Linewidth analysis of the photoluminescence of $In_xGa_{1-x}As/GaAs$ quantum wells ($x = 0.09, 0.18, 1.0$)

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(Received 30 December 1994)

Photoluminescence measurements have been performed at low temperature in $In_xGa_{1-x}As/GaAs$ quantum wells with different well widths, L, and indium concentrations, x. The dependence of the experimental linewidth of the heavy-hole–free-exciton recombination lines on L and x has been compared with existing models of interface and alloy disorder. It has been demonstrated that interface disorder has a crucial role at low L and high x. The estimated values of the interface-roughness size agree well with those found by different techniques.

I. INTRODUCTION

In quantum wells (QW) whose width is comparable with the exciton Bohr radius (R_{ex}), low temperature photoluminescence (PL) spectra critically depend on the quality of the structures in terms of the uniformity of composition and abruptness of the interfaces. In fact, the excitonic recombination is inhomogeneously broadened, because of the disorder due to interface roughness, which includes the island growth and the cation exchange due to atomic interdiffusion. Moreover, because of the alloy disorder, mixed crystals are characterized by exciton linewidths broader than those observed in the pure components. In the case of quantum wells, for well widths larger than R_{ex}, the PL linewidth is determined only by the quality of the small-gap material, while for narrow well widths it is also affected by the quality of the interface and the barrier material, because of the penetration of the exciton wave function into the barrier.

Photoluminescence is, therefore, a very convenient, nondestructive technique for the characterization of the degree of ideality of quantum wells. However, although several theoretical works have appeared which deal with effects of different type of disorder on PL spectra of "real" QW's,[1–5] only two experimental works have been published on this subject, at least at our knowledge.[6,7] The former work, on $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ quantum-well structures, reports data of samples whose linewidth is rather broader than those found in the best samples; the latter work, a report on $In_xGa_{1-x}As/GaAs$ QW's, does not take into account the interface disorder and makes unrealistic assumptions in the analysis of the data.

In this paper, we report a detailed analysis of the full width at half maximum (FWHM) of the luminescence of $In_xGa_{1-x}As/GaAs$ quantum wells. Samples with indium concentrations $x = 0.09$, 0.18, and 1.0 have been studied. It will be shown that the excitonic linewidth is well described only when both interface roughness and alloy disorder are taken into account. An estimate has been obtained of the characteristic parameters entering the theoretical models, namely, the size of the islands at the interface and the effective volume V_{ex} over which the effects of disorder are averaged. The size of islands has been found to be smaller than the exciton Bohr radius, thus accounting for the lack of optical evidence of extended monolayer fluctuations in the spectra of $In_xGa_{1-x}As/GaAs$ quantum wells. Moreover, the effective volume V_{ex} has been found to be roughly equal to the quantum-mechanical expectation value of the exciton volume. The same theoretical model has been also successfully applied, with similar values of the characteristic parameters, to the case of InAs/GaAs quantum wells. In this case, the alloy disorder contribution is absent and interface roughness is the only effect responsible for the reported excitonic linewidth.

In the next section, we will briefly resume the model we have used,[1–4] with some refinements in the treatment of alloy disorder. In Sec. III, we will describe the samples and the experiment, in Sec. IV we shall present the experimental results. Finally, a comparison between the experimental results and theoretical models will be made in Sec. V.

II. THEORETICAL MODEL

A. Interface broadening

A statistical model has been proposed[2] to determine the broadening of the excitonic recombination in a quantum well with potential-energy discontinuity at interfaces of finite size. If $z = 0$ identifies the position of the ideal interface between the compounds $A(z < 0)$ and $B(z > 0)$, in the real interface there are islands of the compound B for $z < 0$ and islands of the compound A for $z > 0$. Therefore, real interfaces involve fluctuations δ_1 of the quantum-well width L. The mean concentrations of the

