Exciton localization by potential fluctuations at the interface of InGaAs/GaAs quantum wells

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Photoluminescence measurements in InGaAs/GaAs quantum wells, both doped and undoped, have provided evidence of a bound exciton of intrinsic origin with peculiar properties. The intensity of this bound exciton exhibits an unusual dependence on the exciting power density. Its relative strength with respect to the free exciton is strongly modified by small changes of the excitation wavelength. Its linewidth closely matches that measured for the heavy-hole free exciton, independent of well width and indium concentration. These features lead one to conclude that the intrinsic bound exciton is localized by a photoassisted interfacial roughness. Possible microscopic origins of such a localizing potential are also tentatively given.

I. INTRODUCTION

The potential fluctuations at interfaces (usually referred to as interface roughness) are an extrinsic and annoying feature of the semiconductor heterostructures since they strongly influence the optical properties of quantum-well (QW) structures. Although interface roughness is supposed to be reduced by improving the growth technique, a certain degree of disorder at the interface is an intrinsic and unavoidable feature of the heterostructures, which makes them different from bulk materials. One of the most important effects induced by the interface roughness is the localization of excitons.[1] So far, the presence of such intrinsic defects in QW structures has been usually inferred from the presence of a Stokes shift between the exciton peak energies in the photoluminescence (PL) and the PL excitation (PLE) spectra,[1,2] although this picture has been recently a matter of debate.[3]

In GaAlAs/GaAs QW's, resonant Rayleigh scattering[4] and four-wave mixing[5] have shown the presence inside of an inhomogeneously broadened excitonic absorption band of a mobility edge with localized states lying on the low-energy side of the band. As a matter of fact, highly spatially resolved PL spectra (resolution better than 1 μm) (Refs. 6 and 7) and have recently shown that the narrow ($\sim$ 1 meV) heavy-hole free exciton (HHFE) band[8] commonly observed in GaAs/AlGaAs QW's results from the convolution of many very narrow lines [full width at half maximum (FWHM) less than one tenth of a meV]. Those lines have been claimed to be associated to excitons localized at the GaAlAs/GaAs interfaces. On the other hand, conventional, i.e., spatially unresolved, PL spectra sometimes exhibit several inhomogeneously broadened exciton peaks. Most authors have attributed those multiplets to recombination of excitons localized in regions of the quantum well, which differ in thickness by one or more monolayers and have lateral extent of the order of, or larger than, the exciton diameter,[9] a picture often questioned.[10] A roughness on a smaller lateral scale, with several well widths within the exciton diameter, is claimed, instead, to give rise to a luminescence with FWHM of several meV. In InGaAs/GaAs quantum wells, however, it has been recently shown that a roughness on a small scale is consistent with narrow ($\sim$1 meV) PL line shapes.[11]

In this paper, we report the systematic observation of a doublet in the conventional PL spectra of a large set of strained $In_xGa_{1-x}As/GaAs$ quantum wells grown by molecular beam epitaxy (MBE). The doublet is made up of the recombination of the heavy-hole free exciton and of a lower-energy line, BE_i (where BE is bound exciton). The comparison with carbon-doped samples allows us to state that the low-energy structure is not associated with an exciton bound to an impurity. The BE_i band shows some peculiar features: (i) its intensity has an unexpected dependence on the exciting power density, quite different from that observed for excitons bound to extrinsic impurities; and (ii) its relative weight in the doublet line shape is strongly modified by slight changes of the excitation energy. The overall behavior of the BE_i features points toward an exciton localization due to disorder at the InGaAs/GaAs interface.

