



OPTICAL CHARACTERIZATION OF INTERFACE DISORDER IN GaAs-Ga_{1-x}Al_xAs MULTI-QUANTUM WELL STRUCTURES

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We report a systematic increase of the linewidth of luminescence, absorption and excitation spectra of undoped GaAs-Ga_{1-x}Al_xAs Multi-Quantum Well structures with decreasing layer thickness. This broadening is interpreted as due to the island-like structure of the interfaces. A very simple model assuming islands with a one monolayer height and a lateral size ~ 300 Å gives very good agreement with the reported linewidths.

The interfaces between III-V semiconductors play an important role in a number of devices such as LED's, lasers, etc. They also provide systems ideally suited for the study of two-dimensional physics. Although the electronic properties of confined carriers at such interfaces are now quite well-known, little attention has been paid to the influence of the topological structure of such interfaces on carrier properties. For such studies, the preparation technique of Molecular Beam Epitaxy (MBE) is very well adapted as it is capable of providing extremely sharp interfaces on a monolayer scale. The purpose of this paper is to report that in MBE grown GaAs-Ga_{1-x}Al_xAs, the topological disorder of the interface produces sizeable optical effects, and that optical methods provide a useful method to characterize this disorder, complementary to the more microscopic methods of Transmission Electron Microscopy (TEM)¹ and X-ray diffraction.²

Our experiments, basically photoluminescence and absorption, are carried out on Multi-Quantum Well (MQW) structures consisting of multiple GaAs-Ga_{1-x}Al_xAs alternate layers.³ The confinement of the carrier wavefunction intensifies the effects of topological disorder as compared to single interfaces. The use of multiple wells also serves to increase the ratio of "interface" to "bulk" phenomena. The experiments are carried out on samples with or without substrate, immersed in superfluid helium or held in a variable-temperature cold-finger cryostat. A cw dye laser is used as a wavelength-tunable excitation source. The samples are prepared by molecular beam epitaxial growth of nominally-undoped GaAs and Al_xGa_{1-x}As layers on polished, etched (100) surfaces of silicon-doped GaAs substrates. Arsenic-rich growth conditions and substrate temperatures of 690°C were employed.⁴

The luminescence and excitation spectra of two samples with respectively 222 Å and 51 Å wells are shown in Figure 1. As is clear from

the spectra, the 51 Å-well sample exhibits spectra which are much broader than those of the 222 Å-well sample. A similar effect is observed on transmission spectra, but the unavoidable random strain present in thin films without substrates diminishes the measurement precision. Therefore, we will restrict the discussion to luminescence and excitation spectra obtained from MQW structures on substrates. As is discussed elsewhere,⁵ - the features observed are due to free excitons and we require an intrinsic mechanism that can explain the exciton broadening when changing the average well-thickness. Two models of exciton linewidth can be invoked (Fig. 2): (i) a layer-to-layer thickness variation in the sample, which leads to different confinement energies for the different layers. A simple estimate of this effect can be drawn from the approximate value of the confinement energy E_{conf} valid for infinitely deep wells, $E_{conf} = \pi^2 \hbar^2 / 2m^* L^2$, where m^* is the conduction electron effective-mass and L the GaAs layer thickness.^{3,6} Assuming a fluctuation ΔL independent of L , the energy fluctuation varies as L^{-3} . (ii) a thickness - fluctuation within each layer, all the layers having the same average thickness. The origin of this fluctuation can be understood by considering the growth mechanism: after a layer (either GaAs or Ga_{1-x}Al_xAs) has been grown, there exist a number of islands at the free interface,¹ as the number of atomic planes in a layer is never exactly an integer number. The next layer grown will imprison these islands, so that the microscopic interface position cannot be defined better than one atomic constant, although the macroscopic average position can be much better defined. The exciton energy is modified by these thickness fluctuations provided that the lateral size of the islands is larger than the exciton diameter (≈ 300 Å in bulk GaAs). If it were smaller, the exciton energy fluctuation, taken as the sum of the fluctuations of the electron and hole confinement energies, would be diminished due to the averaging of fluctua-

