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Localization-enhanced biexciton binding in semiconductors

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The influence of excitonic localization on the binding energy of biexcitons is investigated for quasi-three-dimensional and quasi-two-dimensional $Al_xGa_{1-x}As$ structures. An increase of the biexciton binding energy is observed for localization energies comparable to or larger than the free biexciton binding energy. A simple analytical model for localization in the weak confinement regime ascribes the increase to a quenching of the additional kinetic energy of the exciton-exciton motion in the biexciton. [S0163-1829(99)03524-9]

The binding energy of excitonic molecules in semiconductors has been investigated since the conjectures by Lampert[1] about their existence, and the experimental observation by Haynes.[2] These molecules, also called biexcitons, form in analogy to the hydrogen or positronium molecules. In the hydrogen molecule, the masses of the positive and negative particles differ strongly, while they are equal in the positronium molecule. With increasing mass mismatch, the binding energy of the molecule is increasing.[3,4] While for these molecules the embedding space is isotropic, translationally invariant, and three dimensional (3D), biexcitons in semiconductors can be situated in an anisotropic environment (given by the crystal symmetry), in a fluctuating potential (given, e.g., by composition fluctuations in alloys), and in a reduced dimensionality of free movement (in semiconductor nanostructures). These properties of the embedding space have a strong influence on the binding energies of excitons and biexcitons.

The influence of the dimensionality has been investigated intensively. For quasi-2D quantum wells, the exciton binding energy E_X is enhanced by a factor of 4 compared to three dimensions in the strong confinement limit. The biexciton binding energy E_{XX} is enhanced from $0.1E_X$ in three dimensions[2,5] (Haynes rule) to about $0.2E_X$ (Ref. 6) already in the weak 2D confinement regime,[7] where the exciton binding energy is nearly unaffected. In quantum wires in the strong confinement, E_X scales reciprocally with wire size, while E_{XX} was calculated for strong confinement in square $Al_xGa_{1-x}As$ structures to $0.6E_X$.[8] Experimental results on etched $In_xGa_{1-x}As$ (Ref. 9) and ZnSe (Ref. 10) quantum wires with comparable electron and hole confinement in the weak confinement regime show an enhancement relative to the 2D value up to $0.3E_X$. For differently confined electron and hole, this enhancement is reduced by a repulsive Coulomb potential between the excitons.[9,11] In quantum dots, E_{XX} saturates in the strong confinement limit, while the E_X is diverging.[12–14] This behavior is due to the absence of continuum states. Also here, E_{XX} is enhanced already by weak confinement.[15–17] From these findings, one can speculate that in the weak confinement regime, the biexciton binding energy is enhanced for structures of all dimensionalities, while the exciton binding energy remains nearly unchanged. This enhancement is thus rather due to the restriction of the exciton center-of-mass motion, than to a change of the exciton-exciton interaction potential.

A fluctuating potential, e.g., due to interface fluctuations[18] or statistical disorder in alloys,[19] also restricts the exciton center-of-mass motion. The influence of such a localization on the biexciton binding was not considered theoretically up to now, but can be compared with the binding energy enhancement for biexcitons bound to defects.[20] Experimentally, an enhancement is observed in strongly localized structures.[21–24] We have shown recently[25,26] that the biexciton binding energy is also inhomogeneously broadened by the localization, and that it is dependent on the position of the corresponding localized exciton states relative to the distribution of exciton transition energies.[27] Generally speaking, the localization of electronic states reduces the effective dimensionality of these states towards a quantum dot system.[21] Within this transformation, the relative magnitudes of the exciton binding energy, the biexciton binding energy, and the localization energy determine the localization regime. In the following we will use the width of the excitonic absorption or photoluminescence as localization energy E_{loc}. We investigate the regime of $E_{\mathrm{loc}}<E_X$, and we find that E_{XX} increases for $E_{\mathrm{loc}}>E_{XX}$. We propose a model that explains the enhancement by a reduction of the biexciton binding induced kinetic energy in the exciton-exciton relative motion. The absolute values of E_{XX}/E_X are influenced by the electron-hole mass ratio $\sigma=m_e/m_h$, which is around 1/5 in the $Al_xGa_{1-x}As$, and is varying between 1/3 and 1/6 in quantum wells due to the valence-band mixing. E_{XX} is generally increasing with the mass mismatch[3,28–30] due to the increasing Hartree potential of the charge density.

