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Anisotropy effects on excitonic properties in realistic quantum wells

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The binding energy, lateral and vertical extension, dimensionality, and lifetime of excitons in quantum wells are calculated as a function of well width. A novel variational approach including an elliptically symmetric wave function, finite barriers, and different masses in the well and the barrier is used. Thus, for the first time the geometrical anisotropy and the anisotropy of the physical properties of the quantum well are fully considered. Numerical results are presented for $Al_{0.40}Ga_{0.60}As$/GaAs and $In_{0.53}Ga_{0.47}As$/InP.

Spatial localization of electrons and holes in quantum wells (QW's) causes a dramatic enhancement of excitonic effects. Excitons dominate the optical properties like absorption or luminescence spectra in undoped QW's at all temperatures even at room temperature.[1-4] They lead at room temperature to large optical nonlinearities[4] and to a linear radiative term in the density dependence of the recombination rate,[5] properties not observed for three-dimensional semiconductors. Thus excitons are also of decisive importance for modern photonic devices. Despite this, the understanding of excitons and their properties in real QW's is unsatisfactory from a fundamental point of view. The present theory of real 2D excitons resembles very much the theoretical understanding of 3D excitons[6] before the work of Baldereschi and Lipari[7] beginning in the 1970s. Up to date no quantitatively correct description of such fundamental properties as the binding energy of 2D excitons exists, covering the full QW width range from $L_z=0$ to $L_z\rightarrow\infty$. It is the purpose of this work to present a novel variational treatment of the 2D exciton problem which allows for the first time prediction of such properties as the binding energy, lateral extension of the wave function, penetration into the barrier, and radiative recombination lifetime as a function of the well width over the full L_z range. Numerical results for the two most important class-I QW's, $Al_xGa_{1-x}As$/GaAs and $In_xGa_{1-x}As$/InP, are given.

Before presenting our ansatz and results we briefly review some of the foundations on which our work is based. Miller, Kleinman, Tsang, and Gossard[8] were among the first to calculate the 1S and 2S exciton binding energies using complex one-parameter variational functions and an infinite-barrier model with averaged dielectric constant. Neither for $L_z=0$ nor for $L_z\rightarrow\infty$ correct values are obtained. Bastard, Mendez, Chang, and Esaki[9] briefly later also assume infinite barriers and isotropic masses and do not average over the dielectric constant. Two different trial wave functions are compared with each other, one of them yielding a correct binding energy in the 3D limit. Shinozuka and Matsuura[10] again assume infinite barriers as well and use isotropic masses but make use of a two-parameter trial function.

Greene and Bajaj[11] advanced the theory greatly by taking finite barriers into account and by choosing for the variational function a linear combination of three functions. All of them, however, are isotropic in the exponentially decaying term. Valence-band hybridization has a certain influence on exciton spectra,[12] but some effects cancel each other, resulting only in a slight change of binding energies. In Refs. 11 and 12 the masses and the dielectric constant in the $Al_xGa_{1-x}As$ barrier are taken to be the same as in the GaAs QW, leading to incorrect results for thin wells.

In this paper we use a wave function with elliptic symmetry adapted to the anisotropy of geometry and of physical properties of the QW and take finite barriers into account. This gives correct 3D limits for zero and infinite well width and the correct 2D limits for infinite barriers. Our wave function can continuously interpolate between these extremes and is therefore appropriate to be used for all realistic quantum wells.

Let us assume the origin of our coordinate system in the middle of the quantum well of width L_z. The growth direction of the well is taken as z direction with a (100) interface plane. The positions of the electron and hole are denoted by $\mathbf{r}_e$ and $\mathbf{r}_h$, respectively. Cylindrical relative coordinates ρ, ϕ, z are introduced as usual, for example, $\rho^2=(x_e-x_h)^2+(y_e-y_h)^2$. The potential barriers are assumed to form a square well,

$$V_{\text{confinement}}=\begin{cases}0 \text{ for } |z|<L_z/2,\\ V_0 \text{ for } |z|\geq L_z/2,\end{cases} \tag{1}$$

with different values $V_{0,e}$ for electrons and $V_{0,h}$ for holes in agreement with the relation $V_{0,e}+V_{0,h}=E_{\text{gap}}(\text{barrier})-E_{\text{gap}}(\text{well})$. The Hamilton operator of the relative motion of an exciton in a QW is then given by

$$\mathrm{H}_{\text{ex}}=-\frac{\hbar^2}{2\mu_\pm}\left[\frac{1}{\rho}\frac{\partial}{\partial\rho}\left(\rho\frac{\partial}{\partial\rho}\right)+\frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}\right]-\frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2}-\frac{\hbar^2}{2m_\pm}\frac{\partial^2}{\partial z_h^2}-\frac{1}{\epsilon}\frac{e^2}{|\mathbf{r}_e-\mathbf{r}_h|}+V_{\text{conf.},e}+V_{\text{conf.},h}\,. \tag{2}$$

