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Photoluminescence in heavily doped GaAs. I. Temperature and hole-concentration dependence

Diego Olego and Manuel Cardona
Max-Planck-Institut für Festkörperforschung, 7000 Stuttgart 80, Federal Republic of Germany

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The temperature and hole-concentration dependence of the photoluminescence from the E_0 and $E_0+\Delta_0$ energy gaps have been measured in p-type GaAs. In heavily doped GaAs the emission spectra across the E_0 gap can be described by taking into account band-band optical transitions with and without $\vec{k}$ conservation. The thermal shift of the band gap is the same as for pure GaAs and can be represented by the Varshni equation. The energy-gap shrinkage was measured and compared with the previously determined values of Casey and Stern. The luminescence across the $E_0+\Delta_0$ gap gives evidence for impurity states associated with the spin-orbit-split band. At 2.1 K this luminescence contains two peaks which are interpreted to arise from band-band and band-impurity optical recombination. The energy of these peaks does not depend on hole concentration, hence the $E_0+\Delta_0$ gap is not influenced by the shrinkage affecting the E_0 gap. The luminescence from the sample with 1.7×10^{17} holes cm^{-3} displays at 2.1 K a series of lines corresponding to the emission of LO phonons in the energy range between the E_0 and $E_0+\Delta_0$ gaps. This observation indicates that the holes relax to the top of the valence band by hole-optical phonon scattering.

I. INTRODUCTION

Owing to the technological importance of GaAs the spontaneous radiative recombination of electrons and holes across the fundamental edge (E_0 gap) has been extensively studied during the last two decades. Photo-, cathodo-, and electroluminescence have been the most widely used experimental techniques.

The effects of band-band, band-impurity, and free-exciton transitions on the E_0 gap luminescence are well documented for pure and lightly doped semiconductors; they have been reviewed by Bebb and Williams.[1,2] The influence of high impurity concentrations on the emission spectra[3-7] as well as luminescence in highly excited, nearly intrinsic GaAs have also been studied.[8]

Recently Casey and Stern performed a model calculation for the absorption and emission spectra of heavily doped GaAs, from which the dependence of the energy gap on doping is obtained.[6] The emission spectra are calculated from the absorption coefficient using the detailed-balance approach.[1,6] This procedure is no longer valid for heavily doped semiconductors when the Fermi level lies above the conduction-band minima or below the top of the valence bands: in these cases emission at certain frequencies is possible although absorption is not. In the case of heavily doped p-semiconductors the absorption connects only filled states below the Fermi level with empty conduction-band states, while emission also involves empty valence band states above the Fermi energy.

Most of the luminescence work above the fundamental gap reported for heavily doped GaAs deals with the polarization dependence of the emission in order to determine spin-relaxation processes.[9] However, not much information exists about the origin and concentration dependence of the observed features. In this paper we present a systematic study in GaAs of the temperature and hole-concentration dependence of: (i) the E_0 luminescence and (ii) the luminescence reported earlier in the range between 1.65 to 1.9 eV which was associated with radiative recombinations across the $E_0+\Delta_0$ gap[9-11] (Δ_0 is the spin-orbit splitting).

The observed E_0 luminescence spectra for heavily doped p-GaAs can be interpreted as a mixture of $\vec{k}$-conserving and $\vec{k}$-nonconserving optical transitions. The calculated spectra, obtained by adding these two processes broadened by life-time effects, reproduce quite well the experimental ones. A detailed line shape analysis allows both the temperature and hole-concentration dependence of E_0 to be determined.

The emission from the $E_0+\Delta_0$ gap at very low temperatures shows two peaks whose intensities, but not their photon energies, are dependent on hole concentrations. This emission is thought to arise from transitions from the conduction band to the spin-orbit-split band, as well as to impurity levels associated with the spin-orbit-split band.

The apparent shrinkage observed for the $E_0+\Delta_0$ gap[11] is shown to be independent of doping and is explained by assuming that the recombination takes place near an impurity ion. In one particular sample ($p=1.7\times10^{17}\ \mathrm{cm}^{-3}$) multiphonon processes are observed associated with the $E_0+\Delta_0$ luminescence.