II. RESULTS

Although the results reported in this paper have been observed in tens of samples with different alloy composition, we will refer mainly to samples with indium content in the alloy $x = 0.09$ (well width $1 \leqslant L \leqslant 25$ nm) and $x = 0.18$ (well width $1 \leqslant L \leqslant 14$ nm). These two sets of samples are those used in Ref. 11 to investigate the FWHM of the HHFE. In general, single- and multiple-quantum-well structures

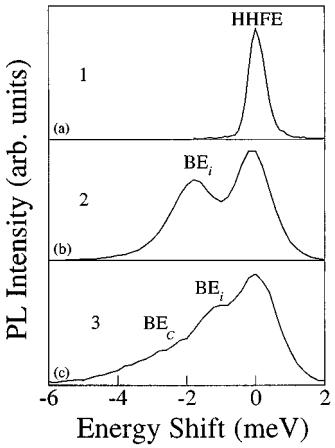

FIG. 1. Photoluminescence at $T = 5$ K of three different quantum wells. The energy is measured with respect to that of the HHFE: (a) $x = 0.09$, $L = 1.5$ nm, undoped (HHFE peak energy = 1.504 eV); (b) $x = 0.11$, $L = 4.0$ nm, undoped (HHFE peak energy = 1.468 eV); (c) $x = 0.10$, $L = 20$ nm, C doped (HHFE peak energy = 1.444 eV).

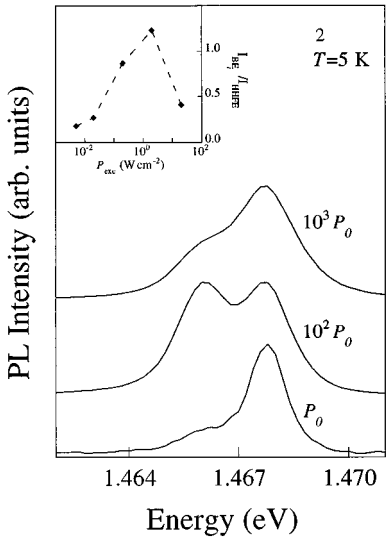

FIG. 2. Photoluminescence spectra of a quantum well with $x = 0.11$, $L = 4$ nm for different values of the exciting power density $P_{\rm exc}$. $P_0 = 2\times10^{-2}$ W cm^{-2}. The ratio between the integrated intensities of the BE$_i$ and HHFE recombinations is reported in the inset vs $P_{\rm exc}$.

have been investigated, undoped and carbon doped. The undoped samples were grown by MBE in a Varian GenII machine at 520 °C on GaAs(100). Carbon-doped samples, grown for other purposes in a different machine of the same type at 580 °C, have been studied in order to verify the intrinsic origin of the bound-exciton recombination object of this paper.

The PL spectrum at $T = 5$ K of sample 1 ($x = 0.09$, $L = 1.5$ nm) is shown in Fig. 1(a). The luminescence consists of a single band due to the recombination of a HHFE with FWHM equal to 0.6 meV. It has been already mentioned that $In_xGa_{1-x}As/GaAs$ quantum wells may have very narrow photoluminescence bands, despite the fact that they are strained and have an alloy as well material. The HHFE origin of the band reported in Fig. 1(a) is supported by the temperature dependence of its peak energy, which closely follows that reported for the free exciton of bulk GaAs. Indeed, when exciton recombination has a strong contribution from a bound exciton, the energy shift of the PL peak with the temperature gets smaller because the PL gradually evolves from being dominated by the BE recombination, at low temperature, to being dominated by the HHFE recombination, at high temperature. No evidence has been found in the spectrum shown in Fig. 1(a) of multiplet structures due to formation of large terraces at the interfaces, or to extrinsic recombination at impurity sites.

The PL line shape at $T = 5$ K changes when the well width and/or the In content increase. In Fig. 1(b), the photoluminescence of sample 2 ($x = 0.11$, $L = 4$ nm) is reported. The photoluminescence of the carbon-doped sample 3 ($x = 0.10$ and $L = 10$ nm) is shown in Fig. 1(c). The emission exhibits a triplet of states, with the C-related emission BE$_C$ 3 meV below that of the HHFE, and 1.8 meV below that of the BE$_i$.

