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Green Luminescence from Solution-grown Junctions in GaP Containing Shallow Donors and Acceptors*

Abstract: Gallium phosphide diodes doped with shallow donors and acceptors that partially compensate one another in both the *n*- and *p*-regions give rise to bright green emission.* This green emission is more efficient at room temperature than the so-called "*A*-line" emission commonly observed in lightly doped diodes. The peak energy of the green emission varies with the donor binding energy, and the emission is interpreted as a radiative donor-acceptor or a donor-valence band transition. Its intensity-voltage dependence and its current-voltage dependence are discussed for room temperature conditions. At 77°K, large shifts in peak energy were observed to accompany changes in applied voltage. Data concerning this peak shift, the intensity-voltage dependence, and the linewidth-voltage dependence are presented and discussed. Time effects and the temperature dependence of the quantum efficiency are also examined.

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* The growth of GaP from a gallium-rich solution is discussed by L. M. Foster, T. S. Plaskett, and J. E. Scardefield in a companion paper (page 114 of this issue). Those authors also treat the morphology and dendritic growth habit of the crystals, describe the production of built-in junctions in crystals doped with Zn and S, Se, or Te, and propose a mechanism for the formation of those junctions.

Introduction

A large number of emission lines with energies near the bandgap energy have been observed in GaP. Located at energies slightly higher than 2 eV, they constitute what is generally called the "green emission" of GaP. These emission lines have been extensively studied in photoluminescence[1] and in electroluminescence.[2]

In general, two types of emission lines are found at low temperatures: (1) a series of bound exciton lines and their phonon satellites and (2) donor-acceptor pair spectra. The exciton line with the highest energy has been termed the "*A*-line." It is particularly easy to observe; its interpretation, however, is not completely clear (recently it was suggested that the *A*-line may be associated with the presence of nitrogen[3]). Donor-acceptor pair spectra consist of a series of sharp lines at high energies and a broad band at low energies. These spectra originate from radiative transitions between donors and acceptors spaced at various distances throughout the lattice. The energy of the emitted photon is modified by the coulomb interaction energy, $e^2/\epsilon r$, between the donors and acceptors (where e is the electronic charge, ϵ is the dielectric constant, and r is the donor-acceptor distance). At 4.2°K and below, recombination over small distances gives rise to the various sharp, high-energy lines, and over large distances, to the broad band.[4,5] At nitrogen temperature, the sharp lines of the pair spectra no longer can be resolved because of phonon broadening, and only one emission band is observed.

At high temperatures the shallow donor and acceptor levels will be ionized to a large extent and shallow donor-acceptor transitions should thus be more difficult to observe. Gershenzon et al.[2] have reported that only band-to-band transitions (or in a different interpretation, the phonon-broadened *A*-line) could be observed in GaP diodes at room temperature. In heavily doped diodes, however, Foster and Pilkuhn[6] found emission lines thought to be due either to donor-acceptor or to donor-valence band recombinations. It is the aim of this paper to present a more detailed analysis of these emission lines and the related properties of the *p-n* junctions from which they are emitted.

Experiment

GaP platelets were grown from a Ga-GaP solution at temperatures little over 1100°C. The platelets contained Zn as a shallow acceptor and Te, S, or Se as shallow donors, in concentrations ranging from 5×10^{18} to 1×10^{19} cm^{-3}. The material was very heavily compensated, and *p-n* junctions could form during the solution growth within the platelets (see Ref. 7, the companion paper). Crystal sections which contained one such *p-n* junction were selected and contacted with Au-Sn and