II. EXPERIMENT

The Zn-doped GaAs samples were cut from single crystals and the Ge-doped one was grown by liquid phase epitaxy on a GaAs substrate. The carrier concentration was determined from Hall-effect measurements. We used either (110) cleavage surfaces or (110) and (100) faces polished and etched with an aqueous solution of NaOCl.

The samples were cooled either by He exchange gas, in which case the temperature could be varied between 300 and 4 K with an accuracy of ±0.5 K, or immersed into pumped liquid He.

The 5145- or 4880-Å lines from an Ar^+-ion laser were used for excitation and the incident light was focused on the surface of the sample with a cylindrical lens in order to avoid the heating. The measurements were performed in the backscattering configuration. The scattered light was analyzed either with a Jarrell Ash 1 m or with a Spex 0.8 m double monochromator.

The detection of the E_0 luminescence was made with an EMI 9684*A* photomultiplier with *S*-1 response and the emission from the $E_0+\Delta_0$ gap with an RCA 31034 photomultiplier. In both cases photon-counting electronics was used. The shape of the recorded spectra depend neither on the crystallographic faces nor on the photon energy of the exciting radiation (provided it lies above the emitted energy).

III. RESULTS AND DISCUSSIONS

A. E_0 luminescence

Typical spontaneous emission spectra as a function of temperature for two different hole concentrations in GaAs are shown in Figs. 1 and 2. As the temperature is decreased a shoulder develops on the high-energy side of the emission line. This change of shape is observed in all the samples with concentrations higher than 10^{19} holes cm^{-3} and has been reported previously by Cusano[3] and Pankove.[4] For the sample doped with Ge the temperature dependence of the line shape is the same as for a Zn-doped material with equal hole concentration.

The luminescence spectra of the samples with 1.6×10^{18} and 3×10^{18} holes cm^{-3} show at very low temperatures a strong emission peaking at 1.49 eV and a small peak at 1.455 eV. The line at 1.49 eV has been attributed to a donor-acceptor transition involving residual Si impurities present in GaAs, and that at the lower energy corresponds to a transition with LO phonon emission.[2] The luminescence from the sample with 1.7×10^{17} holes cm^{-3} consists of three lines, the two mentioned above and a new one at 1.51 eV corresponding to transitions involving exciton complexes.[2] Our results agree with previously published data.[2-4]

In what follows we restrict our discussion to the experimental results obtained from the samples with concentrations higher than 10^{19} holes cm^{-3}, a

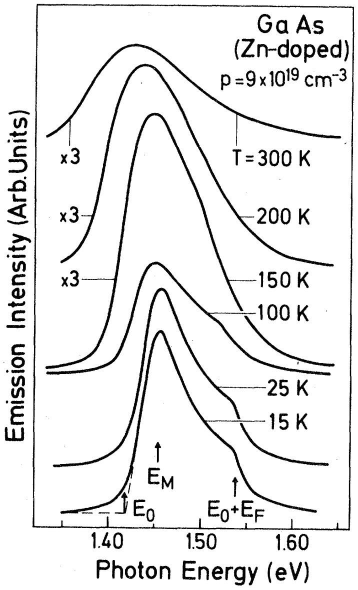

FIG. 1. Typical emission spectra as a function of temperature across the E_0 energy gap of heavily Zn-doped GaAs with 9×10^{19} holes cm^{-3}. A shoulder (E_0+E_F) develops on the high-energy side of the spectra at low temperatures corresponding to the Fermi energy. E_0 and E_M represent, respectively, the actual band edge and the peak maxima.