islands of compounds A and B, and of the flat regions of ideal interface are C_a^0, C_b^0, and C_i^0, respectively, their lateral extensions are δ_{2a}, δ_{2b}, and δ_{2i}. The lateral extent of the exciton wave function is ρ_{ex}. $E_{ex}(L)$ is the exciton recombination energy, with a well known dependence on the well width. $E_{ex}(L)$ diverges for $L \to 0$ in the ideal case of a well of infinite depth, it reaches a finite maximum at $L = 0$ in the case of a well of finite depth, because of a sizeable penetration of the exciton wave function into the barrier material. If $\delta_{2a} \approx \delta_{2b} = \delta_2$ and $\delta_2 < \rho_{ex}$, as it will be shown to hold in our case, and islands occur randomly at the interface, it has been shown that the exciton line shape in the PL spectra is given by a Gaussian function with full width at half maximum,[2]

$$\gamma_{int} = \sqrt{2}\frac{\delta_2}{\rho_{ex}}(1.4C_a^0C_b^0)^{1/2}(C_a^0\Delta^+ + C_b^0\Delta^-)\,, \qquad (1)$$

where

$$\Delta^+ = \left.\frac{\partial E_{ex}}{\partial L}\right|_{L_0+\delta L}\delta_1\,, \qquad \Delta^- = \left.\frac{\partial E_{ex}}{\partial L}\right|_{L_0-\delta L}\delta_1\,, \quad (2)$$

and L_0 is the mean thickness of the quantum well. Δ^+ and Δ^- depend strongly on L, ρ_{ex} depends more smoothly on L. Therefore, for quantum wells of infinite depth, γ_{int} increases monotonically for decreasing L and becomes infinite for $L = 0$, while for quantum wells of finite depth the value of γ_{int} reaches a maximum and then decreases as L approches zero ($\gamma_{int} = 0$ for $L = 0$), because of the increasing penetration of the excitonic wave function into the barrier.

B. Alloy broadening

It can be shown[1,3] that compositional fluctuations give rise to fluctuations ΔE_{ex} of E_{ex}, with respect to its value in the virtual crystal approximation:

$$\Delta E_{ex}(x, x_0; V_{ex}) \cong (x - x_0)\Delta_1\,. \qquad (3)$$

x_0 is the nominal composition of the alloy, $\Delta_1 = \left.\frac{\partial E_{ex}}{\partial x}\right|_{x_0}$, and V_{ex} is a critical volume, of the order of the exciton volume, inside which the $1s$ exciton experiences only a mean crystal potential. If a Poisson distribution gives the probability of finding a defined number n of indium atoms in a given exciton volume in $In_xGa_{1-x}As$, in the limit of large n and large number of anion sites in this volume, one gets a Gaussian function as the distribution function of the exciton energies. The exciton full width at half maximum due to the alloy disorder is then given by[3]

$$\gamma_{all} = 2\left(\frac{V_c}{V_{ex}}1.4x_0(1-x_0)\right)^{1/2}\Delta_1\,, \qquad (4)$$

where V_c is the volume per anion site (0.22^3 nm^3 in GaAs).[8] This formula has been modified[3] in order to take into account the dependence on the well width of the exciton localization, due to the finite value of the confining potential. If P_{ex} is the probability for the exciton to penetrate into the barrier, only the fraction $(1 - P_{ex})$ of the exciton is confined in the well, thus giving rise to γ_{all} values smaller than those given for the bulk by Eq. (4). Taking this correction into account, one finds[3]

$$\gamma_{all} = 2\left(\frac{V_c}{V_{ex}(1-P_{ex})}1.4x_0(1-x_0)\right)^{1/2}\Delta_1\,. \qquad (5)$$

Following the treatment introduced in Ref. 4 to deal with localized exciton wave function in a bulk material, in the present work Eq. (5) has been modified in order to account for the probability $(1-P_{ex})$ that a fluctuation in x produces an energy variation,

$$\Delta E_{ex}(x, x_0; V_{ex}) \cong (x - x_0)\Delta_1(1 - P_{ex})\,, \qquad (6)$$

from which we obtain

$$\gamma_{all} = 2\left(\frac{V_c}{V_{ex}}1.4x_0(1-\omega_0)(1-P_{ex})\right)^{1/2}\Delta_1\,. \qquad (7)$$