The low-energy band of the doublet shown for 2 behaves like a bound-exciton recombination and disappears at temperature around 30 K. However, it is not due to recombination of an exciton bound to unintentional impurities, as becomes clear by comparison with the results obtained in the C-doped samples. We point out that carbon is the typical unintentional impurity in GaAs-based materials grown by MBE. We therefore might expect recombination from excitons bound to extrinsic, unintentional impurities at the energy typical for the BE$_C$.

As far as the intensity of the BE$_i$ is concerned, it is weak for very narrow wells [and even not observed for small values of x—see, e.g., Fig. 1(a)], then it becomes stronger and stronger as the well width increases, and finally it weakens again for very wide wells. For samples with $x = 0.09$, the BE$_i$ recombination is no longer observed for $L > 20$ nm. For $x = 0.18$, the recombination is weaker in the 14-nm-wide QW than in the 7-nm-wide QW.

The dependence of the BE$_i$ intensity on the exciting power density $P_{\rm exc}$ is quite peculiar, as shown in Fig. 2 where $P_{\rm exc}$ changes over three orders of magnitude. At $P_{\rm exc} = P_0 = 0.02$ W cm^{-2}, the HHFE band dominates the spectrum and grows linearly with $P_{\rm exc}$, while the BE$_i$ intensity increases almost quadratically with $P_{\rm exc}$. At power $10^2\times P_0$, the two signals are comparable and both grow linearly with $P_{\rm exc}$. At $P_{\rm exc} = 10^3\times P_0$, the intensity of the BE$_i$ saturates and the HHFE dominates again. A fit to the data has been performed in order to obtain the integrated intensity of the different bands. The ratio between the integrated intensity of the BE$_i$ and of the HHFE is reported vs $P_{\rm exc}$ in the inset of Fig. 2. This behavior of the BE$_i$ recombination with the exciting power has been observed in all samples, undoped and C doped. It has been observed also for several excitation wavelengths, 458 nm, 720 nm, and 830 nm, the latter corresponding to an energy below the GaAs band gap.

The behavior reported in Fig. 2 for medium to high $P_{\rm exc}$ is that typically observed for bound excitons: for increasing excitation power the number of occupied states approaches the number of impurity sites and the intensity of the BE recombination relative to that of the HHFE becomes

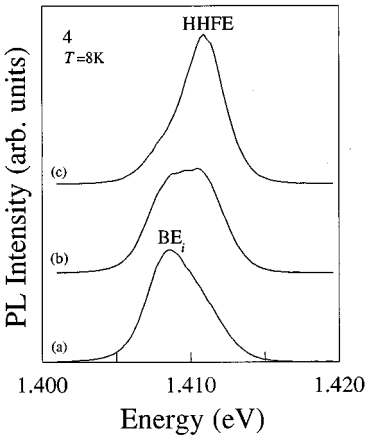

FIG. 3. Photoluminescence spectra of a quantum well with $x = 0.185$ and $L = 4$ nm (sample 4) for three different values of the excitation energy $E_{\rm exc}$. (a) $E_{\rm exc} = 1.473$ eV; (b) $E_{\rm exc} = 1.467$ eV; (c) $E_{\rm exc} = 1.463$ eV.

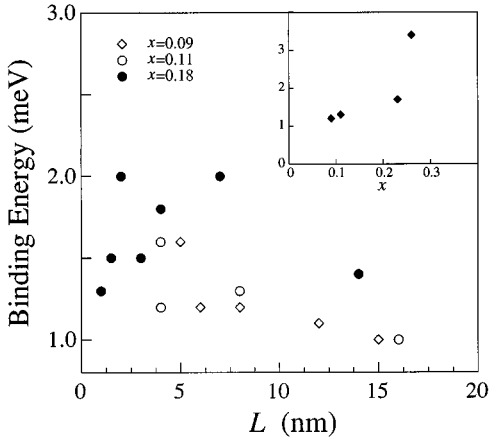

FIG. 4. Binding energy of the BE_i state measured with respect to the HHFE state for samples with $x = 0.09$, 011, and 0.18. In the inset, the BE_i binding energy at fixed well width $L = 8$ nm is reported vs indium concentration x.

smaller and smaller. This is, indeed, the behavior we observe for the C-related recombination in C-doped samples, as will be shown in the following. The behavior reported in Fig. 2 at low $P_{\rm exc}$ is instead unexpected and typical of the BE_i band observed in this work. It is worth mentioning that, in some samples, at very low excitation power the BE_i intensity vanishes.

