Low-temperature exciton trapping on interface defects in semiconductor quantum wells

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We present the results of low-temperature photoluminescence experiments performed on GaAs single quantum wells grown by metal organic chemical-vapor deposition. The luminescence line is down shifted by a few milli-electron-volts below the $n=1$ heavy-hole exciton absorption peak. This behavior is interpreted in terms of exciton trapping on interface defects. A simple model provides reasonable values for the exciton binding energy on these defects as well as insights on the lack of thermalization which characterizes the trapped exciton photoluminescence.

Optical properties of semiconductor quantum wells (QW) are now increasingly documented.[1-5] The low-temperature luminescence of high-quality GaAs-$Ga_{1-x}Al_xAs$ multiple QW grown by molecular-beam epitaxy (MBE) has been attributed to excitonic recombination.[2] The width of the excitation spectrum of the luminescence line has been correlated to interface fluctuations[6,7] of a few hundred angstroms in lateral size. Less interest has been paid to the magnitude of the Stokes shift between absorption and luminescence peaks as well as to the thermalization of recombining excitons. In this Rapid Communication, we report further experimental data on low-temperature photoluminescence and excitation spectroscopy of GaAs-$Ga_{1-x}Al_xAs$ single QW grown by metal organic chemical-vapor deposition (MO-CVD). The use of single QW structures eliminates possible fluctuations of the well thickness in multiple QW structures which may broaden luminescence lines. Our experiments are interpreted in terms of exciton trapping on interface defects. A simple model of exciton trapping provides (i) the dependence of the exciton binding energy on the size of these defects, (ii) the shape of the averaged trapped excitons density of states, and (iii) the evaluation of the acoustical-phonon-assisted hopping of an exciton between interface defects.

Figure 1 presents the experimental results obtained in a sample consisting of an n^+-type GaAs substrate, a 1-μm $Ga_{0.48}Al_{0.52}As$, a 7-nm GaAs, and a 7.5-nm $Ga_{0.48}Al_{0.52}As$ epilayers. The luminescence (solid line) and excitation (dashed line) spectra have been obtained at $T=2$ K, using standard cw dye laser and lock-in techniques. The substrate luminescence which occurs below the GaAs band gap (1.519 eV) is not shown on the figure. The excitation spectrum of the QW luminescence exhibits two well resolved excitonic structures corresponding to electron–heavy-hole $[(HH)_1 \rightarrow E_1]$ and electron–light-hole $[(LH)_1 \rightarrow E_1]$ exciton transitions, respectively. The luminescence line, which is 5.8 meV broad full width at half maximum (FWHM) is Stokes shifted by 4.4 meV from the maximum of the $(HH)_1 \rightarrow E_1$ excitonic peak seen in the excitation spectrum. From comparison with investigations carried out in intentionally doped GaAs-$Ga_{1-x}Al_xAs$ QW, it is clear that this luminescence is not extrinsic. No $\exp(\epsilon/kT)$ behavior has been observed in the high-energy part of the luminescence line. Moreover, neither carrier heating nor saturation effects were detectable: the position and the shape of the luminescence line did not vary when the laser intensity was increased from 10 mW/cm^2 to 10 W/cm^2. The excitation spectrum was also found insensitive to the detection wavelength (within the luminescence line). This shows the lack of inhomogeneity occurring on the scale of a carrier diffusion length in the layer plane. We attempt to explain our experimental findings (luminescence linewidth, Stokes shift between photoluminescence and excitation spectra) in terms of exciton trapping on random interface defects. To model such trapping, consider a GaAs QW of average thickness L between

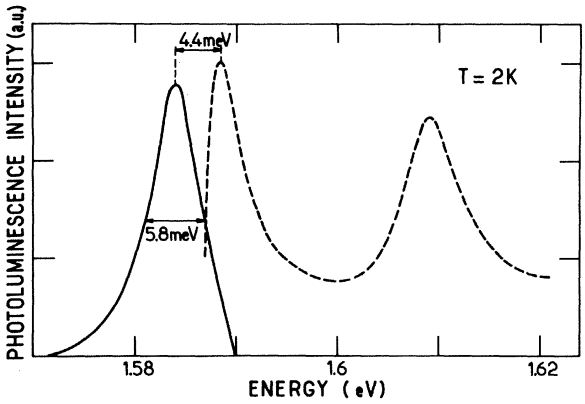

FIG. 1. Photoluminescence spectrum (solid line) and excitation spectrum (dashed line) of a 70-Å-thick single GaAs quantum well at $T=2$ K.

