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Photoluminescence spectroscopy of band-gap narrowing in *n*-type $Al_xGa_{1-x}As$

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We present an experimental investigation of band-gap narrowing in *n*-type $Al_xGa_{1-x}As$ using photoluminescence spectroscopy at room temperature, as a function of electron concentration. The observed systematic shift of the band-to-band transition energy to lower energies with increasing electron concentration has been attributed to band-gap narrowing in $Al_xGa_{1-x}As$. The experimental results have been compared with the available theoretical calculations and interpreted as many-body effects due to exchange and correlation among majority and minority carriers.

In a heavily doped semiconductor, the density of states differs from that of the undoped pure crystal due to many-body effects. The electron-electron interactions shift the conduction and valence bands towards each other and cause band-gap narrowing (BGN). BGN induced by heavy doping has been extensively investigated in elemental and III-V binary compound semiconductor such as GaAs.[1–7] In contrast, no experimental effort has been devoted to study BGN in important ternary alloy semiconductor such as $Al_xGa_{1-x}As$.[7] The effects of heavy doping on the optical and electrical properties of semiconductors have been widely investigated over the years because of two reasons: first the fundamental physics underlying BGN and second their importance in device applications. Heavily doped layers are integrated in many devices and the estimation of BGN is an important input in device simulations. $Al_xGa_{1-x}As$ is the most widely used alloy semiconductor for fabricating heterostructure devices, such as field effect transistors and light emitting devices. Furthermore, several theoretical calculations[3,8–13] with different techniques have been published. Hence more experiments on BGN in different semiconductors with different intervals of doping would therefore appear motivated.

Photoluminescence (PL) spectroscopy has emerged as a standard and powerful technique to study the optical properties of bulk and low-dimensional semiconductor structures. It is highly desirable to adopt a noncontact, nondestructive optical technique to determine the homogeneity of the crystalline quality and distribution of alloy composition in the epitaxial layer. PL spectroscopy is routinely used to characterize the quality of semiconductor substrates and thin epitaxial layers for different device structures and for determining the composition in ternary and quaternary alloy semiconductors. PL spectroscopy is a direct way to measure the band-gap energy as a function of different parameters, such as carrier concentration, excitation intensity, temperature, pressure, and magnetic field. The low-temperature PL spectra from III-V binary and ternary compound semiconductors with reasonably good crystalline quality are dominated by band-edge and near-band-edge transitions. The underlying recombination processes can be identified from the behavior of PL spectra as a function of temperature and excitation intensity. Low-temperature PL spectroscopy is usually performed to study the excitonic recombination mechanisms in semiconductors, but recently room-temperature PL spectroscopy has received attention for different reasons.[14,15] In most cases, band-to-band (BB) transitions can be observed only at room temperature or at higher temperature. Several many-body effects such as BGN, screening, and carrier-exciton scattering can be studied using room-temperature PL spectroscopy.[7] Beside the understanding of basic physics regarding the mechanism of recombination processes at higher temperature, determination of the quantum efficiency of the BB transition at room temperature is extremely important for optoelectronic devices based on $Al_xGa_{1-x}As$.

In this work, the ternary alloy semiconductor $Al_xGa_{1-x}As$ has been used to study the nature of BB radiative recombination at room temperature and the effect of electron population on the unperturbed band gap. Several important band-edge transitions, including excitonic- and impurity-related recombination processes, have been investigated in $Al_xGa_{1-x}As$ epilayers,[7] but the BB transition involving free electrons and free holes has not been studied rigorously in $Al_xGa_{1-x}As$. The goal set in this paper is to study the role of many-body effects on the BGN in *n*-type $Al_xGa_{1-x}As$. $Al_xGa_{1-x}As$ epitaxial layers with different AlAs mole fractions and doping level ranging from 3×10^{16} to $3\times10^{18}\,\mathrm{cm}^{-3}$ have been used in this investigation. To avoid other perturbations, such as electron-donor impurity interaction, band tailing, and band filling effects due to heavy doping on the unperturbed band gap, we have carefully chosen this interval of doping concentrations. The impurity band caused by statistical fluctuations due to impurity-impurity interactions and the random distribution of impurities introduces band tails in the conduction band. A detailed theoretical analysis that rigorously combines many-body effects due to exchange-correlation and statistical fluctuations is extremely complicated problem.[16] As far as our knowledge goes, there is no first-principles calculation available for this problem. This particular range of doping concentration is also important for electronic and optoelectronic devices. We have also used the excitation intensity dependence of the BB transition energy at room temperature to study the BGN in $Al_xGa_{1-x}As$. In summary, the main objective of this paper is to study the effects of exchange and correlation on the BGN in *n*-type $Al_xGa_{1-x}As$.

