescence Measurements

Figure 7 shows a typical recorder trace of the fluorescence emission of Se- and Zn-doped GaAs at 77°K. The solid line represents the emission as ob-
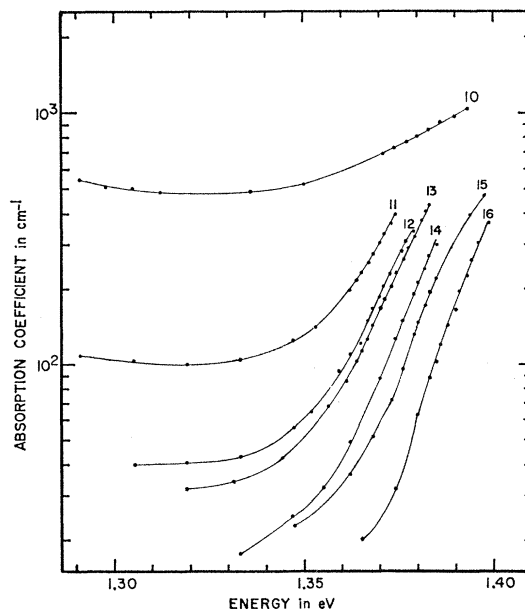

FIG. 6. Optical absorption of p-type GaAs at 300°K.

served from the same surface on which the sample is excited, or front-excited fluorescence. The dotted line represents the same samples excited on the opposite face or back excited. Notice that the back excitation curves show considerable self absorption for the n-type sample but not for the p-type sample even though both are 0.2 mm thick. This shift is consistent with the absorption measurements. The self absorption is in fact present for both n- and p-type GaAs at low carrier concentration but not as significant for p-type material.

The energy of the front-excited fluorescence peaks of n-type GaAs at 77°K is shown in Fig. 8 as data points, along with a solid line which represents the energy at which an absorption coefficient of 300 cm^{-1} occurs for Te- and Se-doped GaAs.
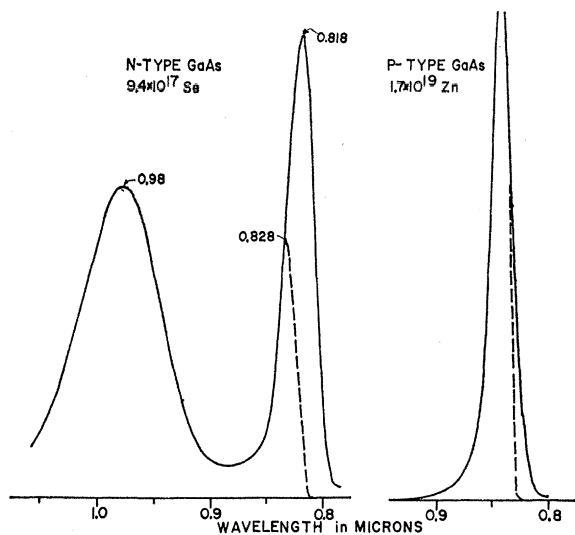

FIG. 7. Typical recorder trace of fluorescence emission for Se- and Zn-doped GaAs.

Note that the peak energy of the fluorescence crosses the line representing an absorption coefficient of 300 cm^{-1} at a carrier concentration of roughly 8×10^{17} electrons/cc indicating the importance of self-absorption for lower carrier levels. One can infer from this crossing that electrons near the Fermi level are not those primarily involved in the recombination resulting in photon emission. For pure GaAs, the emission at 1.508 eV has been identified as arising from exciton recombination.[15] Up to 10^{18} electrons/cc, the emission observed here occurs at nearly the same energy as the exciton emission which is slightly less than the energy gap.[8] As the doping level is increased toward 10^{19} cm^{-3}, the emission energy increases significantly, but not as much as the curve for $\alpha = 300$ cm^{-1}. Since it seems unlikely that the exciton emission would increase in this way, this emission is probably the result of a