In $In_xGa_{1-x}As$/GaAs quantum wells, an increase of the well width for a given x_0 decreases the probability of penetratation into the barrier of the exciton wave function. Correspondingly it increases the contribution of the alloy disorder to the fluctuations of the exciton energy. Assuming a constant exciton volume, the value of γ_{all} monotonically increases and asymptotically reaches its three-dimensional value in the limit of large well widths. This assumption, however, is incorrect in the case of excitons in QW's. In fact, in real QW's the binding energy of the exciton, E_b, shows a maximum (the exciton volume a minimum) due to the competition between the effects of the localization and the penetration into the barrier of the exciton wave function. These effects have been taken into account in Ref. 7, where two strong assumptions have been made: (i) the exciton volume has a linear inverse relationship with L; (ii) the penetration of the exciton wave function into the barrier and its changes in the QW plane can be disregarded. On this ground it has been shown that γ_{all} has a maximum for a finite value of the well width, like the experimental data, and it has been claimed that interface disorder plays a negligible role in the modeling of exciton line shapes.[7]

In our analysis of the exciton line shape for the $In_xGa_{1-x}As$/GaAs system, we have evaluated the exciton volume as a function of L by applying a hydrogenic approximation to the exciton binding energies determined from PL excitation measurements in the very same $In_xGa_{1-x}As$/GaAs quantum wells.[9] Instead, in the case of the InAs/GaAs quantum wells, we have kept the exciton volume constant, equal to the bulk value for GaAs, because it can be shown that 90% of the exciton wave function penetrates into the barrier.[10] In this case, the disorder due to fluctuations of the alloy composition is obviously absent.

III. EXPERIMENT

Three sets of samples have been used for this work. The first set contains samples with an indium concen-

tration $x = 0.09$ and well widths 1.5 nm $\leq L \leq$ 25 nm, the second set samples with $x = 0.18$ and 1 nm $\leq L \leq$ 14 nm. The third set of samples is made by five InAs/GaAs quantum-well structures, with well widths ranging from 0.8 to 1.6 ML in steps of 0.2 ML. The samples have been grown by molecular beam epitaxy on (100)GaAs substrates. The growth temperature for the $In_xGa_{1-x}As/GaAs$ quantum wells was 520 °C. All sample thicknesses are below the critical value for the generation of misfit dislocations. Details on the InAs/GaAs growth are given elsewhere.[10]

The photoluminescence measurements have been performed at 5 K, by using the excitation of an Ar^+ or of a Ti-sapphire laser.

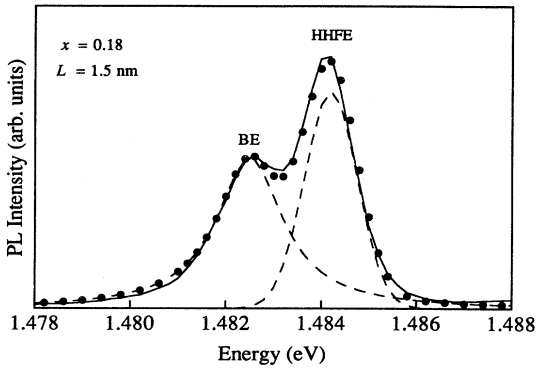

FIG. 2. Photoluminescence at $T = 5$ K (solid curve) of an $In_{0.18}Ga_{0.82}As/GaAs$ quantum well, with $L = 1.5$ nm. Dots are the best fit to the data in terms of heavy-hole–free-exciton and BE contributions (dashed curves). $P = 0.1$ W cm^{-2}.

IV. RESULTS

A typical PL spectrum at 5 K of a $In_{0.09}Ga_{0.91}As/GaAs$ quantum well is shown in Fig. 1. Due to its energy shift with temperature, the observed PL line is attributed to the heavy-hole–free-exciton (HHFE) recombination, whose linewidth is the subject of the present work. For wider wells, and for $x = 0.18$, a low energy PL line is observed at an energy lower than that of the free exciton. This recombination (BE) is a common feature of $In_xGa_{1-x}As/GaAs$ structures[7,11] and it is not related to impurities.[7] In presence of the BE recombination, a two-band fit has been performed in order to obtain the correct line shape of the HHFE, as shown in Fig. 2 for $x = 0.18$.