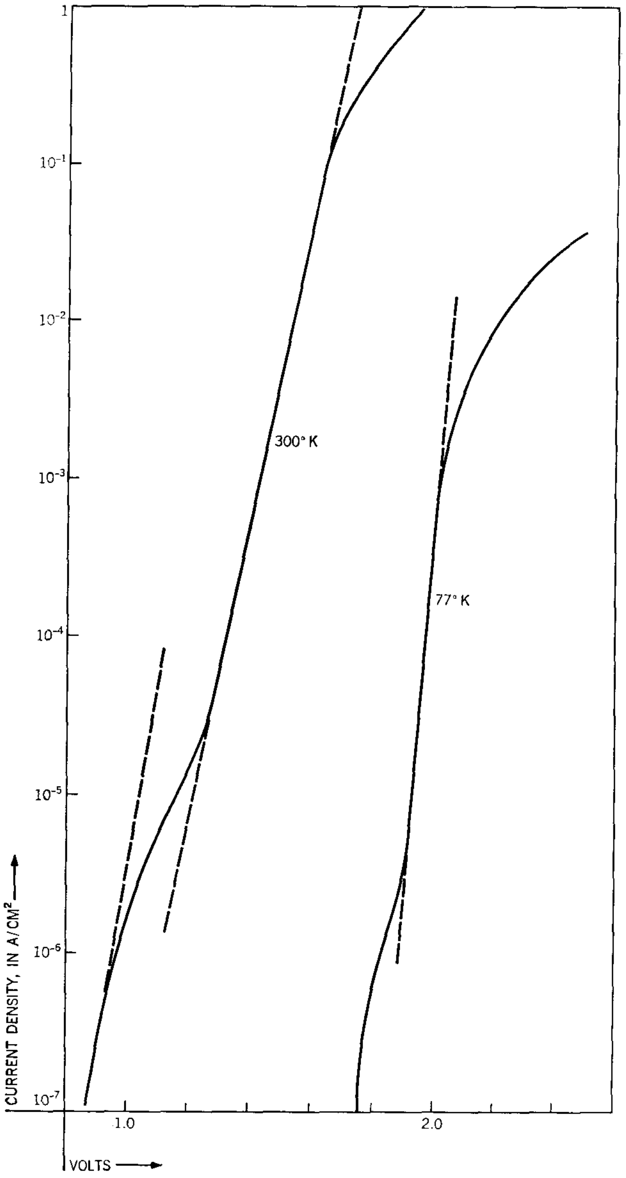


Figure 1 Current density-voltage dependence of a Zn-S doped diode at 300°K and 77°K. Diode area : 1.07×10^{-3} cm².

Au-Zn alloys on the *n*- and *p*-side respectively.[2]

The forward bias electroluminescence was studied under d.c. and pulsed conditions. For spectral measurements, the diodes were immersed in a suitable constant temperature bath with their emission focused on the entrance slit of a Bausch and Lomb grating monochromator, which had a resolution limit of 3 Å. The detector was either an RCA 7265, or a Dumond 6911 photomultiplier (with S-20 and S-1 spectral characteristics). For measurements at continuously variable temperature the diodes were mounted on a metal heatsink inside a variable temperature cryostat.

Results and discussion

- *Electrical properties: I-V characteristics of the junctions*

Figure 1 shows the *I-V* characteristics of a Zn and S doped diode for 77°K and 300°K. Over a large current range, the current density varies as exp $(eV/\beta\, kT)$. In Fig. 1, β is 1.8 for 300°K and is 2.1 for 77°K. This exponential relationship was typical of many diodes, for which β varied between 1.8 and 2.0 at 300°K and was close to 2 or little over 2 at 77°K. We shall interpret this current as being due to recombination in the space charge region of the junction. The current-voltage characteristic can be then expressed in the following form[8]

$$I = I_0 \exp\{-(E_g - eV)/2kT\}, \qquad (1)$$

where E_g is the bandgap energy, V is the applied voltage, and I is the current. I_0 is much less temperature dependent than the exponential factor. From the change in current between 300° and 77°K at constant voltage, a value of 2.31 eV can be derived from the data in Fig. 1 for E_g. This value is close to that for the bandgap of GaP (2.325 eV at 0°K). Other diodes have values for β larger than 2, particularly at low temperatures, indicating the presence of tunneling or other processes that give an additional current component not flowing over the junction barrier.

Excess currents can be found, particularly at low current densities, and some deviations from the exponential voltage dependence are observed in all diodes. A kink in the *I-V* characteristic appears to occur in many cases at a certain voltage (near 1.3 volts at 300°K, and near 1.9 volts at 77°K). These kinks are also mentioned in Ref. 9. No influence of the dopant on the *I-V* characteristic (including the kink voltages) was observed. The excess currents have been interpreted as being due to tunneling because of their temperature insensitivity.[10]

- *Room temperature emission spectra*

A comparison between the emission spectra of three different types of diodes at 300°K is made in Fig. 2. The Type I, undoped, diode was an *n*-type crystal containing a Zn-doped *p*-type region and having a very low concentration of unidentified, unintentional doping. The Type II, Zn-Te doped, diode had the *n*-side Te doped and partly compensated with Zn, and the *p*-side Zn-doped and partly compensated with Te. The Type III diode was Zn-S doped in a similar manner.

The peak of the near bandgap emission of the undoped diode occurs at 2.23 eV; this line can be identified as the so-called *A*-line complex (or band-to-band transition) reported by Gershenzon et al.[2] The peak emission of the Zn-Te doped diode occurs about 36 meV below the *A*-line peak, however, and that of the Zn-S doped diodes (and also that of Zn-Se doped diodes, not shown) occurs