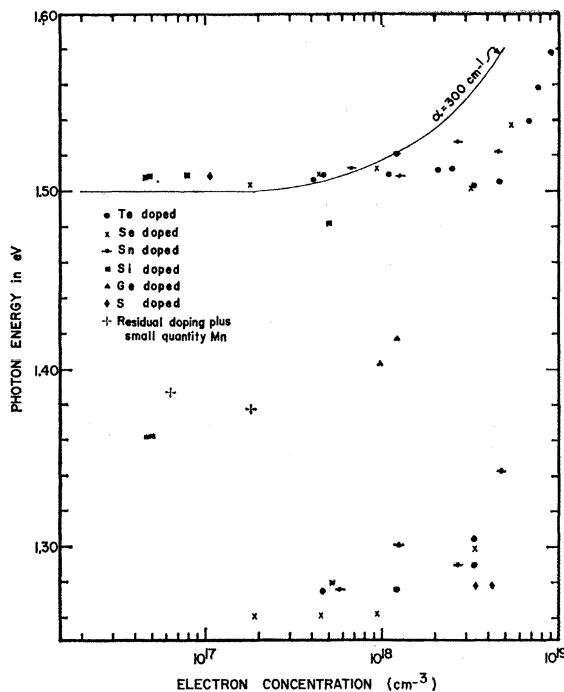[15] M. I. Nathan and G. Burns, Phys. Rev. 129, 125 (1963).

FIG. 8. Energy of fluorescence peak for n-type GaAs at 77°K and energy, at which an absorption coefficient of 300 cm^{-1} occurs for Te- and Se-doped GaAs.

different recombination process starting at an energy below the Fermi level, but above the band edge.

The half-width of the emission increases with doping level from about 0.04 eV at 5×10^{16} electrons/cc to nearly 0.1 eV at the 9×10^{18} electrons/cc level. In addition the intensity of fluorescence decreases at high doping levels becoming difficult to measure accurately at the highest levels.

The lower half of Fig. 8 shows a second peak observed for many samples. This peak is nearly 0.1 eV wide and is not observed for the heaviest doped samples,
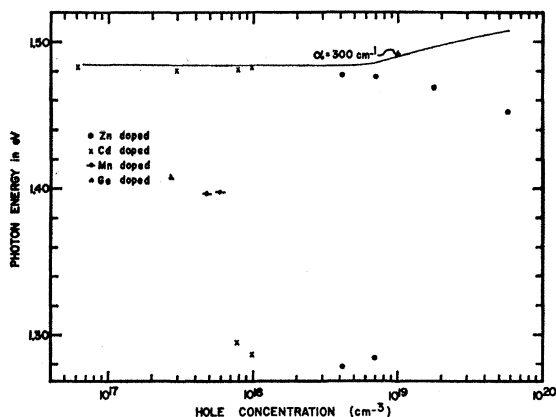

FIG. 9. Energy of fluorescence peak for p-type GaAs at 77°K and energy, at which an absorption coefficient of 300 cm^{-1} occurs for Zn- and Cd-doped GaAs.

while for many lightly doped n-type samples it is considerably more intense than the higher energy peak. The actual energy of this peak may be smaller by as much as 0.05 eV due to the photomultiplier response. The peak is, however, separated from the emission observed by others[15] at 0.95 eV. The origin of this emission is uncertain but the fact that the peak energy varies with both carrier concentration and the particular impurity involved, suggests that the center involved is not a simple one.

Note that the emission for sulfur-doped GaAs occurs at much lower energy than the principal peak for Se- or Te-doped GaAs. This is presumably related to the peculiar absorption edge shift observed for sulfur-doped GaAs.

It is interesting to note that the emission for Ge-doped samples at 77°K occurs at nearly 1.41 eV and still has the width characteristic of the higher energy emission. This is roughly 0.10 eV smaller than that observed for Te- or Se-doped GaAs, while the absorption edge for Ge-doped GaAs is only roughly 0.02 eV lower as discussed earlier. A Ge level roughly 0.08 eV from either the conduction or valence band is indicated by these results and while one might expect the latter, the evidence is insufficient to indicate which.