The $Al_xGa_{1-x}As$ samples were Se doped and grown by metal organic chemical vapor epitaxy (MOCVD) on a semi-insulating and *n*-type GaAs substrate. The active

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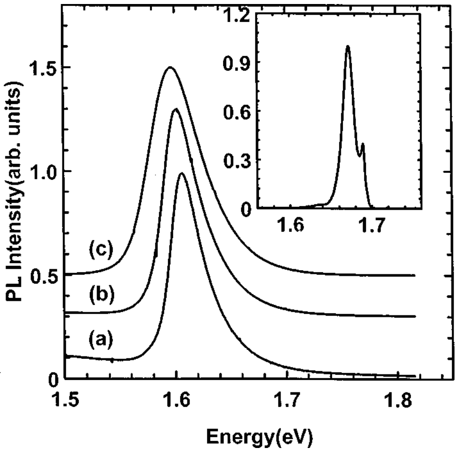

FIG. 1. PL spectra of $Al_{0.11}Ga_{0.89}As$ with (a) $8.2\times10^{16}\,cm^{-3}$, (b) $2.4\times10^{17}\,cm^{-3}$, and (c) $8.9\times10^{17}\,cm^{-3}$ electron concentration at 300 K. An increase of the doping level shifts the band-to-band peak to lower energies. PL spectra of $Al_{0.11}Ga_{0.89}As$ with $2.4\times10^{17}\,cm^{-3}$ electron concentration at 7.8 K are shown in the inset.

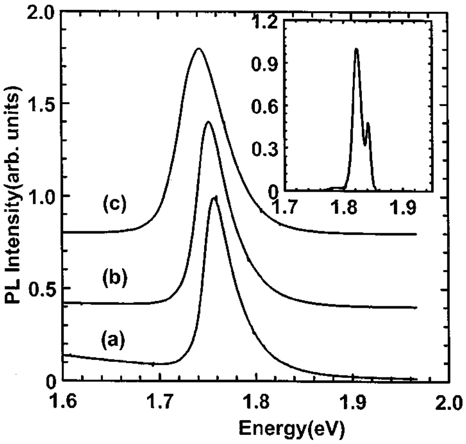

FIG. 2. PL spectra of *n*-doped $Al_{0.22}Ga_{0.78}As$ with (a) $4.5\times10^{16}\,cm^{-3}$, (b) $1.3\times10^{17}\,cm^{-3}$, and (c) $9.6\times10^{17}\,cm^{-3}$ electron concentration at 300 K. An increase of the doping level shifts the band-to-band peak to lower energies. PL spectra of $Al_{0.22}Ga_{0.78}As$ with $1.2\times10^{17}\,cm^{-3}$ electron concentration at 7.8 K are shown in the inset.

