Binding energies and oscillator strengths of excitons in thin GaAs/$Ga_{0.7}Al_{0.3}As$ quantum wells

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We report experimental results of optical-absorption experiments in a waveguiding geometry at low temperature in thin GaAs/$Ga_{0.7}Al_{0.3}As$ quantum wells of widths scaling from 10 to 100 Å. In this experimental configuration, the single quantum-well absorption coefficients are obtained in both polarization directions, parallel and normal to the plane of layers. The binding energy and oscillator strength of the 1*s* heavy and light excitons are determined. For well thicknesses less than 50 Å we observe a decrease of the exciton binding energy and oscillator strength demonstrating the crossover from the two-dimensional to the three-dimensional behavior of excitons.

I. INTRODUCTION

The important carrier confinement in GaAs/$Ga_{1-x}Al_xAs$ quantum-well (QW) heterostructures leads to an enhancement of the electron-hole Coulomb interaction. The consequence of this situation is an important increase of the binding energies (BE's) and the oscillator strengths (OS's) of excitons in QW's as compared to the bulk values. For infinite barrier height, the BE increases in a monotonic way and tends to the two-dimensional (2D) limiting value, which is equal to four times the bulk value (i.e., 16 meV in GaAs). However, if the finite height of the barriers in real QW's is taken into account, the 2D limit can never be reached. When the well width is reduced, the carriers' wave function penetrates into the barrier and the exciton becomes delocalized. The BE, after reaching a maximum smaller than the 2D limit, decreases and tends to the appropriate value of the bulk barrier material when the well width is reduced to zero.

Since the first theoretical approaches which described the increase of BE's in QW heterostructures as due essentially to confinement effects,[1,2] more accurate theories have been recently proposed.[3] These theories take into account additional effects such as valence-band mixing, coupling between excitons from different subbands, nonparabolicity of the bulk material conduction band, and the difference between the barrier and well material dielectric constant. All these effects give comparable contributions and tend to increase the BE and OS of excitons. In the case of GaAs/AlAs QW's the values can even be higher than the 2D predicted limit.[3]

Direct absorption measurements are rather rare in GaAs/$Ga_xAl_{1-x}As$ QW structures and the commonly used techniques for determining BE's of excitons are photoluminescence excitation spectroscopy. In high-quality QW samples the 2*s* exciton state can be resolved and the energy difference between the 1*s* and the 2*s* exciton states gives the BE.[4–6] More indirect methods such as magneto-optical experiments are also used.[7] With these techniques only binding energies of heavy excitons are obtained. The OS of the fundamental 1*s* heavy exciton transition in GaAs/$Ga_xAl_{1-x}As$ multiple-quantum-well (MQW) structures has been determined by means of conventional optical absorption[8–10] and the two-dimensional shrinkage of the exciton wave function in the QW has been reported in GaAs/$Ga_xAl_{1-x}As$ and also in $In_xGa_{1-x}As$/InP MQW structures.[11] The transition from the two- to three-dimensional behavior of heavy excitons was reported in $In_xGa_{1-x}As$/Ga(Al)As MQW's by conventional optical-absorption and photoluminescence spectroscopy.[12]

We present in this paper experimental results on QW absorption in a waveguiding configuration. The absorption coefficients of the QW excitonic transitions are determined in GaAs/$Ga_{0.7}Al_{0.3}As$ QW's with different widths varying from 10 to 100 Å. The BE's as well as the OS's are deduced for both heavy and light excitons.

II. ABSORPTION EXPERIMENTS IN THE OPTICAL WAVEGUIDING CONFIGURATION

The samples in the absorption experiments are optical planar waveguides made of GaAs/AlAs short-period superlattices grown by molecular-beam epitaxy (MBE) at 630 °C on a (001)-oriented GaAs substrate. The single GaAs/$Ga_{0.7}Al_{0.3}As$ QW's are embedded inside the waveguides. The QW thicknesses are 10, 20, 30, 50, and 100 Å and the alloy barrier width is 150 Å. The total thickness of the waveguide is about 2 μm.

Absorption experiments in a waveguiding geometry show several advantages as compared to a conventional absorption configuration. First, it is possible to measure the transmission of a single QW. Second, since light is propagating along the plane of the layers the transmission can be observed in both polarization directions of light parallel and normal to the plane of layers at low temperature. It is also not necessary to remove the substrate. Finally, the absorption coefficient at a given wave-

length can be measured by studying the reabsorption of the guide's luminescence. From the guide's transmission, the absorption of the structure can be deduced in the whole spectral range. This procedure has been described in detail in Ref. 13.