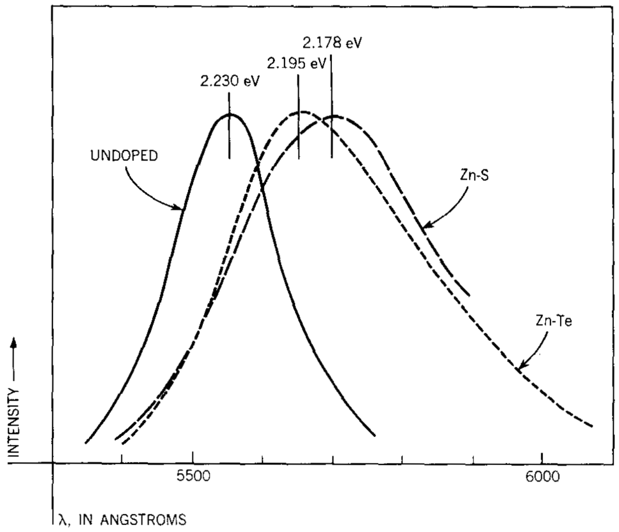


Figure 2 Emission spectra of (I) undoped, (II) Zn-Te doped, and (III) Zn-S doped diodes. All at 300°K. Arbitrary intensity scales are used.

about 53 meV below the A-line peak. There is also a difference in linewidth: the 2.23 eV line has a 180 Å half-width and is narrower than the other two lines in Fig. 2, which have a 340 Å half-width.

These results show that the emission from the doped diodes is different from the A-line emission. It must involve the donor state since it shifts in the expected manner to lower energies as the donor state becomes deeper. The peak difference of the curves for Type II and III diodes, Fig. 2, is 17 meV, which should relate to the difference in donor binding energies between Te and S. From pair spectra data at low temperatures,[5,11] the sum of donor and acceptor binding energies, $E_D + E_A$, can be determined as 0.1402 eV for the case of Zn + Te, and 0.1549 eV for the case of Zn + S. The difference between these values is about 15 meV, which corresponds to the difference in donor binding energies. This has to be compared with the 17 meV value derived from Fig. 2 which, of course, is not too accurate a value because of the width of the lines of Curves II and III. Because the acceptor impurity was not varied in these diodes it cannot be determined whether the radiative transition at 300°K is a donor-acceptor or a donor-valence band transition. This question will be discussed later in connection with the temperature dependence of the emission bands II and III, and that of the bandgap.

Besides this emission near 2.2 eV, all doped diodes emit light at 1.77 eV because of the presence of the deep donor, oxygen, and zinc. This red component could become very small at high current densities in the Zn-S, Zn-Te, or Zn-Se doped materials, where an attempt had been made to exclude oxygen; nevertheless, a certain small amount always remained. The 1.77 eV line could be suppressed to the extent that it contained less than one tenth of the total number of photons. Deliberate addition of oxygen shifts more of the emission into the red, and the green component becomes very small.

- *Dependence of intensity on voltage and current*

We confine ourselves in this section to the high voltage regime where current (and light intensity) varies as exp $(eV/\beta kT)$ with the applied voltage, and where any excess current component due to tunneling is negligible. In the case of a diffusion current, the coefficient β should be unity. Predominant recombination in the space charge region, which occurs with the participation of trap levels, leads to $\beta = 2$.

Recombination through defects in the space charge region was recently treated theoretically by Morgan.[12] According to his calculations, for radiative recombination the intensity should vary as exp $(eV/1kT)$ at *low* voltages, as exp $(eV/2kT)$ at *high* voltages and, eventually, as exp $(eV/\beta\, kT)$ with $\beta > 2$ at *very high* voltages. The kink voltage V_K, at which β changes from 1 to 2, is determined by the energy position of the defect and the ratio of the capture cross sections for electrons and holes C_n/C_p for that center:

$$V_K = \left| V_g - 2(V_t - V_v) - \frac{kT}{e} \ln \frac{C_n}{C_p} \right|, \qquad (2)$$

where V_g is the bandgap, $(V_t - V_v)$ is the distance of trap level from the valence band in volts. Several kinks found in the intensity-voltage relationship of copper-doped GaAs diodes have been interpreted in this manner.[13] The kink voltage, V_K, is approximately twice the distance of the defect level from the midgap (see Eq. 2); i.e., kinks for the emission with lower energy should occur at the lower voltage. This means that the low energy emission will be less intense relative to the high energy emission at high voltages.

In Fig. 3 the intensity-voltage relationship for a GaP diode containing two donor levels (due to sulfur and oxygen) is shown for 300°K. The logarithm of the peak intensity has been plotted against the junction voltage (which was corrected for the voltage drop across the series resistance). At low voltages the red emission is more intense; at high voltages the green emission prevails. The kink voltage for the green emission, where β changes from 1.0 to 1.8, is $V_K = 1.97$ volts. A value of approximately 2.1 volts is estimated from Eq. (2) (with $V_t = 2.18$ volts), if the last term therein is assumed to be small. The red emission shows two kinks, one at 1.86 volts, where β changes from 1.18 to 1.8, and a second one at about 2 volts. (This second kink can be better observed in diodes with less of the 1.77 eV emission). Qualitatively,