$z=0$ and $z=L$. V_e and V_h are the electron and hole confining barriers, respectively. The exciton ground state of the perfect QW is taken as[8]

$$\psi(\vec{R}_\perp,\vec{\rho}_\perp,z_e,z_h) = \frac{1}{\sqrt{S}}\exp(i\vec{K}_\perp\cdot\vec{R}_\perp)\frac{2}{\pi\lambda^2}\exp\left(\frac{-\rho_\perp}{\lambda}\right)f_e(z_e)f_h(z_h) \quad , \quad (1)$$

where S is the sample area, $\hbar\vec{K}$ the center of mass (c.m.) impulsion of the exciton in the layer plane, and $\vec{\rho}$ the in-plane electron-hole position vector ($\vec{\rho}=\vec{\rho}_e-\vec{\rho}_h$). λ is an effective Bohr radius which, for the wells under consideration, is $\sim 0.8a^*$ where a^* is the bulk exciton Bohr radius. $f_e(z_e)$ and $f_h(z_h)$ are the free QW ground-state wave functions for the electron and hole motions along the growth direction. We consider a semi-Gaussian interface defect of radius a and depth b centered at the origin of the coordinates. This gives rise to an additional potential:

$$\mathcal{H}_d = \epsilon V_e Y(\epsilon z_e)\exp\left(\frac{-\rho_e^2}{2a^2}\right)\exp\left(\frac{-z_e^2}{2b^2}\right) + \epsilon V_h(\epsilon z_h)Y(\epsilon z_h)\exp\left(\frac{-\rho_h^2}{2a^2}\right)\exp\left(\frac{-z_h^2}{2b^2}\right) \quad , \quad (2)$$

where $\epsilon=+1$ refers to excess Ga(Al)As protruding into the well and $\epsilon=-1$ to excess GaAs protruding into the barrier. In Eq. (2), $Y(x)$ is the step function [$Y(x)=1$ if $x\geq 0$, $Y(x)=0$ if $x<0$]. The $\epsilon=+1$ defects can only scatter moving excitons and only $\epsilon=-1$ gives rise to bound exciton states. The central assumption of our calculations is that $\mathcal{H}_d$ mostly affects the c.m. degrees of freedom without admixing the ground state (quasi-1S) of the reduced exciton motion with excited states (quasi-2S, . . .) and *a fortiori* without admixing the ground free well wave functions $f_e(z_e)$, $f_h(z_h)$ with excited QW bound and extended states. This is at variance with previous calculations[3] performed for free particles and infinite V_e, V_h. Also this is well suited to the L's of our interest (50 Å $< L <$ 150 Å) where the energy distance $1S$–$2S$ is markedly increased over the bulk value by QW confinement. We have searched for c.m. binding using a simple trial wave function:

$$\psi_{\rm loc}(\vec{R}_\perp,\vec{\rho}_\perp,z_e,z_h) = \frac{\eta}{\sqrt{\pi}}\exp\frac{-R_\perp^2\eta^2}{2}\frac{2}{\pi\lambda^2}\exp\frac{-\rho_\perp}{\lambda}f_e(z_e)f_h(z_h) \quad , \quad (3)$$

where η is the variational parameter. The exciton binding energy on the defects defined in Eq. (2) is shown in Fig. 2 versus a for several values of b and the parameters ($x=0.52$, $L=70$ Å) corresponding to the experimental data shown in Fig. 1. Two features are noticeable. At large enough a ($a\gtrsim 250$ Å), the variational results become exact and $\psi_{\rm loc}$ corresponds to a two-dimensional harmonic oscillator. At small a, the binding energy vanishes below some critical a_c which is weakly b dependent. This last feature is, we believe, incorrect. The resulting error is, however, not too significant since $a_c\sim 40$ Å: the ill-described traps would have a small binding anyway and, in addition, seem to be statistically improbable.[6]

