Polarized hot-electron photoluminescence in highly doped GaAs

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The luminescence polarization is calculated for circularly polarized hot-electron photoluminescence in highly p-doped GaAs. Good overall qualitative agreement with experiments is found. Particular emphasis is put on the explanation of the polarization minimum and its dependence on excitation energy. We find that it is due to excitation of electrons from the light-hole band to the conduction band followed by the recombination with holes in the heavy-hole band, both processes being vertical. We determine the energy position of the minimum as a function of excitation energy, and find good quantitative agreement with experiments. The change in energy dispersions for the conduction and valence bands due to the doping is taken into account in all steps of the calculation.

I. INTRODUCTION

Hot-electron photoluminescence (HPL) is a powerful tool for studying the band structure in pure semiconductors. The energy resolution can be as high as 1 meV.[1] It can also be used to investigate the energy shifts of the electron states due to heavy doping. In the photoluminescence (PL) experiments on heavily p-doped semiconductors some of the photoexcited electrons have time to reach the bottom of the conduction band before recombination. These electrons, collected at the bottom of the conduction band, become thermalized. The luminescence from the thermalized electrons gives information about the shifts of the band edges.[2] The HPL, the luminescence from the fraction of electrons which do not reach the bottom of the conduction band, probes the energy shifts away from the band edges. From HPL one can, furthermore, gain information about the rate for various scattering processes.

The HPL spectra from pure and heavily doped samples look quite different. In pure GaAs or at low doping levels the spectra are characterized by LO-phonon cascades, i.e., the spectra contain series of relatively sharp peaks. These series of peaks correspond to varying numbers of phonon scatterings undergone by the electron before recombination. In the heavily doped samples there are new, dominating scattering processes, viz. electron-electron (e-h pair and plasmon excitations) and electron—dopant-ion scattering. The spectra are more structureless, due to the shorter electron lifetimes. This implies that it is more difficult to extract detailed information. In p-type GaAs one can overcome this disadvantage by studying the luminescence polarization in the HPL when using circularly polarized exciting light.

The electrons excited by circularly polarized light have an anisotropic spin orientation. This causes some circular polarization of the emitted light, which has the degree of $\frac{1}{4}$ when from the recombination with thermalized electrons.[3] The discrepancy between the ideal value $\frac{1}{4}$ and the actual one is a measure of the importance of spin relaxation. The degree of polarization in the HPL is experimentally found to vary with energy and can be much larger than $\frac{1}{4}$. It can even be negative in certain energy regions.[4] This variation occurs because the momentum distribution, as well as the spin orientation, is anisotropic. The maximum theoretical value[3] for the degree of polarization is $\frac{5}{7}$, and is obtained if the electrons are recombining with holes in the same band they were excited from and the recombinations occur before any spin or momentum relaxations take place. If the holes participating in the recombination processes belong to the other hole band the degree of polarization is theoretically $-\frac{5}{19}$. All numbers given are valid for the same direction of propagation for the excitation and luminescence light. The value $\frac{1}{4}$ is obtained after complete momentum relaxation but no spin relaxation, independent of recombination channel. Complete spin relaxation gives an unpolarized luminescence.

These anisotropies in the spin and momentum of the photoexcited electrons are due to the conservation of angular momentum in the excitation process, in combination with the special valence bands in GaAs. The electrons are characterized by spin up and spin down, while a valence-band hole is characterized by the projection of its angular momentum on its momentum. These projections have the values $\pm\frac{1}{2}$ ($\pm\frac{3}{2}$) for the light-hole (heavy-hole) bands. For a more complete description of this effect and to a more extensive list of references to experimental work, we refer the reader to Ref. 5. This possibility to obtain spin polarized electrons is, e.g., also utilized in the GaAs polarized electron source.[6]

The HPL spectra from heavily p-doped GaAs are characterized by a broad shoulder which moves in energy for varying excitation energy.[7] The luminescence polarization in these systems has a distinct minimum.[7] We devote Sec. II to the explanation of these structures. We have a different explanation than the one given in Ref. 7. In Sec. III we calculate the luminescence polarization and make comparison with the experiments. Finally, a summary and conclusion follow in Sec. IV.

II. INTERPRETATION OF THE POLARIZATION MINIMA

In numerous places in the experimental literature on luminescence in heavily doped GaAs one finds that non-k-conserving processes are believed to be important and even dominating. This is assumed to be due to the presence of the randomly distributed impurity potentials, breaking the perfect lattice periodicity.