$Al_xGa_{1-x}As$ layers are separated from the GaAs substrate by an undoped spacer layer to avoid any two-dimensional (2D) effects. The AlAs mole fractions were determined by PL at 7.8 K using the relation[7] $E_g(Al_xGa_{1-x}As)=1.5194+1.48x$. Two sets of samples were grown with AlAs mole fractions of $x=0.11$ and 0.22. Thicknesses of all $Al_xGa_{1-x}As$ layers were about 2–4 μm. The electron concentrations were determined by Hall measurements. The maximum electron concentrations were found to be 2.8×10^{18} and $2.6\times10^{18}\,cm^{-3}$ in $Al_{0.11}Ga_{0.89}As$ and $Al_{0.22}Ga_{0.78}As$, respectively. The samples were housed in optical cryostat with front surface excitation and emitted radiation was collected for PL spectroscopy. The PL spectra were collected in the wavelength region of 600–900 nm. An infrared filter was used to block any light shorter than 600 nm. The samples were kept in a closed-cycle He refrigerator and excited with 457- or 514-nm lines of an Ar^+ ion laser. The PL signal was collected into a 0.46-m Jobin-Yvon grating monochromator and detected either with a liquid-nitrogen-cooled Ge detector or Si detector. The excitation density was varied from 10 $mW\,cm^{-2}$ to 100 $W\,cm^{-2}$ by a neutral density filter or by varying the output power of the laser. The PL data were processed using the lock-in technique. The spectral response of the system was calibrated using a blackbody source.

Typical room-temperature emission spectra in *n*-type $Al_xGa_{1-x}As$ as a function of electron concentration for $x=0.11$ and 0.22 are shown in Figs. 1 and 2, respectively. Room-temperature PL spectra are characterized by a Maxwell-Boltzmann line shape on the high-energy side and a relatively sharp cut off on the low energy side caused by the $(E_g-E)^{1/2}$ dependence, where E_g is the band gap. The insets of Figs. 1 and 2 show the PL spectra at 7.8 K. Low-temperature PL spectra show bound exciton (BE) and donor acceptor pair (DAP) recombination. Figures 1 and 2 show the PL spectra at 300 K for three electron concentrations, and it is clear that BB peaks shift towards lower energy as the electron concentration increases. It has been observed[1–7] that as a function of doping concentration, BB peak maxima in *n*-type GaAs shift to higher energies while the *p* type shifts to lower energies. The contribution due to the Burstein-Moss effect overrules the BGN in *n*-doped GaAs, contrary to *p*-doped GaAs. Hence the band-gap filling is more pronounced in *n*-doped GaAs compared to *p*-doped GaAs due to the small electron effective mass.[5] In case of *n*-doped $Al_xGa_{1-x}As$, the band-gap filling cannot overrule BGN due to a higher electron effective mass compared to that in GaAs. We have also avoided band-gap filling in our $Al_xGa_{1-x}As$ samples by carefully choosing the carrier concentration such that except for few samples with carrier concentration of 8.9×10^{17} and $2.8\times10^{18}\,cm^{-3}$ in $Al_{0.11}Ga_{0.89}As$ and 9.6×10^{17} and $2.6\times10^{18}\,cm^{-3}$ in $Al_{0.22}Ga_{0.78}As$, all samples are nondegenerate. It is clear from the Figs. 1 and 2 that the BB peak broadens as the doping concentration increases, but the typical BB recombination line shape does not change significantly with doping concentration. If the doping concentration is increased beyond $5\times10^{18}\,cm^{-3}$, the line shape of PL spectra at 300 K changes completely and becomes more like a Gaussian line shape and the determination of the band-gap energy from PL spectra becomes doubtful. Figure 3 shows the dependence of the BB emission intensity on laser power. The BB peak at 300 K increases following the relation $P\propto L^m$, where P is the integrated PL intensity and L is the laser power. The BB peak shows an expected[14] linear dependence with $m=1.01$ and 1.09 in $Al_{0.11}Ga_{0.89}As$ and $Al_{0.22}Ga_{0.78}As$ respectively. Impurity-related recombination is generally characterized by $m<1$, which is in disagreement with our experimental observation for BB transitions. The redshifts of BB transitions energies in $Al_{0.11}Ga_{0.89}As$ and $Al_{0.22}Ga_{0.78}As$ at 300 K are shown as a function of the logarithm of electron concentration in Fig. 4. The variation of E_g can be fitted with $E_g=E_g^0-aN^{1/3}-bN^{1/4}$, where E_g^0 is the unperturbed band gap and N is the electron concentration. The coefficients a and b are evaluated by fitting with experimental data (Fig. 4). From experimental data we could derive following empirical relations for BGN:

$$\Delta E_g=-3.2\times10^{-8}N^{1/3}-5.5\times10^{-7}N^{1/4} \qquad (1)$$

for $Al_{0.11}Ga_{0.89}As$ and

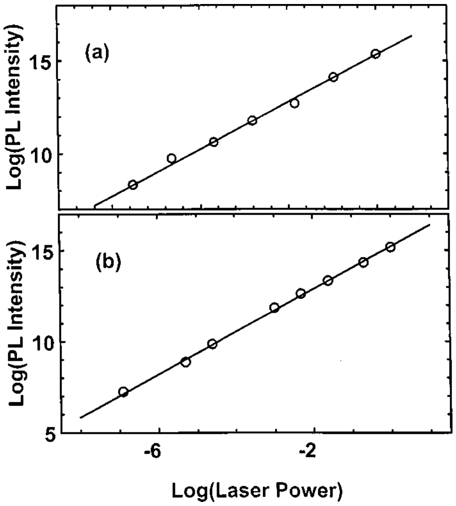

FIG. 3. Log-log plot of integrated PL intensity vs excitation intensity for the BB peak at 300 K in (a) $Al_{0.11}Ga_{0.89}As$ with $2.4\times10^{17}\,cm^{-3}$ electron concentration and (b) $Al_{0.22}Ga_{0.78}As$ with $1.3\times10^{17}\,cm^{-3}$ electron concentration.

$$\Delta E_g=-2.4\times10^{-8}N^{1/3}-6.2\times10^{-7}N^{1/4} \tag{2}$$

for $Al_{0.22}Ga_{0.78}As$. Figures 5 and 6 show the excitation intensity dependence of the position of BB transition energy in $Al_{0.11}Ga_{0.89}As$ and $Al_{0.22}Ga_{0.78}As$, respectively, at 300 K. It is clear from Figs. 5 and 6 that there is strong but continuous shift of the entire spectrum towards lower energies as the laser power increases. We have avoided very high laser power where we start observing a change in the line shape of the PL spectra. Localized heating due to high-intensity laser excitation at the sample surface was thought to be the reason for the redshift of the PL spectra. The same excitation intensity was used at 7.8 K, but no redshift has been observed in the case of BE-related PL peaks, which is evident from the inset of Figs. 5 and 6. Thus the redshift of the BB PL peak position is believed to be related to band-gap shrinkage due to many-body effects.[15] BB PL peaks in other samples with different electron concentrations show a similar trend with excitation intensity. In the case of experiments on BGN as a function of excitation intensity, it can be assumed[15,17] that the electron concentration $N\propto I$, where I is the excitation intensity. The redshift of the BB transition energies in $Al_{0.11}Ga_{0.89}As$ and $Al_{0.22}Ga_{0.78}As$ at 300 K are shown as a function of excitation intensity in Fig. 7. From experimental data we could derive the following empirical relation for BGN:

$$\Delta E_g=-9.3\times10^{-3}I^{1/3}-3.1\times10^{-3}I^{1/4} \tag{3}$$

for $Al_{0.11}Ga_{0.89}As$ and

$$\Delta E_g=-1.1\times10^{-2}I^{1/3}-1.4\times10^{-2}I^{1/4} \tag{4}$$

for $Al_{0.22}Ga_{0.78}As$. The absence of a logarithmic dependence[18] of the BB peak energy with excitation intensity

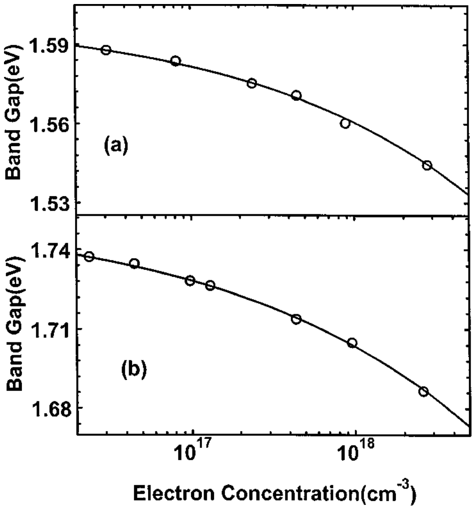

FIG. 4. Band-gap energy determined from the 300 K PL spectra (given in Figs. 1 and 2) shown as a function of logarithm of electron concentration for (a) $Al_{0.11}Ga_{0.89}As$ and (b) $Al_{0.22}Ga_{0.78}As$. Solid lines are the fitted curves.