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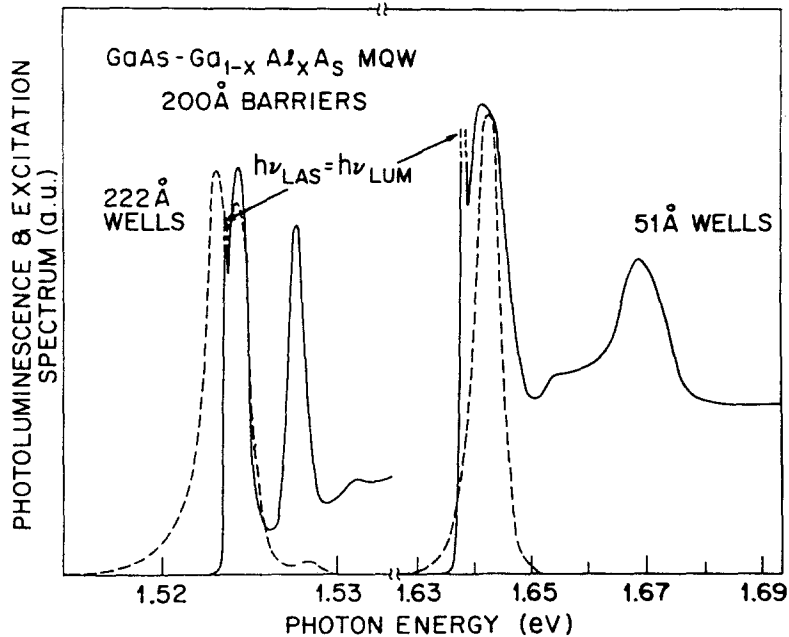


Fig. 1 Photoluminescence (dashed line) and excitation (full line) spectra for two MQW samples with 222 Å and 51 Å wide GaAs-wells respectively. The Ga_{1-x}Al_xAs barriers are 200 Å wide for both samples. Note the change in the horizontal scale. The photoluminescence spectra are obtained under fixed excitation at 1.75 eV. The excitation spectra are recorded while monitoring the luminescence intensity at 1.524 eV and 1.637 eV respectively while varying the energy of the exciting light. Peaks appear at the various characteristic energies of the layers. Only the lowest exciton peaks are shown, corresponding to the $n = 1$ electron to heavy or light-hole transitions. The quantity of interest here is the half-height width of the peaks of the excitation spectra.

tions over the exciton wavefunction. In the large-island approximation, the exciton broadening is calculated exactly like the preceding one. The two mechanisms cannot be distinguished in transmission and luminescence experiments, as both measurements simultaneously probe all the layers. But excitation spectroscopy, i.e. observing the variation of luminescence intensity at a fixed energy while scanning the exciting wavelength allows one to resolve the problem: in the absence of interlayer carrier diffusion, (which certainly is the situation with 200 Å - thick Ga_{1-x}Al_xAs barriers), a layer-to-layer thickness variation [mechanism(i)] would lead to a spatially inhomogeneous luminescence line: the recombination at a given energy originates from layers of a given thickness. The excitation spectrum at this energy would display the energy levels of these layers. Luminescence at other energies, corresponding to other layers and thicknesses, would exhibit different excitation spectra. The actual observation of the same excitation spectrum, regardless of the observation energy means that luminescence originates from identical GaAs wells. Due to their motion through a well, excitons sample out the various sites (i.e. thickness)

described in the broadening mechanism (ii) [see Figure 2, bottom] independently of their recombination energy. One therefore observes the same peaks (energy position, width) in the excitation spectra for the different luminescence energies at which the excitation spectra are recorded. This reproducibility, better than one-tenth of the excitation spectra linewidth, sets a limit to the interlayer thickness fluctuation to be equal or smaller than one-tenth of the intralayer thickness fluctuation. As discussed below, the latter is one monolayer, which indicates that the reproducibility of the average layer thickness during growth is equal or better than 0.1 monolayer. From the reproducibility of the excitation spectra, we deduce the reproducibility of the average layer thickness to be 0.1 atomic layer. We have indeed verified that in imperfect samples with unequal layers, we obtained different excitation spectra at different observation energies.