In Fig. 3, the free-exciton FWHM is reported vs well width for $x = 0.09$ and $x = 0.18$. The absolute values found for our samples are those typically observed for high quality samples of this type.[11] This allows us a comparison with the theory in the absence of undesired and uncontrolled extrinsic broadening mechanisms. Both series of FWHM values show a maximum, for $L = 4$ nm and, less pronounced, for $L = 6$–7 nm in the case of $x = 0.18$ and $x = 0.09$, respectively. This difference can be explained by the different degree of exciton confinement, which gives rise in the latter case to a smoother change from a situation where the exciton is mainly confined in the well to that where it sizably penetrates into the barrier.

A typical PL spectrum of a InAs/GaAs quantum well is shown in Fig. 4. Elsewhere,[10] it has been shown that the high energy peak of the spectrum is due to the HHFE recombination. The FWHM as a function of the well width is reported in the next section together with the fit.

V. DISCUSSION

The experimental values of the FWHM of the two $In_xGa_{1-x}As/GaAs$ sets of quantum wells are now analyzed in terms of Eqs. (1) and (7). The extent of the width fluctuations at the interface has been assumed equal to one monolayer (0.28 nm) in the growth direction (δ_1) and it has been taken as a free parameter in

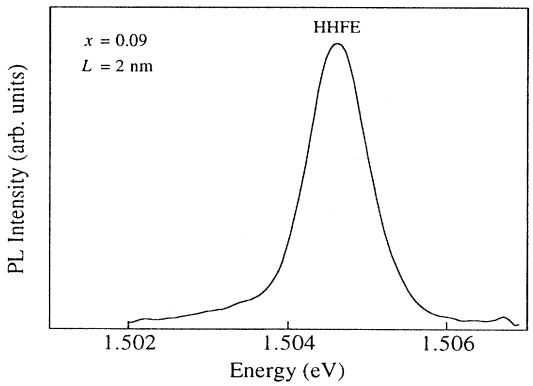

FIG. 1. Photoluminescence at $T = 5$ K of an $In_{0.09}Ga_{0.91}As/GaAs$ quantum well, with $L = 2$ nm. Only the heavy-hole free exciton is observed. $P = 0.1$ W cm^{-2}.

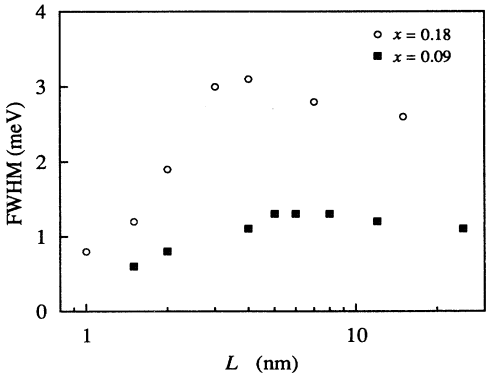

FIG. 3. FWHM at $T = 5$ K for two series of $In_xGa_{1-x}As/GaAs$ quantum wells, with $x = 0.09$ and 0.18 vs the well width L.

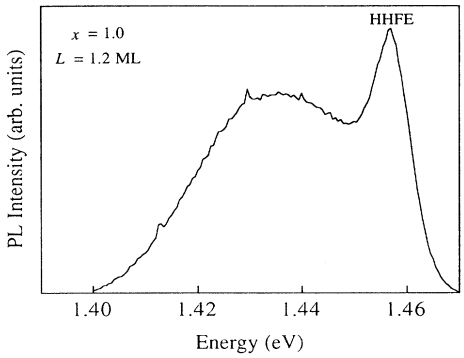

FIG. 4. Photoluminescence at $T = 5$ K of an InAs/GaAs quantum well, with $L = 1.2$ ML. The heavy-hole–free-exciton contribution is indicated.

