Optical Absorption of Gallium Arsenide between 0.6 and 2.75 eV

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The optical absorption coefficient of high-resistivity gallium arsenide has been measured over the range of photon energy 0.6 to 2.75 eV, at temperatures from 10 to 294°K. The main absorption edge shows a sharp peak due to the formation of excitons. The energy gap and exciton binding energy are deduced from the shape of the absorption curve above the edge. Their values at 21°K are 1.521 and 0.0034 eV, respectively. Absorption from the split-off valence band is observed, the spin-orbit splitting being 0.35 eV at the center of the zone. The exciton line shows unexplained structure on the low-energy side. Application of a stress splits the exciton line by 12 eV per unit [111] shear, and shifts it by −10 eV per unit dilation. Absorption due to the ionization of deep-lying impurity levels is observed, with thresholds at 0.70, 0.49, and 0.27 eV from the main absorption edge.

INTRODUCTION

RECENTLY single-crystal gallium arsenide of very high resistivity has become available.[1] An extensive investigation of the infrared lattice bands of this material is being made at this laboratory[2] and it seemed worthwhile to undertake a brief study of the absorption at shorter wavelengths, including the main absorption edge where direct transitions from the valence band to the conduction band begin. The absorption coefficient of mechanically polished specimens of gallium arsenide varying in thickness from 6 mm to 0.6 μ was therefore measured at photon energies from 0.6 to 2.75 eV. Specimens less than 5 μ thick were glued to a glass backing for support. Thicker specimens were suspended freely in the cryostat.[3] Measurements were made at 10, 21, 55, 90, 185, and 294°K.

From 0.6 to 1.4 eV the absorption is small and is due to the ionization of unknown impurities. At low temperatures three broad thresholds are observed. As the temperature is varied, the positions of the thresholds vary but their separation from the absorption edge remains approximately constant. These results are consistent with Hall effect and photoconductivity data on similar material.[1,4] In the region around 1.5 eV, where excitation across the energy gap begins, absorption due to the formation of excitons is observed. By fitting the results to the theory of Elliott[5] (modified to take into account the nonparabolic bands of gallium arsenide) we can find the exciton binding energy and the band gap. The band gap turns out to be somewhat larger than found by previous workers.[6,7] A rather broad threshold for transitions from the split-off valence band to the conduction band is observed, giving a direct measure of the spin-orbit splitting at the center of the Brillouin zone. No further structure is seen up to 2.75 eV, where the absorption constant is 1.4×10^5 cm^{-1}.

A preliminary note describing qualitatively the exciton line has already been published.[8]

EXPERIMENTAL DETAILS

Standard Royal Radar Establishment (R.R.E.) equipment was used and the experimental method was as described in a review article by McLean.[9] The monochromator was a prism-grating combination designed by Roberts[10] based on the Leiss double-prism monochromator.[11] With a 45 000-lines/in. Bausch & Lomb grating, a resolution of about 30 000 could be obtained at 8000 Å. This resolution was not usually required, and the slits were widened to give a resolution of between 5000 and 10 000. A Mullard 61SV lead sulphide detector was used at photon energies below 1.48 eV. Above this an EMI 9558 photomultiplier was used.

The absorption edge of gallium arsenide is extremely steep at low temperatures and, because of the difficulty of preparing thin specimens, small transmission ratios (down to 10^{-4} or less) had to be measured. Spectral purity was therefore essential. The resolution of the prism half of the monochromator was kept as high as possible and baffles were placed in the monochromator to reduce scattered light. Grating ghosts were found to be unimportant except when the highest resolution (better than 2×10^{-4} eV) was required. The apparent transmission of a specimen at a wavelength setting where it should have been opaque was less than 10^{-5}. Care was taken that light scattered around the sides of the specimen was much less than this (except in one case mentioned below).

