Wannier exciton bound states for electron and hole spatially separated in adjacent 2D quantum layers

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We analyze the lowest bound states of a Wannier-Mott exciton in which the electron is constrained to move freely in a 2D quantum layer (2DL) and the hole moves in another parallel 2DL. We assume that in the confinement direction z both electron and hole are in their respective ground state of a harmonic potential which yields a general multipolar expansion of the electron-hole Coulomb potential in terms of Legendre polynomials. The resulting Schrödinger equation can be solved by perturbing 2D exciton states for which both electron and hole are in the same 2DL.

Keywords: Quantum layers; semiconductor heterostructure; Wannier-Mott exciton

Se estudia el estado base y los primeros estados excitados de un excitón de Wannier-Mott, para el cual el electrón está restringido a moverse libremente en una capa cuántica bidimensional (2DL) y el hueco en otra 2DL paralela. Se supone que en la dirección de confinamiento z tanto el electrón como el hueco están en el estado base correspondiente a un potencial armónico, con el cual permite realizar un desarrollo multipolar de la interacción coulombiana electrón-hueco en términos de polinomios de Legendre. La ecuación de Schrödinger resultante se resuelve usando teoría de perturbaciones para los estados excitónicos bidimensionales correspondientes al problema en el cual el electrón y el hueco están en la misma 2DL.

Descriptores: Capas cuánticas; heteroestructuras semiconductoras; exciton de Wannier-Mott

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1. Introduction

Studies of excitons in confined systems are interesting due to the possibility of growing high-quality nanostructures with prescribed configurations. Within the spirit of studying Wannier-Mott excitons in systems that exhibit spatial separation between electron and hole, we investigate from a theoretical point of view a Wannier-Mott exciton in which the electron is confined within a bidimensional layer (2DL) and the hole is confined in another parallel 2DL. The purpose of this paper is to investigate the lowest bound states of this system. Analogous systems were theoretically investigated using variational approaches to calculate groundstate properties [1, 2]. Here we take into account the effects of finite widths of the layers in contrast to Lozovick and Nishanov's model [1], which assumed particles confined in 2DLs with vanishing widths. Bastard *et al.* [2] performed a variational calculation of the exciton binding energy of a type II semiconductor heterostructure consisting of a hole in InAs well confined between two semi-infinite GaSb layers where the electron lied. On the other hand, calculations of the ground-state exciton wavefunctions in layered structures are desired in model calculations of excitons at interfaces, such as that of Kachintev and Ulloa [3], where they investigated the collective modes of diluted 2D exciton gas.

2. Complete multipolar expansion of the inter-layer electron-hole potential

The Hamiltonian in cylindrical coordinates is

$$\hat{H} = \hat{H}_1 + \hat{H}_2 + \hat{V}_{\rm int}, \tag{1}$$

where $\hat{H}_\nu$ (with $\nu = 1, 2 = e, h$) are defined as

$$\hat{H}_\nu = \hat{T}_{\nu x} + \hat{T}_{\nu y} + \hat{T}_{\nu z} + \hat{V}_\nu(z_\nu). \tag{2}$$

being $T_{\nu i}$ (with $i = \rho_\nu, \phi_\nu, z_\nu$) the kinetic and $\hat{V}_\nu(z_\nu)$ the transverse confinement potential of each carrier. The electric interaction potential is

$$\hat{V}_{\rm int}(\vec{r}_1 - \vec{r}_2) = -\frac{e^2/\epsilon}{\sqrt{\rho_1^2 + \rho_2^2 - 2\rho_1\rho_2\cos(\phi_1 - \phi_2) + (z_1 - z_2)^2}}, \tag{3}$$

where ϵ is the appropriate dielectric screening of the semiconductor media.

Let us assume that the transverse dimensions of both 2DLs are small enough so that their associated groundstate energies are the only relevant levels for the energy range we consider. The wave function is variable separable, thus it can be written as

$$\psi = \psi_1^0(z_1)\psi_2^0(z_2)F(\phi_{\rm cm}, \rho_{\rm cm})\Phi(\phi)\Xi(\rho), \tag{4}$$

where ρ, ϕ and $\rho_{\rm cm}$, $\phi_{\rm cm}$ are the usual relative and center of mass cylindrical coordinates

$$\rho = \sqrt{\rho_1^2 + \rho_2^2 - 2\rho_1\rho_2\cos(\phi_1 - \phi_2)},$$

$$\phi = \arctan\left(\frac{\sin\phi_1 - \sin\phi_2}{\cos\phi_1 - \cos\phi_2}\right)$$