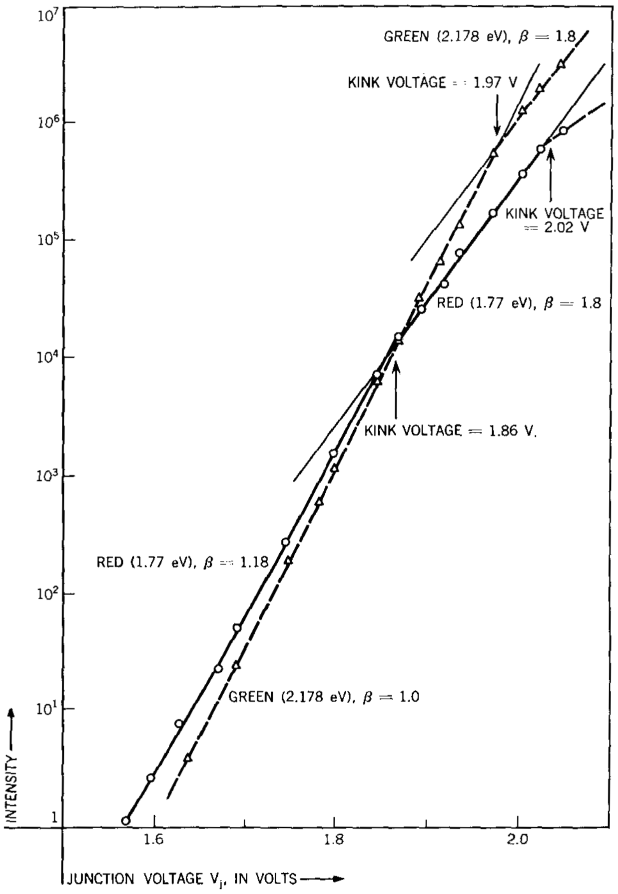
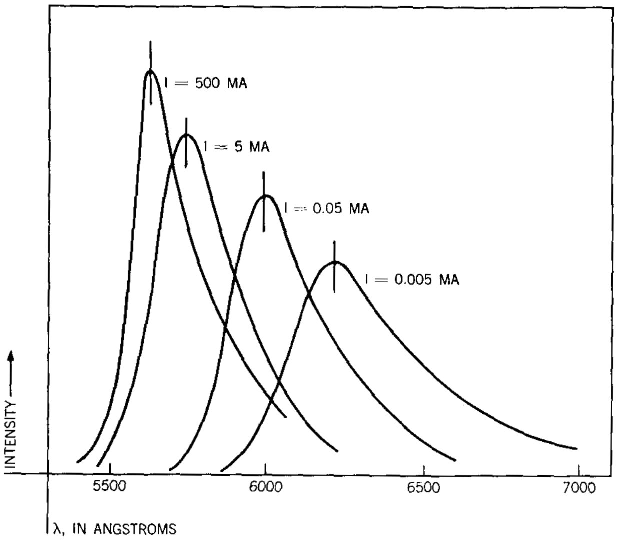


Figure 3 Intensity-voltage relationship for red and green emission at room temperature, for a Zn-S doped GaP diode.

Figure 4 Emission spectra at 77°K for a Zn-Te doped diode at four different currents. Different arbitrary intensity scales are used for each.



the results of Fig. 3 agree with those found for GaAs diodes containing several defects and also with Morgan's model. The kink voltage for the red emission is discussed elsewhere[10] for diodes which emit red light only.

The peak intensity should vary as $I^{1.8/\beta}$ with the current, I, if β is the coefficient of the intensity-voltage relationship just mentioned, and if the current varies as $\exp(eV/1.8\,kT)$ with the voltage. At room temperature this was in fact observed. At low currents, the intensity of the green emission varied with approximately the square of the current (or slightly less); at high currents it was proportional to the current. The intensity of the red emission varied as $I^{1.5}$ at low currents, was proportional to I in an intermediate current range, and to $I^{0.5}$ at high currents.

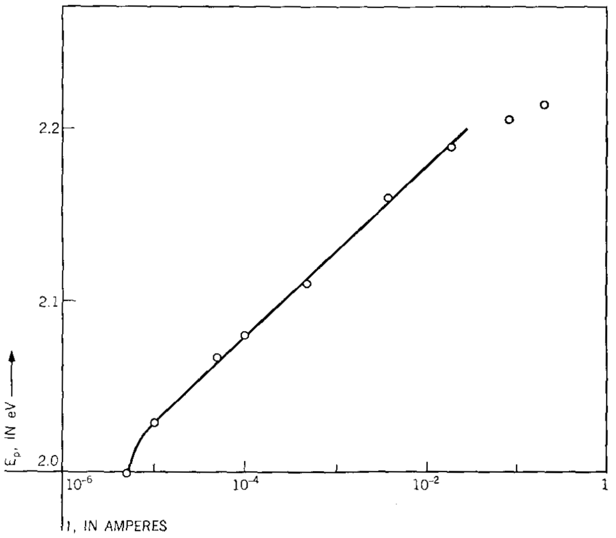
Emission at low temperatures

The number of photons generated in the 1.77 eV line relative to that in the near edge emission decreased monotonically when the temperature was lowered. At 77°K, the red emission was practically absent.

