Photoluminescence of GaAs quantum wells grown by molecular beam epitaxy with growth interruptions

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Single GaAs/$Al_xGa_{1-x}As$ quantum wells, grown by molecular beam epitaxy with growth interruptions at each interface, are investigated using low-temperature photoluminescence. The three clearly resolved photoluminescence peaks are attributed to discrete monolayer thicknesses of the well. The splitting of the peaks is investigated for several hundred points across a 2 in. wafer. The negligible variation of the peak splitting is consistent with abrupt interfaces in the growth direction, atomically smooth interfaces, and discrete thicknesses of the quantum well with changes of only integer multiples of monolayers.

The correlation of the microscopic interfacial structure of the GaAs/$Al_xGa_{1-x}As$ interface with its optical properties is an intriguing question in the characterization of growth-interrupted quantum wells.[1-5] Four simple models of the two interfaces of a quantum well are shown in Fig. 1, along with the emission spectrum expected for each model. Together these four represent the principal classes of interface pairs. An interface configuration which contains steps of monolayer height is shown in Fig. 1(a).[6] The distance between the steps is assumed to be smaller than the excitonic radius ($r_{\mathrm{exc}}^{3\mathrm{D}} \cong 120$ Å). An inhomogeneously broadened luminescence spectrum results, with a linewidth that can be estimated from the thickness variation of the quantum well.[7] The three different interfacial structures shown in Figs. 1(b), 1(c), and 1(d) result in sharp luminescence lines. A quantum well with extended monolayer-flat regions in both interfaces is shown in Fig. 1(b). The spatial extent of the flat regions is assumed to be larger than the excitonic diameter. Several sharp luminescence lines are expected to result from such an interface configuration, with an energy corresponding to a quantum well whose thickness is an integer multiple of GaAs monolayers. However, multiple sharp luminescence lines can be also expected from certain interface configurations containing microroughness ($\ll r_{\mathrm{exc}}^{3\mathrm{D}}$)as illustrated in Figs. 1(c) and 1(d). If a sample contains uniform microroughness in one interface, one smooth interface can yield thickness changes of exactly one monolayer without integral monolayer thickness [Fig. 1(c)]. Discrete changes in thickness of two consistently microrough interfaces would result in discrete luminescence lines, but no particular separation would be expected [Fig. 1(d)].

Unambiguous definitions of interface terms are required for clarity in subsequent discussions. We define the interface between two materials *A* and *B* as *abrupt* if after termination of the epitaxial growth of *A* the newly deposited material *B* does not perturb the former surface atomic configuration of material *A*. It follows that the former surface of *A* and the interface are identical and the transition between *A* and *B* occurs in one atomic layer on any atomic column normal to the interface. The interface between *A* and *B* is *atomically smooth* on a length scale *X* if the interface is abrupt and if the surface of *A* has islands without steps of lateral extent *X*. If material *A* or *B* is an alloy subset of the other, such as $Al_xGa_{1-x}As$/GaAs, the definitions above are still valid. Note that the microroughness illustrated in Figs. 1(c) and 1(d) do not reflect the random fluctuations of the alloy composition[8] found in the ternary $Al_xGa_{1-x}As$. Instead, the barrier alloy is considered in the virtual crystal approximation which allows one to define a clear boundary between the well and the barrier material. We will refer to the models illustrated in Figs. 1(b), 1(c), and 1(d) as the "island model" and the "microroughness" models, respectively.

Although both the island model [Fig. 1(b)] and the microroughness models [Figs. 1(c) and 1(d)] result in sharp luminescence lines, a striking difference is expected from the three interface configurations. The separation between the luminescence lines corresponds to a thickness

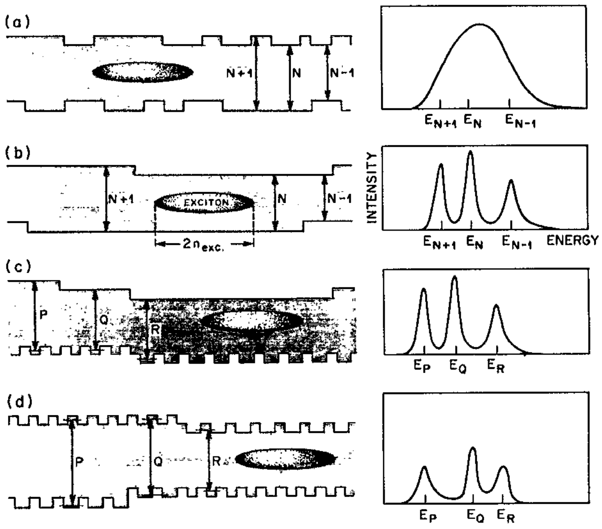

FIG. 1. Schematic illustration of three interface configurations of a quantum well. (a) The rough interface configuration results in broad photoluminescence lines. (b) An interface consisting of large ($\gg r_{\mathrm{exciton}}$) monolayer-flat regions results in several spectrally resolved luminescence lines whose line separations correspond to integer multiples of a GaAs monolayer. (c) and (d) An interface containing microroughness ($\ll r_{\mathrm{exciton}}$) at either or both interfaces also leads to sharp luminescence lines. However, their energetic separation may vary for different locations over the sample.