Another peculiar feature of the PL doublet in the undoped samples is the strong change in the relative intensity of the BE_i and HHFE recombinations for small changes ($\leqslant 10$ meV) of the excitation wavelength, as shown in Fig. 3. The integrated intensity of the three spectra is almost the same. The complicated behavior shown in Fig. 3 does not depend on the sligthly different exciton densities connected to the different excitation wavelengths. In fact, we found essentially the same behavior when the exciting power density was changed within one order of magnitude. Therefore, the dependence of the PL spectra on excitation energy shown in Fig. 3 cannot be ascribed to the dependence of the BE_i/HHFE intensity ratio on the exciting power density.

The value of the binding energy of the BE_i, as measured with respect to the HHFE, is reported in Fig. 4 as a function of L for three values of In concentration ($x = 0.09$, 0.11, and 0.18). Whenever the doublet structure was not well resolved at low T, a deconvolution of the PL spectrum has been done in terms of two bands. Despite a sizable scattering, the data in Fig. 4 show that the BE_i binding energy exhibits a maximum vs L for $x = 0.18$, as expected on the grounds of the dependence on L of two competing effects: carrier localization and wave-function penetration into the barrier. Moreover, from Fig. 4, at all L's the binding energy increases with x. This is confirmed by the inset, where the BE_i binding energy is reported as a function of x for a set of samples with $L = 8$ nm.

Finally, we report in Fig. 5 the FWHM of the BE_i line. The FWHM of BE_i closely matches that of the HHFE recombination (also shown in Fig. 5) and shows a maximum for intermediate well widths. As will be shown in the following section, this feature points toward a direct influence of interface roughness and alloy disorder on the BE_i recombination.[11]

III. DISCUSSION

Let us now discuss the origin of the BE_i recombination. As far as BE_i line shape and binding energy are concerned, recombinations similar to the BE_i have been already reported in the PL spectra of undoped InGaAs/GaAs QW's. Those recombinations have been tentatively attributed to an exciton bound to unintentional doping,[12] to biexcitons,[13] or to bound-to-bound transitions.[14] The presence of exciton localization induced by alloy disorder has been claimed in InGaAs/GaAs QW's on the grounds of an anomalous behavior with temperature, well width, and indium concentration of the linewidth of the HHFE band.[15] A similar kind of exciton localization, also attributed to alloy fluctuations, has been observed in indirect-gap $Ga_xAs_{1-x}P$.[16]

In our samples, it can be ruled out that the BE_i recombination is due to an exciton bound to an unintentional dopant on the grounds of the measurements made in samples intentionally doped with carbon — the typical unintentional impurity in MBE grown GaAs. Photoluminescence spectra

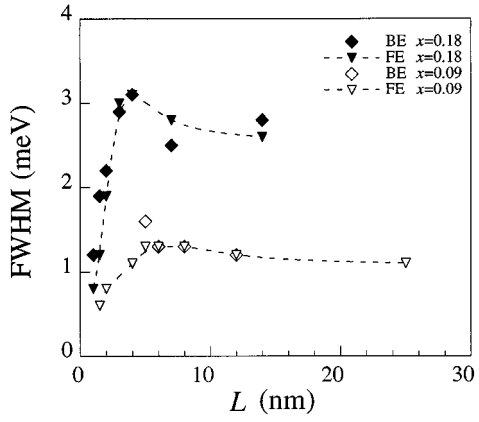

FIG. 5. FWHM of the HHFE and BE_i luminescence vs the well width for $x = 0.09$ and $x = 0.18$.

