Molecular beam epitaxial growth and photoluminescence of near-ideal GaAs-Al$_x$Ga$_{1-x}$As single quantum wells

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Single quantum wells with ~120-Å GaAs wells and Al$_{0.3}$Ga$_{0.7}$As or GaAs-Al$_{0.3}$Ga$_{0.7}$As superlattice barriers were grown by molecular beam epitaxy under conditions known to produce very high-purity material. Low-temperature photoluminescence measurements indicate that the dominant recombination transitions are associated with free and bound excitons involving both light and heavy holes. A forbidden transition, possibly E_{21h}, is also observed. The transition associated with electron-heavy-hole free excitons is most intense and has a linewidth of 0.3 meV at 2 K. The linewidths observed for these samples, grown with As$_4$ species at 630 °C, are the smallest for 120-Å single quantum wells and are close to theoretically calculated limits.

Quantum confinement is an important phenomenon that is being extensively used in making optoelectronic devices with superior properties. In particular, single and multiple quantum wells and superlattices composed of GaAs and Al$_x$Ga$_{1-x}$As are being widely used. Molecular beam epitaxy (MBE) has been very effective in growing such multilayered structures of desirable optical quality. However, effects such as interface roughness, clustering and island formation at the interfaces,[1–4] inter- and intrawell atomic layer fluctuation during growth,[5,6] and interdiffusion of impurities, can severely degrade the optical properties of quantum wells.

In this paper we report the MBE growth and optical properties of high-quality undoped GaAs-Al$_x$Ga$_{1-x}$As single quantum wells (SQW). The samples were characterized by high-resolution photoluminescence (PL) measurements. The excitonic luminescence spectra at 2 K are characterized by transitions which have linewidths (FWHM) ~0.3 meV and large intensity. Almost identical linewidths are measured in SQW with Al$_x$Ga$_{1-x}$As or with GaAs-Al$_x$Ga$_{1-x}$As superlattice barriers. To the best of our knowledge PL transitions with such narrow linewidths in SQW have not been previously reported. In what follows, the growth and PL spectra of the SQW structures are described and discussed.

Undoped SQW samples were grown in a three-chamber RIBER 2300 Modutrac MBE growth system with double load lock. The system preparation is briefly described. The effusion cells without the crucibles are baked at 1400 °C for 10 h. Clean PBN crucibles are next loaded into the cells and are baked at temperatures ranging from 1300 to 1500 °C for 11 h. The growth charges consisting of 8N Ga (Alusuisse), 7N As (Johnson–Matthey) 6N Al (Cominco) and the dopant species Si and Be are loaded into the individual cells and the growth chamber is evacuated to ~10^{-9} Torr. The chamber is then externally baked at 220 °C for 5 days. After removal of the baking shrouds and subsequent cooling, the vacuum in the chamber is usually in the range $(2–4)\times 10^{-11}$ Torr. The source materials are then baked for 6–7 h at 100–200 °C above their normal operating temperatures. The exception is As, which is baked at 40 °C above the operating temperature for 4 h. We find that the steps outlined above are crucial for the growth of GaAs and Al$_x$Ga$_{1-x}$As with desirable electrical and optical characteristics. It has been reported[7] that Al$_x$Ga$_{1-x}$As and GaAs-Al$_x$Ga$_{1-x}$As quantum wells with superior luminescence characteristics can be grown at a substrate temperature ~700 °C using As$_4$ species. We chose to grow the entire multilayered structure at $T_{\text{sub}} = 630$ °C, read by an infrared pyrometer. Two types of SQW samples were grown. The first one consists of a 1-μm undoped GaAs buffer layer on which is grown a 20-period undoped superlattice consisting of 30-Å GaAs and 43-Å Al$_{0.3}$Ga$_{0.7}$As wells and barriers, respectively. This is followed by a 120-Å undoped GaAs layer and a superlattice barrier layer identical to the first one. In the second structure the superlattice barriers are replaced by ternary layers with $x = 0.3$ of approximately the same thickness. All the samples are grown on undoped (001) GaAs substrates.

High-resolution photoluminescence measurements were done in two systems. Measurements with the samples at 2 K were done by providing 6471-Å excitation with a Kr$^+$ laser. The luminescence was analyzed with a 1.26-m Spex spectrometer and was detected with a photomultiplier tube. The spectra were recorded after lock-in amplification. Measurements at higher temperatures were performed with 5145-Å excitation from a Ar$^+$ laser and a 1-m Jarell–Ash spectrometer. The excitation intensity was varied in the range 10^{-2}–10W/cm^2.