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[1] C. M. Gooch, C. Hilsum, and B. R. Holeman, J. Appl. Phys. **32**, 2069 (1961).
[2] W. Cochran, S. J. Fray, F. A. Johnson, J. E. Quarrington, and N. Williams, J. Appl. Phys. **32**, 2102 (1961).
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[9] T. P. McLean, *Progress in Semiconductors* (Heywood and Company, Ltd., London, 1960), Vol. 5, p. 53.
[10] G. G. Macfarlane, T. P. McLean, J. E. Quarrington, and V. Roberts, Phys. Rev. **108**, 1377 (1957).
[11] V. Roberts, J. Sci. Instr. **29**, 134 (1952).

The specimens were all cut from two "semi-insulating" ingots of gallium arsenide[1] in which the carrier concentration was less than 10^{10} per cc, and the mobility in the region 5000 to 6000 cm^2/V-sec, which corresponds to an ionized impurity concentration of between 3×10^{16} and 10^{17} per cc. The absorption in the impurity ionization region below the main edge shows large variations from specimen to specimen indicating variations in impurity content, even within a single ingot. Ten specimens were used, ranging in thickness from 6 mm to 0.6 μ. They were all hand ground and polished as previously described.[10] During polishing, specimens over 5 μ thick were glued to an optical flat from which they were subsequently removed and mounted in the cryostat. Specimens thinner than this broke on removal from the glass. After one side had been polished, they were therefore glued permanently to a flat piece of thin glass with "Araldite" thermosetting epoxy resin. Owing to the difference between the expansion coefficients of glass and gallium arsenide, these specimens were strained at low temperatures. This strain is evidenced by a shift of the absorption edge to shorter wavelengths (compressive strain), a slight broadening and a splitting of the exciton line. The splitting is only seen in glass-backed specimens, but the shift and broadening are apparent in freely suspended specimens less than 25 μ thick, even at room temperature. This effect is demonstrated in Fig. 1, in which the results at 21°K for several freely suspended specimens of the same material but of different thicknesses are shown. (Only representative experimental points are shown. Measurements were made at intervals of approximately 10^{-4} eV in this spectral range.) It appears that the two thinnest specimens contain internal strains, the dilatational strain dominating the optical behavior and shifting the edge to longer wavelengths. The average dilatation, estimated from the shift, is 2×10^{-4} in the 6.5-μ specimen and 5×10^{-5} in the 20.3-μ one. Because of this strain the 6.5-μ specimen curled slightly on removal from its backing. About 5×10^{-5} of the incident radiation found its way round this specimen to the photomultiplier and had to be allowed for in the calculation of large absorption coefficients.

The thickness of specimens over 150 μ was measured with a micrometer, and of those less than 1 mm thick from the spacing of interference fringes due to multiple reflection inside the specimen. The region of overlap provides a check on the refractive index, which was found to be 3.32 at a wavelength of 3 μ, in good agreement with the more precise determination of 3.313.[12] As the absorption edge is approached, the refractive index increases and was found from interference data on thin specimens in which the order of interference was known. The data at room temperature are in good agreement with reflectivity measurements on similarly

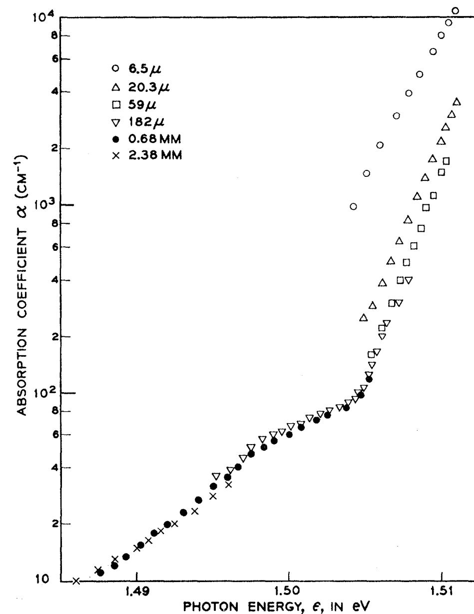

FIG. 1. Absorption of GaAs at 21°K for different specimen thicknesses.

