Exciton Binding Energy in Finite Spherical GaAs/GaAlAs Quantum Dots under an Intense THz Laser Field

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Abstract In this work, we study the ground-state binding energy of heavy-hole excitons in finite spherical GaAs/GaAlAs quantum dots at the very high terahertz laser field. The interaction with the radiation is described by a laser-dressed Coulomb potential in which the electron-hole attraction is modified by the field displacement of the charge carriers. We calculate the excitonic states in the single-band effective mass approximation using a variational method. We find that the electron and hole confinement-energy contributions are initially increasing with the laser-dressing parameter and become saturated. For all the dot radii considered, the exciton binding energy decreases and the decrease is more pronounced in larger quantum dots. Small dots tend to bind with stronger binding because we are in geometric confinement, the electron-hole wavefunction overlap is larger. The statement that the laser "attracts the exciton towards the dot center and stabilizes it" should be removed unless your calculations explicitly show this. Your results suggest that the electron-hole separation is increasing and the excitonic state weakens.

1 Introduction

Confined excitons in quantum dots made from a wide range of semiconductor compounds of II-VI and III-V have been a topic of both theory and practice for two very long years. The interest in these systems is largely due to the technology they offer - single-electron transistors, quantum dot lasers, and more general artificial atoms, artificial molecules and quantum bits. They are known to be very hard-bound in many ways - the electron and hole masses, the size of the dot, the height and shape of the confining potential, and so on - all of which depend on the type of material in the system, and all these properties are well known [3-5]. Beyond these parameters, we have also seen that external forces such as hydrostatic pressure [9], electric fields [7-8] and magnetic fields [6] can significantly affect the exciton behavior. Focusing recently on how intense THz laser fields act on excitons in GaInNAs /GaAs quantum wells and bulk semiconductor material [10-12]. In the near band edge absorption and transient optical spectra, bulk excitons can indeed be ionized to a considerable extent [12], which is supported by F. M. S. Lima and coworkers who calculated the exciton binding energy in bulk GaAs and found it to be near one quarter of the exciton Rydberg energy and falling as the intensity of THz laser field is increased [10]. Similarly, U.I. Yesilgul and coworkers found that in the ground state exciton in GaInNAs /GaAs quantum wells is also confined at different well widths, and this behavior is due to the extra geometric confinement that the THz field acts on [13]. As far as we know, the case we study here has not been investigated in the literature before.

2 Equations

We consider a spherical GaAs quantum dot of radius R, embedded in a GaAlAs barrier. An electron–hole pair X(e,h) is assumed to be confined within the structure. In the absence of an external field, the exciton Hamiltonian is written as:

$$H_0 = T_e + T_h + V_c + V_e + V_h \qquad (1)$$

where T_e and T_h are the kinetic-energy operators of the electron and hole, respectively. The terms V_e and V_h represent the finite confinement potentials produced by the conduction- and valence-band offsets at the GaAs/GaAlAs interface, while V_C describes the attractive Coulomb interaction between the two carriers. The confinement potential for each carrier i=e,h, is defined by:

$$V_{e,h}(\vec{r}_{e,h}) = \begin{cases} 0 & pour\ r_{e,h} < R \\ V_{e,h}^0 & pour\ r_{e,h} > R \end{cases} \qquad (2)$$

$$V_C = -e^2/4\pi\varepsilon r_{eh}$$

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$$T_{e,h} = -\frac{\hbar^2}{2m_{e,h}}\nabla^2_{e,h}$$

with m_e (m_h) the electron (hole) effective mass, $\vec{r}_{e,h}$ the carrier's radial coordinate, R the dot radius and ε the dielectric constant of the host material (material 1).

$$T_e = -\frac{\Delta_e}{1+\sigma_{1,2}}\,; T_h = -\frac{\sigma_{1,2}\Delta_h}{1+\sigma_{1,2}}\,;$$
$$V_C(0) == -\frac{2}{r_{eh}} \qquad (3)$$