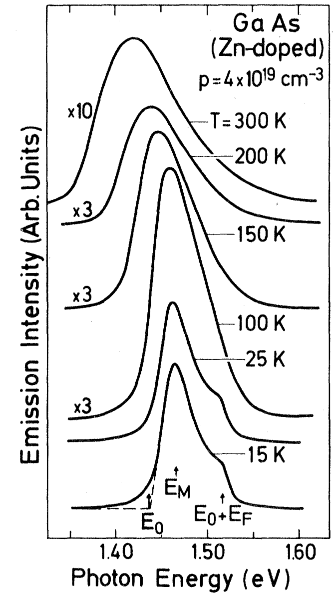

FIG. 2. Typical emission spectra as a function of temperature across the E_0 energy gap of heavily Zn-doped GaAs with 4×10^{19} holes cm^{-3}. A shoulder (E_0+E_F) develops on the high-energy side of the spectra at low temperatures corresponding to the Fermi energy. E_0 and E_M represent, respectively, the actual band edge and the peak maxima.

range for which the details of the observed luminescence have not been satisfactorily explained in the past. Three points are singled out by arrows in Figs. 1 and 2: E_0, E_M, and E_0+E_F. E_0 corresponds to the intersection between the tangent to the low energy tail of the emission band and the background. E_M represents the maximum of the luminescence peak and E_0+E_F refers to the shoulder on the high-energy side. After the temperature dependence is presented, the physical meaning of these points will be discussed.

In Fig. 3 the photon energy of the points labeled E_M and E_0+E_F is shown as a function of doping, together with the data of Cusano[3] and Pankove.[4] All sets of data agree well.

Figures 4 and 5 reproduce the temperature dependence of the three points mentioned. The solid lines are theoretical fits with the Varshni equation[12]

$$E(T)=E(O)-\alpha T^2/(\beta+T) \tag{1}$$

which describes the temperature variation of the band edges in most of the IV and III-V group semiconductors. The parameters α and β, as determined by fitting the experimental points with Eq. (1), are summarized in Table I, where they are compared with Thurmond's parameters for the band gap ($\Gamma_1^c-\Gamma_8^v$) of pure GaAs.[13] From this comparison we infer the meaning of the three points indicated in Figs. 1 and 2: E_0 represents the actual band gap of the material. The assignment is consistent with the fact that in pure semiconductors with parabolic bands the energy gap lies exactly where the emission spectra begins.[1]

E_M, the maximum of the emission peak, cannot be associated with any special point. The shoulder E_0+E_F is caused by indirect transitions between the minimum of the conduction band and the Fermi level below the top of the valence band. The relationship between this shoulder and the Fermi energy was previously recognized,[3,4] and our study of the temperature dependence confirms the earlier assignments. Between 2 and 150 K, a range in which a change in the shape of the E_0 luminescence can be clearly observed, the Fermi energy is independent of temperature. Thus the thermal shift of E_0 and E_0+E_F should be the same, in agreement with our observation (α and β are the same for both, within the experimental error).

With the assignment made for E_0 we can give an accurate value for the band gap and the effective gap shrinkage in heavily doped GaAs. This shrinkage is due mainly to exchange and correlation in the free-hole system.[14] Casey and Stern have determined the gap shrinkage by comparing their calculated absorption curves with the experimental ones and finding the energy shift which allows the best fit to the experimental absorption edges.[6]

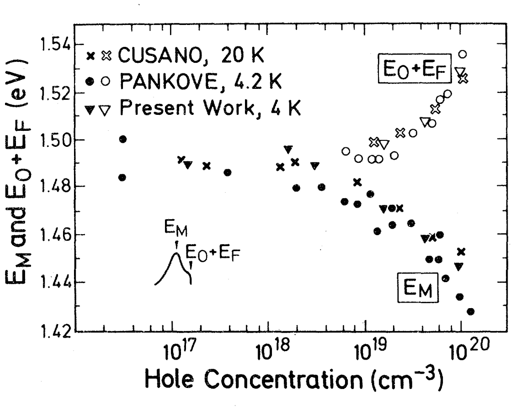

FIG. 3. Hole-concentration dependence of the luminescence peak maxima E_M and E_0+E_F in heavily Zn-doped GaAs.

