Spatial quantization in GaAs–AlGaAs multiple quantum dots

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We present results of the fabrication and investigation of totally spatially localized crystalline structures. Low temperature photoluminescence exhibits structure that is best explained by a bottleneck for hole energy loss. This bottleneck is believed to be a direct consequence of the modification of the band structure by the fabrication-imposed potential and is believed to be the first evidence for total spatial quantization in a fabricated heterojunction system.

The physics of spatially quantized systems has been the subject of intense investigation since molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD) have made possible the fabrication of atomically sharp heterojunction interfaces. Quantum wells have been exhaustively examined since the seminal works of Dingle *et al.*[1] More recently, studies on quantum wires[2] have yielded interesting new properties. Here we present data on a completely spatially quantized system (which by extrapolation we define as "quantum dots") where the carriers have zero degrees of freedom (denoted as "0 DOF"). This paper discloses evidence for the modification of carrier–phonon scattering rates by the imposition of complete spatial quantization on GaAs–AlGaAs multiple quantum wells. The photoluminescence spectrum of the 0 DOF structures exhibits striking structure in the normal intrinsic exciton luminescence of the confined quantum well states. This photoluminescence structure does not occur for structures of higher dimensionality, and is best explained by a bottleneck for electron/hole energy loss. We believe that this bottleneck is a direct consequence of the quantization of the electronic and/or phonon dispersion relations.

The samples used in these experiments were grown by MBE and are shown schematically in Fig. 1(a). The samples were grown on (100) Cr-doped GaAs substrates and consisted of a 0.1 μ GaAs MBE buffer layer followed by a 1 μ $Al_xGa_{1-x}As$ buffer, twenty 20 Å GaAs wells with 100 Å $Al_xGa_{1-x}As(x = 0.3)$ barriers, followed by a 100 Å GaAs cap layer. All samples were nominally undoped. Patterning of the bulk multiple quantum well samples was done by direct e-beam writing in a film of polymethylmethacrylate (PMMA) on the sample surface. Conventional lift-off techniques were used to define 1000 Å thick Au metal patterns on the sample surface. The metal mask (which exhibited little degradation during the fabrication) served as both an etch mask and a semitransparent film. The metal patterns were transferred to the underlying sample by a BCl_3 reactive ion etch that extended into the AlGaAs buffer. The lateral dimension for both the 1 DOF and 0 DOF structures was 0.25 μ. Figure 1(b) shows a schematic of a single quantum dot structure. Quantum wire (1 degree of freedom, or 1 DOF) structures were fabricated simultaneously on the same sample as the quantum dot (0 DOF) structures. Arrays of these structures were fabricated to achieve measurable photoluminescence intensity and the stack of quantum wells was used for sufficient absorption of the excitation radiation.

Photoluminescence measurements were performed (at 45° incidence and detection) in a helium flow Janis optical cryostat. The sample was excited by the 2.54 eV line of a focused Ar^+ laser and the photoluminescence radiation was collected by a 1.0 m Chromatix spectrometer of 7 Å/mm focal plane dispersion. Conventional detection techniques were used.

Figure 2 shows the photoluminescence spectra of sample A for the three distinct DOF cases previously discussed. The 7300 Å luminescence peak is the $(n = 1)$ electron–$(n = 1)$ heavy hole recombination radiation, prominent in all three DOF cases. Note that the quantum well size is adjusted so that the luminescence under investigation is spaced between the GaAs and AlGaAs peaks to eliminate any possibility of impurity effects from the bulk substrates. We have normalized the three different DOF spectra to the same intensity at