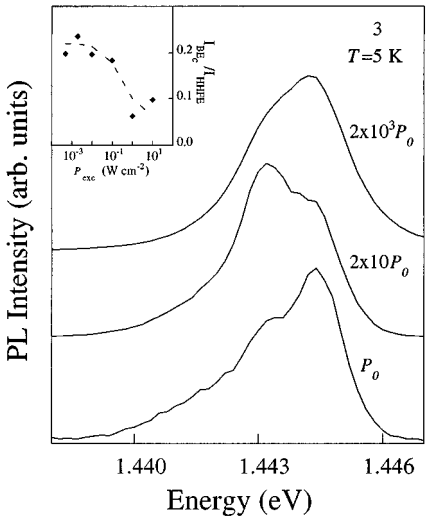

FIG. 6. Photoluminescence spectra of a quantum well with $x = 0.10$, $L = 10$ nm for different values of the exciting power density. $P_0 = 5\times10^{-4}$ W cm^{-2}. The ratio between the integrated intensities of the BE_C and HHFE recombinations is reported in the inset vs P_{exc}.

taken at different values of the exciting power density are shown in Fig. 6 for the *C*-doped sample 3, already shown in Fig. 1(c). The BE_i recombination is clearly resolved and exhibits as a function of P_{exc} the same anomalous behavior observed in the undoped samples. In the spectra taken at the lowest power, the BE_C recombination appears as a shoulder $\sim$3 meV below the HHFE band, a value very close to that reported for excitons bound to neutral carbon in GaAs. As shown in the inset, the intensity of this recombination follows the behavior normally observed for an exciton bound to an extrinsic impurity. At low exciting power density, it increases linearly with P_{exc}, as the HHFE band does, and saturates at the highest value of P_{exc}. It is also worth stressing that the BE_i structure has been systematically found in a very large set of samples grown at different temperatures with different MBE machines, as well as in samples grown with and without the growth interruption technique. Therefore, it is quite unlikely that other less common, unintentional impurities are present in all those different samples.

The measurements made as a function of P_{exc} show that the intensity of the BE_i increases quadratically at low excitation. This might support the attribution of this recombination to biexcitons or to trions, a complex formed by three mutually interacting carriers, which has been often claimed to account for new PL lines.[17] Moreover, the BE_i binding energy measured in the present work is consistent with that of biexcitons in GaAs QW's ($\sim$1 meV),[18] as well as with that of trions ($\sim$2 meV).[19] However, the saturation of the BE_i intensity with exciting power density observed in the present work calls for biexciton (trion) ionization at high particle density or biexciton (trion) localization at a finite number of centers. Those features, as well as the dependence of the BE_i/HHFE intensity ratio on the incident photon energy can be hardly understood in a "simple" biexciton (trion) frame.

As an alternative, more consistent explanation, we suggest that the BE_i recombination arises from excitons weakly localized by the potential fluctuations generated by the interface roughness. As mentioned above, the FWHM of the BE_i recombination closely follows that of the HHFE recombination. In the same InGaAs/GaAs QW's reported here, it has been shown[11] that the FWHM of the HHFE is explained in terms of the disorder induced by the interface roughness and by the random composition fluctuations of the alloy in the well. It is quite likely that the same mechanisms are responsible also for the FWHM of the BE_i. For wide wells the interface roughness gives a minor contribution to the FWHM and we find, indeed, that the contribution to the doublet of the BE_i recombination weakens. This suggests as well that the potential fluctuations responsible for the localization of the BE_i are mainly due to interface roughness. The weakening of the BE_i for very narrow well widths can be attributed, instead, to the great extent of the electronic wave functions in the barriers. Indeed, the intensity of the interface contribution goes to zero for $L = 0$ as it happens for the influence of interface roughness on the FWHM.[11] It may be worth recalling that a weak recombination 0.7 meV below the HHFE peak has been reported in 0.57-nm-wide undoped $In_{0.1}Ga_{0.9}As/GaAs$ QW's.[14] This recombination has been tentatively attributed to a bound-to-bound transition involving a shallow bound electron and a shallow bound hole more closely spaced than they are in the HHFE, although the exact nature of the centers involved has not been clarified. Very recently, an intrinsic bound exciton has been observed for $x \geqslant 0.15$ and attributed to indium accumulation at the InGaAs/GaAs interface.[20] However, the values of the binding energy of the BE_i found in the present work are much smaller than those reported in Ref. 20 (e.g., 7 meV for $L = 8$ nm and $x = 0.2$, to be compared with a value smaller than 3 meV in the present work, see Fig. 5). Moreover, it has to be noticed that the binding energies of the BE_i recombination observed in our work lie within the FWHM of the band attributed to HHFE in Ref. 20.

