Size quantization of an exciton: A toy model of the “dead layer”

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Size-quantization levels of an exciton in large nanocrystals is studied theoretically. For the nanocrystal size, \( L \), much bigger than the Bohr radius, \( a_B \), the level positions do not depend on \( a_B \). The correction to the levels in a small parameter \( a_B/L \) depends on the reflection phase of the exciton from the boundary. Calculation of this phase constitutes a three-body problem: electron, hole, and the boundary. This calculation can be performed analytically in the limit when the hole is much heavier than the electron. Physically, a slow motion of the hole towards the boundary takes place in the effective potential created by the fast motion of the electron orbiting the hole and touching the boundary. As a result, the hole is reflected before reaching the boundary. The distance of the closest approach of the hole to the boundary (the dead layer) exceeds \( a_B \) parametrically.

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I. INTRODUCTION

A concept of the exciton dead layer has emerged in the course of study of the light reflection from a boundary between air and a medium with strong exciton-photon coupling. As a result of this coupling, two (exciton-like and photon-like) waves can propagate in the medium. Thus, in order to find the reflection coefficient, conventional boundary conditions of the continuity of the tangent components of electric and magnetic fields, additional boundary condition is needed. Analysis in the pioneering paper by Hopfield and Thomas suggests that the form of this condition is vanishing of the exciton polarization at certain distance, \( l \), away from the boundary. This distance was estimated in Ref. 2 as \( l = 2a_B \), where \( a_B \) is the Bohr radius of the exciton. In early, see e.g. Refs. 3–8 as well as in recent, see e.g. Refs. 9–11 follow-up papers the concept of dead layer associated with distance, \( l \), was employed.

In the absence of the exciton-photon coupling, the issue of reflection of the exciton from the surface is still important. Namely, the phase of the reflection coefficient defines the positions of the size-quantization level of the exciton in the quantum well.

In particular, in Ref. 4 photoluminescence spectra from thick GaAs quantum well revealed a number of peaks attributed to the size quantization of excitons. Their positions were fit by a formula

\[
E_n = E_x + \frac{\pi^2 \hbar^2 n^2}{2M(L-2l)^2},
\]

where \( L \) is the well thickness, \( M \) is the net mass of the exciton, and \( E_x \) is the position of the exciton line in the bulk GaAs. For the dead-layer thickness, the value \( l = a_B/2 \) was chosen.

Size quantization also manifests itself in the positions of the exciton luminescence lines in nanocrystals. These studies, pioneered in Refs. 12–14, were later conducted on wide variety of nanocrystals. The importance of dead layer for the size-quantization in ZnO nanocrystals was studied in Ref. 14.

Recent revival of interest to excitons in nanocrystals is related to inorganic perovskites, which, due to their exceptionally high photoemission rates, show a good promise for applications. As demonstrated experimentally in Ref. 15 these nanocrystals exhibit a strong size-quantization effect. Further experiments, see e.g. Refs. 16–19 indicated that nanocrystal sizes vary in the range 10nm-50nm, while the exciton Bohr radius was estimated as \( a_B \sim 2 - 6 \text{nm} \).

On the theoretical side, microscopic description of the exciton dead layer poses a challenge, since it is a three-body problem: electron, hole and a boundary. In other words, while the motion of electron and hole can be separated into the motion of the center of mass and the relative motion, this separation is inconsistent with the boundary conditions that the two-particle wave function turns to zero when electron and hole touch the boundary individually. These no-escape boundary conditions constitute a microscopic origin of the dead layer.

Below we consider a toy model which allows to derive the dead layer analytically and confirm Eq. (1) rigorously from the Schrödinger equation. A crucial assumption, which allows us to capture the electron-hole correlation in the presence of the boundary, is that the hole mass, \( m_h \), is much bigger than the electron mass, \( m_e \). Under this assumption, to the first order in \( m_e/m_h \), the fast motion of electron takes place in the field of a “static” hole. Our main conclusion is that, in the formula Eq. (1), the thickness of the dead layer, \( l_N \), is not constant, but depends on the number, \( N \), of the quantization level.