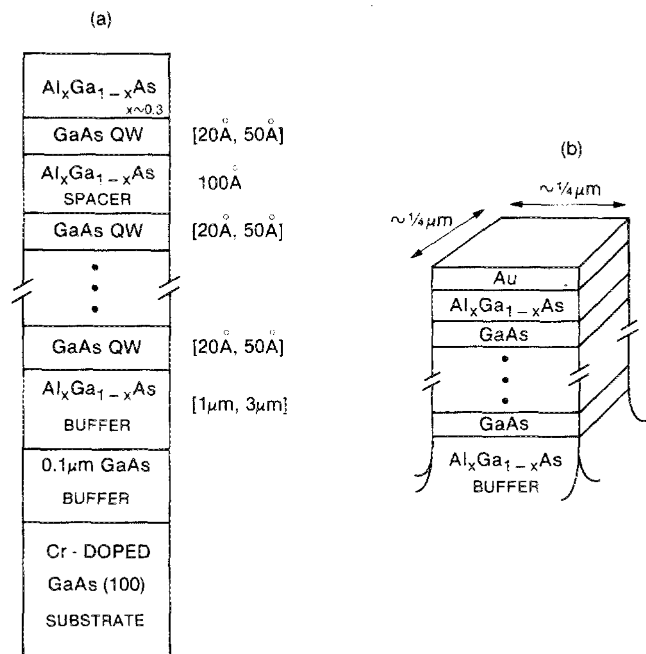

FIG. 1. (a) Schematic cross sectional view of the GaAs quantum well MBE samples. Discussion in the text is on the 20 Å quantum well samples. (b) Schematic cross sectional view of a quantum dot (0 DOF) structure.

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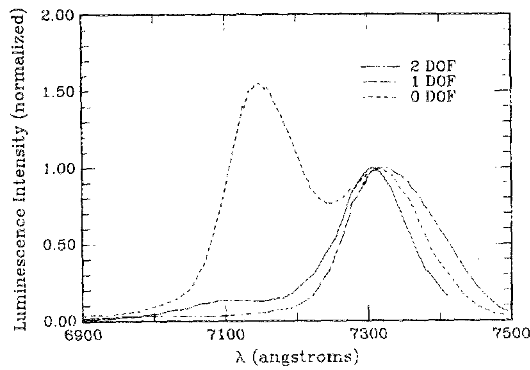

FIG. 2. Photoluminescence spectra of GaAs–AlGaAs quantum wells (2 DOF), quantum wires (1 DOF), and quantum dots (0 DOF) at $T = 4.2$ K. The photoluminescence intensities are normalized for comparison purposes.

this wavelength for comparison purposes. The small shifts in the 7300 Å peak positions are probably due to residual surface damage or strain resulting from the plasma etch. The loss in photoluminescence signal was readily accounted for by loss of sample volume, indicating that nonradiative loss mechanisms due to sidewall damage are not large.

The outstanding structure at ~ 7100 Å in the 0 DOF spectra, also seen at smaller relative intensity in the 2 DOF case, is the ($n = 1$) electron–($n = 1$) light hole recombination radiation. The absence of this structure in the 1 DOF case is probably due to the singularity in the density of states for a 1 DOF structure which depletes holes of higher (than zone minima) energy. The appearance of the light hole peak in the quantum dot case was not a local effect; all sections of the sample investigated exhibited the same spectrum. The 1 DOF and 0 DOF structures were fabricated adjacent to each other on the sample to eliminate any systematic errors due to the plasma etch.

Figure 3 schematically diagrams the relaxation mechanisms in the luminescence process of the ground state of a quantum well. $\tau_{\rm hh,intra}$, $\tau_{\rm lh,intra}$, and $\tau_{\rm e,intra}$ are the intrabranch phonon relaxation times of heavy holes, light holes, and electrons, respectively, down their respective branches. $\tau_{\rm inter}$ is the light-to-heavy hole scattering time. $\tau_{\rm recomb}$ is the electron–hole radiative recombination time. We shall assume that the heavy and light hole–electron recombination times are approximately equal and energy independent. Relaxation mechanisms involving any existing excited states, the split-off band, or interface defects, have been ignored. Carrier–carrier scattering has also been neglected in this treatment, but will be approximately constant in the three different dimension cases. This would not be true for higher excitation intensities.