The PL spectra from the thermalized electrons in heavily doped samples seem to have a common feature, a peak or shoulder on the high-energy side. In the literature this structure is, canonically, interpreted as due to non-vertical transitions, and it is referred to as $E_0 + E_F$. The energy of the shoulder should, with this interpretation, be equal to the energy of the shifted band gap E_0 plus the Fermi energy for the holes E_F (plus the Fermi energy of the thermalized electrons, to be exact). However, we found in an earlier work[2] that the position of this shoulder was far displaced in energy from that which would be expected from this interpretation. Furthermore, we found no indication of contributions from nonvertical transitions in the PL spectra we investigated. In Ref. 7 the shoulder in the HPL and the minimum in the luminescence polarization were attributed to nonvertical transitions. The explanation given was that the electrons were excited from the light-hole band and deexcited to the Fermi level.

We believe that this structure is explained by vertical transitions here as well, vertical excitation from the light-hole band to the conduction band followed by vertical deexcitation to the heavy-hole band.

To test this hypothesis we calculated the change in the energy dispersions in the conduction band and in the light- and heavy-hole bands due to the doping. The energy shifts were calculated as described in Ref. 2. We plotted in Fig. 1 the energy distance between the conduction band and the heavy-hole band (which is the energy corre-

sponding to the position of the polarization minimum) as a function of the energy distance between the conduction band and the light-hole band (which corresponds to the excitation energy). This is plotted for momentum varying between the Fermi momenta for the light and heavy holes, $k_{F,1}$ and $k_{F,h}$, respectively. The result is the ideal luminescence energy (i.e., disregarding broadening effects) as a function of excitation energy for the processes we consider. For excitation energies larger than the vertical distance between the conduction band and the light-hole band at $k_{F,h}$ the luminescence energy stays constant. In this region of momentum there are no holes available with which the excited electrons can recombine. The electrons have to lose energy via inelastic scattering until they reach $k_{F,h}$ before they can be deexcited. The result we obtained is given by the solid curve. The circles represent the position of the experimental polarization minima in Ref. 7 as a function of excitation energy. As a comparison we have plotted (with the dotted curve) the result one would obtain if the deexcitation were to the Fermi level. In that case there is no leveling off for higher excitation energies. The experimental results clearly favor our explanation. Our theoretical curves are valid for an acceptor density of 1.5×10^{19} cm^{-3}. The experimental density was given as $\sim 10^{19}$ cm^{-3}. It turns out that the theoretical E_{min} as function of E_{ex} is rather insensitive to the doping level except for the point of leveling off. The deviation in position of the diagonal part of the curve is less than the thickness of the curve for densities varying between 1×10^{19} and 2×10^{19} cm^{-3}. Thanks to this insensitivity we were able to add an additional experimental point (the solid square) to the plot. It is from Ref. 4 and is obtained for a sample with density 2×10^{19} cm^{-3}. It is quite consistent with the other points. The point of leveling off is rather sensitive to the doping density. Compared to our plotted curve it is roughly 80 meV lower (higher) for 1×10^{19} (2×10^{19}) cm^{-3}. This relative sensitivity might mean that the relation between E_{min} and E_{ex} can be used to get an accurate estimate of the doping density.
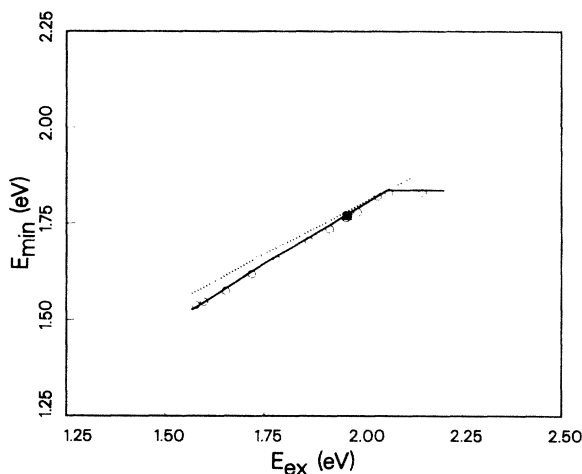

FIG. 1. Energy position of the polarization minimum as function of excitation energy. The experimental result from Ref. 7 is represented by the circles, and the square shows the experimental data point from Fig. 4 in Ref. 4. The solid curve is the theoretical result with our explanation, which should be compared with the dotted curve obtained with the explanation given in Ref. 7.