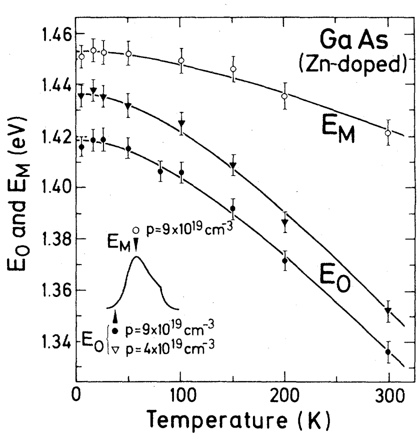

FIG. 4. Temperature dependence of the band gap E_0 and the luminescence peak maxima E_M. The solid lines represent fits with Eq. (1). The resulting parameters are tabulated in Tables I and II.

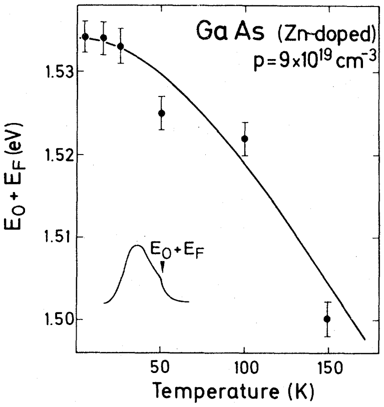

FIG. 5. Temperature dependence of the high-energy shoulder E_0+E_F. The solid line represents a fit with Eq. (1). The resulting parameters are tabulated in Table I.

TABLE I. Coefficients α and β of Varshni's Eq. (1). Also listed are Thurmond's determinations for pure GaAs.[13]

	E_0		E_M		E_0+E_F		Thurmond's determination	
p (cm^{-3})	α (10^{-4} eV K^{-1})	β (K)	α (10^{-4} eV K^{-1})	β (K)	α (10^{-4} eV K^{-1})	β (K)	α (10^{-4} eV K^{-1})	β (K)
9×10^{19}	5.1 ± 0.3	250 ± 20	2 ± 0.3	250 ± 20	5.2 ± 0.3	240 ± 20	5.405	204
4×10^{19}	5.1 ± 0.3	250 ± 20					±0.25	±45

They find that the gap shrinkage with hole concentration p can be represented by

$$E_g(\text{doped}) - E_g(\text{pure}) = -1.6\times10^{-8}p^{1/3} \qquad (2)$$

with E_g in eV and p in cm^{-3}. They have also calculated the exchange contribution to the shrinkage and proved that this amounts to one-half of the total effect. In Table II we list our results for a series of hole concentrations for both, the band gap at $T=0$ K [obtained from the fits with Eq. (1)] and the gap shrinkage determined by assuming that the band gap of pure GaAs is 1.519 ± 0.001 eV.[13] We include also the shrinkage calculated with Eq. (2).

We present next a calculation of the shapes of the emission spectra. The results are displayed in Figs. 6 and 7 for two typical hole concentrations.

The spontaneous recombination spectra were calculated assuming optical transitions between the conduction and the valence bands. Processes with and without wave-vector conservation, as described by Lasher and Stern,[15] were considered in the calculation. No assumption was made about the dynamics of the photoexcited electrons in the conduction band. However, it was assumed that the electrons, and of course the holes, can reach thermal equilibrium within each band. The calculated spectra (unbroadened fit of Figs. 6 and 7) display the contributions of the direct and indirect optical transitions. The narrower line at lower energies represents mainly the $\vec{k}$-conserving processes, while the wider one the $\vec{k}$-nonconserving transitions. The unbroadened fits of Figs. 6 and 7 were convoluted with a Gaussian in order to take into account broadening of the electronic states. The width of the Gaussian was determined by the lifetime obtained from the mobility (very low for these heavily doped p-materials). For the sample with 4×10^{19} holes cm^{-3} that width is 35 meV and for the sample with the largest concentration 46 meV. The calculated curves before and after convolution and the relative contributions of direct and indirect transitions were normalized to the experimental heights of peaks E_M and E_0+E_F. The agreement with the experiments is reasonable, especially if one considers that in our calculation band tail states were neglected and the matrix elements for the optical transitions were taken as constant independent of $\vec{k}$.

