Influence of interface localization on the binding energy of acceptor bound excitons in narrow GaAs/$Al_xGa_{1-x}As$ quantum wells

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The role of interface roughness in bringing about localization of the free exciton in narrow GaAs/$Al_xGa_{1-x}As$ quantum wells is well known. In this paper we investigate the interaction of the localization potential due to interface roughness with that due to the presence of a shallow acceptor impurity. Using a selective excitation technique we are able to accurately measure the bound exciton binding energy in a narrow quantum well, thereby determining the effect of interface localization on the exciton to impurity binding energy. The binding energy decreases with increasing localization strength (due to interface roughness), a localization energy of 3 meV with respect to the free-exciton energy results in a decrease by 1.2 meV in the local binding energy of the bound exciton. The rapid diffusion of weakly localized free excitons is also demonstrated and contrasted with the absence of a similar migration process for bound excitons.

I. INTRODUCTION

The structure of the interface between two sequentially grown semiconductors viewed at the atomic level has been shown to be of primary importance in determining the physical behavior of structures with nanoscale dimensions.[1] An extremely powerful tool in the analysis of this interface has proven to be the measurement of low-temperature photoluminescence (PL).[2-4] The low-temperature emission spectra of these quantum structures are dominated by exciton recombination. A rough interface manifests itself in this recombination as an inhomogeneous broadening of the exciton linewidth. Since the confinement energy of the exciton is dependent on the layer thickness, variations in average thickness due to interface roughness results in a distribution of regions with a specific average thickness (islands) and a spread in the effective band-gap energy. The diffusion of excitons between these islands will favor the lower-energy sites. This description has led to a picture of localization of excitons at interface roughness potentials. Experimentally this localization is most clearly observed as a substantial Stoke's shift between the optical emission and absorption spectra.[5,6] As the dimensions of the structure are reduced, the interface volume (defined by the fluctuation in the position of the interface, for example between the GaAs well and the $Al_xGa_{(1-x)}As$ barrier) forms an increasing proportion of the total active volume, with the result that exciton localization becomes increasingly dominant at low temperatures.

It is important to realize that although the physical dimensions of the roughness may remain comparable (of the order of ± 1 atomic layer perpendicular to the interface), the corresponding fluctuation in potential energy of a confined state due to areas of different average well width L_z will increase with decreasing well width, $dE/dz = -\hbar^2/m^*(1/L_z)^3$, where L_z is the well width and m^* is the effective mass of the confined electron or hole. For narrow quantum wells it is usually no longer possible to resolve the localized free exciton from the impurity bound exciton state in PL spectra. The extent of the interface roughness is found to be strongly dependent on the growth conditions used; in particular, the use of interrupted growth at the interface has been shown to produce extended atomically flat islands (typically >1-μm diameter) which differ in average thickness by a single monolayer.[7] In this case excitation spectra have been obtained with an extremely narrow linewidth, indicating the existence of minimal localization.

The random distribution in local potentials at an interface is reflected in a corresponding distribution of localized states. For a given localized exciton which is not at the minimum potential energy, there exists the possibility of relaxation into lower-lying states via diffusion. The exact mechanism for this migration process has received considerable interest.[8,9] Within each island, the exciton occupies a local minimum in potential energy, and movement to an island defining a lower potential energy will in general require the participation of an acoustic phonon or other scattering mechanisms such as interface rough-

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ness. This mechanism results in the observation of a thermally activated diffusion coefficient.[10]

In addition to the localization potential due to interface roughness there also exists a random distribution of impurity related potentials; in a doped quantum well this reaches a high density within the doped region. The trapping of an exciton at an impurity leads to the conventional picture of the bound exciton (BE). Just as a localized exciton is able to relax through diffusion to states of lower energy, it follows that an exciton can become bound at an impurity during the same mechanism. An impurity can be located at any position in the plane of the quantum well and therefore within any average local potential due to interface roughness. As we shall discuss in this paper, the final extension over which the localization potential is averaged is determined by the bound exciton extension. A distribution of localized free-exciton states necessarily implies a corresponding distribution in impurity bound exciton states. To date, the interaction of the localization potential due to interface roughness with that of an impurity intentionally placed in a narrow quantum well (QW) has not been studied, to our knowledge. In this paper we demonstrate that the interaction of these two potentials has a strong effect on the binding energy for an exciton bound at an impurity, and in addition that the exciton kinetics are also determined by this interaction.