III. THEORETICAL LUMINESCENCE POLARIZATION

In Ref. 3, the following expression is derived for the degree of circular polarization for various luminescence processes in semiconductors with a GaAs-type band structure:

$$P_c = \frac{80 S_0 S + 10 \beta_0 \beta}{20 + \alpha_0 \alpha} . \tag{1}$$

The expression is valid for the emitted light detected in the direction of the exciting light $\hat{n}$. P_c is positive if the emitted light has the same polarization direction (left or right) as the exciting light. The coefficient α_0 characterizes the degree of momentum anisotropy of the photoexcited electrons and β_0 the correlation between the electron spin and momentum. $S_0 \hat{n}$ is the average spin of the electrons at the instant of production. The parameters α, β, and S denote the corresponding values at the moment of deexcitation. If the deexcitation occurs before any momentum or spin relaxation takes place, the two sets of

parameters have the same values. For excitation from the heavy- (light-)hole band the coefficients α_0, β_0, and S_0 have the values -1 (1), -1 (1), and $-\frac{1}{4}$ $(-\frac{1}{4})$, respectively. If the spin relaxation is complete before the deexcitation, S and β are both equal to 0. No spin relaxation but complete momentum relaxation has the effect that α and β vanish and S equals S_0. This means that the polarization depends on the excitation and deexcitation channel and on the momentum and spin relaxation in the way described in Sec. I.

We use the following simple model to calculate the luminescence polarization. We divide the hot electrons into six groups with the occupation numbers $n_k^{p,r}$ for states with wave number k. The index p runs over h and l, specifying from which of the valence bands the electron was excited, the heavy- or light-hole band. The two main groups characterized by the index p are both divided into three subgroups. The first contains electrons with the original anisotropy in both momentum and spin $r=1$; the second has electrons with the anisotropy in spin direction but with isotropic momentum direction $r=2$; and the third has those with isotropic spin orientation r=3. The occupation number can be regarded as averaged over the direction of the wave vector $\mathbf{k}$. The anisotropy lies in the index r.

Assuming a stationary state, we can construct rate equations for the different occupation numbers using the fact that the increase in $n_k^{p,r}$, due to the exciting light and scattering into these states, equals the decrease, due to scattering out. We include electron-ion, electron-electron, and spin relaxation scattering. We assume that the electron-phonon scattering and the electron-hole recombination rates are much lower and can be neglected in the

rate equations. The spin relaxation we assume to be of the D'yakonov-Perel' type.[8] In the electron-ion scattering the degree of randomization of the propagation direction varies with electron momentum. We calculate the mean value X of the projection of a unit vector in the direction of propagation after a scattering event on the direction of propagation before scattering. X as a function of electron wave number is shown in Fig. 2. The wave number k, is expressed in units of $2k_0$, where k_0 is related to the doping density ρ according to $k_0 = (3\pi^2\rho)^{1/3}$. We use X to simulate the fraction of $r=1$ electrons that remains in this subgroup after a scattering event. Similar considerations apply for the electron-electron scattering.

For the $r=1$ electrons the states are filled by the pumping light at the rate $[1-f^p(k)]g^{p,1}(k)$ and by electrons decaying via electron-electron scattering from higher-energy states. A fraction $F_X(k,k_1)$ of the electrons decay into $n_k^{p,1}$ and the rest into $n_k^{p,2}$. The electron states are emptied: through spin relaxation, into $n_k^{p,3}$; through electron-ion scattering, into $n_k^{p,2}$; and through electron-electron scattering to lower-energy states.

For the $r=2$ electrons the states are filled by electron-ion scattering from $r=1$ electrons and by electrons decaying via electron-electron scattering from higher-energy states. The states are emptied through spin relaxation into $n_k^{p,3}$ and through electron-electron scattering to lower-energy states.

For the $r=3$ electrons the states are filled by spin relaxation from the $r=1$ and $r=2$ electrons and from higher-energy states via electron-electron scattering. The states are emptied through electron-electron scattering to lower-energy states.