TABLE II. Band gap and gap shrinkage for p-GaAs at 0 K. The band-gap energy was determined from the fits with Eq. (1). The calculated gap shrinkage was obtained from Eq. (2).

p (cm^{-3})	E_0 (eV)	Measured gap shrinkage (meV)	Calculated gap shrinkage (meV) [Eq. (2)]
9×10^{19}	1.418 ± 0.005	101 ± 6	72
4×10^{19}	1.430 ± 0.005	83 ± 6	55
1.6×10^{19}	1.455 ± 0.005	64 ± 6	40

The calculations just given show that the high-energy side of the emission spectra is determined by indirect transitions. The role of the indirect transitions in the luminescence spectra of heavily doped n-type semiconductors has been also recently recognized.[16] In the case of n-type semiconductors the emission across E_0 can be described exclusively with $\vec{k}$ nonconserving transitions. $\vec{k}$-conserving transitions are enhanced, and thus important, in p-type materials because of the large number of free holes and the asymmetry of electron and hole masses (the heavy-hole mass is roughly ten times as large as the electron mass in

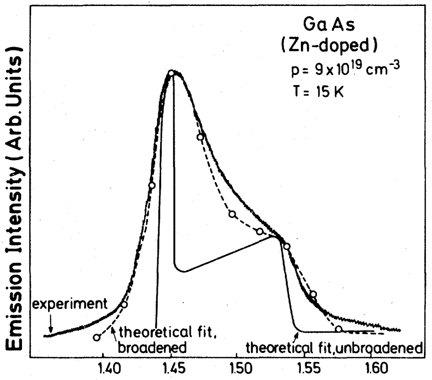

FIG. 6. Experimental emission spectrum of Zn-doped GaAs with 9×10^{19} holes cm^{-3} at 15 K and theoretical calculation of the line shape. The calculated spectrum includes an admixture of $\vec{k}$ conservation and $\vec{k}$ nonconservation. The effect of the broadening due to lifetime of the electronic states is shown by the dashed-dotted line spectrum. The ratio of direct to indirect transitions assumed for this fit is 1:0.42.

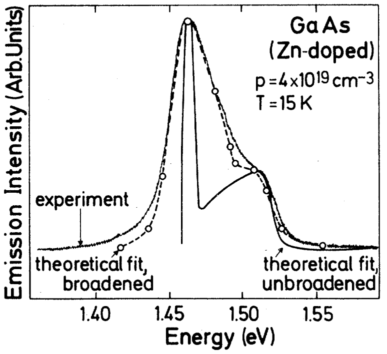

FIG. 7. Experimental emission spectrum of Zn-doped GaAs with 4×10^{19} holes cm^{-3} at 15 K and theoretical calculation of the lineshape. The calculated spectrum includes an admixture of $\vec{k}$ conservation and $\vec{k}$ nonconservation. The effect of the broadening due to lifetime of the electronic states is shown by the dashed-dotted line spectrum. The ratio of direct to indirect transitions assumed for this fit is 1:0.3.

GaAs).

In p-GaAs the breakdown of momentum conservation can be attributed to the carrier scattering by the ionized acceptor impurities. These processes have been previously invoked in order to explain the self-energy of the phonons and the phonon-plasmon coupling measured by Raman scattering in the same materials.[17,18]

Our interpretation of the luminescence of heavily doped p-GaAs as a band-band recombination differs from Pankove's interpretation which related the peak of the emission (E_M) to donor-acceptor transitions.[4] This assignment was based on the observation that in the cathodoluminescence experiments no shift of the emission peak with electron beam current was seen. Such transitions, however, should exhibit the same temperature dependence (not investigated by Pankove) as E_0, contrary to our observations.

B. $E_0 + \Delta_0$ luminescence

We have reported reviously luminescence above the fundamental gap in p-GaAs.[11] From uniaxial stress measurements, we deduced that part of the luminescence was associated with radiative transitions across the $E_0 + \Delta_0$ energy gap (1.859 eV at $T = 0$ K for pure GaAs). We present now additional experimental data in the range between 1.65 to 1.90 eV about the temperature and hole-concentration dependence of the observed luminescence.