The photoluminescence spectrum of a SQW with superlattice barriers recorded with the sample at 4.2 K is shown in Fig. 1. The peak at 1.5443 eV with a linewidth (FWHM) of 0.85 meV arises from light-hole free exciton transitions X_{1l}. The intense peak at 1.5367 eV with a linewidth of 0.4 meV is ascribed to heavy-hole free exciton transitions X_{1h}. When the measurement was done at 2 K with the same sample and the luminescence was analyzed and recorded with a 4-m spectrometer and spectrographic plates, respectively, the linewidth of this transition is 0.3 meV. The peak at 1.5356 eV is due to bound excitons and is really composed of light-hole and heavy-hole donor-bound excitons (D^0X) and free-to-

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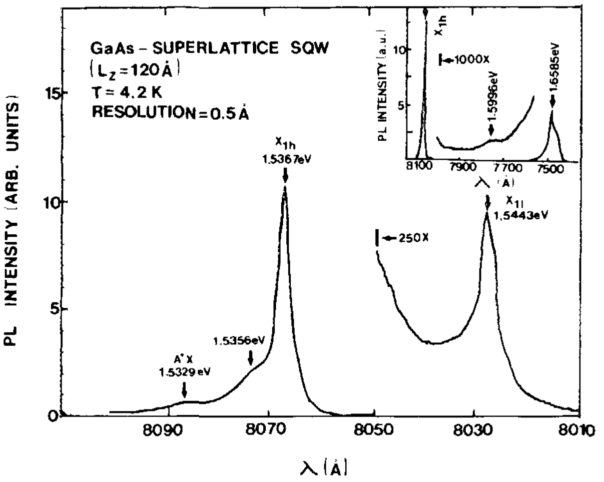

FIG. 1. Photoluminescence in 120-Å GaAs single quantum well with superlattice barriers. The superlattice is composed of 30-Å GaAs and 43-Å $Al_{0.3}Ga_{0.7}As$ wells and barriers, respectively. The main exciton related transitions are seen in the spectrum. The peak at 1.5356 eV is composed of light-hole and heavy-hole donor-bound excitons and D^0h transitions involving heavy holes. The inset shows luminescence from the superlattice and a weak transition at 1.5996 eV, believed to arise from X_{21h}.

bound (D^0h) transitions involving heavy holes. The weak peak at the lowest energy (1.5329 eV) is thought to be due to acceptor-bound (heavy-hole) excitons (A^0X). These assignments are made by comparison of the data with previously reported data on SQW and MQW structures[8-11] and from temperature and excitation-dependent spectra measured in the course of this study. The linewidths mentioned here are for our best set of samples, grown under conditions mentioned earlier. However, even under the same system preparation and growth conditions, the linewidths and peak intensities are strongly dependent on the quality of the source materials, and in particular, As. The linewidth of the 1.5367-eV transition varies from 0.4 to 2 meV.

The inset in Fig. 1 shows the 4 K luminescence of a 120-Å SQW sample at higher energies. The strong peak centered at 1.6585 eV is due to exciton transitions from the superlattice barrier. The weak but distinct peak at 1.5996 eV, separated by ~ 64 meV from the X_{1h} peak, appears to originate from a forbidden ($\Delta n \neq 0$) transition. Comparison with earlier data of Miller *et al.*[8,10] indicates that the transition involved may be X_{21h}, that is from E_{2e} to E_{1h}.

The changes in the PL spectra at 8 K due to variation of excitation intensity are depicted in Fig. 2. Several distinct features are noticeable. The energy position of the various peaks do not shift markedly. The intensity of the peak at 1.5443 eV increases at a larger rate than those of the peaks at 1.5368 and at 1.5356 eV. It is also observed that at the lowest excitation energy, there appears a shoulder to the 1.5356-eV peak at the high spectral energy side. This transition is thought to be due to light-hole D^0X. The excitonic nature of the relevant transitions are confirmed by the plots in Fig. 3, which show the dependence of integrated luminescence intensity on excitation power density and shows, perhaps for the first time, the excitation intensity dependence of light hole free exciton recombinations. The transitions show a linear to superlinear dependence. Additionally, the superlinear