The masses $\mu_\pm$ and $m_\pm$ for the in-plane and z directions for heavy (upper sign) and light holes, respectively, are given in terms of the Kohn-Luttinger band parameters,[13] m_0 being the free-electron mass,

$$\frac{1}{\mu_\pm}=\frac{1}{m_e}+\frac{\gamma_1\pm\gamma_2}{m_0},\quad \frac{1}{m_\pm}=\frac{\gamma_1\mp 2\gamma_2}{m_0}. \tag{3}$$

As elliptically symmetric trial wave function we take

$$\Psi_{\rm ex}(a,\lambda)=\Psi_e(z_e)\Psi_h(z_h)\phi_{\rm ex}(\rho,z),$$
$$\phi_{\rm ex}(\rho,z)=\exp\left[-\frac{1}{a}\sqrt{\rho^2+\lambda^2z^2}\right], \tag{4}$$

with a and λ as variational parameters. λ will be later identified as a measure of the dimensionality and a as a Bohr diameter. Ψ_e and Ψ_h are the unnormalized ground-state wave functions of the finite square well without Coulomb interaction, e.g.,

$$\Psi_e(z_e)=\begin{cases}\cos(k_ez_e) & \text{for } |z_e|<L_z/2\\ A_e\exp(-\kappa_ez_e) & \text{for } |z_e|\ge L_z/2,\end{cases}\quad k_e=\sqrt{2m_eE_{1,e}}. \tag{5}$$

Here $E_{1,e}$ is the energy of the first electron subband, κ_e and A_e are obtained from the continuity condition for Ψ_e and $1/m_e\Psi_e'$ at the interface. Ψ_h is defined in a similar way.

The ground-state exciton binding energy E_b is found according to the Rayleigh-Ritz variational principle[14] by minimizing the energy functional with respect to the two variational parameters a and λ:

$$E_b=E_{1,e}-E_{1,h}-\min_{a,\lambda}\left[\frac{\langle\Psi_{\rm ex}|\mathbf{H}_{\rm ex}|\Psi_{\rm ex}\rangle}{\langle\Psi_{\rm ex}|\Psi_{\rm ex}\rangle}\right]. \tag{6}$$

λ describes the anisotropy of the exciton in the plane and z directions. So our wave function $\Psi_{\rm ex}(a,\lambda)$ interpolates continuously between the 3D case ($L_z\to 0$, $L_z\to\infty$) with $\lambda\to 1$ and the purely 2D case ($V_{0,e},V_{0,h}\to\infty$, $L_z\to 0$) with $\lambda\to 0$. Via

$$P_{\rm barrier}=\left[1-\int_{\rm well}\Psi_{\rm ex}^2dV\right]/\langle\Psi_{\rm ex}|\Psi_{\rm ex}\rangle, \tag{7}$$

we calculate the probability of finding the exciton in the barrier. The radial extension of the exciton in plane is measured by $\rho_{\rm ex}$ and the extension in the z direction is measured by $z_{\rm ex}$ given by

$$\rho_{\rm ex}=\sqrt{\langle\rho^2\rangle},\quad z_{\rm ex}=\langle|z|\rangle. \tag{8}$$

The radiative recombination lifetime $\tau_{\rm rad}$ of an exciton state has been shown to be inversely proportional to the state's oscillator strength f.[15] The oscillator strength per unit cell f^* of an exciton in a QW is proportional[16] to $|\Phi_{\rm ex}(0)|^2/L_z$. The relevant physical property is the oscillator strength per state f which is given by $f=f^*/n$, n being the density of states per unit cell [$n\sim M/L_z$, $M=m_e+m_0/(\gamma_1\pm\gamma_2)$]. Considering $|\Phi_{\rm ex}(0)|^2\sim 1/a^2(L_z)$ in the quasi-2D range, the radiative recombination lifetime shows an implicit dependence on well width L_z,

$$\tau_{\rm rad}\sim Ma^2(L_z). \tag{9}$$

The lifetimes for heavy- and light-hole excitons obtained with formula (9) may not be compared directly because we have to consider different Bloch matrix elements for hh and lh excitons. Group theory gives a well-known additional factor of three for the light-hole lifetime.

Now we turn to numerical results. For the limit of infinite barriers we obtain what we anticipated; in particular, for $L_z=0$ we get $\lambda=0$ and $E_b=4$ Ry. In what follows we will consider real quantum wells of particular relevance: The $Al_{0.40}Ga_{0.60}As$/GaAs and the $In_{0.53}Ga_{0.47}As$/InP systems will serve as models for other presently less important material combinations. The parameters we use are listed in Table I.