Only one emission line was observed at low temperatures, instead of the often complicated spectra of exciton lines which is commonly found in lightly doped diodes. In contrast to the situation at room temperature, it was found that the energy position and the width of the emission line depends on current density at low temperatures. Figure 4 shows emission spectra at 77°K for a Zn-Te doped diode for different current densities. The emission peak shifts considerably to lower energy when the current is lowered, and the line becomes broader.

We consider next some experimental data concerning peak position, intensity, and linewidth as functions of current (or voltage). In Fig. 5 the shift of the emission

Figure 5 Peak shift of the emission from a Zn-Te doped diode with current (77°K).



peak with current is depicted for a Zn-Te doped diode (77°K). The peak shifts from 2.0 eV to a little over 2.2 eV when the current is increased by five decades. At high currents, this shift of the emission peak with current seems to saturate. The high current values for the peak energy agree approximately with the published photoluminescence data[1] for the Zn-S and Sn-Te donor-acceptor pair bands at 77°K. Peak shifts as large as 0.24 eV (~660 Å) have been observed. The peak energy changes as the applied junction voltage at low currents. At high currents the true junction voltage could not be determined because of the series resistance. Capacitance measurements[14] showed that the junction was nearly abrupt for the diode of Fig. 5. Graded junctions were also examined and gave similar results.

Above 10^{-5} A, the intensity of the emission was proportional to the current for the diode of Fig. 5, while below 10^{-5} A it decreased more rapidly with current ($\sim I^4$). An exponential relationship between intensity of the emission and the peak energy, E_p, was found. At 77°K, the intensity varies as exp (αE_p) with the energy and α was found to be 47.7 eV^{-1} in the case of a Zn-Te doped diode. The coefficient α is smaller than $1/2kT$.

The width of the emission line increases with decreasing voltage. For the diode of Fig. 5, the half width, ΔE, increases from 100 to 150 meV as the peak energy (i.e., voltage) decreases from 2.2 to 2.0 eV (77°K).

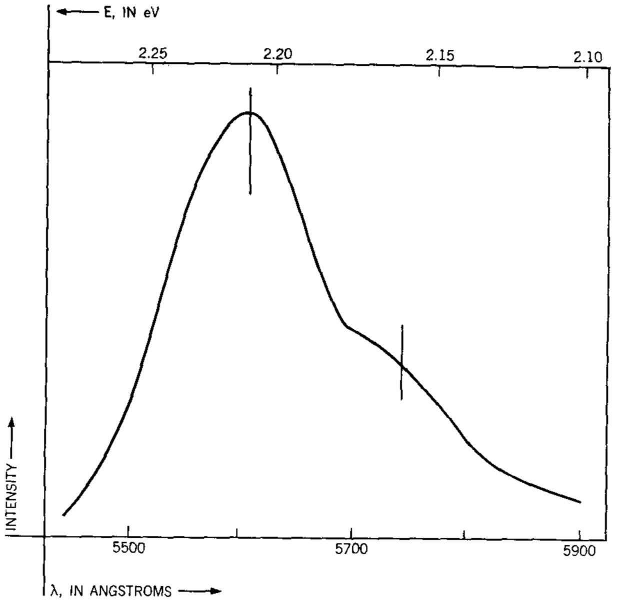
We now look for an interpretation of the data just presented and, in particular, for an interpretation for the strong peak shifts. Slight peak shifts with excitation intensity have been reported in photoluminescence.[5,15,16] Gershenzon et al.[2] have seen a slight peak shift at low temperatures for the Mg-S pair band in electroluminescence. This was interpreted as arising from the fact that recombination through the more distant donor-acceptor pairs is favored when the junction voltage is lowered, which makes the peak emission shift to lower energy. That interpretation does not appear to be valid in our case since, for instance, the low energy limit for the zinc-tellurium pair spectrum ($r \to \infty$) is 2.178 eV at 77°K, but the emission line in Fig. 5 falls considerably below this limit towards lower energy.