prepared specimens.[13] The refractive index n can be represented over the range 0.6 to 1.3 eV, within the experimental accuracy of 2%, by a formula due to Moss[14]

$$n^2-1=A/(\epsilon_0^2-\epsilon^2),$$

where ϵ is the photon energy, $A=53$ eV^{-2}, and $\epsilon_0=2.26$ eV. Above 1.3 eV the data of Morrison were used to calculate the reflectivity correction to the absorption. Because of interference effects, absorption measurements were not made at optical densities less than 0.5 on specimens less than 2-mm thick.

RESULTS AND DISCUSSION

1. Main Absorption Edge

Results for freely suspended specimens in the range 1.37 to 1.52 eV are shown in Fig. 2. The results from the two thinnest specimens have been shifted bodily to higher energies in order to smoothly join the curves for thicker specimens at the same temperature. The amount by which the 6.5-μ results were shifted is recorded in column 4 of Table I. The dashed lines show the absorption at low levels with that due to impurities subtracted. The curve for 55°K is omitted altogether for clarity; it is parallel to the 21°K curve at 0.003 eV lower energy. Figure 3 shows the continuation of Fig. 2 to higher energies, on a linear vertical scale, for the 6.5-μ specimen only. The curves are the fitted theoretical curves [Eq. (3)].

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[13] R. E. Morrison, Phys. Rev. **124**, 1314 (1961).

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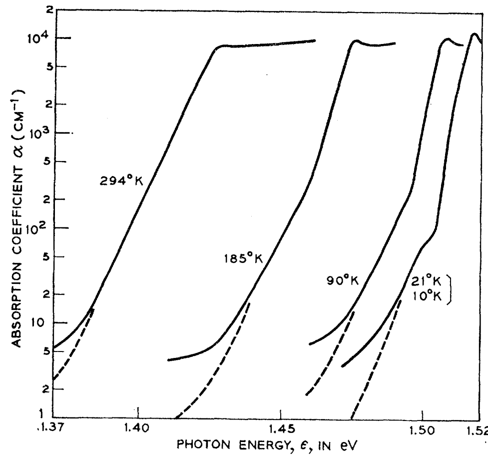

FIG. 2. The absorption edge of GaAs. Solid line, corrected for strain only. Dashed line, corrected for impurity absorption.

The theory of the optical absorption of semiconductors near the band edge, allowing for the Coulomb interaction of the electron and hole produced in the transition and for the consequent formation of excitons, has been worked out in the effective mass approximation by Elliott.[5] For simple nondegenerate parabolic bands at the center of the Brillouin zone, between which transitions are direct and parity-allowed, the absorption coefficient in the region of continuous absorption is given by

$$\alpha=\frac{2\pi e^2\hbar}{m^2c}\left(\frac{2m^*}{\hbar^2}\right)^{\frac{3}{2}}|P_{cv}|^2\frac{\epsilon^{\frac{1}{2}}_x}{n\epsilon}\frac{\exp z}{\sinh z},\quad \epsilon>\epsilon_g \tag{1}$$

$$z^2=\pi^2(\epsilon-\epsilon_g)/\epsilon_x.$$

Here ϵ is the photon energy, ϵ_g the energy gap, P_{cv} the momentum matrix element at $k=0$, m^* the density of states effective mass, and n the refractive index. The exciton binding energy ϵ_x is given in the simple theory by $\epsilon_x=\mu e^2/2\hbar^2\kappa^2$, where $\mu^{-1}=m_e^{-1}+m_h^{-1}$, and κ is the dielectric constant. If we take the degeneracy of the valence band into account[9,15] and use the published band structure constants[16,17] and dielectric constant[12]

TABLE I. Energy gap and exciton binding energy in GaAs.