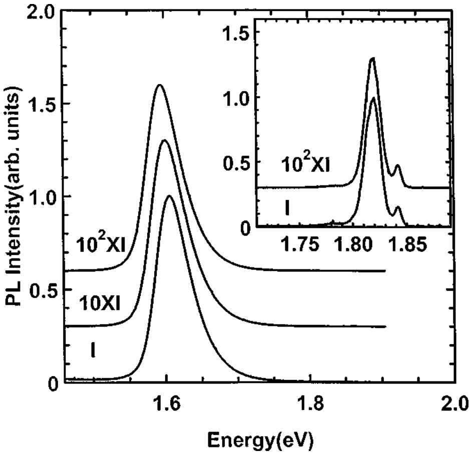

FIG. 5. PL spectra of $Al_{0.11}Ga_{0.89}As$ with $2.4\times10^{17}\,cm^{-3}$ electron concentration at 300 K for three different excitation intensities, $I=100\,mW/cm^2$. An increase of the excitation intensity shifts the band-to-band peak to lower energies. The inset shows the PL spectra for two different excitation intensities at 7.8 K. There is no redshift of the bound exciton peak with excitation intensity.

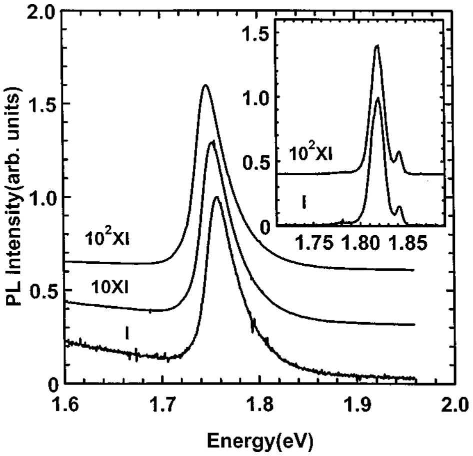

FIG. 6. PL spectra of $Al_{0.22}Ga_{0.78}As$ with $1.3\times10^{17}\,cm^{-3}$ electron concentration at 300 K for three different excitation intensities, $I=100\,mW/cm^2$. An increase of the excitation intensity shifts the band-to-band peak to lower energies. The inset shows the PL spectra for two different excitation intensities at 7.8 K. There is no redshift of the bound exciton peak with excitation intensity.

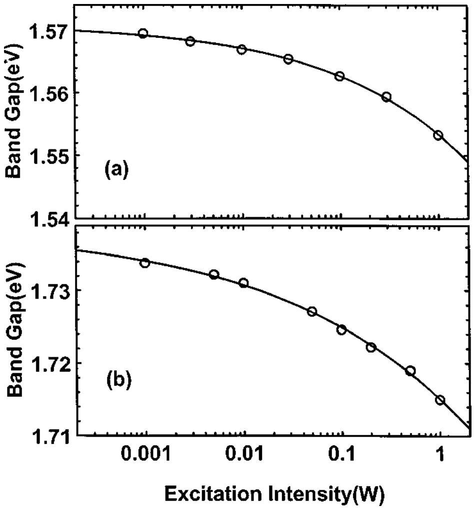

FIG. 7. Band-gap energy determined from the 300 K PL spectra (given in Figs. 5 and 6) shown as a function of logarithm of excitation intensity for (a) $Al_{0.11}Ga_{0.89}As$ with $2.4\times10^{17}\,cm^{-3}$ electron concentration and (b) $Al_{0.22}Ga_{0.78}As$ with $1.3\times10^{17}\,cm^{-3}$ electron concentration. Solid lines are the fitted curves.

rules out the existence of strong band tailing effects in $Al_xGa_{1-x}As$ with the range of doping used for this investigation.