and

$$\rho_{\rm cm} = \frac{\sqrt{m_1\rho_1^2 + m_2\rho_2^2 + 2m_1m_2\rho_1\rho_2\cos(\phi_1 - \phi_2)}}{m_1 + m_2},$$

$$\phi_{\rm cm} = \arctan\left(\frac{m_1\sin\phi_1 + m_2\sin\phi_2}{m_1\cos\phi_1 + m_2\cos\phi_2}\right),$$

with m_ν being the corresponding effective mass), $\psi_1^0(z_1)$, $\psi_2^0(z_2)$, $F(\rho_{\rm cm}, \phi_{\rm cm})$ and $\Phi(\phi)$ are functions such that

$$[\hat{T}_{\nu z} + \hat{V}_\nu(z)]\psi_\nu^0 = E_{\nu 0}^t\psi_\nu^0,$$

(the index 0 denotes the groundstate),

$$\left(\frac{1}{\rho_{\rm cm}}\frac{\partial}{\partial\rho_{\rm cm}}\rho_{\rm cm}\frac{\partial}{\partial\rho_{\rm cm}} + \frac{1}{\rho_{\rm cm}^2}\frac{\partial^2}{\partial\phi_{\rm cm}^2}\right)F = \frac{(\hbar k)^2}{2\mu}F$$

and

$$\frac{d^2}{d\phi^2}\Phi = l^2\Phi,$$

being k the radial exciton wavevector, and l the quantum angular number. Also $\mu = m_1m_2/(m_1 + m_2)$ is the reduced mass and $\Xi(\rho)$ is the wave function part associated with the relative radial coordinate. We calculate the bracket $\langle\psi_1^0\psi_2^0F\Phi|\hat{H}|\psi_1^0\psi_2^0F\Phi\Xi(\rho)\rangle$ to yield

$$\left(\frac{1}{\rho}\frac{d}{d\rho}\rho\frac{d}{d\rho} - \frac{l^2}{\rho^2} + V_{\rm eff}\right)\Xi(\rho) = E^{\rm 2D}\Xi(\rho), \tag{5}$$

where the effective potential $V_{\rm eff}$ for the 2D-dimensional problem is defined as

$$V_{\rm eff}(z) = \int_{-\infty}^{\infty} dz_1 \int_{-\infty}^{\infty} dz_2\,|\psi_0(1)|^2|\psi_0(2)|^2\,\hat{V}_{\rm int}(\vec{r}_1 - \vec{r}_2), \tag{6}$$

and

$$E^{\rm 2D} = \mathcal{E} - E_{10}^t - E_{20}^t - \frac{(\hbar k)^2}{2M} \tag{7}$$

where $\vec{r}_\nu$ are 3D vector, $\mathcal{E}$ is the eigenenergy of the whole system and $M = m_1 + m_2$ is the total mass.

We proceed to perform a multipolar expansion of $V_{\rm eff}$ restricting our model to harmonic potentials in the z-dependent transverse confinement; $\hat{V}_\nu(z_\nu) = k_\nu z_\nu^2/2$. This choice has the following advantages: it could physically represent either soft or hard possible confinements, and as it will be shown, all the moments of the harmonic oscillator calculated in the groundstate can be expressed in terms of the powers of its standard deviation. However, our treatment is valid for any transverse confinement potential whose moment integrals are well defined and known, and for any pair of particles p_1 and p_2 (in this work we will apply it to the exciton problem, so p_1 and p_2 will represent the electron and the hole). It is useful to write the groundstate joint density probability of the bidimensional harmonic oscillator $|\psi_0(1,2)|^2$ in terms of the standard deviations $\sigma_{z\nu} = \langle(z^\nu)^2\rangle_0$ (subindex 0 indicates groundstate), which are of the order of magnitude of the widths of the 2DL and are inversely proportional to the fourth power of the harmonic potential stiffness of the k_ν:

$$|\psi_0(1,2)|^2 = |\psi_0(1)|^2|\psi_0(2)|^2 = \frac{e^{-\frac{z_1^2}{(\sigma_{z1})^2}}}{\sqrt{\pi}\sigma_{z1}}\,\frac{e^{-\frac{(z_2-d)^2}{(\sigma_{z2})^2}}}{\sqrt{\pi}\sigma_{z2}}. \tag{8}$$