T (°K)	Exciton line (eV)	ϵ_x (eV)	$\Delta\epsilon_g$ (eV)	ϵ_g(corrected) (eV)
10	1.5130			
21	1.5130	0.0034	0.005	1.521
55	1.5100	0.0032	0.005	1.518
90	1.5030	0.0033	0.005	1.511
185	1.4722	0.0030	0.004	1.479
294		0.0025	0.006	1.435

we find $\epsilon_x=0.0044$ eV. In addition to the continuous absorption Elliott[5] predicts a series of lines at energies given by $\epsilon_g-\epsilon_x/n^2$, where $n=1, 2, \cdots$, of intensity proportional to n^{-3}. As in germanium[18] only the first of these is resolved.

In gallium arsenide the bands are not simple, in particular the conduction and light hole bands are nonparabolic. An accurate calculation for this case along the lines of Elliott would be impracticable, and instead we resort to the following approximation. Equation (1) may be written

$$\alpha=\alpha' f(z), \tag{2}$$

where α' is the absorption calculated neglecting Coulomb effects, and $f(z)(=z\exp z/\sinh z)$ is a factor taking them into account. We can calculate α' for the actual band structure of gallium arsenide.[6,16,19] We then assume $f(z)$ to be the same as in Elliott's theory. If the variation of refractive index with ϵ is taken into account, we find that the absorption is approximately given by

$$\alpha=\alpha(\epsilon_g)\exp z/2\sinh z, \tag{3}$$

where $\alpha(\epsilon_g)$ is the absorption coefficient at the band edge [obtained by putting $\epsilon=\epsilon_g$ in Eq. (1)]. Comparison of Eq. (3) with the exact results of the calculation shows that the value of ϵ_x found by fitting this equation to the results will not be more than 5% below the correct value. Equation (3) is fitted to some of the results in Fig. 3. The minimum absorption in the "valley" is taken to be $\alpha(\epsilon_g)$; ϵ_x is treated as a disposable parameter to give the best fit, and ϵ_g is defined by the position of the exciton peak which is at $\epsilon_g-\epsilon_x$. At 294°K, where the exciton peak is not resolved, ϵ_g is adjusted to give the best fit. Because of thermal broadening, this fit may be spurious. The values of ϵ_x and ϵ_g are given in Table I, along with the measured positions of the exciton peak and the strain correction $\Delta\epsilon_g$. The fitting error in ϵ_x is ±0.0002 eV (±0.0005 eV at 294°K) and the estimated error in ϵ_g is (chiefly due to $\Delta\epsilon_g$) is ±0.0015 eV (±0.003 eV at 294°K). The variation of ϵ_g with temperature is shown in Fig. 4, along with the previously accepted curve,[7] which is much too low, having been measured on specimens $\frac{1}{2}$-mm thick. It is of interest that the variation of ϵ_g with temperature is about three times that to be expected from the product of the bulk thermal expansion[20] and the change of ϵ_g per unit dilatation.[21,22]

The principal systematic error in this procedure is presumably the assumption that $f(z)$ is the same as in

[15] T. P. McLean and R. Loudon, J. Phys. Chem. Solids **13**, 1 (1960).
[16] H. Ehrenreich, Phys. Rev. **120**, 1951 (1960).
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[20] F. Assmuss (unpublished), quoted by H. Welker and H. Weiss, *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 3, p. 51.
[21] W. Paul, J. Appl. Phys. **32**, 2082 (1961).
[22] T. B. Bateman, H. J. McSkimin, and J. M. Whelan, J. Appl. Phys. **30**, 544 (1959).