The transmission experiments in both polarization directions of light are performed on a broad spectral range with a quartz halogen lamp at low temperature (10 K). The coupling between light and sample is obtained by focusing the incident beam on the cleaved face of the sample with a microscope objective (N.A. 0.6). The transmitted light is collected by a mirror objective (N.A. 0.5) detected through a monochromator by a photomultiplier followed by a lock-in amplifier.

Figure 1 represents the absorption spectra of a 20-Å $GaAs/Ga_{0.7}Al_{0.3}As$ QW at low temperature in both polarization directions superimposed to the photoluminescence spectrum of the sample. In the polarization parallel to the layers (a), we observe the 1*s* heavy- and light-exciton peak at 1.79 and 1.83 eV, respectively. In the polarization normal to the layers (b), only the light-exciton transition is observed at 1.835 eV as expected from selection rules. The QW photoluminescence peaks at 1.77 eV and is shifted by 20 meV from the 1*s* heavy-exciton transition. This Stokes shift is due to the localization of excitons by QW width fluctuations at the interface. At higher energies (1.95 eV) other structures appear in both (a) and (b) absorption spectra. They are attributed to slightly confined states in the $Ga_{0.7}Al_{0.3}As$ alloy. These structures are systematically observed in the thin 10- and 20-Å QW's we have studied. At higher energies (2.1 eV) the increase of absorption is related to the absorption of the superlattice surrounding the GaAs QW and the $Ga_xAl_{1-x}As$ alloy and forming the core of the guide. As is shown in Fig. 1, the photoluminescence of the superlattice consists of a main zero-phonon line at 2.06 eV and

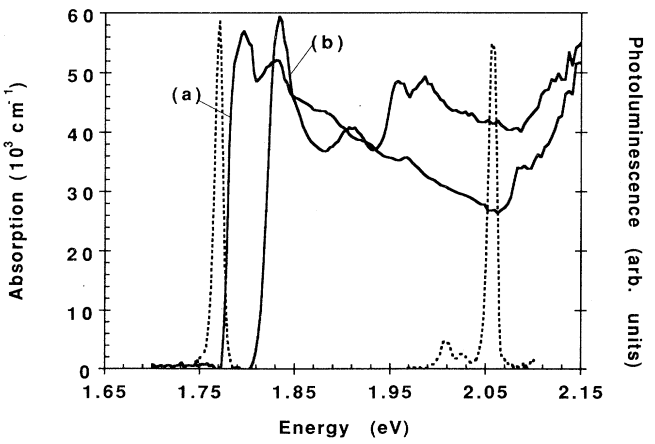

FIG. 1. Absorption spectra at low temperature of the 20-Å $GaAs/Ga_{0.7}Al_{0.3}As$ QW in both polarization directions of light, parallel to the plane of layers (a) and parallel to the growth axis (b). Dotted lines represent the photoluminescence spectrum of the sample.

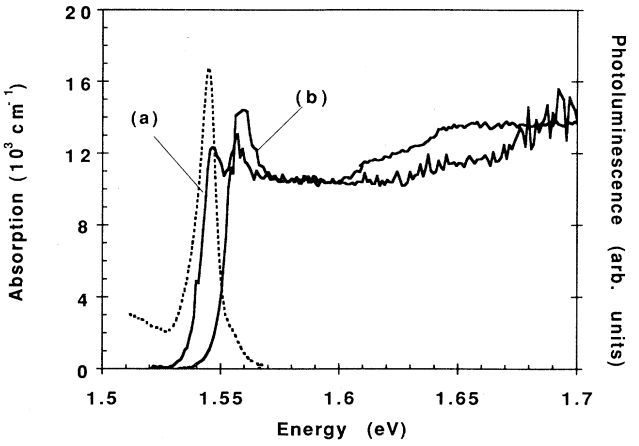

FIG. 2. Absorption spectra at low temperature of the 100-Å $GaAs/Ga_{0.7}Al_{0.3}As$ QW in both polarization directions of light, parallel to the plane of layers (a) and parallel to the growth axis (b). The photoluminescence spectrum of the QW is shown by dotted lines.

two phonon replicas at lower energy. This spectrum is characteristic of a type-II superlattice. The corresponding absorption coefficient is smaller than in a type-I structure and varies smoothly with energy, as observed in Fig. 1.