Here d is the distance between the two particles. Let us recall that the groundstate energy of a harmonic oscillator for a particle of mass m is given by

$$E = \frac{\hbar^2}{2m\sigma^2}. \tag{9}$$

It is well known that $|\psi_0(\nu)|^2$ tends to the Dirac's δ function as $\sigma_{z\nu} \to 0$. Then we can approximate the electrostatic potential as a Taylor expansion around the maximum of $|\psi_0(1,2)|^2$ with respect to its two transverse variables to yield

$$V_{\rm eff}(\rho) = \frac{q_1q_2}{\epsilon}\sum_{ij=0}^{\infty}\frac{I(\sigma_{z1},\sigma_{z2})}{i!j!} \times \frac{\partial^{i+j}}{\partial z_1^i\partial z_2^j}\left[\rho^2 + (z_1 - z_2)^2\right]^{-1/2}|_{z_1=0,z_2=d}, \tag{10}$$

where $I(\sigma_{z1},\sigma_{z2})$ are the moments of the joint density probability of the groundstate given by

$$I(\sigma_{z1},\sigma_{z2}) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} dz_1dz_2\,|\psi_0(1,2)|^2z_1^i(z_2 - d)^j. \tag{11}$$

The calculation of $I(\sigma_{z1},\sigma_{z2})$ is straightforward, and it allows us to rewrite the multiple partial derivative involved in Eq. (10) in terms of orthogonal polynomials as follows

$$\frac{\partial^{i+j}}{\partial z_1^i\partial z_2^j}[\rho^2 + (z_1 - z_2)^2]^{-1/2}|_{z_1=0,z_2=d} = \frac{(-1)^j(i+j)!P_{i+j}(d/\sqrt{\rho^2 + d^2})}{[\rho^2 + d^2]^{(1+i+j)/2}}, \tag{12}$$

where we have used the generating function [4] of the Legendre polynomials P_n $(n = 0, 1, ...)$ and we set $n = i + j$. By substituting Eq. (12) into Eq. (10) and expressing $I(\sigma_{z1}, \sigma_{z2})$ in terms of the gamma function Γ, we arrive to the following general expression for $V_{\rm eff}(\rho)$:

$$V_{\rm eff}(\rho) = \frac{q_1 q_2}{\epsilon} \sum_{n=0}^{\infty} \frac{C_n(\sigma_{z1}, \sigma_{z2}) P_{2n}(d/\sqrt{\rho^2 + d^2})}{(\rho^2 + d^2)^{(2n+1)/2}}, \quad (13)$$

where we have introduced the abbreviation $C_n(\sigma_{z1}, \sigma_{z2})$ defined as

$$C_n(\sigma_{z1}, \sigma_{z2}) \equiv \frac{1}{\pi} \sum_{j=0}^{n} \frac{(2n)!}{(2n-2j)!(2j)!} \times \Gamma\left(n - j + \frac{1}{2}\right) \Gamma\left(j + \frac{1}{2}\right) (\sigma_{z1})^{2n-2j} (\sigma_{z2})^{2j}. \quad (14)$$

Notice that expansion (13) is valid for any values of the length parameters d and any σ's. However, when $d = 0$ and all the σ's are nonvanishing, this expansion is just valid for $z > \max\{\sigma_{z1}, \sigma_{z2}\}$, so that our procedure cannot describe the eigenfunctions in the special case when $d = 0$.

Since we are interested in solving the exciton problem, we set from now on $q_1 q_2 = -e^2$ where e is the electron charge. Whenever that $d > \max\{\sigma_{z1}, \sigma_{z2}\}$ we get to lowest orders in σ_{z1} and σ_{z2}

$$V_{\rm eff}(\rho) = -\frac{e^2}{\epsilon\sqrt{\rho^2 + d^2}} \left[1 + \frac{1}{4} \frac{\sigma_{z1}^2 + \sigma_{z2}^2}{(\rho^2 + d^2)^2} \left(2d^2 - \rho^2\right) + \frac{3}{4} \frac{(\sigma_{z1}^2 + \sigma_{z2}^2)^2}{(\rho^2 + d^2)^2} P_4\left(\frac{d}{\sqrt{\rho^2 + d^2}}\right)\right]. \quad (15)$$

The sign of the second term inside the square bracket has the same sign as $2d^2 - \rho^2$. For small ρ this term is positive and contributes to increase the strength of the "zero-width" Coulomb potential $e^2/(\epsilon\sqrt{\rho^2 + d^2})$. Physically this can be explained in terms of the charge density that occurs when σ grows as follows. For fixed separation of the electron and hole charge distributions (constant d), the negative change in energy caused by the charge density approaching each other dominates the positive change produced by the density spread to the outer side of the layers. Conversely, $2d^2 - \rho^2$ is negative for large ρ and the σ^2 terms contribute to decrease the zero-width Coulomb potential because these terms represents the electrostatic interaction of two charge distributions when one of them is located almost on top of the other. Therefore small growth of σ implies slightly larger interaction distance between charge distributions.