We therefore look for another explanation of the strong peak shifts observed. Since the diodes have an appreciable tunnel current component at low temperatures and low voltages, photon assisted tunneling might be suggested as the recombination mechanism. Photon assisted tunneling has been studied for heavily doped GaAs *p-n* junctions at low forward bias by several workers.[17–20] Qualitatively, the results for GaAs are quite similar to the ones reported here: In GaAs, the energy of the emitted photon, $h\nu$, changes with the applied junction voltage, V, and large peak shifts (up to 0.3 eV) have been observed at low temperatures; the line intensity varies as exp (α E_p) with the peak energy, E_p, and α is related to the junction gradient.[20] As it is in GaP, the coefficient α is smaller than $1/2kT$ at 77°K; in fact, α is practically independent of temperature, which is typical for tunneling. The increase of the linewidth with decreasing voltage is also found for GaAs and is explainable on the basis of the tunneling model.[20]

The similarity between the results found for GaP and GaAs certainly suggest that the same model is applicable, although radiative recombinations involving tunneling should be less probable in GaP because of the larger effective electron mass. At high current densities there should be a smooth transition from the regime where tunnel currents are important to the regime where regular space charge recombination is prevailing. The saturation of the peak shift in Fig. 5 at high currents might be interpreted in this manner (if it is assumed that diode heating is negligible.)

Finally, we consider a fact that is not related to the preceding tunnel model; namely, that in some of the low temperature spectra of Zn-S doped diodes, a low energy shoulder was observed which might be interpreted as a phonon satellite. Figure 6 shows the 4.2°K spectrum, which has, besides the main Zn-S pair band peak at 2.205 eV, a shoulder at about 2.16 eV. The energy difference corresponds approximately to 1 L.O. phonon (47 meV). The low energy shoulder was still seen at 77°K in some instances. These results agree with photoluminescence data[21] where a low energy satellite has been also observed for the Zn-S pair band.

Figure 6 Emission spectrum of a Zn-S doped diode at 4.2°K, showing a donor-acceptor pair band and a low energy shoulder (interpreted as a phonon satellite).



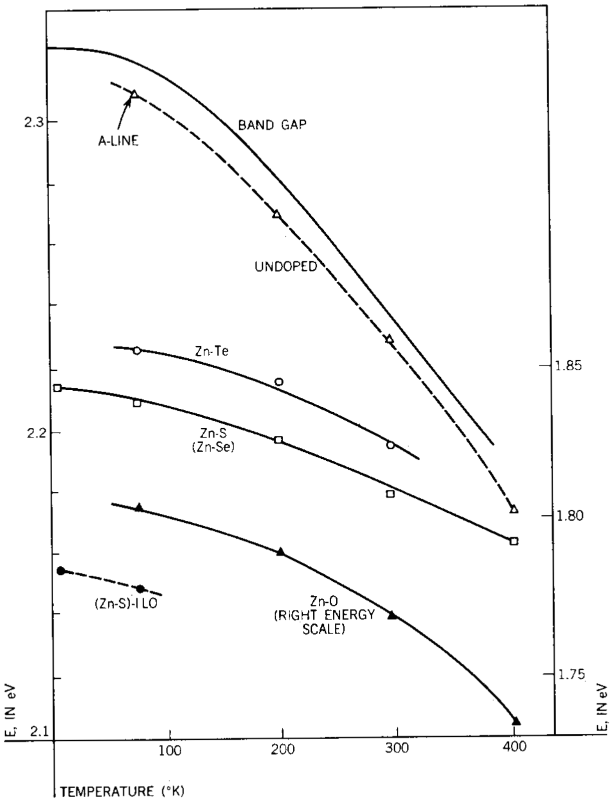


Figure 7 Temperature dependence of the peak energy of various emission lines and of the bandgap.

• *Temperature dependence of the emission peaks*

In Fig. 7, the temperature dependence of the emission from three donor-zinc doped diodes and from an "undoped" diode is compared with that of the energy gap. The temperature dependence of the energy gap is derived from Refs. 22 and 23 (compare, also, Ref. 2). The A-line emission of the undoped material follows the energy gap quite closely, indicating that the sharp exciton line at 77°K (and below) and the broader lines at higher temperatures are, indeed, corresponding radiative transitions. The emission from the donor-zinc doped diodes does not vary with temperature as the gap. At low temperatures, the peak energies for high current densities were taken. The emission from Zn-Se doped diodes virtually coincides with that from Zn-S doped diodes, and hence has not been included in Fig. 7. The temperature dependence of the Zn-O pair band is roughly like that of the shallow donor-zinc bands, as can be seen in Fig. 7.

An explanation for this temperature dependence may lie in the ionization of the shallowest of the levels involved, the Zn level, with rising temperature. The donor-acceptor recombination changes gradually to a donor-free hole recombination as the temperature is raised. The deviation of the observed temperature dependence from that of the gap is approximately 40 meV, if the data for 77°K are compared with that for 300°K. This deviation is approximately the quoted value for the zinc activation energy.[1] Similar arguments also hold true for the shallow donor levels; these, however, are at least twice as deep as the Zn level. In fact, in Fig. 7 there is indication that the emission bands from Type II and III diodes will eventually merge with the *A*-line emission above room temperature.