We have determined the band-gap energy E_g from the PL spectra by a linear extrapolation of the low-energy side of the spectrum to the background level as proposed by Olega and Cardona[4] in case of heavily doped GaAs. PL arising from BB recombination exhibits an emission peak displaced to higher energy than the band-gap value E_g. This occurs due to following line shape of BB transitions assuming k conservation:[19]

$$I_{PL}\propto(E-E_g)^{1/2}\exp\left(-\frac{E-E_g}{k_BT}\right), \tag{5}$$

which corresponds to the BB emission peak maximum at $E_g+\frac{1}{2}k_BT$ (k_B is Boltzmann's constant). We have found that the difference between BB peak maxima and E_g is around 19.4 and 24.4 meV in $Al_{0.11}Ga_{0.89}As$ with $2.4\times10^{17}\,cm^{-3}$ and $Al_{0.22}Ga_{0.78}As$ with $1.7\times10^{17}\,cm^{-3}$ electron concentrations, respectively. This difference increases with doping concentration and becomes around 33.6 and 38.1 meV in the case of $Al_{0.11}Ga_{0.89}As$ with $8.9\times10^{17}\,cm^{-3}$ and $Al_{0.22}Ga_{0.78}As$ with $9.6\times10^{17}\,cm^{-3}$ electron concentrations, respectively. As the doping concentration increases, electron-impurity and electron-electron scattering that relaxes the k-conservation and k-nonconserving BB transitions becomes dominant. If we include this effect, the line shape of the BB transitions can be given by[20,21]

$$I_{PL}\propto(E-E_g)^{\nu}\exp\left(-\frac{E-E_g}{k_BT}\right), \tag{6}$$

where exponent ν is greater than $\frac{1}{2}$ and the BB emission peak maximum occurs at $E_g+\nu k_BT$. We have observed that ν depends on doping concentration and AlAs mole fraction. It has been observed in $Hg_{1-x}Cd_xTe$ (Ref. 20) and $InAs_{1-x}Sb_x$ (Ref. 21) that BB recombination peaks at photon energies at as high as $2k_BT$ above the band-gap energy. The narrow linewidth (~5 meV) of the BE at 7.8 K and strong BB transition at 300 K indicate the high quality of our epitaxial layers. We could observe BB transitions with an excitation intensity as low as $1\ \mathrm{mW\,cm^{-2}}$. Singh and Bajaj[22] have shown that the alloy broadening of exciton is around 2 meV at an Al fraction of 0.11. This means an additional 3 meV broadening in the exciton peak is caused by impurities and local variations in the alloy composition. If thermal- and impurity-related broadening is ignored and it is assumed that 3 meV broadening is caused by alloy fluctuations, the deviation in alloy composition can be calculated and it turned out to be less than 0.003 in the absolute value of the Al fraction. We have also studied the variation of the band gap in $Al_xGa_{1-x}As$ due to alloy fluctuation by scanning the sample with BE peak energy. We have observed that the variation of the BE peak energy is small (~0.1 meV/cm) across the sample. This fluctuation corresponds to a variation of x less than 0.001 per cm and rules out the possibility of an energetic shift of the BB peak as a function of electron concentrations and excitation intensities towards lower energy due to band gap variation caused by alloy fluctuation.

The presence of the large concentration of free carriers can cause a significant reduction of the unperturbed band gap in semiconductors. This reduction is caused by many-body effects and carrier-impurity interactions. As the doping density increases, the conduction band edge moves downward and the valence band edge moves upward. The perturbation to the unperturbed band gap E_g^0 comes from (i) a random distribution of impurities and alloy fluctuations, which are almost independent of doping density,[8] and (ii) an interaction between electrons and donor impurities, which is appreciable only for very high carrier concentration ($>10^{19}\,cm^{-3}$).[8] To avoid these contributions, we have performed our BGN experiments with moderately doped samples with a maximum electron concentration of $10^{18}\,cm^{-3}$. The subject of this investigation is to study the effect of electron-electron interactions on BGN, which has been studied theoretically by several groups.[8–13,23,24] Assuming noninteracting quasiparticles the dispersion for the conduction band is given by