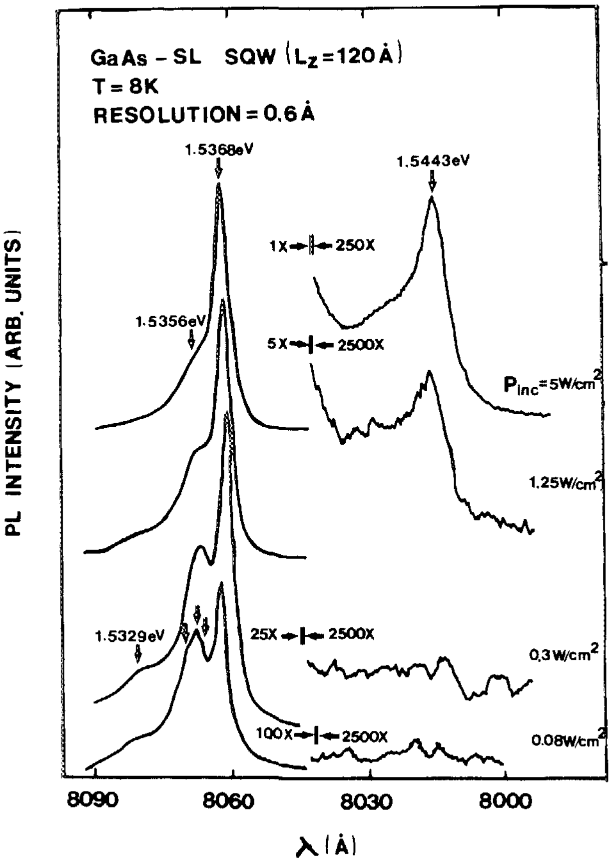

FIG. 2. Excitation dependent photoluminescence spectra in 120-Å GaAs SQW with superlattice barriers. Shoulders on the high and low-energy side of the donor-bound exciton peak appear at the lowest excitation energy, and are due to light-hole D^0X and heavy-hole D^0h transitions, respectively.

behavior is more pronounced for the transitions at 1.5367 and 1.5443 eV than for the transition at 1.5356 eV. It can therefore be concluded that the latter transition is due to bound excitons, while the first two arise from free-exciton recombinations.[2] It may be noted that the main peak is not

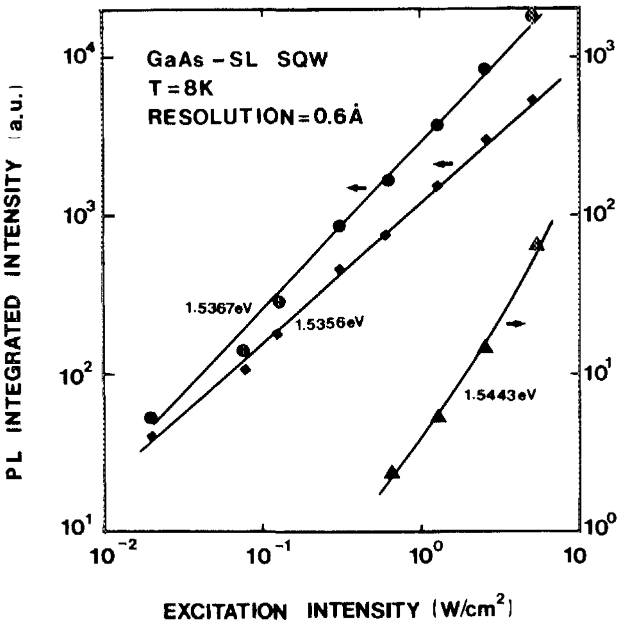

FIG. 3. Dependence of integrated photoluminscence intensity on excitation power for exciton-related transitions in 120-Å GaAs SQW with superlattice barriers.

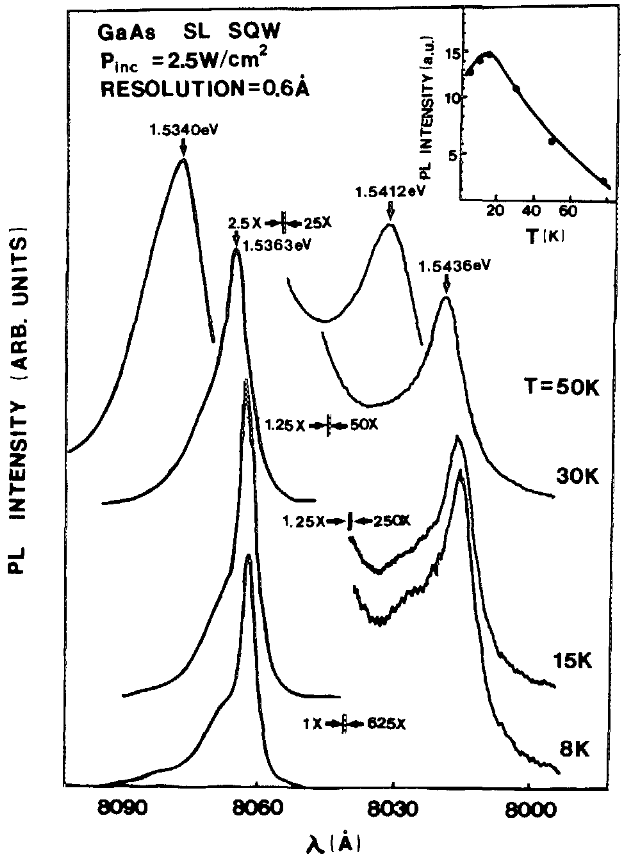

FIG. 4. Temperature dependence of photoluminescence in 120-Å GaAs SQW with superlattice barriers. The variation of the X_{1l} transition peak intensity with temperature is shown in the inset.