• *Quantum efficiency*

External quantum efficiencies were measured with an integrating sphere type of apparatus, which is described in Ref. 24. The efficiency values to be quoted are differential quantum efficiencies taken from the region where the light intensity varies linearly with the current.

At room temperature, the highest quantum efficiency was 1.5×10^{-4} for a green-emitting diode (Zn-S doped). Zn-Te and Zn-Se doped diodes had similar values (10^{-4}, or slightly lower). At high current densities, the total external quantum efficiency came close to the differential quantum efficiency. The highest value of the nitrogen temperature efficiency for the same diodes was 6×10^{-3}.

The temperature dependence of the efficiency, η, of the near-edge emission line of a Zn-S doped diode is shown in Fig. 8, where log η is plotted versus $10^3/T$. Again, this efficiency η is the differential quantum efficiency obtained from the region where the light intensity varies linearly with the current. At high temperatures, the efficiency decreases nearly exponentially with $1/T$, with an activation energy of 43.7 meV. At low temperatures, a constant value is approached.

In order to investigate whether the small value of the external efficiency at room temperature is due to reabsorption in the crystal, the following experiment was performed: the change of the total quantum efficiency with the transmissivity t_{av} of the diode surface was investigated by placing a diode into liquids with different refractive indexes. For small t_{av} and uniform photon distribution within the crystal, we have the following[25]

$$\eta_{\text{ext}} \approx \frac{\eta_i}{1 + 4\bar{\alpha}v/At_{av}}, \qquad (3)$$

where η_i is the internal quantum efficiency, $\bar{\alpha}$ is the effective absorption constant, v is the crystal volume, and A is the crystal surface area. The t_{av}, averaged over all angles of incidence, is given approximately by[26]

$$t_{av} = f(n_1/n_2)\,\frac{4n_1n_2}{(n_1+n_2)^2}\left(\frac{n_2}{n_1}\right)^2, \qquad (4)$$

where n_1 is the refractive index of GaP and n_2 is that of the liquid. The function $f(n_1/n_2)$ is not much different from 1.[28] In Fig. 9, $(1/\eta_{\text{ext}})$ has been plotted as a function of $(1/t_{av})$ for a Zn-S doped diode. This diode was chosen

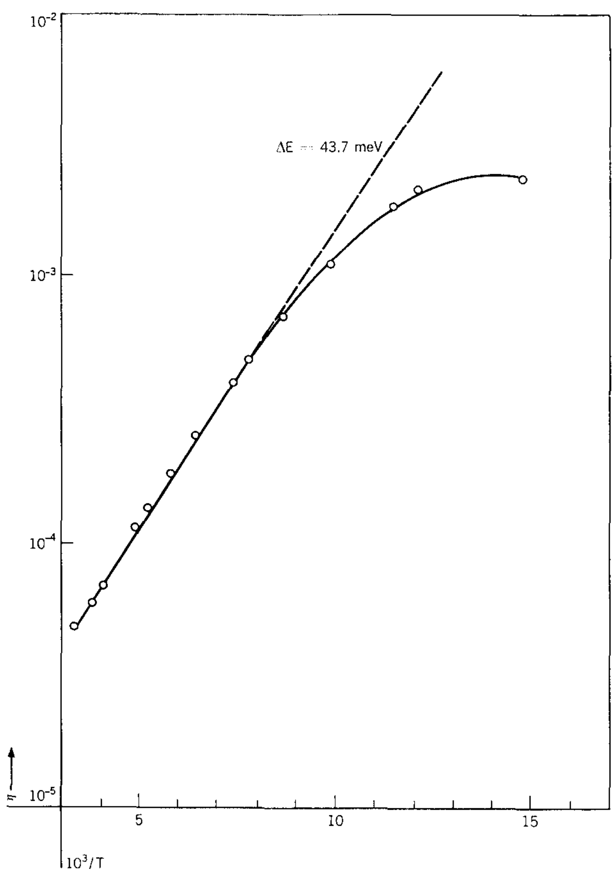
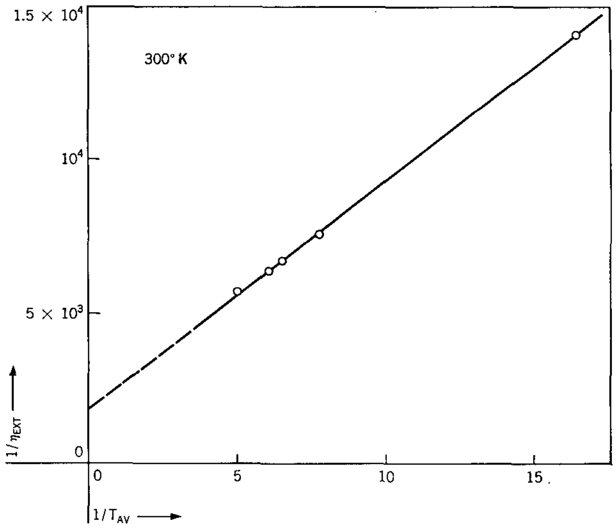


Figure 8 Temperature dependence of the differential external quantum efficiency for the green emission of a Zn-S doped diode.