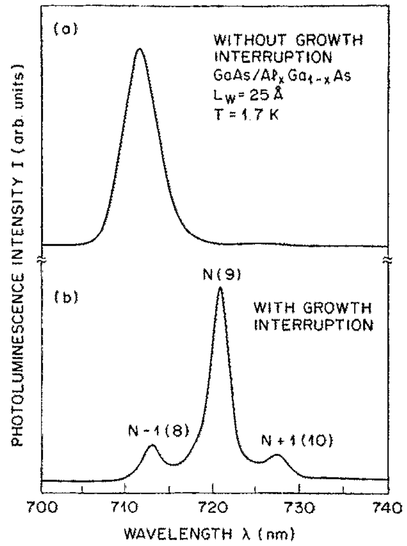

FIG. 2. Low-temperature photoluminescence spectra of a GaAs/Al$_x$Ga$_{1-x}$As single quantum well grown by molecular beam epitaxy (a) without and (b) with growth interruption.

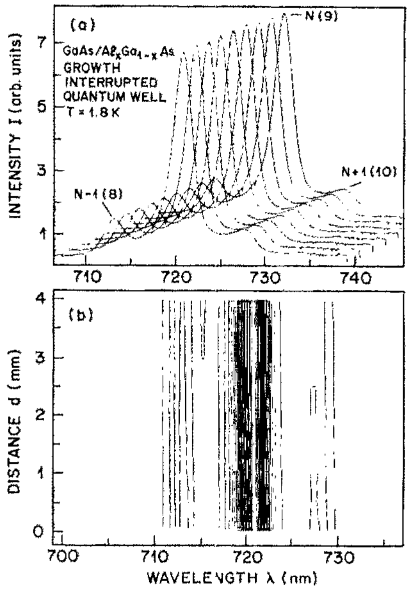

FIG. 3. (a) Photoluminescence spectra with progressive offset for clarity of viewing and (b) contour topography for sequential probing locations on a wafer. The distance between two probing locations is 500 μm. The excitation spot diameter is 100 μm.

difference of exactly one monolayer in the island model. Furthermore, the line separation is *constant* for different lateral probing locations on the wafer. Such a constant line separation is not expected in the microroughness model.

In this letter we report a systematic study of the line separation in growth-interrupted quantum wells using several hundred luminescence scans. Such a large number of data allow us to obtain statistically relevant results. Excitation spectroscopy was performed to identify the nature of the sharp lines.

Two GaAs/Al$_x$Ga$_{1-x}$As($x = 0.35$) single quantum well structures are grown in a Varian Gen II MBE system on 2 in.-diam ⟨001⟩ oriented GaAs substrates at a growth temperature of 580 °C. The layer sequence consists of a 5000 Å GaAs buffer layer, a 1000 Å Al$_x$Ga$_{1-x}$As confining layer, a 25 Å GaAs quantum well, a 1000 Å Al$_x$Ga$_{1-x}$As top confining layer and a 20 Å GaAs cap layer. One wafer is continuously grown and the other has a 120 s growth interruption at each heterointerface of the quantum well. The luminescence experiments are performed at a sample temperature of 1.6 K using an Ar$^+$ laser and a tunable dye laser as excitation sources. The luminescence is detected with a Si charge-coupled device camera array. The luminescence peak energies are evaluated directly by a computer.

Low-temperature photoluminescence spectra of the two single GaAs/Al$_x$Ga$_{1-x}$As quantum well samples are shown in Figs. 2(a) and 2(b) for the uninterrupted and the interrupted growth, respectively. The spectrum obtained from the uninterrupted growth has a single peak. Three clearly resolved lines are observed from the growth-interrupted sample shown in Fig. 2(b). From an assignment of peaks to a discrete number of layers, the center line of the photoluminescence spectra corresponds to a 9 monolayer thick GaAs quantum well, consistent with an anticipated thickness of 25 Å. The shift to higher energy of the continuous growth sample is expected if the barrier-well transition is not abrupt, making the well effectively thinner than a similar growth duration well with abrupt interfaces. Linewidths for these thin QWs are dominated by barrier alloy fluctuations and are comparable to the narrowest linewidths reported for AlGaAs of this composition. Photoluminescence excitation spectroscopy measurements yield a Stokes shift which is smaller than the line width of each of the three photoluminescence peaks.

Several luminescence spectra of the growth-interrupted single quantum well are shown in Fig. 3(a). The spectra are obtained along a 4.0-mm-long radial line close to the center of the wafer. The location of each scan is separated by 500 μm from the next scan. The diameter of the photoexcited spot is 100 μm. Note that the spectra shown are *not* normalized in intensity. The variation of the peak intensity is less than 2%, indicating the superior homogeneity of the wafers. A contour plot is shown in Fig. 3(b) for the luminescence spectra of Fig. 3(a). Inspection of this top-view topography yields that the variation of the peak energy as a function of the location is negligible as compared to the spectral width of the luminescence line. Furthermore, there is no obvious variation of the separation between the three peaks.