The investigation has been carried out using 50 Å QW's doped with Be in the center of the well. The narrow QW implies that the excitons interact strongly with the interfaces, and the optical spectra as a result demonstrate a large linewidth. This linewidth corresponds to an envelope over a large number of transitions corresponding to localization of the exciton in interface potentials of different depth. Using a selective excitation technique, it is possible resonantly to produce excitons with a specific localization energy. In this way we create an exciton population which is well defined in energy, and are able to study the subsequent capture at a correlated impurity potential. There exists an effective spatial correlation between the resonantly excited localized exciton and that subset of impurities which are able to capture it. The correlated, impurity bound exciton population appears in the optical spectra as a strong peak at a separation corresponding to the impurity exciton binding energy from the resonant laser excitation. Using such a technique we are thus able to make a very accurate measurement of the binding energy. Furthermore, we are able to measure the dispersion in exciton binding energy due to the coupling of the impurity potential with the localization potential related to the interface roughness.

Section IV of this paper looks more closely at the kinetics behind the localization and interaction with impurities, using a picosecond-time-resolved photoluminescence technique. In particular we highlight the correlation between capture rate at impurities and the degree of localization of the resonantly excited exciton.

II. EXPERIMENTAL METHOD

Measurements have been carried out on a 50-Å GaAs-$Al_xGa_{(1-x)}As$ multiple-quantum-well sample prepared using molecular-beam epitaxy (MBE) with noninterrupted growth at 680 °C. An undoped GaAs buffer layer followed by a short period superlattice are included before the 50 quantum-well periods, to minimize background impurity level in the active region. The Be dopant was introduced into each well in the central 10 Å only, at a concentration of 3×10^{10} cm^{-2} per well.

Low-temperature time-integrated photoluminescence spectra have been measured using a 1-m Spex double monochromator to disperse the luminescence, which is then detected using a cooled GaAs photomultiplier. The samples are mounted in a helium bath cryostat which is pumped to take the helium below the λ point, giving an approximate measurement temperature of 1.8 K. Tunable excitation is provided by a cw Ti:sapphire solid-state laser pumped using an Ar^+-ion laser. Time-resolved photoluminescence measurements have been carried out using a Hamamatsu synchroscan streak camera. Pulsed excitation is provided from a tunable dye laser with Styryl 8 dye synchronously pumped using a mode-locked Ar^+-ion laser; pulse lengths of around 5 ps are achieved. The resulting luminescence is dispersed through a $\frac{1}{4}$-m monochromator. The overall time resolution for the total system is approximately 20 ps.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A series of PL spectra illustrating the development in the emission as the laser excitation photon energy is scanned through the localized exciton emission band is shown in Fig. 1. The top curve in the series shows the broad exciton transition typically observed for nonresonant excitation, and lower curves illustrate spectra obtained for excitation photon energies progressively deeper within the localized exciton band. Excitation resonant with the high-energy tail of the emission results in relatively little change in the observed spectra; excitons excited with this energy remain mobile and able to diffuse among localization islands despite a total energy that is lower than the $K=0$ free exciton. The redistribution of

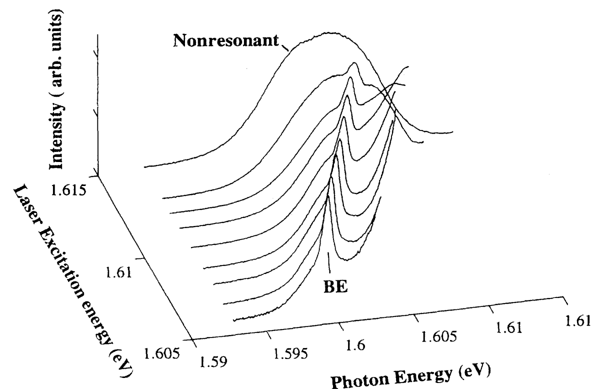

FIG. 1. Series of PL spectra for nonresonant excitation and for a number of energies within the localized exciton (LE) band. The selective population of a subset of bound exciton states results from the resonant excitation technique.

excitons among different islands before recombination results in a line shape which is not significantly perturbed in comparison to the nonresonant case.