The exciton linewidth has been systematically studied in a series of samples with 200Å-Ga_{0.76}Al_{0.24}As barriers and varying-width GaAs wells, all grown under the same conditions (substrates cut from the same ingot, no opening of the MBE chamber during the whole sequence)

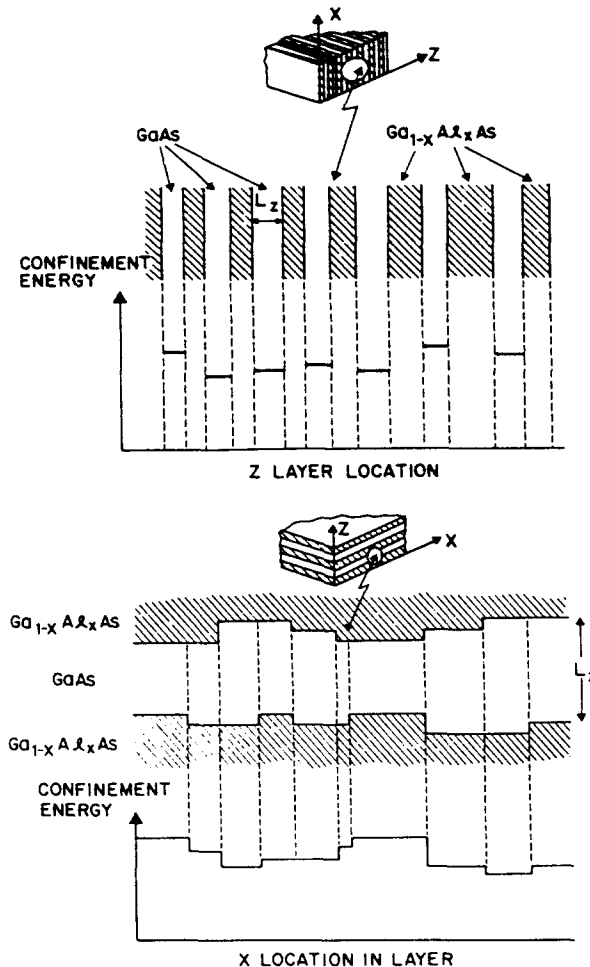


Fig. 2 Two types of width fluctuations and resulting confinement energy fluctuations in MQW structures. Top: layer-to-layer fluctuations, due to an imperfect control of layer thickness in the growth process. Bottom: thickness-fluctuations within one layer, due to the island-like structure of the interface between Ga_{1-x}Al_xAs barriers and GaAs wells. The important parameters are the step-height and island lateral size.

in order to eliminate as much as possible spurious effects. The results are shown in Figure 3, with the solid line giving the result of the calculation taking into account the finiteness of the well-depth for electrons and holes^{3,6} and assuming total layer thickness fluctuations of one and two monolayers. Actually the simple infinitely-deep well approximation gives almost the same curve. The very good agreement of the calculation (which has no adjustable parameter) with experiment in the 150-80 Å range is a strong support both for the model and the estimate of the island-height as one monolayer. No attempt to fit the lineshape of absorption or excitation spectra was made as this would require a detailed statistics of the shapes and heights of the islands, on which we lack too much information at this point. The disagreement at both ends of the well-width range is not surprising: at small confinement energies (wide wells) the

intrinsic broadening mechanism discussed here becomes relatively unimportant when compared to other broadening mechanisms, such as those due to impurities. For such MQW's, the measured linewidth is an overestimate of the layer-fluctuation induced linewidth and all the experimental points should lie above the theoretical value. At the other end of the range, large confinement energies (narrow wells), our simple approximations may break down: (i) disorder-induced energy fluctuations are of the same order as the exciton binding energy and can modify both its energy and wavefunction. (ii) the electrons and holes, which have a large confinement energy, have wavefunctions which penetrate several atomic layers in the Ga_{1-x}Al_xAs barrier and will therefore be less sensitive to the effect of interface compositional disorder. In this limit a more detailed theoretical analysis is clearly needed.

