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Photoluminescence of GaAs-$Al_xGa_{1-x}As$ multiple quantum well structure under high excitations

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Allowed transitions between the $n = 1$, 2, and 3 subbands of the conduction and valence bands of a multiple quantum well heterostructure of GaAs-$Al_{0.6}Ga_{0.4}As$ are seen in spontaneous emission under high excitations. The observed peaks agree very well with the calculated locations of the peaks when the finite depth of the potential well and the nonparabolicity of the conduction band are taken into account. The same basic features are seen under cw or picosecond pulse excitation and at room temperature, 77 K, or 4.2 K.

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There have been a number of reports on the photoluminescence spectra of multiquantum well structures of GaAs-$Al_xGa_{1-x}As$. Striking features related to the steplike density of states of such structures have been revealed in optical experiments such as absorption[1] and photoluminescence excitation spectroscopy.[2] There are significant differences in the reported photoluminescence (PL) spectra of samples grown by different methods from different laboratories, however, particularly in the case when the excitation level is high. Holonyak *et al.*[3] have performed a series of PL experiments on materials grown by metalorganic chemical vapor deposition (MOCVD). A variety of stimulated emission peaks was observed in these experiments. There was, however, no apparent correlation between the reported spontaneous emission peaks and the calculated quantum well states for $n \geqslant 2$. More recently, Miller *et al.*[4] reported on the observation of parity-forbidden transitions ($\Delta n \neq 0$) in GaAs wells under high photoexcitations in materials grown by molecular beam epitaxy (MBE). Allowed transitions between $n \neq 1$ states in the conduction and valence bands were not observed, however.

In this letter we report the observation of allowed transitions ($\Delta n = 0$) between the $n = 1$, 2, and 3 subbands of the conduction band and the heavy hole band of multiquantum well structures grown by MOCVD in conventional photoluminescence spectroscopy under high photoexcitations. No forbidden transition was seen in our sample. The observed peaks are in very good agreement with the calculated locations of the $\Delta n = 0$ peaks when the finite potential well depth and the nonparabolic nature of the GaAs Γ_6^c conduction band are taken into account. The same basic spectral features were seen with cw or picosecond-pulse excitation and at room temperature, 77 K, or 4.2 K. With ps photoexcitation, the initial amount of carriers generated in the quantum well states and, hence, the corresponding quasi-Fermi level can be estimated more accurately.

The sample used in the experiments was grown by a conventional low-pressure (76 Torr) MOCVD reactor using trimethylgallium (TMG), trimethylaluminum (TMA), and arsine (AsH_3) as the sources. The structure consists of seven undoped 145-Å GaAs wells between the 400-Å $Al_{0.6}Ga_{0.4}As$ barriers grown in a single growth cycle at 650 °C. A 300-Å-thick GaAs buffer layer was first grown on a Cr-O-doped semi-insulating GaAs substrate. Next, a 2000-Å layer of $Al_{0.6}Ga_{0.4}As$ was grown followed by the quantum well structure. In the low-temperature experiments, the sample was mounted on a large copper heat sink which was immersed in liquid nitrogen (77 K) or liquid helium (4 K). No heating effects were observed during the experiment.

The sample was photoexcited by a DCM dye laser at 650 nm operating in either the cw mode or synchronously pumped mode-locked mode producing a train of 5-ps pulses at a repetition rate of 10^8/s. The light was focused onto the sample with a microscope objective. Excitation densities of up to 10^{20} electron-hole pairs per cm^3 per pulse could be achieved in the sample. Since the sample was not in the form of an optical cavity, the sample did not operate as a laser. The spontaneous emission spectra were analyzed by a double-monochromator and detected by a cooled GaAs photomultiplier tube.