We arrive at the following rate equations for $n_k^{p,r}$:

$$n_k^{p,1} = \frac{[1-f^p(k)]g^p(k,\omega)\hbar/2 - \int_k^\infty dk_1 \frac{k_1^2}{k^2}F(k,k_1)F_X(k,k_1)n_{k_1}^{p,1}}{\mathrm{Im}\{-\Sigma^s(k)-\Sigma^e(k)-[1-X(k)]\Sigma^i(k)\}} , \tag{2}$$

$$n_k^{p,2} = \left[\int_k^\infty dk_1 \frac{k_1^2}{k^2}F(k,k_1)\{n_{k_1}^{p,2}+[1-F_X(k,k_1)]n_{k_1}^{p,1}\}+n_k^{p,1}[1-X(k)]\Sigma^i(k)\right]\Big/ \mathrm{Im}[\Sigma^s(k)+\Sigma^e(k)] , \tag{3}$$

and

$$n_k^{p,3} = \left[\int_k^\infty dk_1 \frac{k_1^2}{k^2}F(k,k_1)n_{k_1}^{p,3}+(n_k^{p,1}+n_k^{p,2})\Sigma^s(k)\right]\Big/ \mathrm{Im}[\Sigma^e(k)] . \tag{4}$$

The function $F(k,k_1)$ is defined in such a way that $-2dk\,F(k,k_1)/\hbar$ equals the probability per unit time for an electron in state k_1 to decay via electron-electron scattering into the wave number element $(k, k+dk)$. It is related to the imaginary part of the self-energy shift according to

$$\int_0^{k_1} dk\,F(k,k_1) = \mathrm{Im}\Sigma^e(k_1) = -\frac{\hbar}{2\tau_{k_1}^e} . \tag{5}$$

The function $F_X(k,k_1)$ is defined in analogy with X, but now for transitions via electron-electron scattering

from state k_1 to k. The functions F and F_X are shown in Figs. 3 and 4, respectively, as functions of k, for a set of k_1 values. The k_1 values are given by the k values at the right-hand onset of the curves. F is expressed here in units of $\mathrm{eV}/2k_0$, which means that the area under the curve equals $\mathrm{Im}\,\Sigma^e(k_1)$ in eV. It should be noted that F is limited to a rather narrow k region and that the plasmon excitations give contributions in an even more narrow region. The plasmon contribution sets in with a threshold on the right-hand side and then drops off gradually towards smaller k values. These sharp structures cause
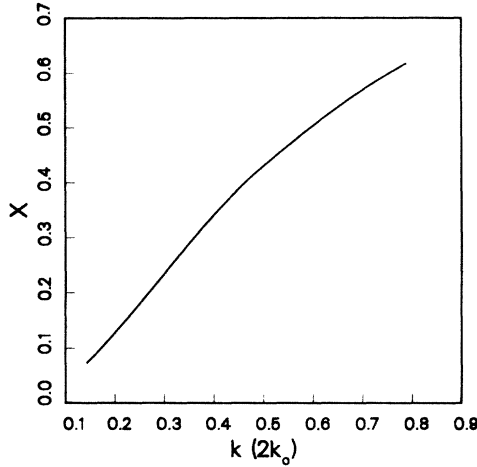
FIG. 2. Mean value X of the projection of a unit vector in the direction of electron propagation, after an ion-scattering event, on the direction of propagation before scattering, as a function of electron wave number.
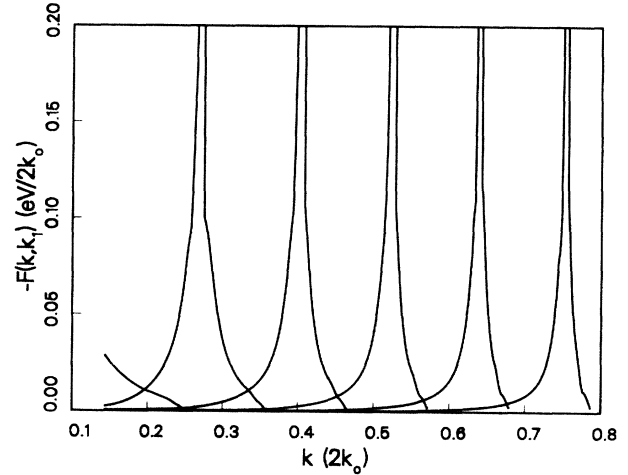

FIG. 3. $F(k,k_1)$, described in the text, as function of k for a set of k_1 values. The k_1 values are the k values at the right-hand onset of the curves.