$$E_c(\mathbf{k},\omega)=\epsilon_c^0(\mathbf{k})+\hbar\Sigma_c(\mathbf{k},\omega), \tag{7}$$

where $\epsilon_c^0(\mathbf{k})$ is the conduction band energy and $\hbar\Sigma_c(\mathbf{k},\omega)$ is the self-energy of the electrons. As the valence band is completely filled, the Hatree-Fock exchange interaction $\hbar\Sigma_v^{\mathrm{HF}}(\mathbf{k})$ is included in the unperturbed band energy $\epsilon_v^0(\mathbf{k})$. Hence the dispersion for the valence band can be given by

$$E_v(\mathbf{k},\omega)=\epsilon_v^0(\mathbf{k})+\hbar\Sigma_c(\mathbf{k},\omega)-\hbar\Sigma_v^{\mathrm{HF}}(\mathbf{k}). \tag{8}$$

Hence the BGN ΔE_g is given by

$$\Delta E_g=\Delta E_c(\mathbf{k},\omega)-\Delta E_v(\mathbf{k},\omega). \tag{9}$$

If we neglect the electron-donor interaction, two terms contribute in ΔE_c: those are exchange and correlation energies of the majority carriers (electrons). The only contribution to the hole energy ΔE_v in case of n-type semiconductors (when holes are minority carriers) comes from the correlation energy due to its interaction with its own screening cloud. Hence there are three energy terms which contribute to the reduction of the band gap with electron population and it has been shown[23] that the effective change in the band gap due to many-body interactions can be given by

$$\Delta E_g = -aN^{1/3} - bN^{1/4}, \tag{10}$$

where the first term is due to the exchange interaction, which comes from the spatial exclusion of the like spins away from each other. The second term is due to the correlation energy, which comes from the repulsion of the like charges, so that they do not move independently, but in such a way so as to avoid each other as far as possible. It has been argued against the assertion given by Inkson[24] that the contribution of the correlation energy to ΔE_g is not important, because correlation energies cancel from the conduction and valence bands. This would be possible if correlation energies have opposite sign and carriers have the same effective mass with the same **k**. These assumptions are not valid in real systems. Furthermore, the correlation energies of both electrons and holes are negative and they actually add and do not cancel each other.

In case of very high doping, strong band tailing and lifetime broadening distort the density of states of the conduction band and cause band-gap shrinkage. This causes the determination of the band gap from the room-temperature PL spectra unreliable. This might be one of the reasons for observing[1–6] different dependences of ΔE_g on the majority carrier density. Moreover, in the case of ternary semiconductors like $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$, alloy fluctuation can also cause band tailing. Experimentally, it is extremely difficult to separate the different contributions in the band tailing from heavy doping and alloy fluctuations. We have observed a complete change in the line shape of the room-temperature PL spectra of $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ when the doping is increased from 2×10^{18} to $5\times10^{18}\ \mathrm{cm}^{-3}$. For determining the role of many-body effects due to exchange and correlation, it is desirable to restrict the doping concentration in such a way that the typical BB line shape can be observed in the PL spectra. Our assertion of ignoring the electron-impurity interaction and strong band tailing is justified by the results of experiments on the dependence of the band gap on excitation intensity. This is a much cleaner experiment to study the role of exchange and correlation on the reduction of the band gap. In both experiments on BGN, i.e., the variation of the band gap as a function of electron concentration and excitation intensity, the experimental results coincide with the theoretical predictions[23] [Eq. (10)] based on second-order perturbation theory with the random phase approximation. It is not possible to make any comments on the value of coefficients a and b, because neither detailed theoretical nor experimental results are available on BGN in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$.

In conclusion, band-gap narrowing effects have been studied in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ as a function of electron concentration and excitation intensity. The band-to-band transition shows a redshift due to band-gap narrowing as the electron concentration or excitation intensity increases. It has been found that exchange and correlation among majority and minority carriers causes band-gap narrowing in moderately doped *n*-type $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$.

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