Upon initial excitation, the three DOF cases have similar thermalization dynamics. The crossover from essentially three-dimensional behavior to the specific DOF case is complex and will not be treated here. When the carriers reach the region $E \sim \hbar\omega_{\rm LO}$, the crossing from the light hole to heavy hole branch can occur.[3] The interbranch scattering time, $\tau_{\rm inter}$, will be limited by the phase space available at the crossover. If $\tau_{\rm inter}$ is sufficiently large, a "bottleneck" results and an appreciable population of light holes can be formed.

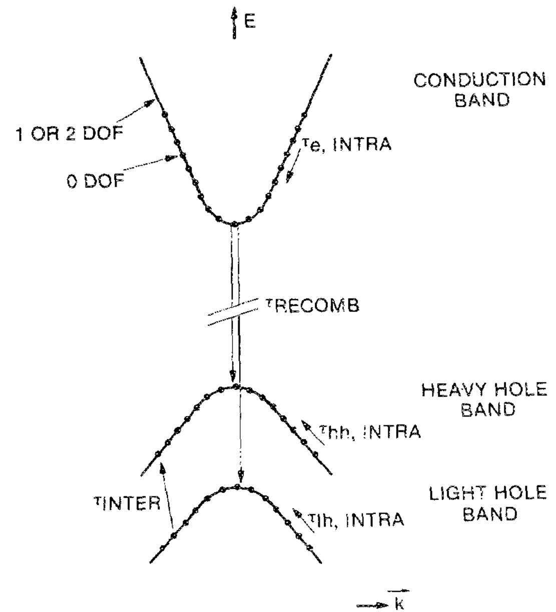

FIG. 3. Electronic dispersion relation of the $n = 1$ ground state for quantum wells and wires (solid lines) and for quantum dots (points). The notation is defined in the text.

Thus the formation of discrete points in the dispersion relation due to the 0 DOF boundary conditions increases $\tau_{\rm inter}$ sufficiently to make the light hole peak observable. Additionally, the elimination of selected small k modes in the phonon spectrum will enhance the light hole transient population.

Observation of this bottleneck in light-to-heavy hole scattering by photoluminescence in bulk GaAs is impractical since $\tau_{\rm recomb}/\tau_{\rm inter} \sim 10^3$. We have reduced this rate not only by the fabrication-imposed increase of $\tau_{\rm inter}$ but also by the decrease of $\tau_{\rm recomb}$ due to the enhancement of the radiative transition probability in a quantum well with sufficiently narrow well thickness.[4] Band filling can be eliminated as an alternative explanation since the linewidths are approximately the same, do not exhibit high energy tails, and the excitation power dependence exhibited no change in the heavy hole peak : light hole peak ratio. Bound impurity effects can be eliminated by the 1 DOF control, while nonradiative recombination can be eliminated as a cause by the normalized luminescence intensity.

An intriguing possibility is the direct observation of spatial quantization from the discrete (multiply degenerate) electronic levels in the 0 DOF quantum dots. The spectrum was investigated in sufficiently high resolution to observe fine structure in the quantum well luminescence with the sample immersed in superfluid helium. The negative results for this search are best explained by an effect analogous to inhomogeneous line broadening; the variation of the dot size across the array will produce fluctuations in the discrete line positions. For the experimental resolution of 0.1 Å and the above dot parameters, a fluctuation of $\sim 1\%$ is sufficient to mask the discrete lines.

A possible solution to the inhomogeneous line broadening problem is to observe the photoluminescence from a single

quantum dot. Such a structure was fabricated and investigated, but the attempt was unsuccessful due to insufficient photoluminescence intensity.

In summary, we have observed the photoluminescence from a series of spatially quantized quantum structures and believe we have observed the first indirect evidence for total spatial quantization by a fabrication-imposed potential. The effect of the discrete dispersion relations of the electrons and/or phonons is to modify the relaxation kinetics of carriers in the quantum structures, and this is observable in the photoluminescence spectrum.

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