In the case of InGaAs/GaAs coupled-quantum-well structures, a disorder in the growth direction, due to indium flux fluctuations during the growth of different wells, has been claimed to give rise to localized holes, which then recombine with free electrons.[21] This picture cannot apply in our case. Indeed, we have observed exciton localization also in single QW's, where disorder can only occur in the growth plane, due to interface roughness.

As for the lateral extent of the potential fluctuations giving rise to the BE_i, an exciton localization of the type described in this work can be only understood in terms of a fluctuation scale much smaller than the exciton diameter. Potential fluctuations on a scale greater than the exciton diameter would bring us, indeed, to the observation of several peaks due to HHFE recombination in different parts of the sample, as reported, e.g., in Ref. 9. A roughness on a small scale is consistent with the analysis of the FWHM of the HHFE, from where a lateral extent of the interface roughness of about 2 nm has been inferred,[11] i.e., a value smaller than the exciton diameter ($\sim$30 nm).

The microscopical origin for the BE_i recombination remains to be explained. The quadratic dependence of the BE_i intensity on the exciting power density suggests that photogenerated carriers might induce a lattice readjustment that gives rise to centers capable of localizing an exciton.

These centers are most likely formed at the interfaces, where the lattice mismatch and the ensuing strain favor the formation of weak bonds. This picture could also explain the formation of deeper fluctuations for increasing x, at fixed L, reported in Fig. 4. It may also be worth mentioning here that near-edge states have been invoked to explain a PL band observed in hydrogenated bulk GaAs, from 50 to 90 meV below the GaAs band edge.[22] In that case, the insertion into the GaAs lattice of an H atom gives rise to a lattice deformation and to the insurgence of weak bonds, the possible origin of near-edge donorlike and acceptorlike states. In the present case, photogenerated carriers and weak bonds should give rise to donorlike and acceptorlike states localized at the interface where excitons (or electrons and holes) could be trapped. The possibility that the superlinear behavior at low exciting power density could be due to an initial saturation of nonradiative states can be ruled out, instead, on the grounds of the observations made on the BE_C state. In fact, in the same samples where the BE_i/HHFE intensity ratio shows a maximum with P_{exc}, the BE_C/HHFE ratio exhibits the standard behavior for extrinsic impurities: it remains constant at low P_{exc} and decreases at high P_{exc}.

We have in progress extensive PLE and time-resolved measurements, which should help us to clarify the origin of the BE_i, as well as to explain its quite complex carrier dynamics — see, e.g., the dependence of the PL spectra on excitation wavelength reported in Fig. 3.

IV. CONCLUSIONS

An intrinsic PL band, BE_i, has been observed on the low-energy side of the heavy-hole free exciton in the low-temperature photoluminescence spectra of InGaAs/GaAs quantum wells. On the grounds of a strong analogy between the line shape of this recombination and that of the heavy-hole free exciton, we suggest that the BE_i band is due to an exciton bound to potential fluctuations at the interface whose detailed microscopic origin remains to be understood. The peculiar dependence of the bound exciton intensity on the exciting power density indicates that exciton localization centers are activated by the carriers photoexcited in the quantum-well structure.

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