As model systems for the transformation from free to localized biexcitons, we have investigated the biexciton binding in two series of $Al_xGa_{1-x}As$ structures grown by molecular beam epitaxy on (100) GaAs substrates, all nominally undoped. The first series consists of 200 nm thick $Al_xGa_{1-x}As$ mixed crystal layers embedded in AlAs barriers, in the composition range x from 0.00 to 0.35 in steps of 0.05. They represent the transition from weakly to strongly localized quasi-3D systems. The second series consists of GaAs quantum wells (QWs) with thicknesses between 35 nm and 4 nm, embedded in $Al_{0.3}Ga_{0.7}As$ barriers. They represent the transition from weakly to strongly localized quasi-2D systems. Additionally, a 15 nm thick $Al_{0.1}Ga_{0.9}As$ QW, having a comparable 2D confinement as a 15nm GaAs QW, but a larger localization energy due to random alloy fluctuations, is investigated. The biexciton binding energy is determined

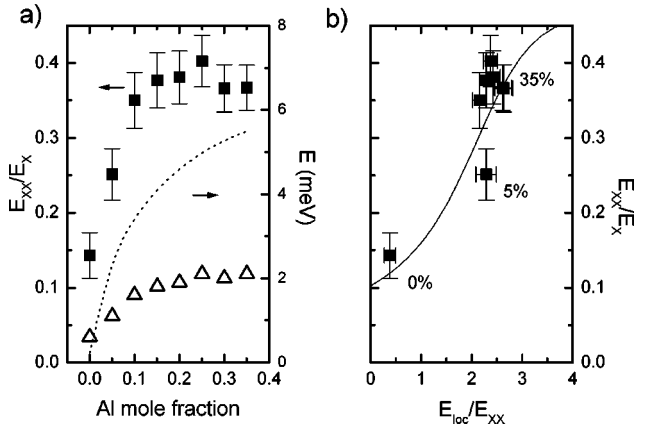

FIG. 1. (a) Biexciton binding energy E_{XX} (triangles), photoluminescence linewidth E_{loc} (dotted line), and ratio E_{XX}/E_X (squares) for the $Al_xGa_{1-x}As$ mixed crystal samples as a function of the Al mole fraction x. (b) E_{XX}/E_X as a function of the localization ratio E_{loc}/E_{XX}. Line: calculated dependence [Eq. (5)] for $n=3$, $E_{\mathrm{loc}}=4\hbar\omega_{\mathrm{loc}}$, $\alpha=0.312$, $\beta=2.4$, and $\sigma=0.2$.

by polarization dependent four-wave mixing (FWM). We perform spectrally resolved, time-integrated FWM in reflection or transmission geometries. A spectrally narrow (≈ 1 meV) pulse of direction $\vec{k}_1$ excites excitons with a well defined energy. The nonlinear response of these excitons is probed by a spectrally broad (≈ 10 meV) pulse of direction $\vec{k}_2$, creating a FWM signal in the $2\vec{k}_2-\vec{k}_1$ direction.[25,27] The biexciton binding energy is given by the difference between the photon energy of pulse $\vec{k}_1$ and that of the biexcitonic FWM response. These binding energies are in agreement with the quantum-beat period observed using a spectrally broad pulse $\vec{k}_1$.[22,25] All experiments are performed at 5 K lattice temperature.

The biexciton binding energies in three-dimensional (bulk) crystals have been investigated intensively for binary materials,[3,31] whereas to our knowledge the only experimental result for alloys is given for $CdS_{0.3}Se_{0.7}$,[32] with a localization energy comparable to the biexciton binding energy. Here we present measurements of biexciton binding energies in $Al_xGa_{1-x}As$ mixed crystals. Experimental details are given in Ref. 27. A strong enhancement of the biexciton binding energy from 0.5 meV for GaAs ($x=0$) to about 2 meV for $x>0.15$ is observed [Fig. 1(a)]. The resulting ratio E_{XX}/E_X is given in Fig. 1(b) as a function of E_{loc}/E_{XX}. The exciton binding energy E_X is estimated scaling the GaAs value of 4.2 meV, using a linear interpolation of the inverse effective masses and the dielectric constant.[19] As localization energy E_{loc} we take the photoluminescence width of the bound exciton.[33] E_{XX}/E_X changes from about 0.1 for GaAs to 0.4 for $x>0.15$. The enhancement of E_{XX}/E_X occurs at $E_{\mathrm{loc}}/E_{XX}\approx 1$. This is evidence for the influence of the localization on the biexciton binding energy. Since the shape of the exciton wave function is not significantly changed by the localization, this has to be mainly attributed to the restriction of the exciton-exciton motion rather than to a change of the biexciton binding potential.