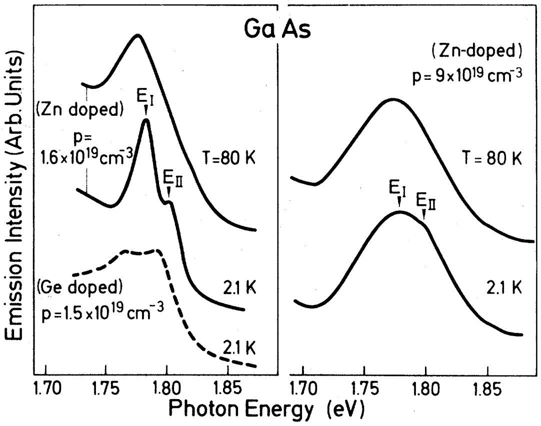

FIG. 8. Typical emission spectra as a function of temperature across the $E_0 + \Delta_0$ energy gap of heavily doped Zn and Ge-doped GaAs. At very low temperatures two peaks labeled $E_{\rm I}$ and $E_{\rm II}$ are observed. They are attributed to band-impurity and band-band optical transitions, respectively.

Figure 8 shows typical emission spectra at low temperatures in the range mentioned above for two different concentrations. For the sample with 9×10^{19} holes cm^{-3} the spectrum at 80 K reproduces the published results,[11] and at 2.1 K a change of shape is detectable in the high-energy side of the emission. The spectrum at 80 K for the sample with 1.6×10^{19} holes cm^{-3} (Zn acceptor impurities) resembles the spectrum of the sample with the largest concentration at the same temperature. That is to say, the frequency of the peak is the same but the line becomes narrower in the latter case. At 2.1 K the change of shape observed with the heaviest doped sample develops cearly into two peaks, labeled $E_{\rm I}$ and $E_{\rm II}$ in the figure. The Ge-doped sample with 1.5×10^{19} holes cm^{-3} exhibits also two peaks at very low temperatures. However, the energy position of these peaks differs slightly from the case of Zn-doped materials. At 2.1 K, $E_{\rm I}$ and $E_{\rm II}$ are seen for the whole range of hole concentrations.

We have attributed the emission in this energy range to transitions associated with the $E_0 + \Delta_0$ gap. In what follows we discuss their origin. In Fig. 9 we have plotted the energy position at 2.1 K of $E_{\rm I}$ and $E_{\rm II}$ as a function of hole concentration for the Zn-doped samples. We cannot observe any concentration dependence of the peak positions within our experiment error. A mean value of 1.778 eV can be determined for $E_{\rm I}$ and 1.800 eV for $E_{\rm II}$. In the case of the Ge-doped sample we measure 1.765 eV for $E_{\rm I}$ and 1.792 eV for $E_{\rm II}$.

We propose the following recombination mechanisms in order to explain the two peaks observed. $E_{\rm II}$ corresponds to the band-band transitions between the conduction band (Γ_1^c) and the spin valence band (Γ_7^v). We assume that the recombination takes place near an acceptor ion and samples a local band structure with a spin-orbit splitting influenced by the presence of the Zn or Ge impurities. We have calculated the contribution to the spin-

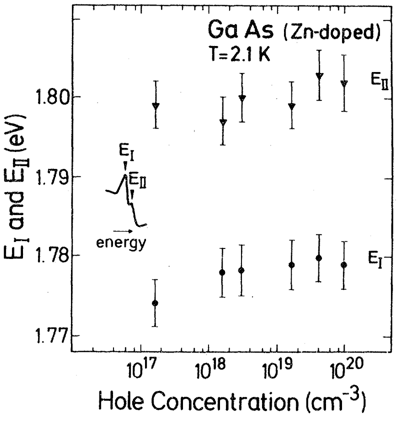

FIG. 9. Hole-concentration dependence of the energy position of E_I and E_{II} at 2.1 K.

orbit splitting Δ_0 of Zn in Ga sites and Ge in As sites according to the procedure described in Ref. 19. In both cases a reduction of the $E_0+\Delta_0$ gap is expected because the contributions to Δ_0 of the Zn and Ge ions are smaller than those of the Ga and As ions, respectively. In Table III we compare the experimental data with the results of this estimate.