plasmon replicas of the minimum in the luminescence polarization and of the weak shoulder in the HPL. The function F_X is plotted in Fig. 4 for the same set of k_1 values as F was in Fig. 3. As can be noted the plasmon scatters the electron almost completely in the forward direction. For higher k_1 values there are regions in k where the scattering is weakly dominated by backward

scattering, i.e., F_X is slightly negative. The functions Σ^s, Σ^e, and Σ^i are the self-energy shifts for the electron due to spin, electron-electron, and electron-ion scattering, respectively. The first term in the numerator of Eq. (3) represents the pumping rate. The first factor is the Fermi occupation number for the holes in the valence band p. For the function $g^p(k,\omega)$ we use the following formula:

$$g^P(k,\omega) \sim \frac{-\pi^{-1}\,\mathrm{Im}[\Sigma^e(k)+\Sigma^i(k)+\Sigma^P(k)]}{\mathrm{Im}^2[\Sigma^e(k)+\Sigma^i(k)+\Sigma^P(k)]+\left[\hbar\omega-\left(E_g+\dfrac{\hbar^2k^2}{2m_e}+\mathrm{Re}\Sigma^e(k)+\mathrm{Re}\Sigma^i(k)+\dfrac{\hbar^2k^2}{2m_p}+\mathrm{Re}\Sigma^P(k)\right)\right]^2} \quad (6)$$

where E_g denotes the unperturbed band gap and $\hbar\omega$ is energy of the exciting photon. $\Sigma^P(k)$ is the self-energy shift in the valence band p and contains contributions from both electron-electron and electron-ion scattering.

We gave in Ref. 2 a detailed derivation of the complex-valued self-energy shifts, entering the present calculation, and we refrain from repeating it here. We briefly mention that the calculation was performed with the Rayleigh-Schrödinger perturbation theory and within the random-phase approximation. The valence-band coupling was taken into account, giving rise to inter-valence-band scattering. However, in the screening of the scattering matrix elements we used decoupled valence-band polarizabilities. The reader is referred to Ref. 2 for more details.

In Fig. 5 are shown the imaginary parts of the self-energy shifts, which are proportional to the scattering rates $\tau^{-1} = -2\,\mathrm{Im}\Sigma/\hbar$. The dotted and dashed curves represent the contribution from electron-ion and electron-electron scattering, respectively. The solid curves show
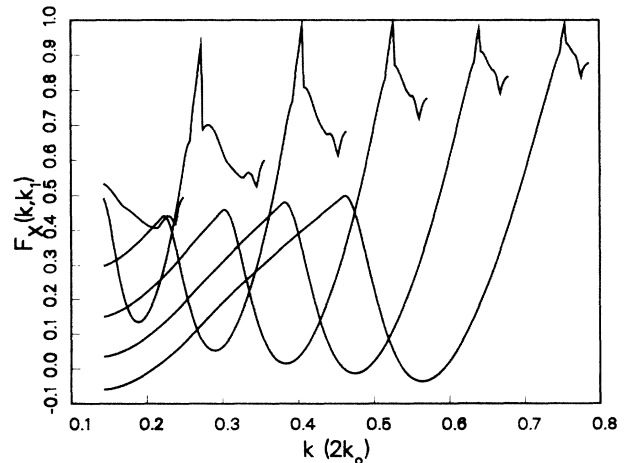

FIG. 4. $F_X(k,k_1)$, described in the text, as function of k for the same set of k_1 values as in Fig. 3.
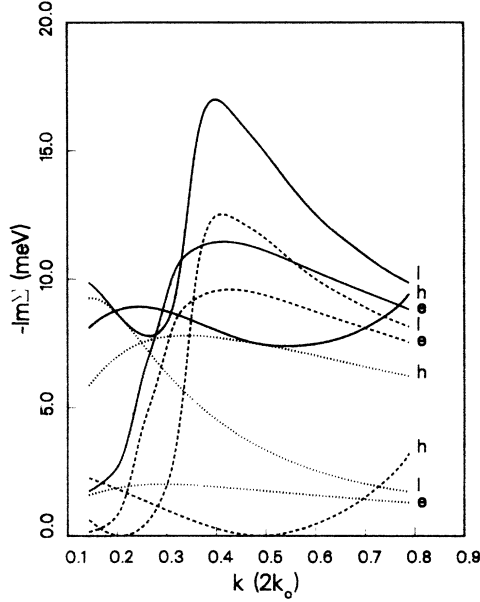
FIG. 5. Minus the imaginary part of various self-energy shifts as functions of particle wave number. The indices e, h, and l denote electron, heavy hole, and light hole, respectively. The dotted curves show the contributions from electron-ion scattering while the electron-electron scattering contributions are represented by the dashed curves. The full curves give the sum of the two contributions.

