Direct evidence of the indirect energy gap in InAlAs/AlAsSb multiple quantum wells by time-resolved photoluminescence

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Time-resolved photoluminescence spectroscopy has been applied to determine the nature of the energy gap of InAlAs/AlAsSb multiple quantum well structures. While the InAlAs buffer layer exhibits a decay time of the order of 1 ns, which is typical for direct gap semiconductors, the decay time of the InAlAs/AlAsSb multiple quantum well structures is prolonged by more than two orders of magnitude. This observation is direct evidence for the presence of an indirect energy gap. The decay time increases with increasing InAlAs layer thickness indicating the decreasing overlap of electron and hole wave functions. © *1998 American Institute of Physics.* [S0003-6951(98)00213-7]

Type II staggered quantum well structures exhibit unique optical properties due to confinement of electrons and holes in the different layers. In these structures, tunneling-assisted electron-hole recombination occurs across the type II heterointerface, resulting in light emission below the energy gaps of the constituent materials. The respective optical transition is indirect in real space, but direct in reciprocal space. InAlAs/InP quantum well structures lattice matched to InP substrates represent the most widely studied type II material system.[1–5] Electric-field induced excitons and a new type of optical bistability were observed for InAlAs/InP type II multiple quantum well (MQW) diodes.[6,7] Recently, the enhancement of the recombination lifetime has been reported for several type II single quantum well systems, e.g., for GaP/AlP/GaP[8] and tensile strained InGaAs.[9]

$In_{0.52}Al_{0.48}As/AlAs_{0.56}Sb_{0.44}$ quantum well structures lattice matched to InP substrates are also expected to exhibit a type II staggered band configuration. In this material system, electrons are confined to the InAlAs layers, while the holes are localized in the AlAsSb layers as indicated in Fig. 1. Recently, we reported the successful growth of high-quality $In_{0.52}Al_{0.48}As/AlAs_{0.56}Sb_{0.44}$ type II MQW layers by molecular beam epitaxy (MBE).[10] Light emission at about 1.28 eV, which is below the energy gaps of InAlAs and AlAsSb, was observed and identified to occur near the InAlAs/AlAsSb heterointerfaces. However, the type II character of the emission was only inferred from the spectral position of the emission lines. The optical properties of type II emission are expected to differ considerably from that of bulk material and/or type I quantum well structures. In particular, the type II emission should exhibit a prolonged electron/hole recombination lifetime in comparison with type I structures, because the overlap of the electron and hole wave functions is strongly reduced.

In this letter, we present direct evidence of the type II character of the InAlAs/AlAsSb MQW obtained from time-resolved photoluminescence (PL) spectroscopy. The radiative lifetime was strongly increased in the type II MQW compared to the InAlAs bulk-like buffer layer. Furthermore, the lifetime increases with increasing InAlAs layer thickness.

The InAlAs/AlAsSb MQW structures consist of ten periods grown by MBE on Fe-doped (100) InP substrates. A 0.3 μm thick, undoped InAlAs buffer layer was incorporated between the substrate and the MQW structure. In and Al metals were used to generate the group III beams, while the tetramers As_4 and Sb_4 were emitted by the group V beam sources. The group III beam and the Sb beam were supplied using conventional effusion cells (K cells), while the As beam was supplied using a valved As cell. The substrate temperature during the growth was 505 °C, which was monitored by a calibrated infrared pyrometer. Prior to the growth, the InP substrate surface was thermally cleaned at 515 °C for 1 min under an arsenic vapor pressure of 1×10^{-3} Pa. The

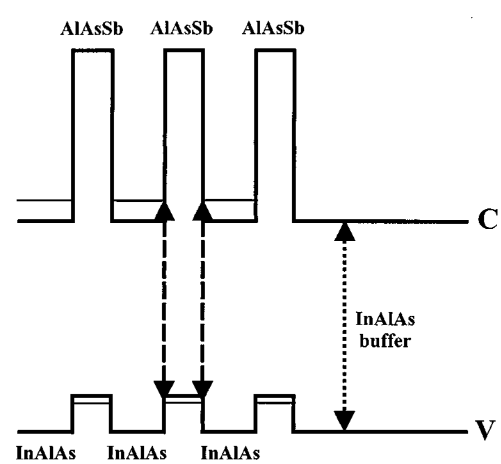

FIG. 1. Schematic diagram of the type II band alignment in InAlAs/AlAsSb multiple quantum well structures. The letters C and V indicate the edge of the conduction and valence band, respectively. The vertical arrows indicate the spatially indirect recombination at the InAlAs/AlAsSb heterointerfaces and the direct recombination in the InAlAs buffer layer.