II. THE MODEL

To maximally simplify the calculations, we assume that the confinement is one-dimensional in the form of two hard walls at \( x = \pm l \), see Fig. A. Moreover, we will replace the Coulomb attraction between the electron and hole by a short-range attraction. Under these assumptions, the Hamiltonian of the model reads
where function Ψ(hole, respectively; v_nian Eq. (2) only as a parameter, which can be viewed

The discontinuity of the derivative at \( x_c = x_h \) yields the second condition

\[
\kappa_h \alpha \cosh \left[ \kappa_h \left( \frac{L}{2} + x_h \right) \right] - \kappa_h \beta \cosh \left[ \kappa_h \left( x_h - \frac{L}{2} \right) \right] = -\frac{2m_e v_0}{\hbar^2} \alpha \sinh \left[ \kappa_h \left( \frac{L}{2} - x_h \right) \right].
\]

Combining Eqs. (5) and (6), we get the equation for \( \kappa_h \)

\[
\sinh \kappa_h L = \frac{2m_e v_0}{\hbar^2 \kappa_h} \sinh \left[ \kappa_h \left( \frac{L}{2} - x_h \right) \right] \sinh \left[ \kappa_h \left( \frac{L}{2} + x_h \right) \right].
\]

This equation defines the dependence \( \kappa_h(x_h) \), i.e. the dependence of the electron energy on the position of the hole.

The concept of a dead layer is meaningful only when the “nanocrystal” is big, namely, \( L \) is much bigger than the size of the exciton. In the limit \( \kappa_h L \gg 1 \) and \( \left( \frac{L}{2} - x_h \right) \ll L \), we can replace \( \sinh \kappa_h L \) by \( \frac{1}{2} \exp(\kappa_h L) \) and \( \sinh \left[ \kappa_h \left( \frac{L}{2} + x_h \right) \right] \) by \( \frac{1}{2} \exp(\kappa_h L) \). Then Eq. (7) assumes the form

\[
\kappa_h = \frac{m_e v_0}{\hbar^2} \left\{ 1 - \exp \left[ -2\kappa_h \left( \frac{L}{2} - x_h \right) \right] \right\},
\]

where the exponent in the square brackets accounts for electron “hitting” the right wall and does not “sense” the left wall. When the exponent is small, Eq. (8) yields

\[
\kappa_h \approx \frac{m_e v_0}{\hbar^2} \frac{1}{a_B}.
\]

and, correspondingly, \( E_B = \frac{\hbar^2}{2m_e a_B^2} \) for the binding energy.

As \( x_h \) approaches the right (or left) wall, the bound state disappears at critical \( \frac{L}{2} - x_h = \frac{a_B}{2} \). Near the threshold, the solution of Eq. (8) behaves linearly with \( x_h \)

\[
\kappa_h = \frac{\left( \frac{L}{2} - x_h - \frac{a_B}{2} \right)}{\left( \frac{L}{2} - x_h \right)^2} \approx \frac{4}{a_B^2} \left[ \frac{L}{2} - x_h - \frac{a_B}{2} \right].
\]

The corresponding normalized wave functions have the form

\[
\varphi_{x_h}(x_e) = \frac{(2\kappa_h)^{1/2}}{a_B} \exp \left[ \kappa_h (x_e - x_h) \right], x_e < x_h.
\]
With $E_e(\kappa_h)$ depending on $x_e$ via Eq. (8), a hole with a finite mass will move slowly in the effective potential created by a fast-moving electron. We will establish the form of this potential in the next Section.

### III. EFFECTIVE POTENTIAL FOR A HOLE

To incorporate the hole motion, we search for the solution of the Schrödinger equation $\hat{H}(x_e, x_h)\Psi = E\Psi$ in the form

$$\Psi(x_e, x_h) = \varphi_{x_e}(x_e)\Phi(x_h).$$

Substitution of this form into the Schrödinger equation yields

$$-\frac{\hbar^2}{2m_e} \left[ \frac{\partial^2 \varphi_{x_e}(x_e)}{\partial x_e^2} + 2 \frac{\partial \varphi_{x_e}(x_e)}{\partial x_e} + \frac{\partial^2 \phi_{x_h}(x_e)}{\partial x_h^2} - E\varphi_{x_e}(x_e) \right] = \left[ E - E_e(\kappa_h) \right] \varphi_{x_e}(x_e)\Phi(x_h).$$

In deriving Eq. (13) we took into account that $\varphi_{x_e}(x_e)$ satisfies the equation

$$\left[ \frac{\hat{p}_e^2}{2m_c} - v_0 \delta(x_e - x_h) \right] \varphi_{x_e}(x_e) = E_e(\kappa_h) \varphi_{x_e}(x_e).$$