Figure 9 The reciprocal of the external quantum efficiency as a function of the reciprocal of the average transmissivity (300°K).



to have well cleaved and clean surfaces, but it was not one with the highest efficiency. The refractive index of GaP was assumed to be 3.4 at 5700 Å and room temperature.[29] From Fig. 9, an internal efficiency of 5.5×10^{-4} can be estimated by using Eq. (3). This value is about eight times higher than the external efficiency value in air.

The number derived for the internal quantum efficiency is still quite low. One may therefore conclude that re-absorption alone does not account for the small magnitude of the external efficiency and that competitive recombinations become important at high temperature.

- *Time effects*

If the diodes were operated with current pulses (50 to 500 nsec duration), time effects in the light emission were observed. As an example, Fig. 10 shows an oscillographic display of both the current pulse and the corresponding light pulse for a Zn-S doped diode. It was found that RC time effects were unimportant in these experiments. The rise time of the light pulse was usually a little greater than the decay time (up to a factor of 1.6 greater) but there were also diodes for which they were equal. In the following, we shall consider only the emission decay and compare its time constants for various types of emission lines.

The decay of the light intensity was nonexponential with time, which is analogous to results reported in the case of photoluminescence.[28] The shortest half-times were observed for the *A*-line (10 nsec or below at room temperature). The emission from Zn-Te doped diodes (2.195 eV peak energy) had time constants between 50 and 60 nsec, and from Zn-S doped diodes (2.17 eV peak energy) between 70 and 100 nsec. This demonstrates again that the emission from diodes doped with shallow donors and acceptors is different from the usual *A*-line emission. The half-time for the red emission (1.77 eV) was 90–110 nsec, in agreement with reported results.[29]

If the temperature is lowered, the time constants increase, which is in qualitative agreement with cathodoluminescence data.[30] Quantitatively, there is agreement only with the low temperature data of Reference[30]. At high temperatures the time constants derived from electroluminescence are greater. The decay time for the Zn-Te doped diodes becomes about 200 nsec (at 5660 Å) at 77°K. It was found that the time constant varied with wavelength for a given emission band. Short wavelength corresponds to the shorter time constant. In the case of the Zn-Te band, for instance, the half-time changed from 100 to 230 nsec as the wavelength changed from 5500 to 5700 Å. A similar behavior reported in photoluminescence,[16,31] has been interpreted through the decay kinetics of variably spaced donor-acceptor pairs. The time constants for the exciton lines were again the shortest at 77°K (~20 nsec).

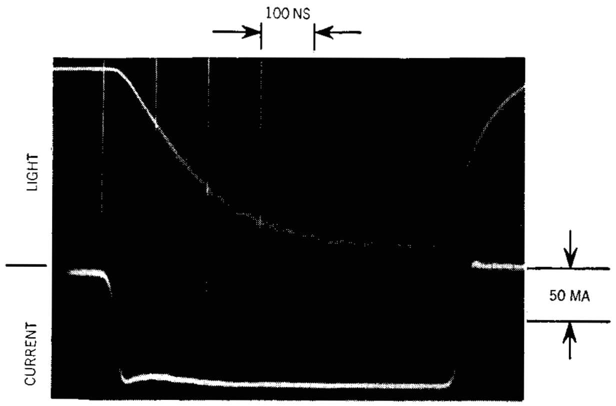


Figure 10 Time effects in the light emission from a Zn-S doped diode (at room temperature). Vertical divisions represent 50 mA for the current pulse; horizontal divisions, 100 nsec.

Conclusions

The broad green emission from GaP diodes that are heavily doped with shallow donors and acceptors has been studied as a function of voltage and temperature. The peak energy varies with the donor binding energy, and thus the emission is interpreted as a radiative donor-acceptor transition. It was found that the peak energy does not change in the same manner with temperature as the bandgap. This finding is interpreted through the ionization of the Zn-level at high temperatures, which has the effect that the donor-acceptor transition becomes a donor-valence band transition.

At room temperature, the recombination in the *p-n* junction is thermally activated and the dependence of current and intensity on voltage indicates predominant recombination in the space charge region. At low temperatures, a tunnel current component becomes important. The emission peak shifts strongly with voltage; peak shifts as large as 0.24 eV have been seen. The emission is interpreted as photon assisted tunneling in this regime.

The quantum efficiency decreases significantly with increasing temperature, and an activation energy of 43.7 meV is derived from the data. Nonetheless, the efficiency at room temperature is still high enough (1.5×10^{-4} at 5700 Å, in the best case) for the diodes to appear bright enough to be considered as useful semiconductor sources of green light.

Acknowledgments

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