Figure 9 shows the front excited fluorescence peak for p-type at 77°K as a function of doping level. The energy at which the absorption coefficient of 300 cm^{-1} occurs is shown as a solid curve. The linewidth for p-type fluorescence is somewhat smaller than for n-type as seen in Fig. 7. Note that the fluorescence and absorption curves in Fig. 9 diverge at higher carrier concentration. The increasing energy exhibited by the absorption curve is a reflection of the Fermi level shift as discussed earlier. The decrease of the fluorescence energy is related to the shrinking of the energy gap for high doping levels. This latter effect has been observed by Nathan et al.[16] whose data include higher zinc concentration than that presented here.

The energy for p-type emission is somewhat less than that for n-type material, even at low doping levels. In addition, the fluorescence of the Mn-doped samples occurs at very nearly the energy which would be predicted from the activation energy determined from electrical measurements.[17] Such emission occurs even for samples in which Mn is not the major impurity (see Fig. 8), indicating that the recombination process at 77°K involves the Mn acceptor center. This result along with the similar conclusion of Nathan et al.[16] for the case of Zn- and Cd-doped GaAs adds weight to the speculation that the Ge acceptor level is about 0.08 eV from the valence band and the Ge donor is close to the conduction band, rather than the
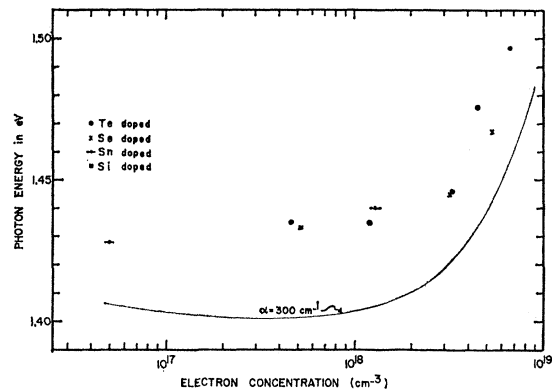
[16] M. I. Nathan, G. Burns, S. E. Blum, and J. C. Marinace (to be published); see also M. I. Nathan and G. Burns, Bull. Am. Phys. Soc. Ser. 2 8, 201 (1963); and M. I. Nathan and G. Burns, Appl. Phys. Letters 1, 89 (1962).
[17] L. J. Vieland, J. Appl. Phys. 33, 2007 (1962).

FIG. 10. Energy of fluorescence peak for n-type GaAs at 300°K and energy, at which an absorption coefficient of 300 cm^{-1} occurs for Te- and Se-doped GaAs.

reverse. The fluorescence of p-type Ge-doped GaAs is similar to n-type Ge-doped GaAs, as would be expected on the basis of the estimates of self-compensation. The room temperature fluorescence for n-type GaAs is given in Fig. 10. The most obvious difference between these data and the 77°K data is that the 300°K fluorescence and absorption data do not cross at high doping levels. This indicates that the recombination process leading to the fluorescence is different at the two temperatures, at least for heavily doped samples.

The corresponding data for p-type GaAs is given in Fig. 11. Note that the fluorescence peak for Mn-doped GaAs now occurs at the same energy as Cd-doped GaAs. This is further evidence that the recombination process leading to fluorescence at 300°K is different than the 77°K process. Since a large portion of the Mn centers will have trapped a hole even at 300°K one would expect that the state of ionization of the centers is not an important factor in the differences. It is also interesting to note that the diode emission which occurs at the same energy as the Zn fluorescence[16] at 77°K occurs at considerably lower energy[1] than the Zn fluorescence at 300°K.
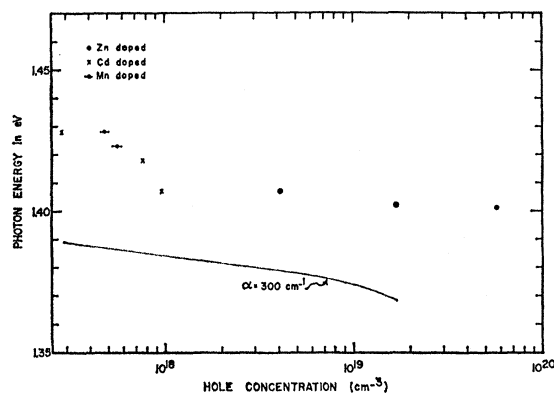

FIG. 11. Energy of fluorescence peak for p-type GaAs at 300°K and energy, at which an absorption coefficient of 300 cm^{-1} occurs for Zn- and Cd-doped GaAs.