To explain the experimentally observed dependence of the binding energy of the biexciton on the localization, we propose a simple model that takes into account the system dimensionality and localization strength. We first treat the biexciton binding under negligible localization. We describe the exciton-exciton interaction using an interaction potential V_{XX} that is a function of the distance of the exciton center-of-mass coordinates only. Its shape is in general dependent on σ, and it contains two contributions: A correlation part due to the change of the excitonic internal motion, and a Hartree part due to the potential of the e-h charge density for $\sigma\neq 1$. First, we approximate the exciton-exciton interaction for small distances by a spherically symmetric harmonic potential in the exciton center-of-mass distance $\vec{r}_{XX}$ in the units of the excitonic Rydberg E_X and the excitonic Bohr radius a_B:

$$V_{XX}=\beta E_X(\alpha|\vec{r}_{XX}/a_B|^2-1). \tag{1}$$

Here, α and β are dimensionless fitting parameters, describing the extension and the depth of the potential, respectively. Using the relation $a_B=\hbar/\sqrt{2m_em_hE_X/M_X}$, valid for three- and two-dimensional Wannier excitons ($M_X=m_e+m_h$ is the exciton mass), we get the corresponding harmonic energy $\hbar\omega_{XX}$ of the exciton-exciton relative motion as $\hbar\omega_{XX}=E_X\eta$ with $\eta=\sqrt{2\alpha\beta/\xi}$, and $\xi=(1+\sigma)(1+\sigma^{-1})/4$. Taking into account the ground state zero-point energy of $\frac{1}{2}\hbar\omega_{XX}$ for each dimension of free motion, the biexciton binding energy E_{XX}^{free} for n dimensions of exciton center-of-mass motion is $E_{XX}^{\mathrm{free}}=E_X(\beta-n\eta/2)$. The mass ratio σ not only explicitly enters here, but also influences the interaction potential, i.e., the parameters α,β. For mass ratios different from unity, the binding potential acquires a repulsive kernel due to the Hartree terms. In order to keep the analytical treatment, we take this into account considering a repulsive term $\Delta E_X\delta(\vec{r}/a_B)$ in first-order perturbation, where δ is the Dirac delta distribution, and get

$$E_{XX}^{\mathrm{free}}/E_X=\beta-n\eta/2-\Delta(\xi\eta/\pi)^{n/2}. \tag{2}$$

The Hartree potential between two three-dimensional excitons shows a repulsive kernel with an area $\Delta\approx 8(\xi-1)/(2\xi-1)$, which we will use in the following. For $\sigma=0.2$, the experimental values for E_{XX}/E_X of 0.1 in quasi-3D and of 0.2 in quasi-2D are reproduced choosing $\alpha=0.312$ and $\beta=2.4$. We now introduce the localization of the excitons by a harmonic potential V_i^{loc} in the center-of-mass coordinates of the excitons $\vec{R}_i(i\in\{1,2\})$, with the harmonic frequency ω_{loc}:

$$2V_i^{\mathrm{loc}}=M_X\omega_{\mathrm{loc}}^2|\vec{R}_i|^2-n\hbar\omega_{\mathrm{loc}}. \tag{3}$$

The constant potential $-n\hbar\omega_{\mathrm{loc}}$ has been added to compensate for the localization-induced zero-point energy of the exciton motion, normalizing the ground state energy of the localized exciton to zero. To include the exciton-exciton interaction potential V_{XX}, we separate the motion of the two excitons into their center-of-mass coordinate $\vec{R}_{XX}=(\vec{R}_1+\vec{R}_2)/2$ and relative coordinate $\vec{r}_{XX}=\vec{R}_1-\vec{R}_2$. The total biexciton potential without the repulsive kernel then reads