We assume that the electron wave function $\varphi_{x_e}(x_e)$ is normalized $\int dx_e \left( \varphi_{x_e}(x_e) \right)^2 = 1$. To obtain a closed equation for the hole wave function, $\Phi(x_h)$, we multiply Eq. (13) by $\varphi_{x_e}(x_e)$ and integrate over $x_e$. This yields

$$-\frac{\hbar^2}{2m_h} \left[ \frac{\partial^2 \phi_{x_h}(x_e)}{\partial x_h^2} + 2I(x_h) \frac{\partial \phi_{x_h}(x_e)}{\partial x_h} + J(x_h) \Phi \right] = \left[ E - E_e(\kappa_h) \right] \Phi,$$

where the functions $I(x_h)$ and $J(x_h)$ are defined as

$$I(x_h) = \int dx_e \varphi_{x_e}(x_e) \frac{\partial \varphi_{x_e}(x_e)}{\partial x_e},$$

$$J(x_h) = \int dx_e \varphi_{x_e}(x_e) \frac{\partial^2 \varphi_{x_e}(x_e)}{\partial x_e^2} - \frac{\partial \varphi_{x_e}(x_e)}{\partial x_e} \frac{\partial \varphi_{x_e}(x_e)}{\partial x_e}.$$

Normalization condition ensures that $I(x_h) = 0$ and that the first term in the right-hand side in the expression for $J(x_h)$ is zero. The fact that the equation for $\Phi(x_h)$ is closed is a consequence of our choice Eq. (12) of the wave function $\Psi(x_e, x_h)$ in the form of the product. By making this choice, we neglected the excited states of the electron wave function. This choice is justified if the typical electron energy, $E_e(\kappa_h)$, is much bigger than the size-quantization energy, $\sim \hbar^2/m_h L^2$, of the hole. Upon setting $I(x_h) = 0$ we cast Eq. (13) into the form of the Schrödinger equation

$$-\frac{\hbar^2}{2m_h} \frac{\partial^2 \Phi}{\partial x_h^2} + V(x_h)\Phi = E\Phi,$$

with effective potential, $V(x_h)$, representing the sum

$$V(x_h) = \frac{\hbar^2}{2m_h} \int dx_e \left( \frac{\partial \varphi_{x_e}(x_e)}{\partial x_h} \right)^2 - \frac{\hbar^2}{2m_c} \left( k_h(x_h) \right)^2.$$

### IV. COMPARING CONTRIBUTIONS TO THE EFFECTIVE POTENTIAL

At distances much bigger than $a_B$ from the boundary the second term in $V(x_h)$ dominates, so that $V(x_h) = -E_B$. Upon approach to the right boundary $x_h = \frac{L}{2}$ the term $\hbar^2 k_h^2/2m_c$ falls off and finally vanishes at $x_h = \frac{L-a_B}{2}$. Near this point the first term in $V(x_h)$ dominates the effective potential. To trace the crossover history of the first and second terms, it is sufficient to use the asymptotic expressions Eqs. (10) and (11). Differentiating Eq. (11) with respect to $x_e$ we get

$$\frac{\partial \varphi_{x_e}(x_e)}{\partial x_h} = \left[ 1 + 2\kappa_h(x_e - x_h) \right] \exp \left[ \kappa_h(x_e - x_h) \right] \frac{d\kappa_h}{dx_h}.$$

Expressing $d\kappa_h/dx_h$ from Eq. (10) we have

$$\left( \frac{\partial \varphi_{x_e}(x_e)}{\partial x_h} \right)^2 = \frac{8}{\kappa_h a_B^2} \left[ 1 + 2\kappa_h(x_e - x_h) \right]^2 \exp \left[ 2\kappa_h(x_e - x_h) \right].$$

Then the integration over $x_e$ yields

$$\frac{\hbar^2}{2m_h} \int dx_e \left( \frac{\partial \varphi_{x_e}(x_e)}{\partial x_h} \right)^2 = \frac{2\hbar^2}{m_h \kappa_h^2 a_B^4} = \frac{\hbar^2}{8 \kappa_h^2 a_B^4} \left[ L - a_B - 2x_h \right]^2.$$

Thus, the first term in $V(x_h)$ falls off quadratically away from the threshold $x_h = \frac{L-a_B}{2}$. It should be compared to the second term, $\hbar^2 k_h^2/2m_c$. Using the threshold behavior Eq. (10) of $\kappa_h$, we have

$$\frac{\hbar^2 \kappa_h^2}{2m_c} = \frac{2\hbar^2}{m_c a_B^4} \left[ L - a_B - 2x_h \right]^2.$$
Comparing Eqs. (22) and (23) we conclude that the crossover from the first to the second term in \( V(x_h) \) takes place at

\[
L - a_B - 2x_h = \frac{a_B}{2} \left( \frac{m_e}{m_h} \right)^{1/4}. \tag{24}
\]

We see that crossover takes place in the threshold region, i.e. at \( (L - a_B - 2x_h) \ll a_B \). This is ensured by the smallness of the ratio \( m_e/m_h \). Note, that this smallness also justifies the above use of the asymptotic expressions for \( \varphi_h(x_e) \) and for \( \kappa_h(x_h) \).