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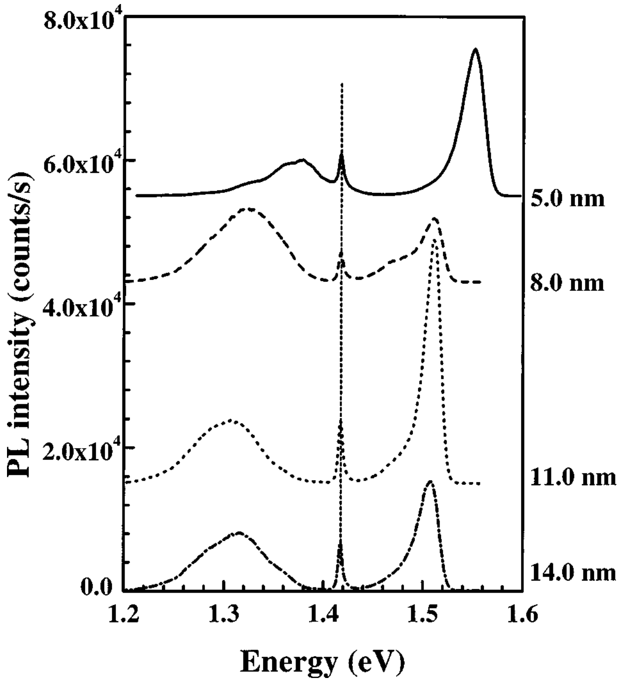

FIG. 2. Steady-state photoluminescence spectra of all four InAlAs/AlAsSb samples for a temperature between 7 and 10 K, excitation energy of 1.676 eV, and laser power of 50 μW. The different samples are labeled by their respective InAlAs layer thickness. The vertical line marks the position of the InP:Fe substrate peak. The spectra for the 11.0, 8.0, and 5.0 nm samples have been vertically shifted by 1.5×10^4, 4.3×10^4, and 5.5×10^4 counts/s, respectively.

growth rate of the InAlAs and the AlAsSb layer was 1.4 and 0.67 μm/h, respectively. Four samples with different InAlAs layer thickness of 5.0, 8.0, 11.0, and 14.0 nm were investigated. The AlAsSb layer thickness was held constant at 5.0 nm. Further details of the growth conditions are described in Ref. 10.

For the cw and time-resolved PL experiments, the samples were mounted on the cold finger of a He-flow cryostat with the temperature adjusted between 5 and 10 K. In the cw measurements, the optical excitation was achieved with a tunable Ti:sapphire laser pumped by an Ar^+ laser. The PL signal was dispersed in a 1 m monochromator and detected with a cooled charge coupled device (CCD) detector. The laser power was adjusted to 50 μW with the beam focused to a diameter of about 50 μm. The time-resolved PL measurements were performed using a streak-camera system (Hamamatsu) in synchro-scan or single-shot operation in conjunction with a mode-locked Ti:sapphire laser with 150 fs laser pulses, a repetition rate of 76 MHz in the synchro-scan modus and down to several 10 kHz in the single-shot modus, and a photon energy of 1.676 eV. The luminescence is dispersed by a 22 cm monochromator with a 300 lines/mm grating and focused onto the photocathode of the streak tube (S25). The streak images are recorded by a cooled CCD array. The spectral and synchro-scan mode minimum temporal resolution amount to about 0.8 nm and 2 ps, respectively.