3. Numerical results and discussion

By employing the eigenenergies and eigenfunctions of the 2D exciton when both electron and hole lie in the same 2DL $(d = 0)$ [5], we can calculate the groundstate of our system by solving Eq. (5) by common time-independent perturbation theory. The perturbed exciton states will tend to the unperturbed states in the limits vanishing thicknesses $(\sigma_{z\nu} \to 0)$ and a single 2DL $(d \to 0)$. The unperturbed eigenenergies and eigenfunctions satisfy the same equation as (5) but with a screened two-particle Coulomb potential instead of $V_{\rm eff}(\rho)$. The equation for the unperturbed states is

$$\left(\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2} - \frac{l^2}{\rho^2} - \frac{e^2}{\epsilon\rho}\right) f_{n,l}^{(0)}(\rho, \phi) = E_{n,2\rm D}^{(o)} f_{n,l}^{(o)}(\rho, \phi), \quad (16)$$

and the corresponding bound states eigenenergies and eigenfunctions are [5],

$$E_{n,2\rm D}^{(o)} = -\left|E_o^{3\rm D}\right| \frac{1}{\left(n + \frac{1}{2}\right)^2} \quad \text{with} \quad \mathrm{n} = 0, 1, \ldots, \quad (17)$$

$$f_{n,l}^{(o)}(\rho, \phi) = \Xi_{n,l}^{(o)}(\rho) \exp(il\phi) = A_{n,l} \rho'^{|l|} \exp(-\frac{\rho'}{2}) L_{n+|l|}^{2|l|}(\rho') \exp(il\phi), \quad (18)$$

where $V_o(\rho) = -e^2/(\epsilon\rho)$ is the Coulomb interaction for a single 2DL, $E_o^{3\rm D} = -e^2/(2\epsilon a_o^{3\rm D})$ is the 3D exciton groundstate binding energy, $a_o^{3\rm D} = \hbar^2\epsilon/(e^2\mu)$ is the 3D exciton Bohr radius, $\mu = m_1 m_2/(m_1 + m_2)$ is the reduced mass, $\rho' = \frac{2\rho}{(n+1/2)a_o^{3\rm D}}$, $L_q^p(\rho') = \sum_{\nu=0}^{p-q}(-1)^{\nu+p} \frac{(q!)^2(\rho'^{\nu})}{(q-p-\nu)!(p+\nu)!\nu!}$ are the associate Laguerre polynomials [6] and $A_{n,l}$ is a normalization constant. Notice that $E_{o,2\rm D}^{(o)} = 4E_o^{3\rm D}$. Obviously, the eigenenergies of the continuum 2D exciton states take non-negative values.

Therefore, to solve Eq. (5) perturbatively, we make use of Eqs. (5), (17) and (18) where the perturbing operator term is $V_p(\rho) = V_{\rm eff}(\rho) - V_o(\rho)$. In order to explore the effects of our multipolar expansion of the Coulomb interaction potential, we proceed to present calculations for InAs layers, where for simplicity we have used the InAs 3D exciton bulk values [7], namely, $\epsilon = 14.55$, $m_1 = 0.026\, m_e$, $m_2 = 0.025\, m_e$ for the electron and light hole (m_e is the electron mass), respectively, yielding for the 3D exciton the values $E_o^{3\rm D} = 0.819$ meV and $a_o^{3\rm D} = 604$ Å. For $\sigma_{z1} = \sigma_{z2} \equiv \sigma$ we plot in Fig. 1 exciton groundstate energies in terms of the 2D exciton groundstate $E_{o,2\rm D}^{(o)}$ and in Fig. 2 the corresponding ρ-dependent density probability. As shown in Fig. 1, the exciton binding energy increases as the separation between layers also increases, as compared with the 2D exciton, as expected. On the other hand, the change in the binding energy decreases as σ increases for all values of d. Figure 2 shows how the height of the normalized wavefunction gets monotonically lowered as a function of d due to a weaker interlayer Coulomb interaction for larger d. We also found that for fixed values of d the wavefunction does not change much when σ varies. For these parameters, we found that the perturbative first-order contribution is much larger than the remaining contributions, and that the contribution of the last term of the square bracket of (15) (proportional to $\sigma_{z\nu}^4$) to both groundstate energy and