To investigate the variation of the peak energy and the peak separation further, a square-shaped array, approximately 5×5 mm^2, of 100 different measurement locations is evaluated. The separation between the measurement locations is 500 μm. The peak-wavelengths of the three luminescence lines are illustrated in Fig. 4. Inspection of the diagram reveals that the peak energies and separations are constant and independent of the measurement location. A scan from the wafer center to the edge shows similar uniformity to Fig. 4.

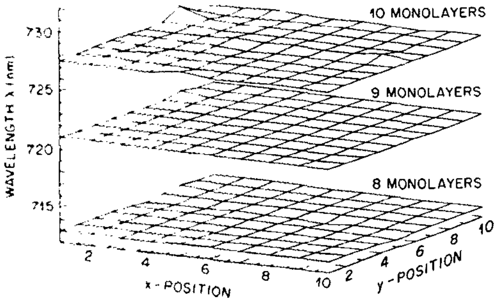

FIG. 4. Energy and line separation of an array of 100 probing locations of a growth interrupted GaAs/Al$_x$Ga$_{1-x}$As single quantum well. The area of the array is 5 × 5mm^2.

The constant separation between the peaks clearly shows that the configuration of the interface does not change over macroscopic distances. It is reasonable to assume that the relationship between any interface configuration and the resulting energy of the radiative transition is unique, i.e., a certain interface configuration results in a well-defined energy. The preferred interface configuration is most likely a thermodynamically stable one. It is well known that a surface configuration which minimizes the number of atomic steps, i.e., a monolayer-flat interface, is thermodynamically stable.[11]

The energies of the luminescence lines are calculated assuming that the lines correspond to discrete thicknesses. The calculation is performed in terms of the one-dimensional Schrödinger equation in the envelope function approximation. The calculated energies[12] agree within 1% with the measured energies. Despite the good agreement no quantitative conclusions can be derived from the comparison between calculated and measured energies due to the uncertainty of several parameters, e.g., the band offsets. However, any thickness or interface variation of the quantum well results in a large energy variation for the thin quantum wells employed in our study, since the quantization energy is approximately inversely proportional to the square of the thickness of the quantum well.

In a separate study, we measured the homogeneity of the Al content in Al$_x$Ga$_{1-x}$As. For an Al$_x$Ga$_{1-x}$As sample with a nominal Al mole fraction of $x = 30\%$, the mole fraction was found to change by less than 0.1% over the center circular area with radius of 20 mm. Note that this Al content variation translates into an energy variation of $\cong 1.5$ meV.

While these spectral data argue strongly for smooth interface island model, the only atomic resolution probe now available is scanning tunneling microscopy (STM) of surfaces. Consequently, a sample was grown identical to the interrupted growth 25 Å well except in place of the second Al$_x$Ga$_{1-x}$As barrier, the sample was cooled to 0 °C and a 1000 Å As cap layer was evaporated onto the GaAs layer. The sample was transferred in air to an ultrahigh vacuum STM system, and the As was removed by heating to 365 °C. The atomic resolution STM image of the resulting surface was atomically smooth with a single terrace, one monolayer lower in the 1000 Å by 1000 Å imaged area. This image provides unequivocal proof of the stability of an atomically smooth As-terminated GaAs surface with islands much larger than an exciton diameter. This surface becomes one of the two quantum well interfaces. Since interdiffusion of the quantum well-barrier interface is negligible at this growth temperature, an abrupt, atomically smooth quantum well interface is expected. The viable models for the interface structure are thus narrowed to the island model of Figs. 1(b) or 1(c). A systematic STM study of the relevant interfaces in this system is under way and will be published separately.

In conclusion, low-temperature luminescence spectra of growth-interrupted wells exhibit clearly resolved lines. The separation of the luminescence lines is constant and independent of the position of the wafer. STM images of interrupted growth GaAs surfaces show atomically smooth islands larger than the exciton diameter. These results argue strongly for the model of extended monolayer-flat regions of the interfaces, i.e., abrupt, atomically smooth interfaces and discrete thicknesses of the quantum well.

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[12] The parameters used for the calculation are $m^e_{\mathrm{well}} = 0.067m_0$, $m^e_{\mathrm{barrier}} = 0.093m_0$, $m^{\mathrm{hh}}_{\mathrm{well}} = 0.45m_0$, $m^{\mathrm{hh}}_{\mathrm{barrier}} = 0.55m_0$, $\Delta E_c/\Delta E_v = 70/30$, $\Delta E_g = 1.455\mathrm{eV}\cdot x$, $E_g^{\mathrm{GaAs}} = 1.519\mathrm{eV}$, $x = 31.7\%$, $a_0 = 2.827$Å. For $N = 8,9$, and 10 molecular layers are calculated transition energies of 1.752, 1.730, and 1.711 eV, respectively. The calculated energies agree with the measured ones within 1%.