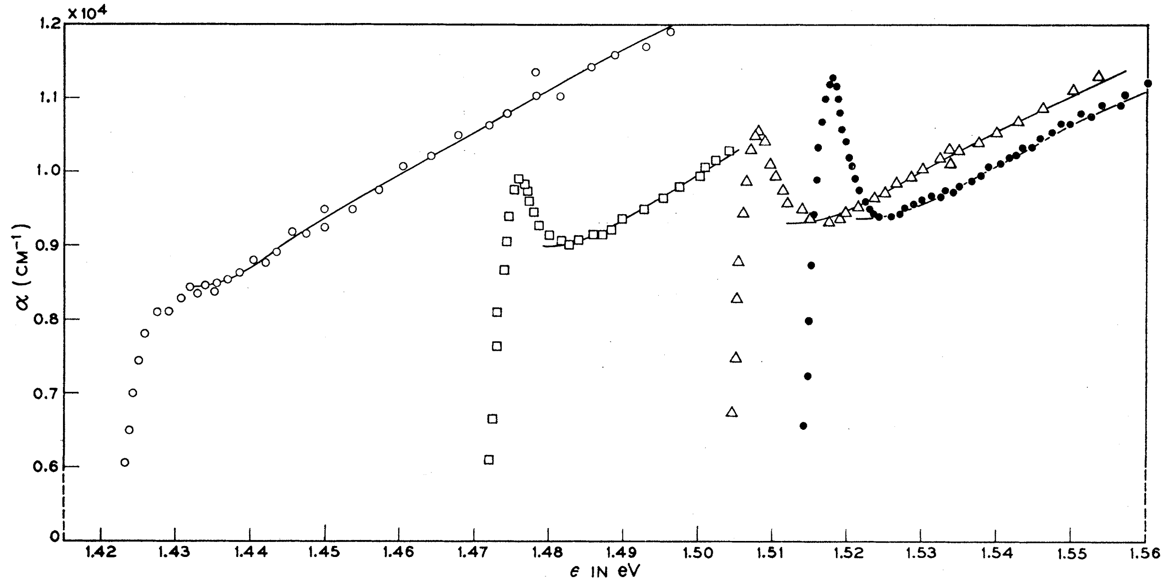

FIG 3 Exciton absorption in GaAs; ○ 294°K, □ 186°K, △90°K, ● 21°K.

the case of simple bands. As the effective mass in the conduction and light hole bands increases with ϵ, ϵ_x might also be expected to increase. Equation (1) shows that the absorption increases with ϵ_x, so the slope of the absorption curve will be greater than the theory predicts. When Eq. (3) is fitted to the experimental results the fitted value of ϵ_x, which depends inversely on the slope, will be too small. This may at least partially explain the fact that as in germanium[18] ϵ_x found by this method is below the theoretical value.

The absolute value of $\alpha(\epsilon_g)$ below 90°K is 9400 ± 400 cm^{-1}. In Eq. (1) P_{cv} can be calculated from the known band structure constants if exchange is neglected,[16,19] and we find $\alpha(\epsilon_g)=8900$ cm^{-1}. As in the case of germanium,[9] the agreement is excellent, justifying the neglect of exchange.

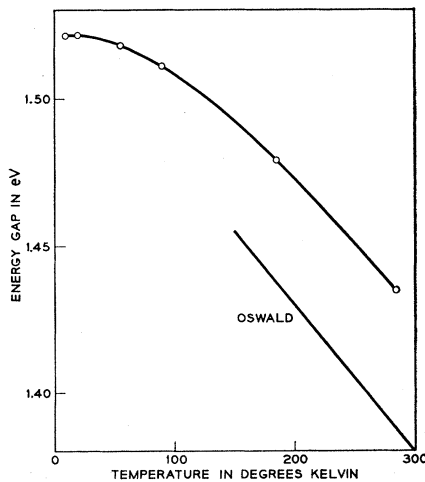

FIG. 4. Energy gap in GaAs. Upper curve, present measurements. Lower curve, reference 7.