Knowing the functional dependence of $\epsilon(a,b)$, we can evaluate $\langle\rho(\epsilon)\rangle$, the trapped exciton density of states averaged over the sizes a and b assuming diluted enough interface defects. Very little is known about the statistical distribution of the sizes defects. We have assumed a Gaussian probability density $P(a,b)$, taking a as a continuous variable and b as a discrete one: $b_n=nb_0$ where n is an integer and $b_0=2.86$ Å. We took

$$2\pi a\,da\,P(a,b_n)=2\pi a\,da\,\mathcal{N}\exp\left(\frac{-(a-a_0)^2}{2\alpha^2}\right)\exp\left(\frac{-b_n^2}{2\beta^2}\right) \quad , \quad (4)$$

where $\mathcal{N}$ is a normalization coefficient, a_0 some kind of mean lateral size for a defect, and α,β measure the spread of the distribution. Figure 3 shows the behavior of $\langle\rho(\epsilon)\rangle$ vs ϵ for the same QW parameters as used in Figs. 1 and 2. We have taken $a_0=200$ Å, $\alpha=150$ Å, $\beta=5.72$ Å. These values are not completely arbitrary: sharply peaked distributions which would single out a definite pair (a,b) and thus lead to a highly peaked $\langle\rho(\epsilon)\rangle$ are excluded by our experiments as well as others. As for the value $a_0=200$ Å, it is consistent with previous estimates[6] of the lateral defects size. The choice of β makes the deep protrusions (e.g., $n=4$) rather insignificant. We see in Fig. 3 that $\langle\rho(\epsilon)\rangle$ displays structures associated with the b discreteness. A continuous variation of defect depth, which is in fact not inconsistent with our effective-mass-like treatment, would wash out the structures. Such a smoothened curve is ~ 2.5 meV broad (FWHM) and its maximum occurs ~ 4.5 meV below the edge of the exciton continuum ($\epsilon=0$ in Fig. 2). These calculated values are in qualitive agreement with experiments. (FWHM ~ 5.8 meV; Stokes shift ~ 4.4 meV.) It should be stressed that we compare a theoretical density of states with an experimental luminescence line. This pro-

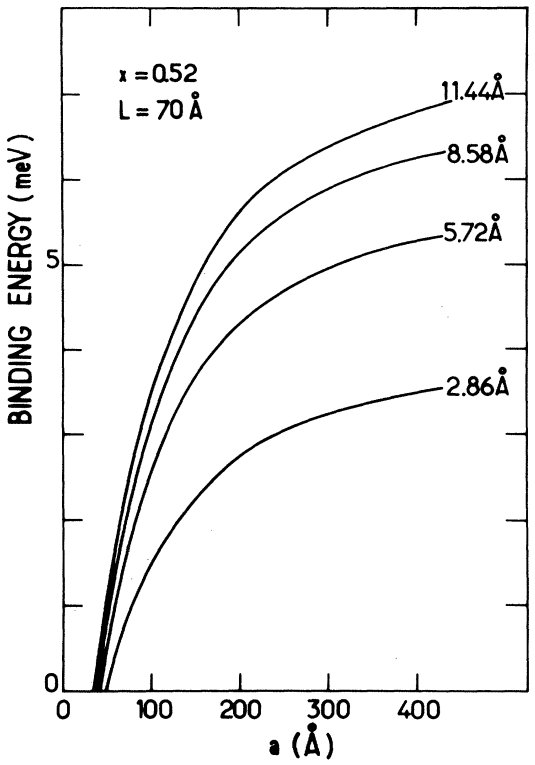

FIG. 2. Exciton binding energy on semi-Gaussian interface defects is plotted vs the lateral size a for different values of the defect depth b in a GaAs quantum well of thickness $L=70$ Å.