In Fig. 2 are shown the absorption spectra of a 100-Å $GaAs/Ga_{0.7}Al_{0.3}As$ QW at low temperature in both polarization directions superimposed to the luminescence of the QW. In the polarization parallel to the layers (a) we observe the 1*s* heavy- and the 1*s* light-excitonic transitions at 1.546 and 1.558 eV, respectively. At higher energies the signature of higher-order transitions in the QW is evidenced, i.e., the $E1$-HH3 and the $E2$-HH2 transitions at 1.608 and 1.644 eV, respectively. The QW luminescence peak is shifted by 2 meV from the energy position of the 1*s* heavy-exciton transition. This shift corresponds to the variation of the transition-energy position when the QW width varies by one monolayer. For all the studied QW's similar spectra are obtained.

III. DETERMINATION OF EXCITONIC PARAMETERS

The obtained absorption spectra are decomposed using a phenomenological expression in a 2D model. The 1*s* excitonic peak is fitted by a Gaussian line shape with an inhomogeneous broadening factor. The band-to-band absorption is fitted with a broadened 2D continuum, taking also into account the Sommerfeld factor,[14] which describes the electron-hole pair interaction in the continuum. The excited excitonic states $n > 1$ are considered phenomenologically in the broadening parameters. Such a description of the continuum is only valid in the vicinity of the absorption band gap, since other effects such as valence-band mixing are not included in this simplified description.

The decomposition of the absorption spectrum in the polarization along the growth axis is straightforward

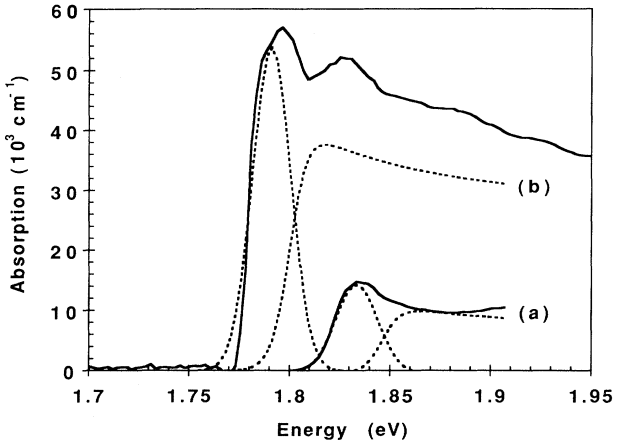

FIG. 3. Decomposition of the 20-Å GaAs/$Ga_{0.7}Al_{0.3}As$ QW absorption spectrum for light polarized along the growth axis (a) and parallel to the layers (b). The absorption spectrum (a) has been divided by 4 in order to show the decomposition of the absorption spectrum (b) in heavy exciton, light exciton, and continuum.

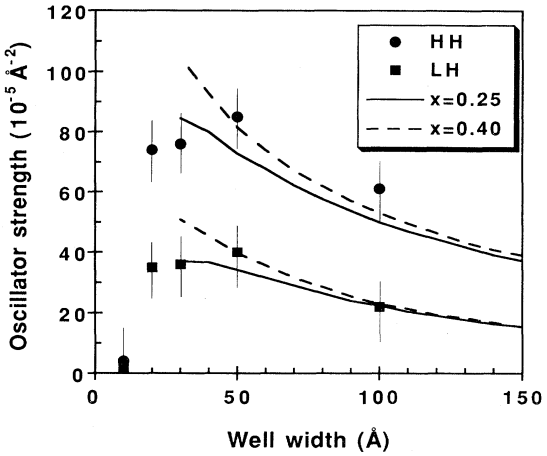

FIG. 5. Oscillator strengths for the polarization parallel to the plane of layers of the 1*s* heavy and light excitons as a function of the QW width, compared to the theoretical curves of Ref. 3. The two upper (lower) curves correspond to the calculated heavy- (light-) exciton OS for a 25% (solid line) and 40% (dashed line) Al concentration in the barriers. The experimental oscillator strength of the heavy and light excitons in the 10-Å QW are 4×10^{-5} and 2×10^{-5} Å^{-2}, respectively.

with such a description as is shown in Fig. 3(a). By using the predicted selection rules for polarized light (the light-exciton OS for light polarized along the growth axis is four times that for light polarized along the plane of the layers) the contribution of heavy excitons can be deduced when the light exciton contribution is subtracted (b).

From this analysis two parameters can be deduced: the BE and the OS of both heavy and light excitons. The BE is directly determined from the difference between the energy position of the 1*s* excitonic peak and the position of the continuum. The OS per unit area is calculated from the integrated absorption over the exciton peak.