Excitonic units (e.u.) are adopted throughout the calculations. The unit of length is the three-dimensional effective exciton Bohr radius, $a^* = \frac{\varepsilon_r \hbar^2}{\mu k^2 e^2}$ whereas the unit of energy is the effective three-dimensional exciton Rydberg, $R^* = \mu\, k^2 e^4/2\varepsilon_r{}^2\hbar^2$ with $k = \frac{1}{4\pi\varepsilon_0}$. For material j (j=1,2), the electron-to-hole effective-mass ratio is defined as : $\sigma_{1,2} = \frac{m_e^{1,2}}{m_h^{1,2}}$, $\mu = \frac{m_e^1 m_h^1}{(m\_e\hat{}1+m\_h\hat{}1)}$, $m_e^{1,2}$ ($m_h^{1,2}$) is the electron(hole) effective mass in the material 1,2.

Within the laser-dressed-potential approach, the Coulomb interaction under an intense high-frequency laser field is expressed as [10]:

$$V_c(\alpha) = -\left(\frac{1}{\left|\vec{r}_e - \vec{r}_h + \alpha\vec{k}\right|} + \frac{1}{\left|\vec{r}_e - \vec{r}_h - \alpha\vec{k}\right|}\right) \qquad (4)$$

where $\alpha = eF_0/\mu\omega^2$ is the laser field parameter and $\vec{k}$ is the unity vector along the field's polarization direction. Let's assume that the confinement potentials (V_e and V_h) do not surpass the laser field parameter for computational ease.

3 Solutions

The ground state of the exciton is to satisfy the Schrödinger equation:

$$H_\alpha \psi(\vec{r}_e, \vec{r}_h) = E\psi(\vec{r}_e, \vec{r}_h) \qquad (5)$$

with

$$H_\alpha = H_0 + V_c(\alpha) - V_c(0) \qquad (6)$$

Since the coordinates in this problem do not separate, the solution is not exact and we have to deal with the problem in an approximate way. We get the binding energy E_β through the variational process minimizing the expectation $\langle\psi|\tilde{H}|\psi\rangle$ in a family of trial wavefunctions ψ chosen to be the ground state and to be varied. As an example of the method, we use the following trial function:

$$\psi = \begin{cases} A\,\varphi_e{}^{in}(r_e)\varphi_h{}^{in}(r_h)(1+\beta\,\alpha\,Z)\varphi_{eh} & ;r_i < R \\ B\,\varphi_e{}^{out}(r_e)\varphi_h{}^{out}(r_h)(1+\beta\,\alpha\,Z)\varphi_{eh}; & r_i > R \end{cases} \qquad (7)$$

with $i = e,h$ and $Z = z_e - z_h$ the relative electron–hole separation along the polarization axis of the field; $\varphi(r)$ is the single-particle wavefunction defined inside this dot, and A, B are normalization coefficients fixed by $\int\psi^2 = 1$ over all space. The factor $\varphi_{eh} = \exp(-\lambda r_{eh})$ encodes the electron–hole spatial correlation, with λ and β serving as the variational parameters; the term $(1+\beta Z)$ is included specifically to capture the distortion the laser field imposes on the wavefunction. The exciton's ground-state binding energy then follows from:

$$E_b = E_e + E_h - min_{\beta,\lambda}\frac{\langle\psi|H|\psi\rangle}{\langle\psi|\psi\rangle} \qquad (8)$$

where E_e (E_h) is obtained by solving the corresponding single-particle problem:

$$-k^i_{out}R = k^i_{in}Rcotg(k^i_{in}R) \qquad (9)$$

with

$$k_{out}^{i} = \sqrt{\frac{2m_{out}^{i}(V_0^i - E_i)}{\hbar^2}} \quad ; \qquad i = e,h \qquad (10)$$

$$k_{in}^{i} = \sqrt{\frac{2m_{in}^{i}E_i}{\hbar^2}}$$

corresponding to the wave function:

$$\varphi_i(r_i) = \frac{A_{0ut}}{r_i}\exp(-k_{0ut}r_i)\Theta(r_i - R) + A_{in}\frac{sink_{in}r_i}{r}\Theta(R - r_i) \qquad (11)$$