E_I is related to optical transitions between the conduction band and impurity levels associated with the spin-split band. The presence of such levels have been well documented for acceptor impurities in Si.[21] For the III–V semiconductors we could not find any references in the literature. However, resonant impurity levels associated with secondary conduction band minima and degenerate with a continuum of band states have been reported for GaSb, InP, GaP, and GaAs.[22]

The energy difference $E_{II}-E_I$ is tabulated also in Table III and compared with the activation energy of the Zn and Ge acceptor states associated with the top of the valence band (Γ_8^v). If our interpretation is correct, this would be the first report of impurity states related with the spin-split band Γ_7^v in a III–V compound. The measured $E_0+\Delta_0$ gap (renormalized due to impurities) is independent of hole concentrations as one can deduce from Fig. 9, and thus the shortening of this gap reported previously[11] cannot be attributed to an exchange-correlation shrinkage as in the case of the E_0 gap: exchange and correlation due to free holes do not affect the Γ_7^v states because of the orthogonality of the wave functions (the free holes occupy the Γ_8^v valence band states).

In Fig. 10 we show that the ratio of the E_{II} to E_I intensity increases with increasing hole concentration. This behavior is consistent with the assignment made to E_I and E_{II}. Like E_0 gap luminescence, band-impurity recombinations dominate the emission spectra at lower concentrations: the intensity of the E_I peak is stronger than that of the E_{II} peak. When the hole concentration increases, the band-band component of the luminescence gains importance, perhaps due to the increasing probability of indirect transitions, as discussed in the case of the E_0 luminescence for heavily doped samples. The influence of indirect transitions in the line shape of the emission across $E_0+\Delta_0$ has been recognized in the case of *n*-type GaAs by Zakharchenya *et al.*[9] In our case the increase of the linewidth with hole concentration (Fig. 8) gives one more indication of the presence of indirect transitions. One must, however, raise the question of whether shallow bound states associated with the Γ_7 valence band are possible in *heavily doped* samples, i.e., in the presence of strong screening by the Γ_8 free holes. While we do not have a satisfactory answer to this question we mention an alternative explanation of peaks I and II. Such explanation is based on the presence of free holes at the top of Γ_7 in a *local enviroment* around the impurities. Such free holes would be expected to produce two structures related to $E_0+\Delta_0$, similar to the E_M and E_0+E_F we discussed for E_0. We are, however, not able to present a quantitative discussion of these alternatives.

The temperature shift of the $E_0+\Delta_0$ gap luminescence is shown in Fig. 11 for the sample with 9 $\times 10^{19}$ holes cm^{-3}: Only for this sample the emission from $E_0+\Delta_0$ can be resolved at room temperature. The solid line represents a fit with Eq. (1). The parameters obtained from the fit are $\alpha=4.1 \pm 0.5\times10^{-4}$ eV/K and $\beta=250\pm20$ K. The α coef-

TABLE III. Band impurity (E_I) and band-band (E_{II}) recombinations across $E_0+\Delta_0$. Renormalized $E_0+\Delta_0$ gap calculated according to Ref. 19 (see text). The energy difference $E_{II}-E_I$ is compared with the activation energy of the acceptor associated with the Γ_8^v states.

Dopant	E_I (eV)	E_{II} (eV)	Renormalized $E_0+\Delta_0$ gap (eV)	$E_{II}-E_I$ (meV)	Activation energy of acceptor levels (meV)
Zn	1.778 ± 0.003	1.800 ± 0.003	1.830	22	29 [a]
Ge	1.765 ± 0.003	1.792 ± 0.003	1.799	27	38 [a]

[a] Reference 20.