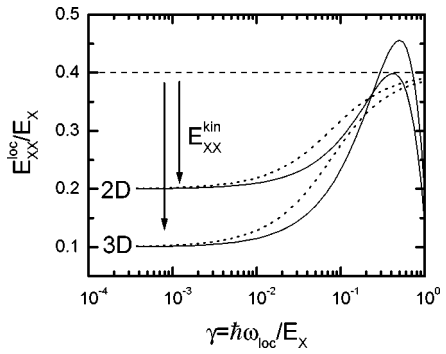

FIG. 2. Calculated ratio between biexciton and exciton binding energy E_{XX}/E_X as a function of the ratio γ between the localization energy and the exciton binding energy [Eq. (5)] for quasi-2D ($n=2$) and quasi-3D ($n=3$), using $\sigma=0.2$, and without repulsive kernel (dotted): $\alpha=0.09, \beta=0.4$ or with repulsive kernel (solid): $\alpha=0.312, \beta=2.4$.

$$V_{XX}^{\mathrm{loc}}=M_X\omega_{\mathrm{loc}}^2|\vec{R}_{XX}|^2 + \frac{1}{4}M_X(\omega_{\mathrm{loc}}^2+\omega_{XX}^2)|\vec{r}_{XX}|^2-n\hbar\omega_{\mathrm{loc}}-\beta E_X. \quad (4)$$

Introducing the localization parameter $\gamma=\hbar\omega_{\mathrm{loc}}/E_X$, and again taking into account the repulsive kernel in first-order perturbation, the resulting binding energy of the localized biexciton is

$$\frac{E_{XX}^{\mathrm{loc}}}{E_X}=\beta-\frac{n}{2}(\sqrt{\eta^2+\gamma^2}-\gamma)-\Delta\left(\frac{\xi}{\pi}\sqrt{\eta^2+\gamma^2}\right)^{n/2}. \quad (5)$$

The result is plotted in Fig. 2 for three- and two-dimensional systems. Without the repulsive kernel ($\Delta=0$), the additional kinetic energy due to the biexciton binding, given by the difference to the constant biexciton binding energy in 0D, is vanishing for $\omega_{XX}\ll\omega_{\mathrm{loc}}$ (corresponding to $\gamma\gg0.2$ in Fig. 3), and E_{XX}^{loc} approaches the depth βE_X of the biexciton binding potential. Including the repulsive kernel, the biexciton binding also increases for $\gamma\ll1$, but decreases for $\gamma>0.5$, i.e., outside the weak confinement limit. Within the weak confinement limit, both calculations show a comparable increase of the biexciton binding energy.

To compare Eq. (5) with the experimental data, we have to relate the quantity $\hbar\omega_{\mathrm{loc}}$ with the inhomogeneous linewidth E_{loc}. In a random localization potential with a correlation length much smaller than the localization radius, the average kinetic and potential energies are scaling equally. All the investigated samples are grown under optimum growth conditions, for which clustering can be neglected, and the disorder, given by random alloy broadening and interface roughness, has a short correlation length. In this case the influence of the disorder on the exciton motion can be described by a single broadening parameter. We take here $\hbar\omega_{\mathrm{loc}}=E_{\mathrm{loc}}/4$, a choice that is giving the best quantitative agreement between the model and the experimental data for both quasi-2D and quasi-3D cases. The calculated dependence is compared with the quasi-3D $Al_xGa_{1-x}As$ system in Fig. 1. It shows good agreement with the experimental data, indicating that the theoretical description catches the essential features of the problem.