By varying the values of the parameters which enter in the decomposition we can estimate the accuracy of the energy difference between the exciton and the onset of the continuum to be 1–2 meV. However, the exact determination of the band gap may be more ambiguous since we do not have a complete description of the excited states and the continuum. This might give rise to a scale error for the BE. The determination of the exciton OS is done with good accuracy (estimated to be 10%) since the two parameters, excitonic peak absorption and exciton linewidth, are determined directly from the spectrum without any adjustable parameter.

It is interesting to note that in order to properly decompose the absorption spectrum in the polarization parallel to the layers it is always necessary to take into account a shift of the energy position of the light-exciton transition, i.e., the *ZT* splitting due to exchange interaction in QW's. This excitonic parameter is rarely measured experimentally because of the difficulty in a conventional geometry to propagate light along the QW layer.[15,16] The light-exciton *ZT* splitting in the waveguiding geometry can be directly measured from the difference in the light exciton peak energy position between the two polarizations. The observed *ZT* splitting for the 20- and 30-Å QW's is of the order of 4–5 meV. In the 10-Å QW the transitions are too broadened and the exciton peaks cannot be observed, making the determination of the *ZT* splitting not obvious. On the other hand, for wider QW's the *ZT* splitting has the same order of magnitude as the experimental inaccuracies. The theoretical value can be calculated by using expression (14) in Ref. 17, taking the experimental OS and calculating the overlap integral and

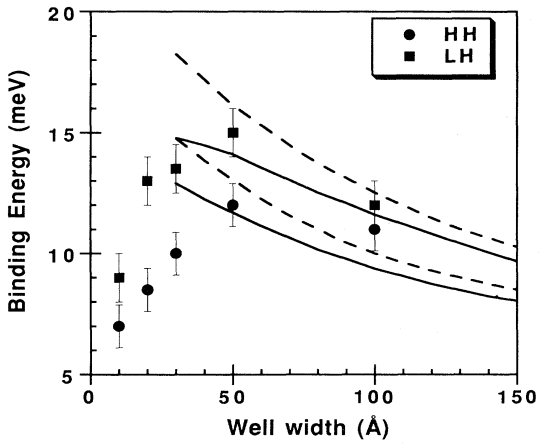

FIG. 4. Exciton binding energies of the 1*s* heavy and light exciton as a function of the QW width compared to theoretical calculations of Ref. 3. The two upper (lower) curves correspond to the calculated light- (heavy-) exciton binding energies for a 25% (solid line) and 40% (dashed line) Al concentration in the barriers.

the integral J. In the case of the 20-Å $GaAs/Ga_{0.7}Al_{0.3}As$ QW we find a ZT splitting equal to 2 meV, a value which is smaller than the experimental one by a factor of 2.

The experimental results for BE and OS of excitons are plotted as a function of the QW thickness in Figs. 4 and 5, respectively. For large QW's the BE's of both heavy and light excitons first increase as the QW width decreases. After reaching a maximum, the BE's decrease for a QW thickness less than 50 Å. Indeed, when the QW width is reduced the wave functions of excitons become delocalized in the barriers and the 3D character of excitons is progressively recovered. Then the BE's tend to the value of the bulk $Ga_{0.7}Al_{0.3}As$ barrier. The experimental data are compared to the theoretical results of Ref. 3 and a good agreement is obtained with the calculated values, i.e., for QW widths larger than 30 Å. Unfortunately, the approximations made in the theory are clearly not adequate when the QW width is further reduced and no exact calculations exist in the limit of narrow QW's, where a decrease of the BE's is expected. Let us note that the light-exciton BE is expected to decrease at a larger QW width than the heavy-exciton BE since light excitons are more rapidly delocalized in the barriers. Such a tendency is not observed experimentally.

A similar behavior for the exciton OS is observed in Fig. 5. The ratio between light- and heavy-exciton OS's in the polarization parallel to the layers is equal to 2, as predicted by theoretical results,[3] which took into account the valence-band mixing for QW widths larger than 30 Å.

IV. CONCLUSION

The optical-absorption experiments in a waveguiding geometry allowed us to determine accurately the heavy- and light-exciton binding energies and the oscillator strengths in $GaAs/Ga_{0.7}Al_{0.3}As$ QW's with thicknesses ranging from 10 to 100 Å. Our experimental results are in agreement with the recent theoretical calculations made for QW widths larger than 30 Å and experimental data are obtained for very thin QW widths. The binding energies that have been determined are strong even for narrow QW's. A decrease of both excitonic BE and OS is observed with decreasing well width, demonstrating the crossover from the 2D to the 3D character of excitons.

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