TABLE I. Material parameters of $Al_{0.40}Ga_{0.60}As$, GaAs, $In_{0.53}Ga_{0.47}As$, and InP used in the calculations (4 K values).

Material	$E_{\rm gap}$ (eV)	$V_{0,e}/\Delta E_g$	γ_1	γ_2	m_e	ϵ
GaAs[a]	1.518	60%[d]	6.85	2.1	0.0665	12.5
$Al_xGa_{1-x}As$[b] [Al] = 0.40	2.163		4.67	1.17	0.0895	11.5
InP[a]	0.813	40%[e]	5.15	0.94	0.0803	12.6
$In_xGa_{1-x}As$ [In] = 0.53	1.423[c]		11.01[f]	4.18[f]	0.041[f]	13.9[g]

[a]Reference 18.
[b]Reference 19.
[c]Reference 20.
[d]Reference 21.
[e]Reference 22.
[f]Reference 23.
[g]Reference 24.

In Fig. 1 the hh- and lh-exciton binding energies are plotted as a function of well width. The correct bulk values are approached for $L_z \rightarrow 0$ and $L_z \rightarrow \infty$. For both limiting cases the values for hh and lh excitons are identical as expected from a correct theoretical treatment. Heavy- and light-hole excitons are degenerate in the bulk material. Between these limiting cases the binding energies reach a maximum value for a QW width close to 1 nm. We note that the hh-exciton binding energy is always appreciably smaller than the lh-exciton binding energy for $L_z > 1$ nm. This is a result of the larger in-plane mass of the lh excitons. It differs qualitatively from that stated by Greene *et al.*[11] and is not related to different valence-band discontinuities used in our work and Ref. 11, as a calculation shows, but is a result of the anisotropic properties of excitons in quantum wells. The probability of finding the exciton in the barrier is plotted in the insets of Fig. 1. The probability to be found in the barrier is greater for lh excitons than for hh excitons for all well widths.

$d = \lambda + 2$ can be interpreted as the dimensionality of the exciton. For $L_z \rightarrow 0$ and $L_z \rightarrow \infty$ λ approaches unity, the excitons are three dimensional. For a wide range of well thicknesses 1 nm $< L_z \sim 20$ nm we find λ to be close to zero; this means the excitons are indeed almost ideally two dimensional. For this range we have tested the rule of thumb $E_b a_{\text{Bohr}} = \text{const}$. This rule is correct for the 3D and 2D limits. For $\lambda = 0$ $a/2$ is just the 2D Bohr radius. We find that $E_b a/2$ varies by less than 10% from the 3D or 2D limit. So the customary rule is quite a good estimate. The relation $E_b a^2 = \text{const}$ which is sometimes used[17] is not valid.

In Fig. 2 the lateral extension ρ_{ex} and the extension in z direction z_{ex} defined by Eqs. (8) are plotted versus the well width. In quantum wells the exciton is compressed laterally and in z direction. The z extension of the excitons is always well below $L_z/2$, a rather remarkable result. The excitons reach their minimum extension for approximately the same well width for which the maximum bind-

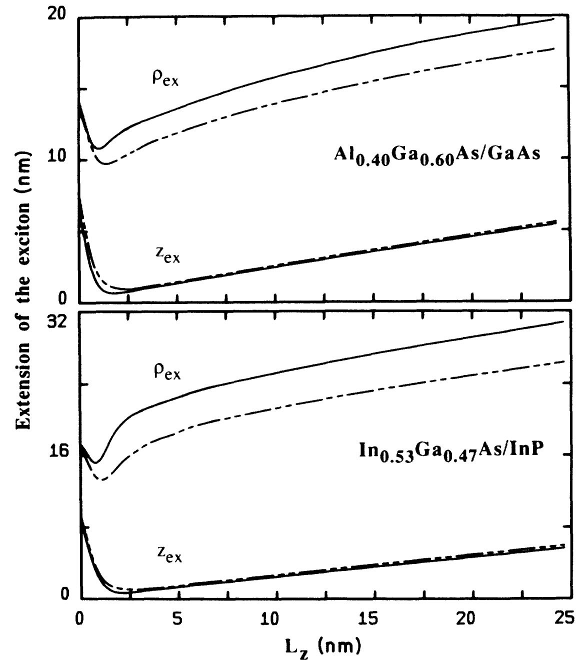

FIG. 2. Extension of heavy (solid line) and light (dashed line) hole excitons in plane and in z direction for $Al_{0.4}Ga_{0.6}As/GaAs$ and $In_{0.53}Ga_{0.47}As/GaAs$.