2. Effect of Strain

Results at 21°K for a 4.7-μ specimen cut parallel to a (111) plane and glued to a glass backing are shown in Fig. 5. Also shown for comparison is the corresponding curve for a freely suspended specimen. The strain in the specimen was found by comparing the measured contraction of the glass between 50°C (the setting temperature of the adhesive) and 77°K (below which contraction is negligible) with that to be expected for gallium arsenide. The expansion coefficient of gallium arsenide at room temperature is 5.7×10^{-6},[20] which is

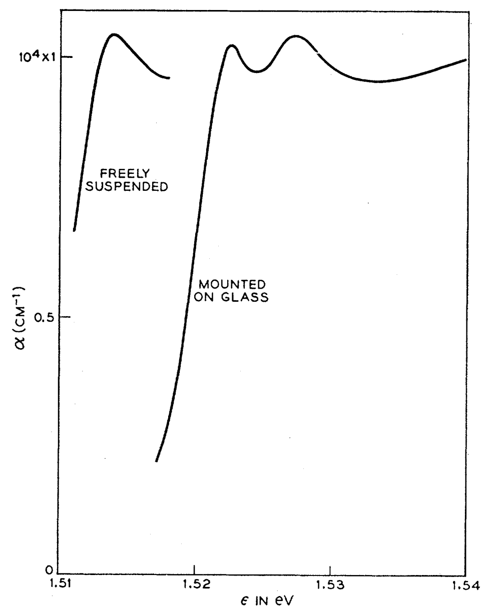

FIG. 5. The effect of strain on the exciton line in GaAs at 21°K.

equal to that of germanium.[23] It seems reasonable to assume that this equality holds at all temperatures, in which case we can calculate the contraction of the gallium arsenide to be 1.1×10^{-3}. That of the glass was 1.5×10^{-3}, giving a strain of $(4\pm1)\times10^{-4}$,[24] which is assumed to have been homogeneous. The increase in the band gap δE and the splitting 2Δ of the valence band under the conditions of this experiment are given by[25]

$$\delta E=(2-\lambda)(D_d{}^c-D_d{}^v)S,\quad 2\Delta=\tfrac{4}{3}(1+\lambda)D_u{}'S,$$

where $D_u{}'$ is the shift of the valence band for unit shear in the (111) plane, $D_d{}^c$ and $D_d{}^v$ the shifts in the conduction and valence band edges for unit dilatation, and $\lambda=2(c_{11}-2c_{12}-2c_{44})/(c_{11}+2c_{12}+4c_{44})=0.465$ in gallium arsenide.[22] The conduction band is nondegenerate and so cannot be split, and if we assume that the exciton binding energy is the same for each of the two valence bands we may equate 2Δ with the splitting of the exciton peak, 0.0045 ± 0.0003 eV, and δE with the mean shift of the energy gap from the unstrained position, 0.006 ± 0.001 eV. We find

$$|D_u{}'|=6\pm1.5 \text{ eV},\ D_d{}^c-D_d{}^v=-10\pm3 \text{ eV}.$$

The value for $D_d{}^c-D_d{}^v$ is in good agreement with value of -9 eV found from the shift of the edge with hydrostatic pressure.[21,22]

The value of ϵ_x found by fitting Eq. (3) to the experimental results in this specimen is 0.0025 eV, appreciably lower than that for a freely suspended specimen. It appears from the results on several specimens that the more strained the material the lower the fitted value of ϵ_x. This does not appear to be due to fitting errors caused by broadening or splitting of the exciton line, which would both tend to reduce the slope of the $\alpha(\epsilon)$ curve and to give, therefore, too high a value of ϵ_x. It could be due to a genuine change of ϵ_x with strain, but failure of the theory to hold for strained material seems the more probable explanation. It appears that in strained specimens the value of ϵ_g deduced by fitting the theory to the results in the continuous absorption region is consistently higher by up to 0.001 eV than that found by adding ϵ_x to the mean position of the exciton line. This suggests that ϵ_x may be low by up to 0.001 eV.