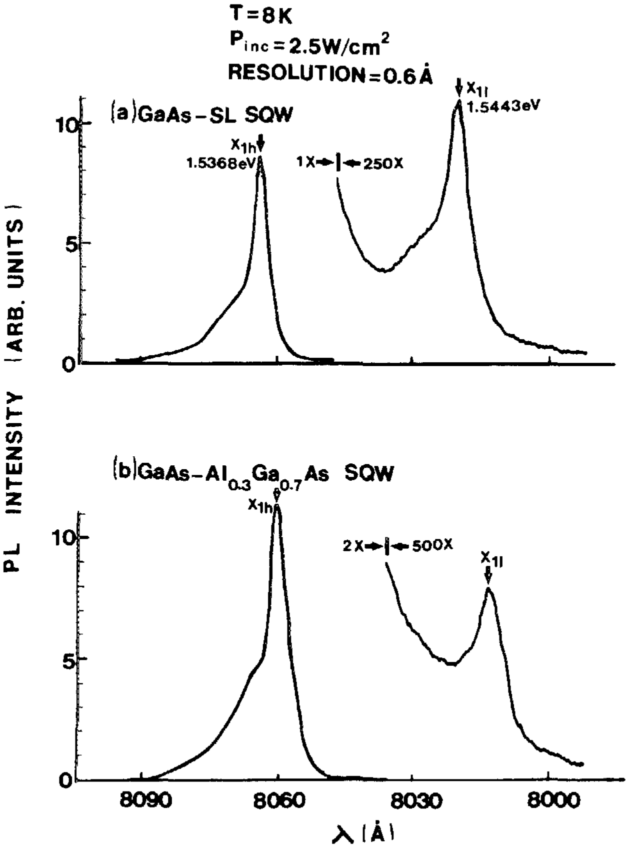

FIG. 5. Photoluminescence spectra of 120-Å GaAs SQW with (a) superlattice barriers and (b) $Al_{0.3}Ga_{0.7}As$ barriers. It is evident that the photoluminescence from the two samples are comparable.

split, even at the lowest excitation intensities. This confirms that biexcitons and bound states do not contribute to this transition. The energy separation $X_{1h} - X_{1l} = 8.4$ meV at 4.2 K is close to the value reported by Miller *et al.*[10] and by Masselink *et al.*[2] or that which can be obtained from a Kronig-Penney potential well-type model.

Temperature dependence of photoluminescence at constant excitation intensity is illustrated in Fig. 4. It is clear that the free-exciton transition at the highest energy becomes the dominant peak as temperature increases, confirming the recombination with light holes as its origin. In general, the recombination intensity of all the heavy-hole excitonic peaks decreases with increasing temperature, being at a more rapid rate for the bound excitons. In some cases the intensity of the heavy-hole free-exciton transition increases slightly at first and then decreases, as shown in the inset. This phenomenon has also been reported by Masselink *et al.*[2] and is attributed to carrier trapping and detrapping at the superlattice barriers.

Figure 5 shows the measured PL excitonic spectra at 8 K in a GaAs-$Al_{0.3}Ga_{0.7}As$ SQW with a well width of 120 Å. The linewidth of 0.9 meV in this sample and PL intensity lower than the SQW with superlattice barriers by only a factor of 2 indicate the superior quality of the interfaces. Singh and Bajaj[4] have recently calculated the effects of alloy disorder and interface roughness, caused by lateral fluctuations in the position of the barrier and island formation, on the linewidths of excitonic emission spectra. Comparison of our experimental data with their results show that linewidths ~0.3–0.8 eV measured at $T = 2$–8 K almost represent the theoretically calculated limit and are comparable to the best results published to date.[9,12] The important point is that the results were achieved for a 120-Å well, with the use of As_4 species, growth at low substrate temperatures, and without any substrate prebaking.

In conclusion, we have demonstrated that extensive baking of the MBE growth chamber, effusion cells, and effusing species are essential for the growth of GaAs and $Al_xGa_{1-x}As$ of desirable optical quality. Measured photoluminescence linewidths in 120-Å GaAs-$Al_{0.3}Ga_{0.7}As$ and GaAs- superlattice (barrier) SQW structures are among the narrowest ever reported. These characteristics could be obtained with growth at 630 °C and by using As_4 species.

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