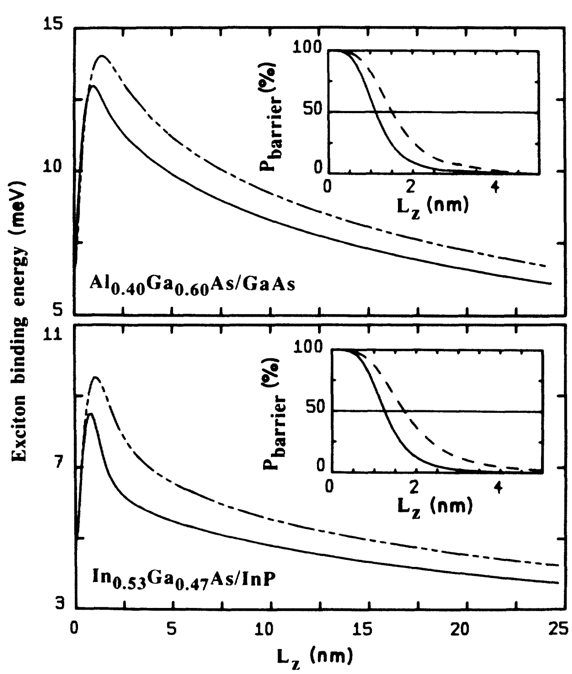

FIG. 1. Heavy (solid line) and light (dashed line) hole exciton binding energies for $Al_{0.40}Ga_{0.60}As/GaAs$ and $In_{0.53}Ga_{0.47}As/InP$ quantum wells. Insets: probability of finding the excitons in the barrier.

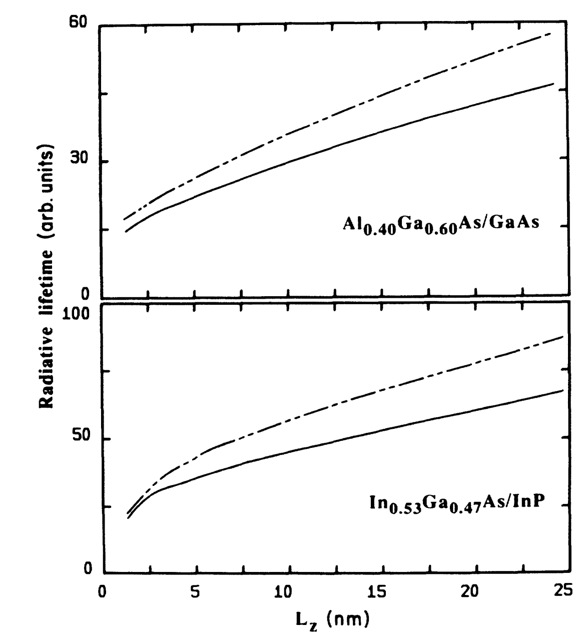

FIG. 3. L_z dependence of the radiative recombination lifetime of heavy (solid line) and light (dashed line) hole excitons for $Al_{0.40}Ga_{0.60}As/GaAs$ and $In_{0.53}Ga_{0.47}As/InP$ in arbitrary units. The scale for the heavy-hole exciton is smaller by a factor of 3 than the scale for the light-hole exciton.

ing energy is reached. It should be noted that in the 3D bulk limit the trial wave functions for hh and lh excitons are slightly different, which does not affect the degeneracy of binding energies.

In Fig. 3 the L_z dependence of the radiative recombination lifetime of heavy-hole excitons in the quasi-2D regime as given by Eq. (9) is plotted versus the well width. The scales for heavy- and light-hole excitons are different by a factor of 3 as mentioned above. Thus heavy-hole lifetimes are shorter. The lifetime is given in arbitrary units because thermal effects cause a proportionality factor[17] without changing the L_z dependence. The decrease of lifetime for small well widths is more pronounced in the $In_xGa_{1-x}As/InP$ system than in the $Al_xGa_{1-x}As/GaAs$ system.

In summary, for the first time binding energies, lateral and vertical extensions, barrier penetration depths, and relative lifetimes of excitons in quantum wells are calculated with an anisotropic wave function and finite barriers, thus giving a maximum of physical significance. Our model yields correct values for the 3D and 2D limits. The two variational parameters λ and a are identified as measures of dimensionality and the Bohr radius. We have presented numerical results for the technologically relevant systems $Al_xGa_{1-x}As/GaAs$, [Al] $=0.40$, and $In_xGa_{1-x}As/InP$, [In] $=0.53$. The results give a realistic insight into the behavior of excitons over the whole range of quantum-well thicknesses from $L_z = 0$ to $L_z \rightarrow \infty$.

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