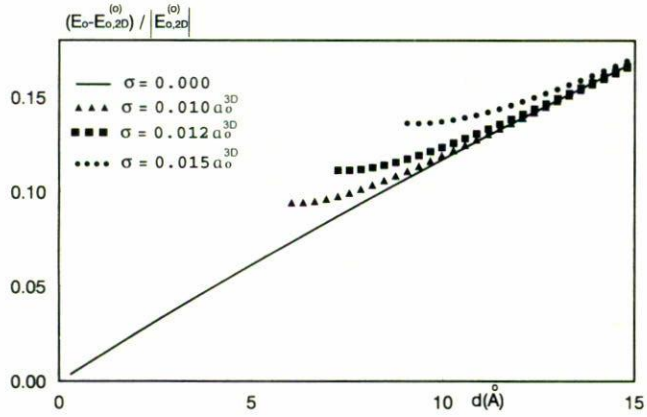

FIGURE 1. Exciton groundstate energies E_o for InAs layers when $\sigma_1 = \sigma_2 \equiv \sigma$ as function of d for various values of σ/a_o^{3D}. The range of all curves start at $d = \sigma$. We use $E_{o,2D}^{(o)} = 4E_o^{3D} = -3.276$ meV as reference and normalization constant in our plots.

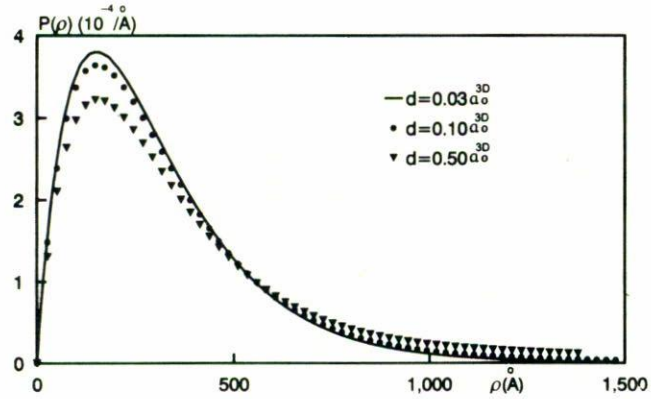

FIGURE 2. Normalized groundstate radial probability density $P(\rho) = \rho|\Xi_{o,o}(\rho)|^2$ of the exciton wavefunctions for InAs layers for various values of d and $\sigma = 0.01a_o^{3D}$. Integration over ρ yields $1/2\pi$ since integration over ϕ is not included.

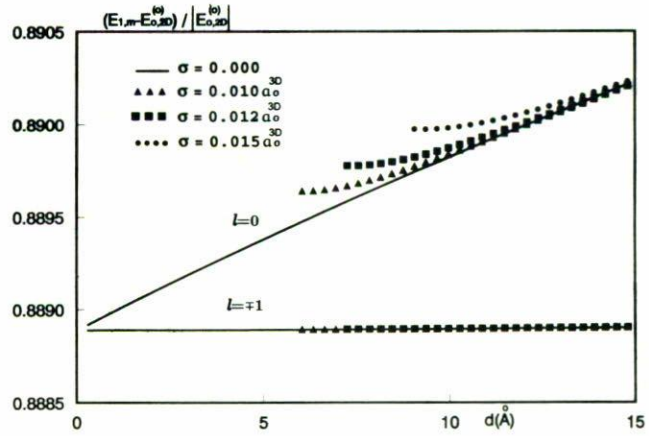

FIGURE 3. Same as Fig. 1 but for excited states and with a different scale in the ordinate axis. Exciton energies $E_{1,l}$ with $l = 0, \pm 1$ are labeled analogously to 2D exciton since former energies were obtained from the 2D system by first-order degenerate perturbation theory.