FIG. 6. Real part of various self-energy shifts. The notations are the same as in Fig. 5.

the total results. The parameters e, h, and l denote states in the conduction, heavy-role, and light-hole bands, respectively. The dashed curves for l and h touch the abscissa for the wave numbers $k_{F,l}$ and $k_{F,h}$, respectively, the Fermi wave numbers in the valence bands. The steep increase in the dashed curves is due to the onset of plasmon excitations. For the heavy holes this onset occurs at higher energies, outside the range of the figure.
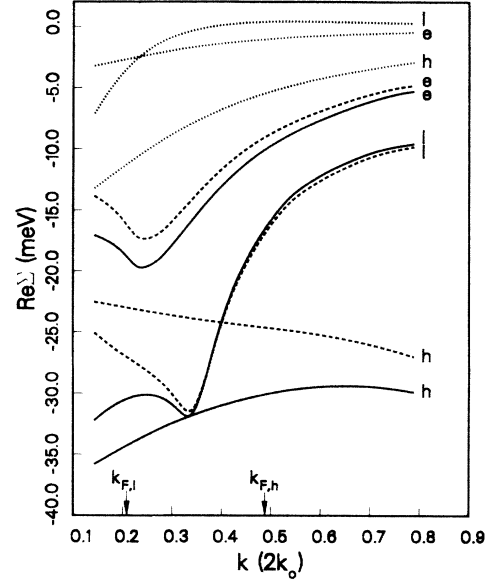
In the region of plasmon excitation the real parts of the self-energy shifts have a dip as can be seen in Fig. 6. The exact position and strength of these plasmon structures should not be taken too seriously as they could be modified by the inclusion of the inter-valence-band polarizability, which we have left out.[2]

We take the lifetime broadening into account in the expression for the emission intensities, the same way as in the expression for the pumping. From the occupation numbers which are obtained $n_k^{p,r}$, we calculate the emission intensities for the two polarization directions according to

$$I^+(\omega) \sim \int_0^\infty dk\, k^2 \{ f^h(k) g^h(k,\omega)[6n_k^{h,1}/7 + 7n_k^{l,1}/19 + 5(n_k^{h,2} + n_k^{l,2})/8 + (n_k^{h,3} + n_k^{l,3})/2]$$
$$+ f^l(k) g^l(k,\omega)[6n_k^{l,1}/7 + 7n_k^{h,1}/19 + 5(n_k^{l,2} + n_k^{h,2})/8 + (n_k^{l,3} + n_k^{h,3})/2] \} , \qquad (7)$$

and

$$I^-(\omega) \sim \int_0^\infty dk\, k^2 \{ f^h(k) g^h(k,\omega)[n^{h,1}/7 + 12n_k^{l,1}/19 + 3(n_k^{h,2} + n_k^{l,2})/8 + (n_k^{h,3} + n_k^{l,3})/2]$$
$$+ f^l(k) g^l(k,\omega)[n_k^{l,1}/7 + 12n_k^{h,1}/19 + 3(n_k^{l,2} + n_k^{h,2})/8 + (n_k^{l,3} + n_k^{h,3})/2] \} . \qquad (8)$$

The luminescence polarization is defined as

$$P_c(\omega) = [I^+(\omega) - I^-(\omega)]/[I^+(\omega) + I^-(\omega)] . \qquad (9)$$

Our results for the luminescence polarization are displayed in Fig. 7. The position and size of the minima agree quite well with the experimental results in Fig. 2 of Ref. 7. However, we do not reproduce the steep increase towards, and the high values at, the excitation energies. One possibility is that these high polarization values are from recombination with the photocreated holes. To

simulate this contribution we arbitrarily let the temperature for holes be 100 instead of 2 K. The results in Fig. 7 are from that calculation. The only important effect of the enhanced temperature is for excitation energies 1.782 eV and larger, in which case a new peak develops, centered around the excitation energy. In the results from the calculation at 2 K, the curves turn over at the shoulder to the left of the peak.