As the excitation photon energy is decreased, excitons are excited within increasingly strongly localized regions from which they are less able to diffuse; as a result, a distinct bound exciton (BE) peak emerges as excitons are trapped at acceptors only within these regions. The extent to which localized excitons are able to move between islands is illustrated by the low-energy tail to the BE. Stronger localization results in a sharper BE peak. The transition between a delocalized exciton for which there is no distinct bound exciton peak resolved and the first appearance of such a peak therefore approximately corresponds to the exciton mobility edge.[5,11] A distinction for the mobility edge defined in this way results from the fact that we observe the combined mechanisms of diffusion and capture to an impurity. A weakly diffusing exciton can be captured before it has time to move to a neighboring island. The mobility edge identified in this way should therefore be at higher energies than that identified in the work of Hegarty, Goldner, and Sturge.[5] We shall return to this discussion later in the paper. The significance of this work can be summarized in that we identify discrete populations for both the localized exciton and the correlated bound exciton, and thereby determine the perturbation of the impurity-exciton binding energy due to the interface localization potential containing the acceptor impurity.

Figure 2 illustrates the significant decrease in binding energy for the acceptor bound exciton as the excitation photon energy is decreased through the localized exciton band. The effect of stronger exciton localization is to play an increasing role in the binding of the bound exciton, while the impurity potential no longer entirely determines the extension of the bound exciton. The calculations of Bastard *et al.*[12] associate increasing exciton localization energy with a corresponding increase in the radius of the localization island. For decreasing exciton localization energy the island size becomes comparable to the spatial extension of the acceptor bound exciton. In this limit, the total confinement in the plane of the QW is

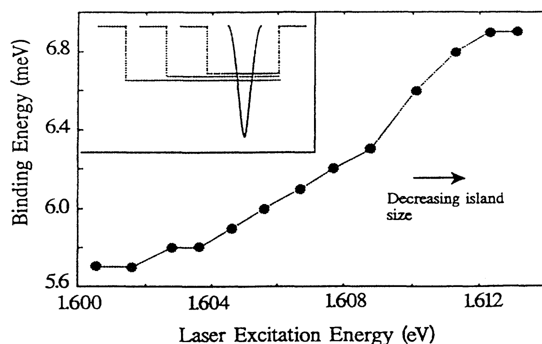

FIG. 2. Dispersion in bound exciton binding energy with laser excitation energy. The binding energy shows a significant decrease for bound excitons formed from strongly localized free excitons.

increasingly influenced by the contribution of the localization potential over that due to the impurity potential (see Fig. 2, inset). Using the calculations of Bastard *et al.* as an estimate, an island size corresponding to the approximate extension of the acceptor bound exciton (70 Å) would imply a localization energy of 2–3 meV, depending on the exact island depth. This corresponds to the limit at which the measured binding energy reaches its maximum in Fig. 2. For stronger localization (i.e. larger island size) of the free exciton the confinement of the bound exciton is defined by the acceptor potential. At this limit the further lowering in energy of the free exciton, due to stronger localization, has in contrast little effect on the impurity bound state. The relative binding energy, measured from the localized exciton to the BE is therefore decreased. Following this argument through, a still smaller island dimension should further increase the confinement of the acceptor. However, it is not possible to accurately determine from these measurements whether the binding energy has a maximum at nonzero localization energy.

It is important to note the consequences this model would have for a donor-doped system. The larger Bohr radius of the donor implies that an interface localization potential which already strongly localizes the free exciton can also still affect the binding energy of the exciton at the donor. As a consequence the donor exciton binding energy may actually be increased for weak localization. This result may explain why there has been some discrepancy in the measured value for the donor bound exciton reported in the literature.[13–15]

Finally we discuss the capture and relaxation mechanisms observed during recombination of the localized and impurity bound excitons. The process of relaxation corresponds to the migration of excitons among island structures of successively lower potential energies. The energy mismatch is compensated for by the emission of acoustic phonons during the transfer process. The scattering mechanism responsible for the movement of excitons occurs via interaction with acoustic phonons or interface roughness; this results in an activated behavior for the diffusion of excitons. Correspondingly, the relaxation rate, or the rate of further localization, is a relatively slow process. The decreasing rate of relaxation with increasing localization is responsible for the logarithmic dependence typically observed in systems for which strong localization plays a role; see, for example, Ref. 16.