Figure 1 shows the PL spectra of a GaAs-$Al_{0.6}Ga_{0.4}As$ multiple quantum well heterostructure at 77 K under a low excitation level of $4\times10^{17}/cm^3$ (photogenerated carriers in each well). Only two peaks were observed with photon energies of 1.513 and 1.544 eV. The 1.513-eV peak is the expected contribution of the near-band emission from the GaAs buffer layer, denoted as BB in Fig. 1. The observation of BB emission rather than bound exciton emission BE can be attributed to thermal dissociation of the excitons at 77 K; in addition, the high excitation level led to a photoexcited carrier density greatly exceeding that of the excitons. The dominant peak at 1.544 eV is due to the GaAs well, and corresponds to the transition between the $n = 1$ electron subband and the $n = 1$ heavy hole subband (referred to as E_{1h}). On closer scrutiny, a small splitting of the 1.544-eV peak can be seen, with a shoulder peak at 1.555 eV. A similar structure has been observed both in absorption and PL excitation spectroscopy of high-quality GaAs-AlGaAs quantum well structures[1,2] and it has been attributed to the transitions (denoted as E_{1l}) between $n = 1$ bound electron states and $n = 1$ bound light hole states. As the excitation intensity is increased to $1\text{–}1.3\times10^{18}/cm^3$ and $3.6\text{–}4.4\times10^{18}/cm^3$ two lower intensity peaks appear at 1.610 eV (E_{2h}) and 1.705 eV (E_{3h}), respectively. This could be explained as the band fill-

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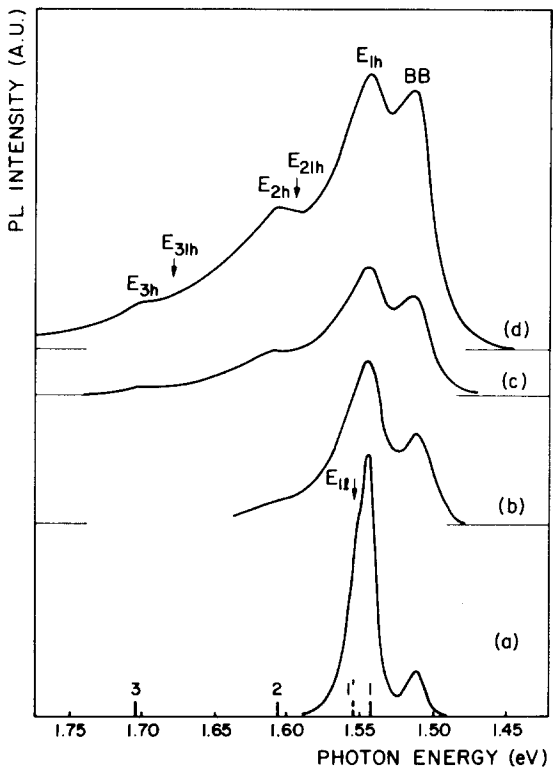

FIG. 1. Photoluminescence spectra of a GaAs-$Al_{0.6}Ga_{0.4}As$ multiple quantum well structure at 77 K when excited at different levels: (a) $4\times10^{17}/cm^3$, (b) $1.2\times10^{18}/cm^3$, (c) $4\times10^{18}/cm^3$, (d) $1.6\times10^{19}/cm^3$. The numbered vertical lines (solid for heavy hole, dashed for light hole) on the horizontal scales indicate the calculated energy positions of E_{1h}, E_{2h}, E_{3h}, and E_{1l}.

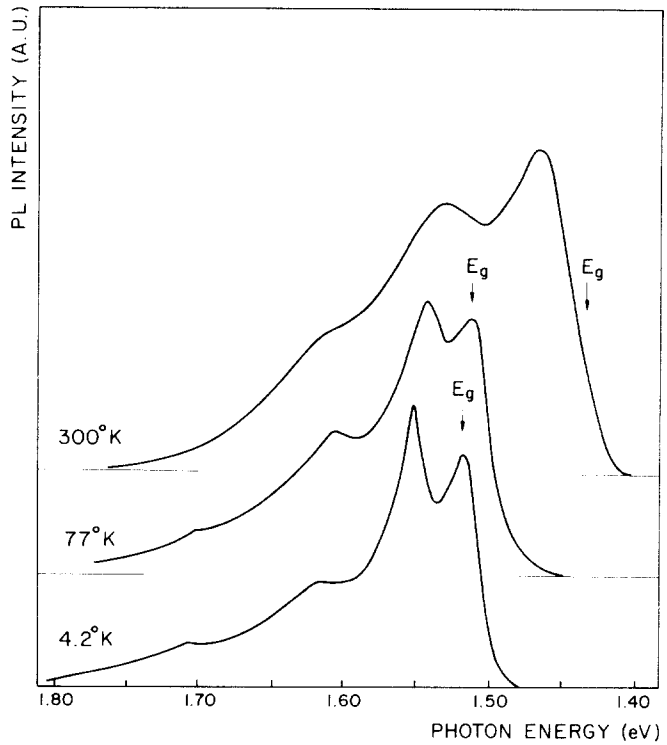

FIG. 2. Typical PL spectra at different temperatures under excitation level $\sim10^{19}/cm^3$. Arrows labeled E_g represent the band edges at different temperatures.

ing effect and will be discussed later. The peak energy positions of these two peaks do not change significantly with excitation level, as shown in Fig. 1.