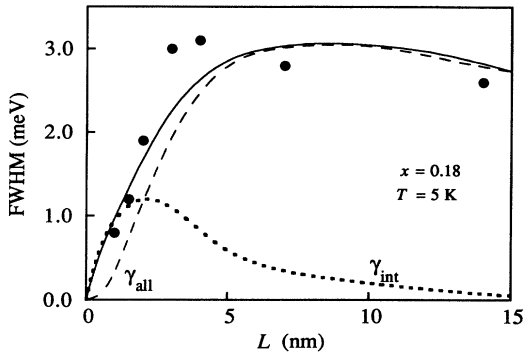

FIG. 6. Theoretical FWHM of the free-exciton recombination line of $In_{0.18}Ga_{0.82}As/GaAs$ quantum wells (solid line) together with the experimental results (dots). The separate contributions of the interface disorder (γ_{int}, dotted line) and of the alloy disorder (γ_{all}, dashed line) are also reported. $\delta_2 = 1.3$ nm, $\beta = 2$.

the growth plane (δ_2). The values of the lateral extent ρ_{ex} of the exciton wave function and of the effective exciton volume V_{ex} have been derived from the experimental values of the exciton binding energy by using a three-dimensional spherical exciton model in the hydrogenic approach, thus neglecting the exciton ellipsoidicity. The exciton effective volume has, therefore, been evaluated in terms of the Bohr radius R_{ex}, as obtained from the exciton binding energy E_b by means of the relationship

$$R_{ex}E_b = \frac{13.6}{\epsilon}0.053 \text{ eV nm}. \tag{8}$$

Different values for V_{ex} have been assumed in the theoretical models: $V_{ex}=V_B = 4/3\pi R_{ex}^3$, in the simplest model of Ref. 3, while, on the basis of different full quantum-mechanical approaches, $V_{ex} = 7.5V_B$ and $V_{ex} = 9.35V_B$ in Refs. 1 and 4, respectively. In view of the arbitrary nature of the definition of V_{ex} (and of V_c), we have assumed $V_{ex} = \beta V_B$, where β is a fit parameter.

The parameters Δ_1, Δ^+, Δ^-, and P_{ex}, that appear in Eqs. (1) and (7), have been calculated neglecting the electron-hole interaction. P_{ex} has been determined as the product of the probabilities of finding the two particles outside the quantum well, Δ_1, Δ^+, and Δ^- by standard methods.[12] The experimental full widths at half maximum have then been fitted to a theoretical total linewidth given by

$$\text{FWHM}_{th} = \sqrt{\gamma_{int}^2 + \gamma_{all}^2}\,. \tag{9}$$

In Fig. 5, the same experimental data for $x = 0.09$ shown in Fig. 3 are presented, together with the fit to the data (solid curve). The two dashed curves represent the separate contributions of the alloy disorder and of the interface disorder. The alloy disorder contribution alone accounts well for the experimental data in wide wells. However, the interface disorder contribution is crucial in order to fit the data obtained in narrow quantum wells: the alloy disorder contribution cannot reproduce the present experimental data at low L, even for different choices of β. The value of δ_2 (2.4 ± 0.3 nm) is smaller

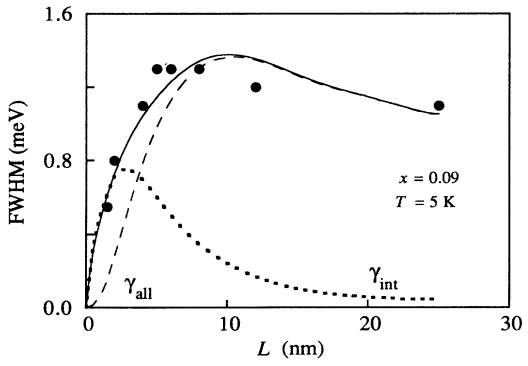

FIG. 5. Theoretical FWHM of the free-exciton recombination line of $In_{0.09}Ga_{0.91}As/GaAs$ quantum wells (solid line) together with the experimental results (dots). The separate contributions of the interface disorder (γ_{int}, dotted line) and of the alloy disorder (γ_{all}, dashed line) are also reported. $\delta_2 = 2.4$ nm, $\beta = 6$.