Figure 3(a) is a projected three-dimensional (3D) image which illustrates the development in time and energy of the PL spectra following resonant excitation in the high-energy tail of the emission. The relaxation of the excited exciton population via localized states is clearly observed; similarly, the direct excitation of weak BE's can be seen at lower energies. Figure 3(b) summarizes the same data in the form of a contour plot, which directly allows a more quantitative analysis to be made. A small population is found to remain at the free-exciton energy, but decays rapidly as further localization and radiative recombination occurs. As indicated by the dotted line, there is no shift in energy of the free-exciton transition. In contrast, the peak energy of the localized exciton moves rap-

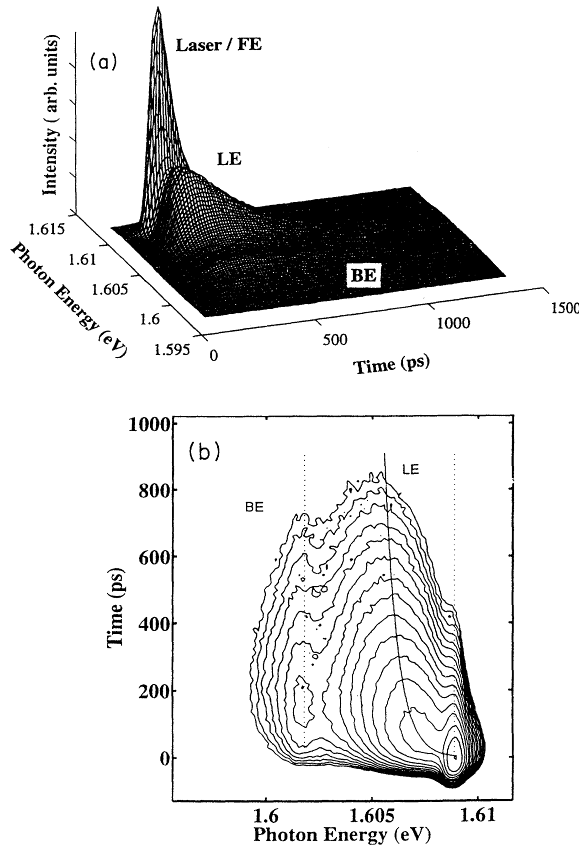

FIG. 3. (a) Projected 3D image illustrating the development in time of the PL spectra following resonant excitation in the high-energy tail of the emission. (b) Contour plot of the data in (a). The solid line is a fit to the shift in peak energy using an equation of the form $E = E_0 - \alpha \ln(t+1)$, where E_0 is the peak position at $t=0$, and illustrates the logarithmic shift in peak energy due to an acoustic-phonon-assisted migration to lower energies.

idly to lower energy as excitons diffuse by hopping to regions of lower potential energy, thereby emitting an acoustic phonon. The solid line is a fit of the logarithmic dependence expected for an acoustic-phonon-assisted migration to lower energies.[16] Finally, the BE peak again shows no shift in energy, although a low-energy tail is seen to develop. This demonstrates two important points: (i) Once trapped, the impurity bound exciton no longer diffuses, since at this doping level there is no interaction between impurities allowing the exciton to relax via diffusion in an impurity band. (ii) The most efficient capture to impurities occurs for the more weakly localized excitons, i.e., localization inhibits impurity capture. A detailed discussion of this important result is given in Ref. 17.

IV. SUMMARY AND CONCLUSIONS

In a narrow quantum well the broad distribution of excitons localized due to interface roughness results in a broad PL emission spectrum for nonresonant excitation, where impurity bound excitons are unresolved from the localized free exciton. The use of selective excitation within the localized exciton band limits the distribution of possible states occupied. The resulting luminescence occurs only from this subset of states, and allows distinct components within the original nonresonant spectra to be resolved. In this way bound excitons associated with impurities in different average local potentials are identified and the influence of the coupling of the two potentials investigated. Using this technique we have measured the dispersion in binding energy of the bound exciton due to the effect of localization. The origin of this dispersion is the difference in the effect of localization on the free and bound excitons. Since the bound exciton is additionally confined by the impurity potential, localization in the 2D plane only plays a significant role when the dimensions of this localization are comparable to the excitonic extension. Enhanced electron-hole correlation due to small localization dimensions may result in an increased binding energy of the bound exciton. For islands larger than the free-exciton volume the effect is equivalent for both free and bound excitons and there is no further dispersion in relative binding energy. An analogy with this mechanism can be made by comparing the dependence of the interaction of an impurity potential with the confinement potential in the growth direction. The decreasing binding energy of the bound exciton relative to the free exciton associated with impurities placed further from the center is due to the increasing difference in the interaction with the confining potential between free and bound excitons. The exciton dynamics observed using a picosecond-time-resolved measurements illustrate the diffusion of weakly localized excitons, but the absence of significant hopping for bound excitons.

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