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Free excitons in room-temperature photoluminescence of GaAs-$Al_xGa_{1-x}As$ multiple quantum wells

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We report a room-temperature study of the photoluminescence observed from a high-quality, molecular-beam-epitaxy grown GaAs-(AlGa)As multiple-quantum-well structure. The spectrum is dominated by free-exciton emission. This assignment is inferred by the use of photoluminescence excitation spectroscopy.

INTRODUCTION

The refinement of the semiconductor thin-film growth techniques of metal organic chemical vapor deposition (MOVCD)[1] and molecular-beam epitaxy (MBE)[2] has made it possible to grow complex structures with abrupt changes of doping level and composition leading to a new generation of optoelectronic devices utilizing quantum size effects.[3] The most widely studied materials system has been (AlGa)As, and in this paper we discuss the origin of the room-temperature radiative emission observed from a high-quality (AlGa)As multiple-quantum-well (MQW) structure grown by MBE.

Measurements of the absorption spectra of (AlGa)As-GaAs MQW's at 2 K emphasized the strong free-exciton character of the observed peaks.[4] The photoluminescence from high-quality, nominally undoped GaAs-(AlGa)As multiple-quantum wells has subsequently been identified as being intrinsic in its nature at temperatures up to about 50 K.[5] Photoluminescence excitation spectroscopy has indicated that the recombination process at these temperatures involves radiative decay of the ($n=1$ electron)–heavy-hole and the ($n=1$ electron)–light-hole free excitons. There are only a few reports of room-temperature studies of the radiative transitions in quantum-well structures which we compare with our results below.[6-8]

EXPERIMENTAL DETAILS

The sample used in this present study was grown by MBE in a noncommercial, two-chamber, oil diffusion pumped system equipped with full computer control of effusion cell temperatures and beam shutter operations. The epitaxial layers were deposited on a semi-insulating, (100) oriented GaAs substrate. The growth sequence was as follows: (a) a 0.5-μm GaAs buffer layer deposited at a substrate temperature of 620 °C, (b) 1400 Å of $Al_{0.33}Ga_{0.67}As$, (c) 60 periods of alternate wells of 55-Å GaAs and 170-Å barriers of $Al_{0.33}Ga_{0.67}As$, and (d) 1400 Å of $Al_{0.33}Ga_{0.67}As$. Layers (b)–(d) were not intentionally doped and deposited at a substrate temperature of 700 °C as measured with an optical pyrometer. Supplementary measurements on single, thick layers of GaAs, also not intentionally doped, show a residual p-type carrier density of approximately 10^{16} cm^{-3}.

The same optical system was used for recording the photoluminescence and the excitation spectra. The excitation source was made from a 150W Philips 7158 quartz halogen lamp whose output was collected and focused on the input slits of a Spex Minimate grating spectrometer whose bandpass at full width half maximum (FWHM) was 40 Å. The light output from the Minimate was directed and focused onto the sample being studied at an angle to its surface of about 40°. The spot size on the sample was 1×4 mm which gave an excitation density of 8×10^{-8} W/cm^2 at a wavelength of 6000 Å. The photoluminescence was collected in a direction normal to the sample surface and focused onto the slits of a computer-controlled, 1-m Jobin-Yvon HR1000 spectrometer whose resolution was 24 Å. The photoluminescence was detected by a cooled (-40 °C) GaAs photomultiplier (RCA C31034) used in conjunction with a Brookdeal 5C1 photon counting system. The Minimate wavelength scale was calibrated using the Jobin-Yvon spectrometer after recording the excitation spectrum.

RESULTS AND DISCUSSION

The photoluminescence and excitation spectra from the sample are shown in Figs. 1 and 2, respectively. The same sample was used for photoluminescence and excitation spectroscopy, thereby eliminating problems with strain encountered in preparation of samples for absorption measurements.[5] The spectra have not been corrected for the system response. The photoluminescence spectrum shows the double-peak structure which has been seen before at room temperature.[6] It is dominated by a peak at 1.513 eV with a smaller peak on the high-energy side. The best estimate of the energy of the smaller peak is 1.532 eV. In the excitation spectrum of Fig. 2, curve (a), the luminescence is being observed in the low-energy tail at 1.499 eV. The two peaks seen at 1.512 and 1.534 eV in Fig. 2, curve (a), can be assigned to the ($n=1$ electron)–($n=1$ heavy-hole) and the ($n=1$ electron)–($n=1$ light-hole) free excitons.[4] The signal-to-noise ratio is improved by detecting the luminescence at the energy of one of the peaks. Excitation spectra obtained in this way are shown in Figs. 2(b) and 2(c). Because of the closeness of the positions of the two peaks in luminescence to those in excitation we can conclude that the luminescence peaks are due to the free excitons. This is unusual; free excitons are not normally seen in the room-temperature photoluminescence of bulk semiconductor crys-

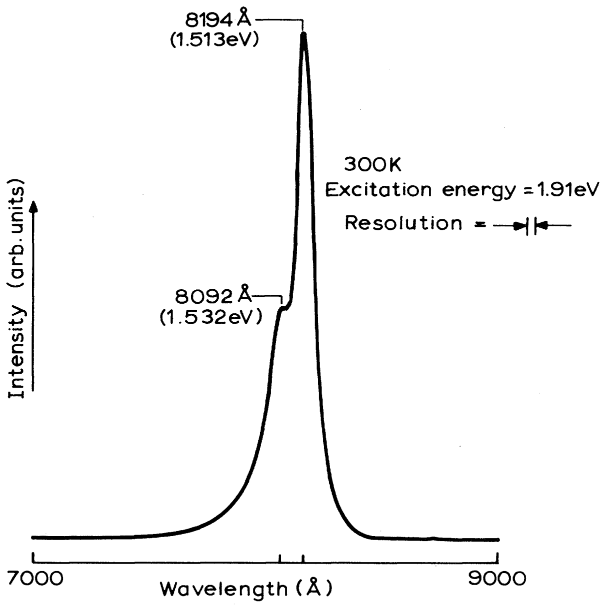

FIG. 1. Room-temperature photoluminescence of the sample described in the text.