The interface disorder of GaAs-Ga_{1-x}Al_xAs

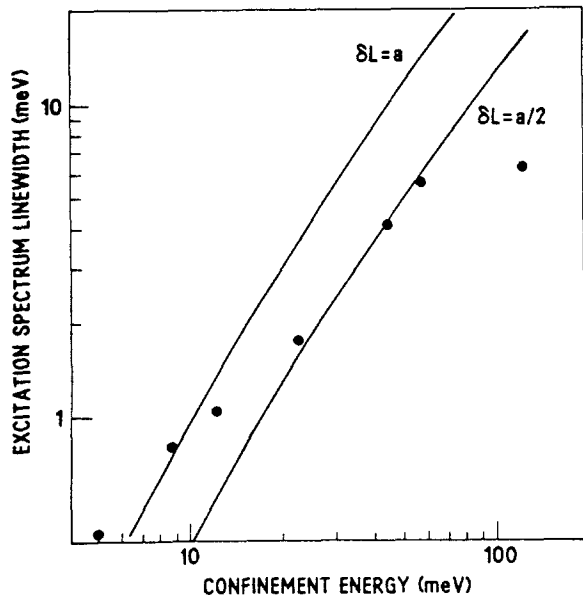


Fig. 3 Variation of the excitation spectrum line width with the confinement energy for a series of samples grown under similar conditions. The two calculated curves correspond to layers with a thickness fluctuating between $L \pm a/2$ and $L \pm a$, L being the average layer thickness and a the lattice constant. Sample characteristics: $\text{Ga}_{1-x}\text{Al}_x\text{As}$ thickness 200 Å, $x = 0.3$; GaAs layer thickness: 327, 222, 205, 145, 102, 81 and 51 Å respectively; substrate temperature during growth $T_S = 690^\circ\text{C}$.

interfaces has also been directly observed by TEM¹ and X-ray diffraction² for samples grown under conditions similar to those reported here, but with thinner wells and barriers. In TEM,

island-like structures several hundred Angstroms wide have been observed at the interfaces.¹ In X-ray diffraction, the island-like clusters at the interfaces were revealed by observation of weak diffuse X-ray scattering.⁷ A detailed comparison between the optical method and these more microscopic characterization schemes is difficult to make because they are not carried out in the same samples: TEM and X-ray diffraction work best for thin layers (a few atomic layers thick) where large diffraction angles are obtained, allowing a good precision in linewidths measurements. On the contrary, optical methods are limited to MQW structures with GaAs wells more than ≈ 15 GaAs atomic layers thick so that the electron (i.e. excitonic) levels are confined in the well and experience the interface disorder. The TEM and X-ray diffraction methods clearly give a more detailed image of the island structure and also permit, in the case of TEM, to observe spatial variations of this structure on the micron scale. On the other hand, the optical method has the advantages of being nondestructive and requiring no sample preparation. It also yields the layer-to-layer reproducibility and is sensitive enough to characterize disorder in layers where the relative thickness fluctuation is only $\approx 2\%$. It only characterized the wider islands, however, as those much smaller than the exciton Bohr radius do not affect the confinement energy.

In conclusion, the present experiments show that the intrinsic, unavoidable topological disorder at the hetero-interfaces of semiconductors can produce sizeable optical effects. We interpret the results in terms of potential fluctuations due to an island-like structure of the $\text{GaAs-Ga}_{1-x}\text{Al}_x\text{As}$ interface in MQW structures, islands which are one monolayer high and with a lateral dimension ≈ 300 Å. These measurements confirm, for larger layer thicknesses, the results reported previously by the TEM and X-ray diffraction techniques.

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