Figure 2 shows the cw-PL spectra for all four InAlAs/AlAsSb MQW structures recorded between 7 and 10 K. Three distinct peaks can be clearly identified in all samples. The peak at 1.507 eV is identified as the emission peak of the InAlAs buffer layer (cf. Fig. 1). For the 5.0 nm sample, the InAlAs buffer layer peak is shifted towards higher energies, which is probably due to an Al-rich composition of this layer for this particular sample. The narrow peak at 1.418 eV, which is located at exactly the same energy for all four samples as shown by the vertical dashed line, originates from the InP:Fe substrate. The peak lowest in energy between 1.30 and 1.38 eV is assigned to the below band-gap emission of the type II InAlAs/AlAsSb MQW layers (cf. Fig. 1). It is well known that the type II emission exhibits a rather large linewidth compared to the one of the InAlAs buffer layer. Such spectral broadening has also been observed in the emission of type II InAlAs/InP MQW layers,[4] which is probably due to the interface roughness at the type II heterointerfaces. The similar broadening of the MQW PL signal in all samples may indicate the importance of interface roughness in the recombination process. Note that the peak for the 14.0 nm sample is slightly shifted to higher energies compared to the 11.0 nm sample, which may be due to a small increase of the As composition in the AlAsSb layer. For the other three samples, there is a clear blueshift of the emission energy with decreasing InAlAs layer thickness. The cw-PL spectra did not change significantly when the excitation energy was increased to 1.771 eV. Using a He-Ne laser at 1.959 eV, the PL spectra changed in terms of the intensity distribution between the different peaks due to the reduced penetration depth of the excitation, i.e., the intensity from the MQW structure increased with respect to the buffer layer and the substrate peak. In all PL spectra (cw and time-resolved), carriers are only excited in the InAlAs layer (energy gap near 1.5 eV) and not in the AlAsSb layer (energy gap near 2.4 eV) of the MQW structure. This is in contrast to the previously published PL spectra at 77 K, where the green line of an Ar^+ laser (2.41 eV) was used for excitation. Due to the shorter absorption length at the respective photon energy, the buffer layer and substrate peak were not observed.[10]

The PL-intensity transients of the InAlAs buffer layer and of the 5.0 nm InAlAs/AlAsSb MQW structure recorded at 6 K are shown in Figs. 3(a) and 3(b), respectively. The buffer layer PL transient was recorded at 1.55 eV, the MQW-PL transient at 1.38 eV. The buffer layer exhibits a mono-exponential decay with a time constant of 1.7 ns as obtained by a linear fit to the transient on a semilogarithmic scale indicated by the solid line in Fig. 3(a). The InP:Fe-substrate peaks decay even faster for all four samples with a time constant in the range of 200–300 ps. The decay times for both peaks are typical for recombination across a direct energy gap. The faster recombination of the substrate peak is due to the larger doping density. The decay of the MQW structure exhibits a bi-exponential decay on a much longer time scale than the buffer layer peak. The time constant obtained from a linear fit as shown by the two solid lines in Fig. 3(b) are 0.19 and 1.2 μs. The initial decay time of the MQW structure increases by more than two orders of magnitude in comparison to the buffer layer. This observation is direct evidence for the type II nature of the band alignment and the resulting indirect energy gap, since the transition probability of a type II transition is reduced by about two orders of magnitude in comparison to a type I transition.[11] The bi-exponential decay of the MQW structure could be due to the

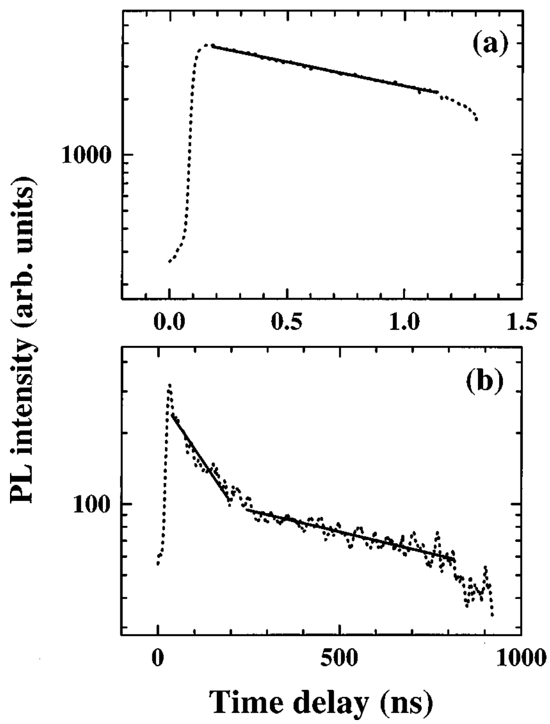

FIG. 3. Photoluminescence transients of the InAlAs buffer layer (a) and the MQW structure with 5.0 nm InAlAs (b) for a temperature of 6 K detected at 1.55 and 1.38 eV, respectively. Note the very different time scales in (a) and (b). The solid lines indicate linear fits corresponding to exponential decay times with values of 0.19 and 1.2 μs.