V. ENERGY LEVELS

Semiclassical quantization condition for the particle with mass, \( m_h \), moving in the potential, \( V(x_h) \), see Fig. 4, reads

\[
2 \left( \frac{2m_h}{\hbar^2} \right)^{1/2} \int_0^{x_t} dx_h \left[ E_N + \frac{\hbar^2 (\kappa_h(x_h))^2}{2m_e} \right]^{1/2} = \pi \left( N + \frac{1}{2} \right), \tag{25}
\]

where \( x_t \) is the turning point defined as \( \hbar \kappa_h(x_t) = (2m_e|E_N|)^{1/2} \). Using Eq. (8) we get the following expression for \( x_t \)

\[
x_t = \frac{L}{2} + \frac{a_B}{2} \left( \frac{E_B}{|E_N|} \right)^{1/2} \ln \left[ 1 - \left( \frac{|E_N|}{E_B} \right)^{1/2} \right]. \tag{26}
\]

Since \( \kappa_h \) changes only in the vicinity of the upper limit and is equal to \( \frac{1}{a_B} \) otherwise, it is convenient to isolate the constant part

\[
\left[ E_N + \frac{\hbar^2 \kappa_h^2}{2m_e} \right]^{1/2} + \frac{\hbar^2}{2m_e} \left( \kappa_h - \frac{1}{a_B} \right)^{1/2} = \left[ E_N + \frac{\hbar^2}{2m_e a_B^2} \right]^{1/2}.
\]

Integration of the first term is elementary. To perform integration of the second term it is convenient to switch from the variable \( x_h \) to the dimensionless variable \( u = \kappa_h a \). To do so, we differentiate both sides of Eq. (8). This yields

\[
\frac{du}{2u^2} \left[ \ln (1 - u) + \frac{u}{1 - u} \right] = -\frac{dx_h}{a_B}. \tag{28}
\]

Then the condition Eq. (25) takes the form

\[
\frac{\pi}{2} \left( N + \frac{1}{2} \right) \left( \frac{m_e}{m_h} \right)^{1/4} = (1 - q_N)^{1/2} \left[ \frac{L}{2a_B} + \frac{\ln \left( 1 - q_N^{1/2} \right)}{2q_N^{1/2}} \right] - I(q_N). \tag{29}
\]

where the function \( I(q_N) \) is defined as

\[
I(q_N) = \int_{q_N^{1/2}}^1 \frac{du}{2u^2} \left[ \ln (1 - u) + \frac{u}{1 - u} \right] u^2 - q_N^{1/2} + (1 - q_N)^{1/2}, \tag{30}
\]

The parameter

\[
q_N = \frac{|E_N|}{E_B} \tag{31}
\]

is the dimensionless exciton energy, so that \( 1 - q_N \) has a meaning of the dimensionless size-quantization energy. In the limit of large \( L \) the size-quantization energy is much smaller than \( E_B \), so that \( (1 - q_N) \ll 1 \). This, in turn, means that the lower limit in the integral Eq. (30) is close to 1. Then the integral \( I(q_N) \) can be evaluated asymptotically in small parameter \( 1 - q_N \), which yields

\[
I(q_N) = \frac{(1 - q_N)^{1/2}}{2} \ln 2. \tag{32}
\]

Substituting this expression in the right-hand side of Eq. (29), we arrive at a closed equation for \( q_N \), which can be also viewed as the equation for the electron energies, \( E_N \). Solving this equation in the limit of large \( L \), we get

\[
E_N = -E_B + \frac{\hbar^2 \pi^2 n^2}{2m_h (L - l_N)^2}, \tag{33}
\]

where \( l_N \) is given by

\[
l_N = a_B \ln \left[ \frac{L}{\pi a_B (N + \frac{1}{2})} \right] \frac{\pi a_B^2}{2m_h L^2} \left( N + \frac{1}{2} \right)^2
\]

\[
= 2a_B \left[ \ln \frac{L}{\pi a_B (N + \frac{1}{2})} + \frac{1}{2} \ln \frac{m_h}{m_e} \right]. \tag{34}
\]

We see that the result Eq. (33) essentially reproduces Eq. (1) but with the width of the dead layer specified. We see that the width, \( l_N \), grows with \( L \), which is somewhat non-trivial.
VI. DISCUSSION

It is seen from Eq. (34) that the arguments of both logarithms in the right-hand side are large. This suggests that the size of the dead layer exceeds the exciton radius parametrically. On the other hand, it follows from Eq. (22) that the effective repulsive potential for a hole diverges at \( x_h = \frac{1}{2}(L - a_B) \), i.e. at a distance \( \frac{a_B}{2} \) from the “hard wall” at \( x_h = \frac{L}{2} \).