3. Region above 1.5 eV

The absorption at 21°K of specimens 1.4 and 0.67 μ thick glued to a glass backing is shown in Fig. 6. The surface damage produced by polishing presumably extends right through these specimens, broadening the exciton line at 1.53 eV almost to vanishing point. A weak absorption edge is clearly visible in the region of 1.88 eV due to transitions from the third valence band (split off by spin-orbit coupling) to the conduction band. This edge is very much broader than the main edge, presumably because the lifetime of the exciton is much shortened by autoionization. Because of this broadening and the uncertainty in the background absorption, it is not possible to fit Eq. (3) to the results. However, by inspection of Fig. 6 one can estimate the spin-orbit splitting to be 0.35 ± 0.01 eV. Certainly the value of 0.33 eV, obtained from the infrared absorption due to free holes[26], seems too low to be consistent with the present data.

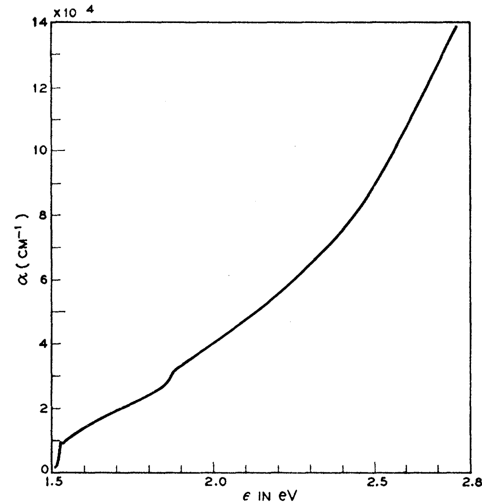

FIG. 6. Absorption of GaAs at 21°K.

4. Broadening of the Exciton Line

The low-energy side of the absorption edge (Fig. 2) shows the exponential broadening known as the Urbach effect.[27] Only at high temperatures can this be described by a single exponent: 145 eV^{-1} at room temperature. At low temperatures, the curve breaks up into two parts. The upper portion which is roughly Gaussian, with a limiting width at low temperatures of 3×10^{-3} eV. If this width is attributed solely to lifetime broadening[28] the exciton lifetime is 1.3×10^{-12} sec (this is presumably a lower limit). Below 100 cm^{-1}, there is a shoulder whose intensity remains constant and finite at low temperatures and is therefore difficult to explain in terms of the simultaneous absorption of a phonon and an optical phonon,[29] which should vanish at temperatures well below the Debye temperature. The large impurity content of these specimens, up to 10^{17} per cc,

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[24] This is lower than the figure given in reference 8, as the contraction of the glass has been remeasured.
[25] W. H. Kleiner and L. M. Roth, Phys. Rev. Letters **2**, 334 (1959).
[26] R. Braunstein, J. Phys. Chem. Solids **8**, 280 (1959).
[27] F. Urbach, Phys. Rev. **92**, 1394 (1953).
[28] G. G. Macfarlane, T. P. McLean, J. E. Quarrington, and V. Roberts, Phys. Rev. **111**, 1245 (1958).
[29] W. P. Dumke, Phys. Rev. **108**, 1419 (1957).

might produce a "tail" in the density of states at the band edge[30] but quantitative agreement cannot be obtained. If this were the correct explanation, it would be difficult to understand why the absorption in this region varies little if at all between specimens (see Fig. 1), whereas the absorption which is unambiguously due to impurities varies by a factor of over 2 (Fig. 7).