In order to identify these two peaks, we have calculated the theoretical values for bound electron and hole states in the well. In the calculations, the effective masses for the ternary alloy $Al_xGa_{1-x}As$ were determined by assuming a linear dependence on composition (x) between the measured values[5] in GaAs and AlAs

$$m_e = 0.0665 + 0.0835x,$$
$$m_{hh} = 0.45 + 0.302x, \quad (1)$$
$$m_{lh} = 0.08 + 0.057x.$$

The total barrier height $\Delta E_g = E_g(Al_{0.6}Ga_{0.4}As) - E_g(GaAs)$ is split between the conduction band and valence band in the ratio $\Delta E_c = 0.85\Delta E_g$ and $\Delta E_v = 0.15\Delta E_g$. Most important, however, the nonparabolicity of the GaAs Γ_6^c conduction band has been taken into account in our calculations. This leads to an effective mass dependence on electron energy in the GaAs conduction band[5]

$$m_e^*(E) = 0.0665 + 0.0436E + 0.236E^2 - 0.147E^3, \quad (2)$$

where E is in eV. The net effect of an increasing effective mass on the bound state energies of a finite well is to lower the upper states while having a lesser effect on the lower states. The calculated energy positions of the allowed transitions ($\Delta n = 0$) are marked on the horizontal scale in Fig. 1 to facilitate comparison with experimental results. The agreement between the calculated and experimental values is excellent. In Fig. 1 we also show the positions of the parity-forbidden transitions, E_{21h} and E_{31h}, which are responsible for the transitions between E_{2e} and E_{1hh}, and E_{3e} and E_{1hh} as reported by Miller *et al.*[4] It is clear that our lower intensity peaks do not correlate with E_{21h} and E_{31h} transitions. Also, in our experiments, no evidence of phonon-assisted transitions has been found.

Similar data have been obtained both at room temperature and at 4.2 K, and are shown in Fig. 2. At 300 K the contribution from the GaAs buffer layer can hardly be seen because of spectral broadening. We have determined the first luminescence peak as being the allowed transition from the $n = 1$ subband by using temperature-dependent experiments. At 4.2 K, a GaAs buffer layer emission has been seen at 1.519 eV instead of 1.515 eV, a well accepted exciton emission in pure GaAs at 4.2 K. This and the fact that the photoexcited carrier density was much higher than the exciton density convinced us that at such high excitation levels, all peaks appearing in the spectra are not related to excitons; therefore, there is no reason to take the exciton binding energy into account in our calculations.

The variation of the spontaneous PL spectra with excitation intensity can be understood in terms of the band-filling effect.[3] With ps-pulse excitation, the initial position of the quasi-Fermi level of the electrons in the conduction band can be calculated in terms of the total number of photons, N, deposited in the sample per unit area per well:

$$N = g\sum_i \int_{E_{ie}}^{\infty} f(E - E_F)\,dE, \quad (3)$$

where $f(E - E_F) = [e^{(E-E_F)/kT} + 1]^{-1}$ is the Fermi distribution and $g = m_e^*/\pi\hbar^2$ is the two-dimensional density of

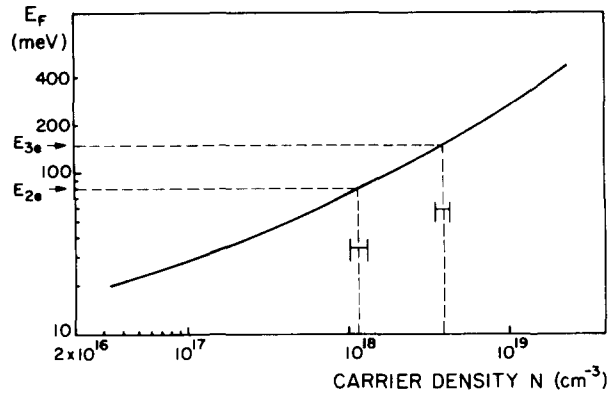

FIG. 3. Calculated electron Fermi level as a function of the photoexcited carrier density N at 77 K. The bracketed regions correspond to the excitation densities where the E_{2h} and E_{3h} transitions first occurred.