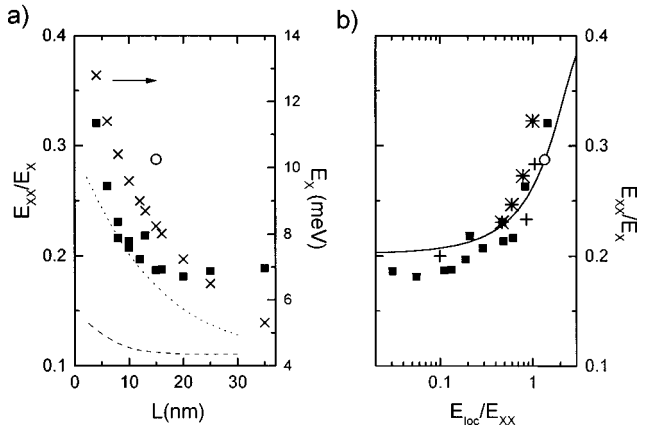

FIG. 3. Experimental and calculated data for quasi-2D samples. (a) Symbols: experimental data for $GaAs/Al_{0.3}Ga_{0.7}As$ QWs as a function of the well thickness L. Crosses: Exciton binding energy; squares: experimental ratio E_{XX}/E_X. Circle: E_{XX}/E_X for an $Al_{0.1}Ga_{0.9}As/Al_{0.3}Ga_{0.7}As$ QW. Lines are calculations (Refs. 30, 35 and 36). (b) E_{XX}/E_X versus localization strength E_{loc}/E_{XX} for different quasi-2D material systems: Open diamonds: this work; filled squares: $GaAs/Al_xGa_{1-x}As$ QWs; crosses: $ZnSe/Zn_xMg_{1-x}S_ySe_{1-y}$ (Refs. 25, 37, and 38); stars: $In_{1-x}Ga_xAs/GaAs$ (Ref. 39). Line: calculated with Eq. (5) for $n=2$, other parameters as in Fig. 1.

For the quasi-two-dimensional systems the transition from free to localized biexcitons is traced by the GaAs QW series, for which the experimental results are given in Fig. 3(a). E_X (crosses) is determined from the absorption spectra, and changes from 5.3 meV for 35 nm well width to 13 meV for 4 nm well width, in agreement with published data.[34] E_{loc} is taken from the full width at half maximum of the heavy-hole exciton absorption line. The measured biexciton binding energy E_{XX} is increasing with decreasing well width, reaching 4.1 meV in the 4 nm QW. The ratio E_{XX}/E_X (squares) is constant at about 0.2 for well widths from 35 nm to 8 nm, for which E_{loc} is lower than E_{XX}, while E_X is increasing from 4.8 meV to 11 meV. This implies that the change of the exciton wave function from quasi-3D to quasi-2D is not influencing E_{XX}/E_X significantly. The change of this ratio from about 0.1 in quasi-3D structures[2] to about 0.2 in quasi-2D structures[6] is due to the reduction of the dimensionality of the relative exciton-exciton motion in the biexciton, which occurs in GaAs QWs already between 50 nm and 100 nm well thickness.[7] Below 8 nm well thickness E_{loc} is larger than E_{XX} and an enhancement of E_{XX}/E_X is observed. The behavior in quasi-2D is thus qualitatively comparable to the quasi-3D case, showing an enhancement for $E_{\mathrm{loc}}>E_{XX}$, with the difference that one dimension of exciton-exciton motion is already quenched by the 2D confinement.

The variational calculation of Kleinman[30] (dashed line) is far below the experimental data, while an improved calculation of Liu, Kong, and Liu[35] (dotted line) is close to the measured ratio for QW thicknesses around 10 nm. For larger well widths the ratio is underestimated, probably due to restrictions in the used quasi-2D trial function. Also at smaller well widths, for which the trial function should be well adapted, their results are below the experiment, indicating the importance of exciton localization. This importance is evident when comparing the 15 nm GaAs QW with the 15

nm $Al_{0.1}Ga_{0.9}As$ QW (circle). The latter has a larger localization energy at equal well width, and shows a significantly higher biexciton binding.

If we instead of the well width use the localization ratio $E_{\rm loc}/E_{XX}$ as coordinate [Fig. 3(b)], all the QW E_{XX}/E_X data show a common dependence. In addition to the $Al_xGa_{1-x}As$ QW data, results from literature on $In_xGa_{1-x}As/GaAs$ (stars) and $ZnSe/Zn_{1-x}Mg_xS_ySe_{1-y}$ (crosses) QWs are displayed. This indicates that the localization ratio is the crucial parameter determining E_{XX}/E_X, rather than the actual quantum well confinement potential. The result of Eq. (5), using the same parameters α,β as in the quasi-3D case, is given as a solid line. It reproduces the experimental data. The model thus captures the main mechanisms leading to the enhancement of E_{XX} also in localized quasi-2D systems.

In summary, we have shown that localization of excitons leads to an enhanced binding energy of the biexciton. In the proposed analytical model, this is attributed to a quenching of the exciton-exciton motion in the biexciton. The predicted increase of the biexciton binding energy due to localization is stronger for quasi-3D than for quasi-2D structures. The model shows qualitative agreement with the experimental data for the $Al_xGa_{1-x}As$ material system. All the presented data are on type-I material systems. In type-II systems, the spatial separation between electron and hole leads to additional Coulomb repulsion, reducing the biexciton binding.

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