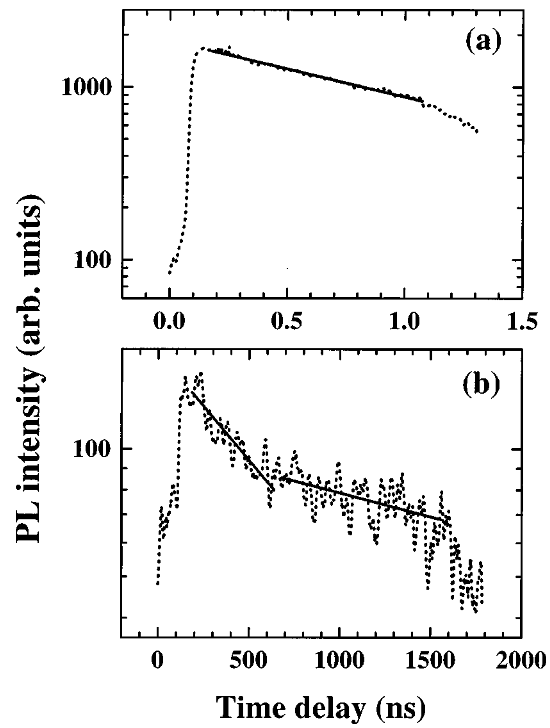

FIG. 4. Photoluminescence transients of the InAlAs buffer layer (a) and the MQW structure with 8.0 nm InAlAs (b) for a temperature of 6 K detected at 1.51 and 1.32 eV, respectively. Note the very different time scales in (a) and (b). The solid lines indicate linear fits corresponding to exponential decay times with values of 0.86 and 3.9 μs.

presence of two different recombination channels or relaxation mechanisms.

For the 8.0 nm MQW structure, the decay of the buffer layer PL recorded at 1.51 eV is comparable to the one in the 5.0 nm sample and monoexponential with a time constant of 1.4 ns as shown in Fig. 4(a). The PL transient of the MQW structure recorded at 1.32 eV shows again a bi-exponential behavior as indicated in Fig. 4(b). However, the initial time constant is significantly increased to a value of 0.86 μs. The decay time at longer times is also increased to 3.9 μs. Since with increasing InAlAs layer thickness the spatial overlap between the electron and hole wave functions is reduced, the increased decay time in the wider sample gives additional evidence for the spatially indirect recombination process. For the 11.0 nm sample, the PL transient shows essentially a constant signal within the time range of 2 μs. The PL transient of the 14.0 nm sample was smaller than the detection limit of our streak-camera system, although the time-integrated spectrum could be observed in the so-called focus mode of the streak camera. However, as mentioned above, an increasing layer thickness should result in a further increase of the time constant. Since all samples show a cw-PL signal of the same order of magnitude, we can only conclude that the PL signal in the time domain is spread out over an even longer time scale so that eventually the time-resolved PL signal is reduced below the detection limit. The buffer layer and substrate PL transients, however, could be observed in all four samples so that the excitation intensity was sufficiently high.

In conclusion, we have investigated InAlAs/AlAsSb MQW structures with different InAlAs layer thickness by time-resolved PL spectroscopy. The PL decay time of the MQW structure is prolonged by more than two orders of magnitude compared to the PL decay time of the InAlAs buffer layer. The latter is typical for a direct-gap semiconductor. Furthermore, the decay time increases with increasing InAlAs layer thickness indicating the reduction in the spatial overlap of electron and hole wave functions. The prolonged decay time and the InAlAs layer thickness dependence of the time constants give direct evidence for the type II nature of the band alignment in the InAlAs/AlAsSb MQW structure resulting in spatially indirect recombination.

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