The fact that the turning point, \( x_t \), for the hole motion, given by Eq. (26), is parametrically further away from the boundary than \( a_B \) suggests the following picture of the exciton reflection. As a hole slowly moves towards the boundary, it is orbited by a fast-moving electron. The energy of the system is \(-E_B\). Upon the approach to the boundary, the electron “senses” the boundary by virtue of the no-escape boundary condition. As a result, the energy of the system electron+hole increases. This increase acts as a barrier for the hole, from which the hole is reflected being accompanied by the electron.

Our conclusion that \( t \gg a_B \) suggests that the above assumption about the short-range character of the electron-hole attraction can be relaxed. Moreover, we can extend the above consideration to the 3D case. In the Appendix we derive the form of potential barrier, \( \tilde{V}(x_h) \), which enters into the quantization condition

\[
2 \left( \frac{2m_e}{\hbar^2} \right)^{1/2} \int_0^{x_t} dx_h \left[ E_N + E_B - \tilde{V}(x_h) \right]^{1/2} = \pi \left( N + \frac{1}{2} \right),
\]

With the help of Eq. (A5) we find the positions of the turning points

\[
x_t = \frac{L}{2} + \frac{a_B}{2} \ln \left( 1 - \frac{|E_N|}{E_B} \right),
\]

which appears to be similar to the 1D expression Eq. (26). Repeating the steps in the previous section, it can be readily shown that our main result Eq. (34) for the width of the dead layer remains valid in three dimensions.

Appendix A: Effective potential in 3D

In this Appendix we calculate the correction to the ground state energy of electron in the field of a hole due to the presence of a boundary at \( x = \frac{L}{2} \). We start by writing down the Schrödinger equations with and without the boundary

\[
- \frac{\hbar^2}{2m_e}\Delta\psi_0 - \frac{e^2}{|\mathbf{r}_e - \mathbf{r}_h|}\psi_0 = -E_B\psi_0, \quad (A1)
\]

where the function \( W(x_e - \frac{L}{2}) \) describes a barrier which ensures that electron does not penetrate into the region \( x_e < \frac{L}{2} \).

Multiplying Eq. (A1) by \( \psi(r) \) and Eq. (A2) by \( \psi_0(r) \), subtracting the two, and integrating over the domain \( x_e < \frac{L}{2} \), we find

\[
E(x_h) + E_B = -\frac{\hbar^2}{2m_e} \int dy_e dz_e \left( \frac{\partial \psi_0}{\partial x_e} - \psi_0 \frac{\partial \psi}{\partial x_e} \right)_{x_e = \frac{L}{2}} \\
\approx \frac{\hbar^2}{4m_e} \int dy_e dz_e \frac{\partial \psi_0^2}{\partial x_e}. \quad (A3)
\]

In Eq. (A3) we took into account that the first term in the brackets is zero, since \( \psi(r) = \psi_0(r) \) at the boundary. We have also set \( \psi(r) = \psi_0(r) \) in the second term. Integration over the plane \( x_e = \frac{L}{2} \) in the right-hand side yields the correction to the binding energy of the exciton due to the presence of the boundary.

The ground state wave function of an electron in the field of a hole which is located at \((x_h,0,0)\) has the form

\[
\psi_0(r_e) = \frac{1}{(\pi a_B^3)^{1/2}} \exp \left\{ -\frac{\left[ y_e^2 + z_e^2 + (x_e - x_h)^2 \right]^{1/2}}{a_B} \right\}.
\]

With \( \psi_0(r_e) \) given by Eq. (A4), the integration in Eq. (A3) can be performed explicitly in polar coordinates

\[
E(x_h) + E_B = \frac{\hbar^2}{2m_e a_B^3} \int_0^{\infty} dr^2 \exp \left[ -\frac{2}{a_B} \left( \rho^2 + \left( \frac{L}{2} - x_h \right)^2 \right)^{1/2} \right] \\
= \frac{\hbar^2}{2m_e a_B^3} \exp \left[ -\frac{2}{a_B} \left( \frac{L}{2} - x_h \right) \right] = \tilde{V}(x_h). \quad (A5)
\]

The result Eq. (A5) defines the form of the barrier from which the exciton is reflected.

Appendix B: Acknowledgements

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