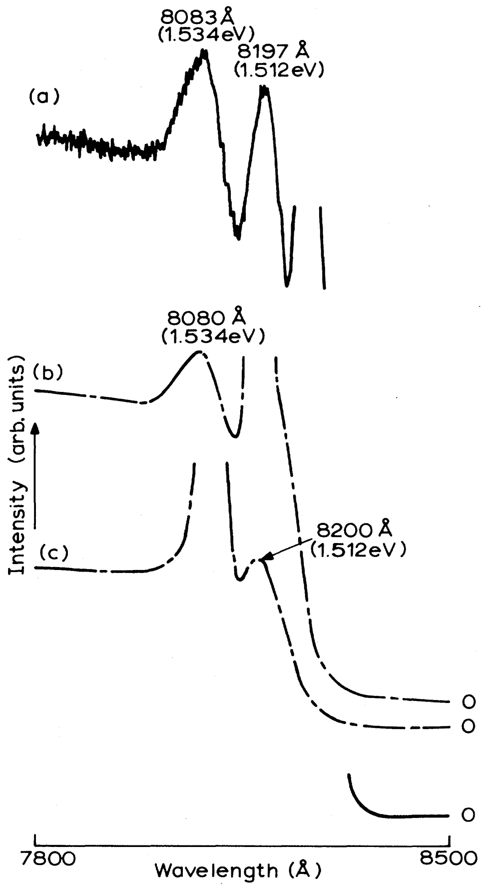

FIG. 2. Room-temperature photoluminescence excitation spectra of the same sample as used for Fig. 1. The detection wavelength was (a) 8272, (b) 8197, and (c) 8083 Å.

tals. They might be expected to show up in quantum wells, however, because of their prominence in the absorption spectrum. In the high-energy tail of the spectrum, the luminescence decreases by a factor of 2 for an increase in energy of about 220 meV. This is just what is expected from exciton theory for free-carrier recombination just above the band edge[9] so that we can conclude that the high-energy tail is due to the free carriers.

The three curves in Fig. 2 show that the excitation spectrum is independent of the energy at which the luminescence is detected. This indicates that the excitons become thermalized with the free carriers in a time much shorter than the free-carrier recombination and the exciton annihilation times. In this situation the excitation spectrum should mimic the absorption spectrum rather closely. The room-temperature absorption spectrum obtained by Miller *et al.*[10] shows the heavy-hole exciton peak to be much higher than that of the light-hole exciton, whereas we find their heights to be similar. We think, however, that there is not necessarily a discrepancy here. The carriers in this system can be expected to exhibit a strong two-dimensional character. The absorption spectrum due to excitation across a gap from a simple valence band to a simple conduction band in such a system is expected to consist of a peak due to exciton creation in the 1*s* state followed by an absorption nearly constant with energy due to free-carrier absorption with possibly some structure due to the excited exciton states in between. In a quantum well we get the superposition of two spectra of the type described above because of the splitting of the valence band. If the hole were truly split into light and heavy holes, the ratio of the component involving the heavy hole to that involving the light hole would be 3:1. The absorption due to free-carrier creation can be expected to have the same magnitude in crystals with sharper exciton transitions as in those with broader exciton transitions. In crystals with broad transitions, the weaker light-hole exciton peak, which is superimposed on a background of heavy-hole free-carrier creation, will have a height similar to that of the heavy-hole peak, which does not. In crystals with sharper transitions the heavy-hole exciton peak will be much higher than the light-hole exciton peak. Although the quantum well will mix the heavy and light holes, we expect some effect to be seen. Absorption spectra reported by Dingle[4] show this effect.

We are now able to attempt interpretations of what previous authors have seen in room-temperature photoluminescence. The double-peak structure seen by Laidig, Lee, Wortman, and Littlejohn[6] in their samples grown at their lower temperatures is the same as we observe and must represent decay of the light- and heavy-hole excitons with the high-energy tail coming from free-carrier recombination. Kawabe, Kondo, Matsuura, and Yamamoto[7] obtain only a single broad peak at room temperature. Using their quoted aluminium concentrations and layer widths in a Kronig-Penny calculation, we find that the light- and heavy-hole bands are split by only about 4 meV. The light- and heavy-hole excitons should exhibit a similar splitting. We find also that their room-temperature peak position is 22 meV higher than that calculated. With so small a splitting the occupation of the light-hole exciton states should be approximately half that of the heavy-hole excitons at 77 K. The relative populations of the light- and heavy-hole excitons and free carriers do not change a great deal between 77 and 300 K so that it is difficult to understand why the luminescence linewidth increases from 18 to 47 meV between these temperatures. One explanation is that the well width is smaller than the authors think. The exciton peak would then appear at a higher energy and the splitting between the light and heavy holes would be larger. The large increase in linewidth between 77 and 300 K would then be due to the change in population of the light-hole exciton relative to the population of the heavy-hole exciton.

CONCLUSION

In conclusion, we have shown that the room-temperature photoluminescence from a high-quality, MBE grown GaAs-(AlGa)As MQW is dominated by free-exciton recombination. This assignment has been inferred by room-temperature photoluminescence excitation spectroscopy on the same sample.

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