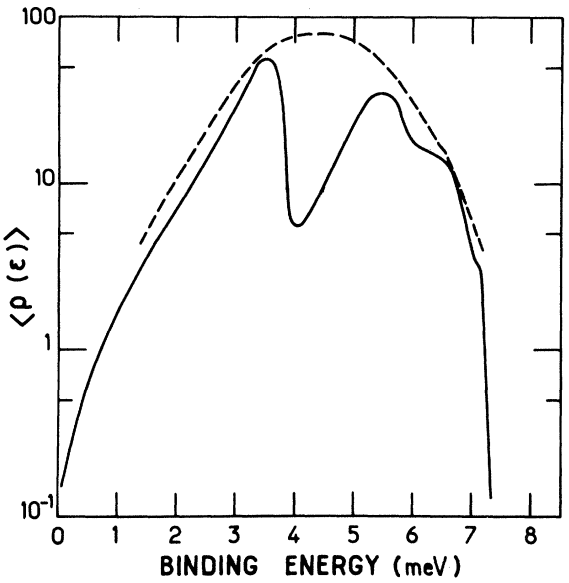

FIG. 3. Averaged density of states of trapped excitons on interface defects vs the trapped exciton binding energy. The solid line corresponds to a discrete distribution of defect depth b. The dashed line is the expected curve for a continuous distribution of defect depth b. The parameters of the density distribution $P(a,b)$ are given in the text.

cedure amounts to assuming that there is no energy dependence of the recombination matrix element and also implies that there is no thermalization of the recombination exciton. As for the last point, we note that at low temperature, the only possible motion for a trapped exciton is to hop from site to site by emitting an acoustical phonon. The transition probability per unit time W_{hop} that such a phonon-assisted hopping takes place from a site $\vec{R}_0$ to a site $\vec{R}_i$ has been calculated using Eq. (3) and the deformation potential approximation for the hole-phonon and electron-phonon interaction. The result is proportional to a Gaussian function of $\vec{R}_i - \vec{R}_0$. Taking $|\vec{R}_i - \vec{R}_0| = 400$ Å, $a(\vec{R}_0) = 110$ Å, $a(\vec{R}_i) = 190$ Å, $b(\vec{R}_0) = 2.86$ Å, $b(\vec{R}_i) = 5.72$ Å, and $C_e = 8.6$ eV, $C_v = 3.5$ eV for the deformation potentials,[9] we have obtained $\tau_{\text{hop}} = W_{\text{hop}}^{-1} \sim 3 \times 10^{-9}$ s. Since $|\vec{R}_i - \vec{R}_0|$ is far smaller than the mean defect separation ($N_d^{-1/2}$) for reasonable defect concentration N_d, we are led to conclude that a trapped exciton almost never hops before it recombines radiatively (the exciton recombination lifetime is estimated[10] to be 4×10^{-10} s). We have also calculated the trapping time τ_{trap} for a delocalized exciton with $\vec{K} = \vec{0}$ to become trapped on interface fluctuations (binding energy E_i) by acoustical phonon emission. A representative value is $\tau_{\text{trap}} \sim 4 \times 10^{-10}$ s if $N_d = 10^{10}$ cm^{-2} and $E_i = 3$ meV.

From sample to sample, the unknown density and size of the defects change. The Stokes shift and the luminescence linewidth scatter accordingly. Small figures for these quantities have been reported in high-quality MBE samples (4 and 2.5 meV in $L = 51$ Å multiple QW sample of Ref. 11, or even less[1]). The MO-CVD sample whose results have been shown in Fig. 1 is of comparable quality. We believe our model of exciton trapping at interface defects to be capable of *qualitative* agreement with the experiments done on high-quality samples. More quantitative descriptions are excluded before information on the statistical distribution of the radius a and the depth b of the defects are available. Our calculations give information about the binding energy of exciton trapped on "intrinsic," short-ranged interface defects. For the well under consideration ($x = 0.52$, $L = 70$ Å), these calculations exclude that the luminescence line is separated from the absorption peak by more than ~ 10 meV. Larger Stokes shifts and broader luminescence lines (FWHM ~ 14 meV) have been observed on less well-characterized structures. To interpret these features, intrinsic interface defects are insufficient. Impurity trapping near interfaces is likely to be involved in the observed large Stokes shift and broad luminescence. Calculations are in progress to ascertain whether the addition of a Coulombic potential to the defect Hamiltonian [Eq. (1)] is capable of interpreting the luminescence data of samples exhibiting broad luminescence lines.

To conclude, we would like to emphasize that these investigations show that exciton trapping on interface defects is certainly an important effect in reasonably good quantum wells ($L \lesssim 150$ Å). They show also that there are experimental observations in some samples which cannot be accounted for by exciton trapping, leading us to suggest that, even in nonintentionally doped structures, impurities associated with interface defects are likely to be involved.

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