These results indicate that the 300°K fluorescence for p-type GaAs does not involve the acceptor states. The fluorescence for lightly doped n- and p-type GaAs occurs at nearly the same energy as the energy gap. However, for heavily doped n-type GaAs, the recombination leading to the fluorescence seems to originate at an energy 1 to 2 kT below the Fermi level as computed from the absorption measurements.

IV. SUMMARY AND CONCLUSIONS

From the absorption measurements it can be seen that in general the shifts of the absorption edge with increased doping level occur in either direction and can be divided into two components. The shifts to higher energy predominate in n-type GaAs at 77°K and the shifts to lower energy predominate in p-type GaAs at 300°K. The shifts to higher energy are caused by the change of the Fermi level with doping. The shifts to lower energy can probably be explained as an effective decrease in the band gap. An additional phenomena which should be taken into account is the change in energy gap which occurs when the lattice parameter changes with doping level. The Burstein type shifts are not satisfactorily explained in detail by Eq. (1). This is not too surprising for such a simple theory, but it is of interest to know where the principal

difficulties lie. Part, but not all of the problem lies in not accounting for the nonparabolic nature of the conduction band. The effect of impurities in decreasing the band gap mentioned in the previous paragraph if understood and properly accounted for may explain most of the remaining differences. The shifts observed for heavily sulfur-doped GaAs are anamolous and not understood at the present time.

From the fluorescence measurements we see that the acceptor state is involved in the recombination which leads to fluorescence at 77°K, but not at 300°K. In addition we have seen that Ge as a dopant in GaAs produces a deep level, presumably an acceptor state roughly 0.08 eV above the valence band edge. The n-type fluorescence occurs essentially at the band gap energy for low doping levels and although the fluorescence energy increases at the highest doping levels, the recombination resulting in this emission does not primarily involve carriers in the neighborhood of the Fermi level.

ACKNOWLEDGMENTS

Conduction Band Minima of Ga(As$_{1-x}$P$_x$)†

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Photoresponse of surface barriers on samples of Ga(As$_{1-x}$P$_x$) covering the range $0 \leq x \leq I$ has been measured. Thresholds corresponding to both direct and indirect band-to-band excitations within the semiconductor and also photoinjection from the metal have been identified. The threshold of the direct transition varies with composition from 1.37 eV in GaAs to 2.65 eV in GaP. The indirect transition was followed for $x \gtrsim 0.38$ and again varied linearly from 2.2 eV in GaP to an extrapolated value in 1.62 eV in GaAs. The energy separation of the two conduction band minima in GaAs is in disagreement with previously reported values.

I. INTRODUCTION

IT has been demonstrated in recent work, that when the photovoltaic response is measured for a Schottky barrier at a clean metal-semiconductor interface, the response has two distinct spectral regions. The first region is where the photon energy exceeds E_g the energy gap of the semiconductor, and electron transitions take place from the valence to the conduction band. The second region, where $h\nu < E_g$ is generally a region where

the response per photon is much smaller, and a number of recent studies[1–9] have attributed this response to

[1] C. R. Crowell, W. G. Spitzer, L. E. Howarth, and E. E. LaBate, Phys. Rev. **127**, 2006 (1962).
[2] C. A. Mead and W. G. Spitzer, Appl. Phys. Letters **2**, 74 (1963).
[3] G. W. Mahlman, Phys. Rev. Letters **7**, 408 (1961).
[4] R. Williams, Phys. Rev. Letters **8**, 402 (1962).
[5] W. G. Spitzer and C. A. Mead, J. Appl. Phys. (to be published).
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[7] C. R. Crowell, W. G. Spitzer, and H. G. White, Appl. Phys. Letters **1**, 3 (1962).
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[9] D. Kahng, Solid State Electron. **6**, 281 (1963).