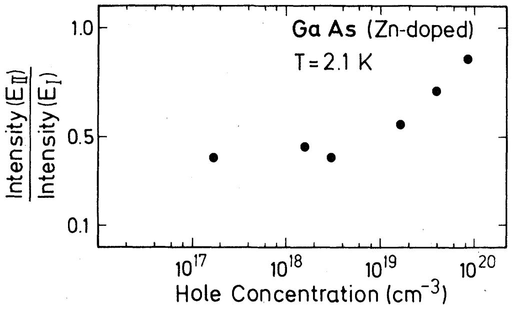

FIG. 10. Hole-concentration dependence of the intensity of E_{II} normalized to the intensity of E_I at 2.1 K.

ficient for $E_0+\Delta_0$ so obtained is somewhat smaller than that for the E_0 gap (Table I). However, in view of the scarcity of experimental points in Fig. 11 we hesitate to attribute any real significance to this difference: more detailed measurements are needed. Earlier work has always yielded a negligible temperature dependence for Δ_0.[23]

An interesting experimental observation at 2.1 K for the sample with 1.7×10^{17} holes cm^{-3} is reported in Fig. 12. Besides the two peaks E_I and E_{II} one can see, at lower photon energies, a series of lines separated from each other by 37 ± 3 meV. This separation corresponds to the LO phonon energy. The photoexcited holes at the top of the spin-orbit-split band relax to the light and heavy hole bands and then to the band edge by emission of longitudinal-optical phonons. This behavior is similar to the dynamics of hot carrier cooling[24,25] as determined, for example, with psec spectroscopy.[26,27] With samples of larger concentrations we have failed, however, to detect these structures in the energy range between the E_0 and $E_0+\Delta_0$ gap. With increasing concentration the indirect transitions become important and can obscure the presence of such structures and, perhaps most important of all, the phonons and the free holes are strongly coupled with the result that the polar optical modes are screened. This effect can reduce the efficiency of the photoexcited carriers—phonon interaction as was proved for hot carriers in GaAs.[26]

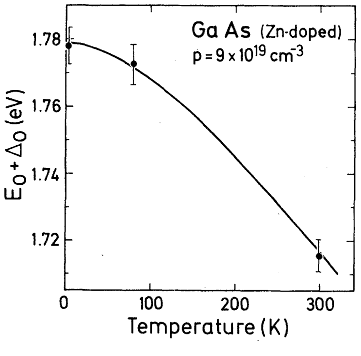

FIG. 11. Temperature dependence of $E_0+\Delta_0$ for Zn-doped GaAs with 9×10^{19} holes cm^{-3}. The solid line represents a theoretical fit with Eq. (1).

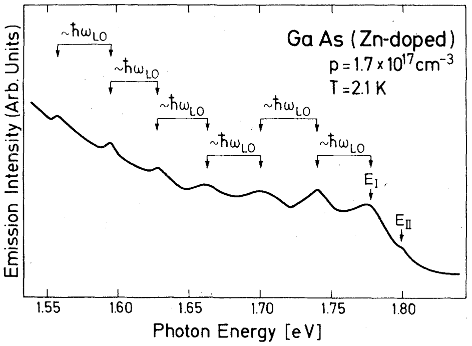

FIG. 12. Typical luminescence spectrum of Zn-doped GaAs with 1.7×10^{17} holes cm^{-3} at 2.1 K showing the E_I and E_{II} peaks and a series of lines identified as a relaxation of the photoexcited holes with emission of LO phonons.

IV. CONCLUSIONS

We have studied in p-type GaAs the influence of temperature and hole concentration on the radiative recombination across the fundamental gap E_0 and its spin-orbit-split gap $E_0+\Delta_0$. Both the temperature dependence and the line shape analysis of the emission from the E_0 gap allow a precise determination of the band edge for heavily doped p-GaAs and, at the same time, a direct measurement of the band-gap shrinkage. The line shape was calculated assuming a combination of direct and indirect optical transitions between the conduction and valence bands.

From the luminescence of the $E_0+\Delta_0$ gap at very low temperatures we found experimental evidence for the presence of impurity states associated with the spin-orbit-split band. This gap should not show any shrinkage due to either the free holes or the impurities. However, a narrowing of this edge was measured, which was interpreted as a local band-structure effect due to the smaller contribution of the impurities to the spin-orbit splitting.

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