5. Impurity Ionization

The absorption at levels below 4 cm^{-1} varies substantially from specimen to specimen and is presumably due to impurities. At energies below 0.7 eV, all specimens have an absorption coefficient of less than 0.03 cm^{-1}. The absorption in the region between 0.7 and 1.4 eV is shown for two specimens at two temperatures in Fig. 7. The absorption coefficient rises from a threshold energy ϵ_i according to the empirical law $\alpha \propto (\epsilon - \epsilon_i)^2$. At room temperature only one threshold is observed, but at lower temperatures two further thresholds appear at higher energies. In both cases, the initial rise is as $(\epsilon - \epsilon_i)^2$ when the (more or less constant) background due to lower thresholds is subtracted. Extrapolation of this square law to zero α gives the empirical values for ϵ_i, $i = 1, 2, 3$, given in Table II. The third threshold shows a variation between specimens greater than the estimated error of ± 0.02 eV. This could be due to several transitions contributing to the absorption in this region, due to different impurities which might have different relative concentrations in different specimens. As well as the ϵ_i, the differences $\epsilon_g - \epsilon_i$ are tabulated, where ϵ_g is the band gap at that temperature. It will be seen that for the first two threshold $(\epsilon_g - \epsilon_i)$ remains constant with temperature within experimental error, while ϵ_i varies considerably. This is to be expected if the absorption is due to the ionization of an electron (hole) from an occupied acceptor (empty donor) level to the conduction (valence) band. The slow rise in α near threshold

TABLE II. Thresholds for impurity ionization in GaAs.

T (°K)	Specimen	ϵ_1 (eV)	$\epsilon_g - \epsilon_1$ (eV)	ϵ_2 (eV)	$\epsilon_g - \epsilon_2$ (eV)	ϵ_3 (eV)	$\epsilon_g - \epsilon_3$ (eV)
21	1	0.80	0.72	1.04	0.48	1.27	0.25
	2	0.83	0.69	1.03	0.49	1.23	0.29
77	2	0.80	0.71	1.00	0.51	1.18	0.33
185	2	0.80	0.68	0.97	0.50		
294	1	0.73	0.70				
	2	0.72	0.71				

[30] D. L. Dexter, Nuovo cimento 7, 245 (1958).

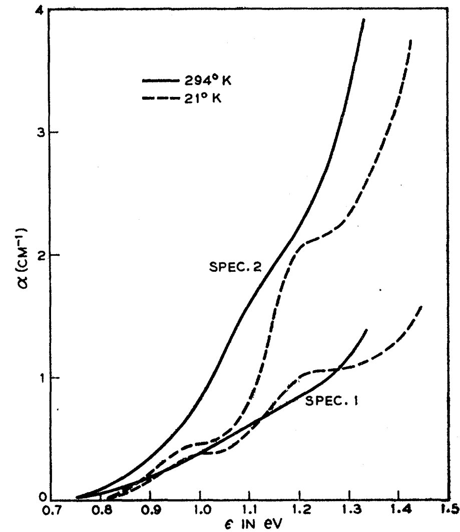

FIG. 7. Absorption due to impurities in GaAs.

is characteristic of excitation from charged centers.[31] The three levels observed may perhaps be identified with the levels at approximately 0.70, 0.47, and 0.30 eV observed (among others) in photoconductive and related experiments by the RCA group.[4] In this case the 0.70- and 0.47-eV levels would be donors, in agreement with the suggestion[32] that a donor at about 0.70 eV is responsible for the high resistivity of this material. On the other hand, Gooch *et al.*[1] believe the high resistivity to be due to an acceptor at 0.76 eV from the conduction band. In this case, the absorption observed here would be due to excitation of electrons to the conduction band, and the other two levels would be acceptors at 0.49 and 0.27 eV from the valence band. These have no counterpart in Bube's experiments.

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[31] E. Burstein, G. S. Picus, and N. Sclar, *Proceedings of the Conference on Photoconductivity, Atlantic City, November 4–6, 1954* (John Wiley & Sons, Inc., New York, 1956), p. 353; R. Newman and W. W. Tyler, *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1958), Vol. 8, p. 83.
[32] J. Blanc and L. R. Weisberg, Nature **192**, 155 (1961).