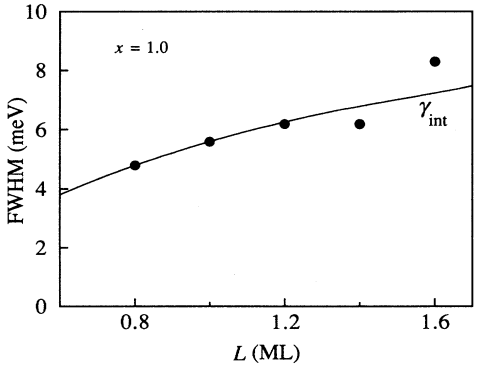

FIG. 7. Experimental FWHM at $T = 5$ K of the free-exciton recombination line of InAs/GaAs quantum wells. The solid line has been obtained by a fit of Eq. (1) to the data. $\delta_2 = 2.9$ nm.

than the Bohr radius (6.5 ± 1 nm in the range of well width investigated here), as assumed in deriving Eq. (1). This also accounts for the lack in these strained structures (see Fig. 1) of the number of free-exciton peaks observed in $Al_xGa_{1-x}As/GaAs$ quantum wells and attributed to monolayer fluctuations in the well width.[13] The value of β (6 ± 1) is in good agreement with those assumed in Refs. 1 and 4 (7.5 and 9.35, respectively), thus showing that (i) those theoretical models are quite adequate in this case of low strain and low disorder, (ii) the choice of the other parameters has been correct, (iii) the quality of these samples is very high.

The results of the same analysis applied to the set of samples with $x = 0.18$ are shown in Fig. 6. Although the fit is quite good, it has to be noticed that different values for the fitting parameters have been found, $\delta_2 = 1.3 \pm 0.2$ nm and $\beta = 2 \pm 0.4$. The value of δ_2 is once more smaller than R_{ex} and adequately matches the length scale of the interface roughness (2 nm), as measured in similar structures with $x = 0.2$ by a scanning tunneling microscope.[14] The lower value of β, with respect to the $x = 0.09$ case, can have different explanations. On the experimental side, the deconvolution of the data and the hydrogenic approximation may have introduced an experimental uncertainty larger than in the previous cases, because of the higher degree of exciton localization at higher indium concentration. On the theoretical side, our results could indicate a degree of alloy disorder in samples with $x = 0.18$ higher than that predicted by the $x(1-x)$ simple law used in the models. This would imply that the small perturbation limit is already overcome for $x \simeq 0.18$. On the other hand, this feature could be due to an incipient columnar growth.

The analysis made on the FWHM of the InAs/GaAs quantum wells is reported in Fig. 7. In this case the alloy broadening is absent ($x = 1$) and only the interface roughness has been considered as a broadening mechanism. The volume of the exciton in bulk GaAs has been used through the whole analysis, a good approximation in this system.[10] The best fit to the data is once more very good, thus confirming that interface roughness plays a central role in determining the exciton linewidth. This fit has been obtained with $\delta_2 = 2.9 \pm 0.3$ nm, a value close to that found for the $In_{0.09}Ga_{0.91}As/GaAs$ quantum wells in the opposite limit of low indium concentration.

VI. CONCLUSIONS

The experimental linewidth of $In_xGa_{1-x}As/GaAs$ and InAs/GaAs quantum wells have been well reproduced by simple theoretical models, which take into account the disorder generated by both interface roughness and random alloy composition. The interface contribution has been shown to be relevant not only for InAs/GaAs QW's, but also for $In_xGa_{1-x}As$ QW's with narrow well widths. The value found for the lateral extent of the interface disorder is smaller than the exciton extent, in agreement with the observation of a single free-exciton recombination in $In_xGa_{1-x}As/GaAs$ structures and with the dimension of In clusters in similar structures, as measured by a scanning tunnel microscope. An estimate of the volume, where the exciton samples the alloy disorder, has been obtained. This value agrees with that estimated in a full quantum-mechanical treatment of the problem.

ACKNOWLEDGMENTS

We thank D. Orani for her help in the determination of the exciton binding energy. The work at Fondazione Bordoni has been carried out within the agreement with the Italian P.T. Administration.

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