In our results we find a tendency towards a second minimum, to the left of the main minimum. This is the
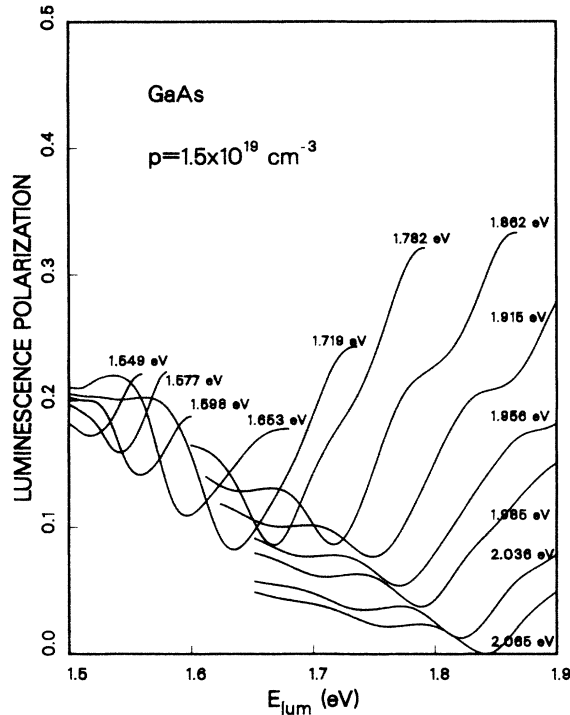
FIG. 7. Theoretically obtained luminescence polarization as function of luminescence energy. The curves are labeled by the excitation energies.
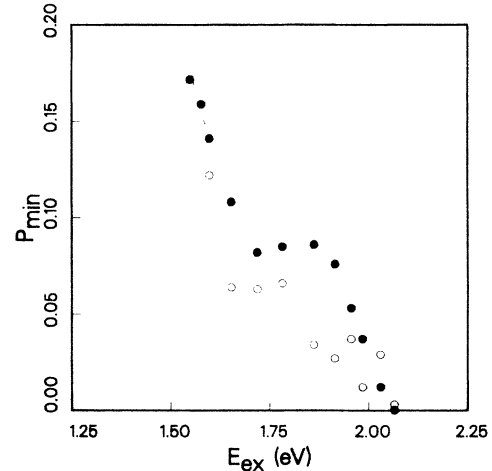

FIG. 8. Polarization at the polarization minimum as function of excitation energy. The solid circles are the theoretical result from Fig. 7. The experimental result from Ref. 7 is shown as open circles.

plasmon replica mentioned before. These structures are also found in the experimental curves, but weaker.

In Fig. 8 we show the theoretical (solid circles) and experimental (open circles) polarization values at the minima. The two sets of data points have some trends in common. They first decrease steeply, then have a flat region and finally decrease steeply again.

IV. SUMMARY AND CONCLUSIONS

We have performed a calculation of the luminescence polarization for p-type GaAs at the doping density 1.5×10^{19} cm^{-3}. The shift of the states in the conduction and valence bands, due to the doping, was taken into account as well as the effect of lifetime broadening in the excitation and luminescence processes. We found, from the variation in its position with excitation energy, that the minimum in the luminescence polarization is caused by electrons, promoted from the lighthole band to the conduction band, recombining with heavy holes. Both processes are k conserving. The variation of the position of the minimum with excitation energy was reproduced quite well theoretically, indicating that the calculated doping-induced energy shifts were good for the states in the whole momentum range bracketed by the two-hole Fermi wave numbers.

Our calculated luminescence polarization showed an overall good agreement with experiments, except for the energy region close to the excitation energy. The experimental curves show a broad peak with rather sharp edges centered around the excitation energy.[4] The shape was

not reproduced theoretically and the obtained peak value was only roughly half the experimental one. One possible explanation for this peak is recombination with the photo-created holes, not included in the calculation. To include these in a realistic way one has to calculate hole occupation numbers similar to the electron occupation numbers we obtained here, but one needs in this case absolute numbers. This is because these photocreated holes take part in the luminescence processes in competition with the thermalized holes.

A second possible cause of the discrepancy is the corrugation of the equienergy surfaces in the valence bands, due to the warping of the bands. They were found to be important for linear luminescence polarization,[9] but we have neglected them here.

A third, rather speculative, possibility is that part of the detected light in the region of excitation energy has been scattered inelastically through inter-valence-band excitations.

In conclusion we found that the minimum in the luminescence polarization, and the corresponding shoulder in HPL, were explained by k-conserving processes. These structures are due to contributions from electrons, promoted from the light-hole band, recombining with heavy holes.

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