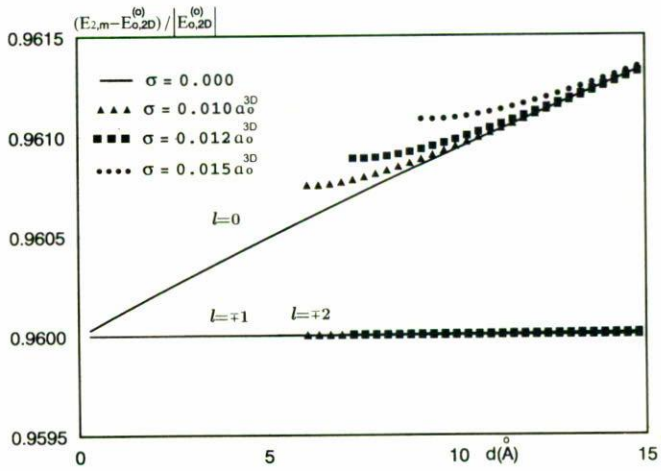

FIGURE 4. Same as Fig. 3, but with $E_{2,l}$ with $l = 0, \pm 1, \pm 2$.

wavefunction is negligible, as compared with those of the other terms of Eq. (15). First excited states were calculated by making use of first-order degenerate perturbation theory since 2D unperturbed excited states are degenerated. For instance $f_{1,l}^{(o)}$ and $f_{2,l}^{(o)}$ exhibit three- and five-fold degeneracy, respectively. The ρ-dependence of the unperturbed $\pm l$ states is the same and since V_p depends only on ρ, then $< f_{n,l}^{(o)} \mid V_p \mid f_{n,l'}^{(o)} >$ is diagonal in both the $n = 1$ and $n = 2$ states basis and perturbation removes degeneracy partially; $E_{1,0} \neq E_{1,1} = E_{1,-1}$ and $E_{2,0} \neq E_{2,1} = E_{2,-1} \neq E_{2,2} = E_{2,-2}$ where $E_{n,l} = \langle f_{n,l}^{(o)} \mid V_p \mid f_{n,l}^{(o)} \rangle$. Figures 3 and 4 show that excited-state energies are, unlike groundstate energy, almost independent of σ that is, these states could be calculated by approximating Eq. (15) by $V_{\text{eff}}(\rho) = -e^2/\epsilon\sqrt{\rho^2 + d^2}$. This is more evident for $l \neq 0$ states and in Fig. 4 the $l = \pm 1$ and $l = \pm 2$ curves are practically indistinguishable.

Since we assumed that in the confinement direction z both electron and hole are in their respective groundstate, let us discuss two physical consequences of considering confinement excited states ($E_{\nu s}^t$ with $s \neq 0$). In the first place, it is well known that in highly confined electronic systems (as in our case), energy separation between adjacent levels of the exciton is much smaller than the corresponding energy separation due to transverse confinement. For instance, for $m = 0.025\ m_e$ and $\sigma = 100$ Å Eq. (9) yields a groundstate energy $E_{\nu 0}^t = 15.24$ meV and a separation between this state and the first excited state of $3E_{\nu 0}^t$. Therefore, if electron or hole (or both) were in any excited state of the harmonic confinement, the total energy of the system Eq. (9) would increase in a much larger scale than the scale of the excitonic energies. Secondly, in regards to the continuum exciton states, it was shown in Ref. 8 that high-energy bound states (corresponding to large values of the principal quantum number n) have a spectrum of negative values of the excitonic energy E which approaches zero as n increases. This result was obtained assuming that in the confinement direction both electron and hole are in their respective ground-

state. That is, there is a discrete (although infinite) energy spectrum for $E < 0$ and a continuum spectrum for $E > 0$ when $s = 0$ in both E^t_{1s} and E^t_{2s}. Thus, a confinement excited state corresponds to an unbound state of the system immersed in a continuum spectrum, in other words, it corresponds to a resonant scattering state of the electron and the hole -not an exciton anymore. In Ref. 8, the WKB approximation (or Bohr-Sommerfeld quantization of the action integral) was employed to calculate approximately the eigenenergies and eigenfunctions of the bound states of the exciton. This approximation provides convenient analytical expressions as well as an overall picture of the exciton spectra, but it appropriately describes only high-energy states.

In summary, we developed an exact multipolar expansion of the interlayer electron-hole potential for arbitrary values of layer widths σ_{z1} and σ_{z2} [Eq. (13)]. Our results go beyond Lozovick and Nishanov's calculations [1] which are variational with $\sigma_{z1} = \sigma_{z2} = 0$. As an application of this expansion we chose to perturbatively calculate the lowest states of a system consisting of two adjacent 2DLs for a set of feasible parameters. From our results it can be noticed that finite width effects can not been neglected . This works on the theory of the lowest states of our system and presents a general analytical multipolar expansion of the interlayer potential. We hope that our efforts can stimulate further experimental and theoretical work on the study of heterostructure systems that exhibit spatial separation between the electron and the hole.

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