states of the well. E_{ie} is the quantized energy of the electron due to the well. In Eq. (3), because of the large effective mass of the heavy hole, we neglect the band-filling effect in the valence band. Integrating this expression, one obtains

$$N = g\sum_i \{E_F - E_{ie} + kT\ln|1 + e^{(E_{ie}-E_F)/kT}|\}. \quad (4)$$

Assuming that most of the electrons in the conduction band are confined in the well, the sum over i in these expressions can be limited to the bound states in the well. For our experimental conditions, it is sufficient to sum over the first three levels. Substituting in the values of g, E_{1e}, E_{2e}, E_{3e}, and kT, one can calculate E_F for each value of N. Figure 3 shows our calculated results for 77 K under ps-pulse excitation. The horizontal scale is expressed in terms of the photoexcited carrier density. The bracketed regions correspond to the excitation densities where the E_{2h} and E_{3h} transitions (E_{ih} refers to E_{ie} to $E_{i,\text{heavy-hole}}$) were first observed. These correspond roughly to where the calculated Fermi levels are within kT of E_{2e} and E_{3e} as one would expect according to the band-filling model. Also, experimentally where the different bound states first became observable did not vary significantly with temperature. This agrees with the above theory. The details of the spectral shapes depend upon the time variations of the quasi-Fermi levels in the case of pulse excitation and are more difficult to calculate; no attempt is made here to do so.

We have also studied the dependence of the luminescence peak intensity on excitation level for $n = 1,2$ states in the well. The data are shown in a log-log plot in Fig. 4. For comparison, the near-band emission from the GaAs buffer layer is also shown in the same figure. The slope is 2 at relatively low excitation intensities and 1.2 at higher excitation levels. These dependences for the near-band emission of bulk GaAs have been explained on the basis of nonradiative linear and quadratic recombination mechanism.[6] It is possible that the same mechanisms are responsible for the bound states in the quantum well.

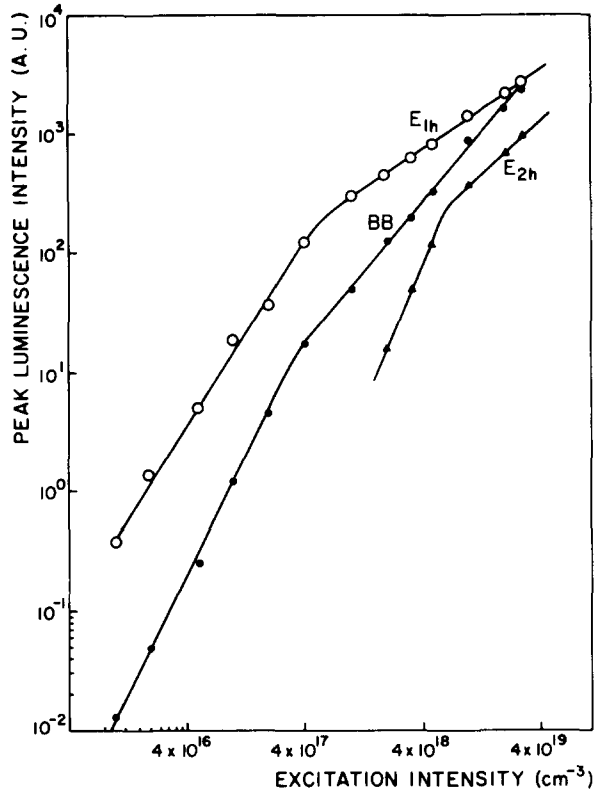

FIG. 4. Dependence of the luminescence intensities of peaks BB, E_{1h}, and E_{2h} on the excitation levels. The excitation level is expressed in terms of the photoexcited carrier density in the sample.

In conclusion, photoluminescence measurements on a high quality GaAs-$Al_{0.6}Ga_{0.4}As$ multiple quantum well structure at high excitation levels have shown $n = 1$, 2, and 3 bound state recombination in spontaneous emission. Consideration of the conduction-band nonparabolicity and the finite depth of the potential well have greatly improved the agreement between the measured transition energies and the calculated allowed transitions. Numerical calculations show that the variation of PL spectra with excitation intensity can be explained in terms of the band-filling effect.

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