where the constants A_{0ut} and A_{in} are deduced from the normalization condition which writes:

$$A_{in}^{i} = \left[2\pi\left(\frac{(k_{in}R - sink_{in}Rcosk_{in}R)}{k_{in}} + \frac{sin^2k_{in}R\exp(-2k_{0ut}R)}{k_{0ut}}\right)\right]^{-1/2}$$

$$A_{out}^{i} = A_{in}^{i}sink_{in}^{i}\exp\left(k_{out}^{i}R\right)$$

$$k_{out}^{i} = \sqrt{\frac{2m_{out}^{i}(V_0^i - E_i)}{\hbar^2}} \; ; \; k_{in}^{i} = \sqrt{\frac{2m_{in}^{i}E_i}{\hbar^2}} \; ; \; \text{i=e,h}$$

m_{in}, m_{0ut} being the particle's effective mass inside the dot (material 1) and outside the dot (material 2), respectively. We employ the full Hylleraas coordinates [14]. $(r_e, r_h, r_{eh}, z_e, z_h)$. The elementary volume in these coordinates is determined by:

$$d\tau = \frac{8\pi r_h dr_h r_e dr_e r_{eh} dr_{eh}\, dz_e dz_h}{\sqrt{4(r_e^2 - z_e^2)(r_h^2 - z_h^2) - (r_e^2 + r_h^2 - r_{eh}^2 - 2z_e z_h)^2}} \qquad (12)$$

And $\Delta_i = \frac{\partial^2}{\partial r_i} + \frac{2}{r_i}\frac{\partial}{\partial r_i} + \frac{\left(r_i^2 + r_{ij}^2 + r_j^2\right)}{r_i r_{ij}} \cdot \frac{\partial^2}{\partial r_{ij}\partial r_i} + \frac{2}{r_{ij}}\frac{\partial}{\partial r_{ij}} + \frac{\partial^2}{\partial z_i^2} + \frac{2z_i}{r_i}\frac{\partial^2}{\partial z_i \partial r_i} + 2\frac{(z_i - z_j)}{r_{ij}}\frac{\partial^2}{\partial r_{ij}\partial z_i}$ (13)

The integrations are evaluated over the domain: $0 < r_e, r_h \leq \infty$; $|r_e - r_h| \leq r_{eh} \leq r_e + r_h$; $-r_e \leq z_e \leq r_e$; $Z_1 \leq z_h \leq Z_2$ where Z_1 and Z_2 denote the two roots obtained by setting the denominator of Eq. (12) equal to zero and solving with respect to z_h . Accordingly, the volume element $d\tau$ can be written as:

$$d\tau = \frac{4\pi r_h dr_h dr_e r_{eh} dr_{eh} dz_e dz_h}{\sqrt{(z_h - Z_1)(Z_2 - z_h)}} \qquad (14)$$

With:

$$Z_1 + Z_2 = S_h = z_e\left(1 + \frac{r_h^2 - r_{eh}^2}{r_e^2}\right) \qquad (15)$$

$$Z_1 Z_2 = P_h = \frac{r_e^2}{4z_e^2}S^2 - \left(1 - \frac{z_e^2}{r_e^2}\right)r_h^2$$

4 Results and discussion

The exciton binding energy in GaAs/Ga_{1-x} Al_xAs quantum dots is determined by a variational scheme based on the effective mass approximation as well as non-perturbative treatment of the intense high-frequency laser drive. All numerical results below are given in excitonic units in terms of the effective Rydberg energy R^* and the effective Bohr radius a^* introduced earlier; for a GaAs/Ga_{1-x} Al_xAs dot with $x = 0.3$, which means $a^* = 99.6$ Å and $R^* = 6.6$ meV. Each dot has the ratio σ 1,2 and the excitonic units R^* and a^*. As shown in Fig. 1, the exciton binding energy E_β drops almost exponentially as the dressing parameter α_0 is swept from 0 to 10 excitonic units, and the number of dots does not vary.

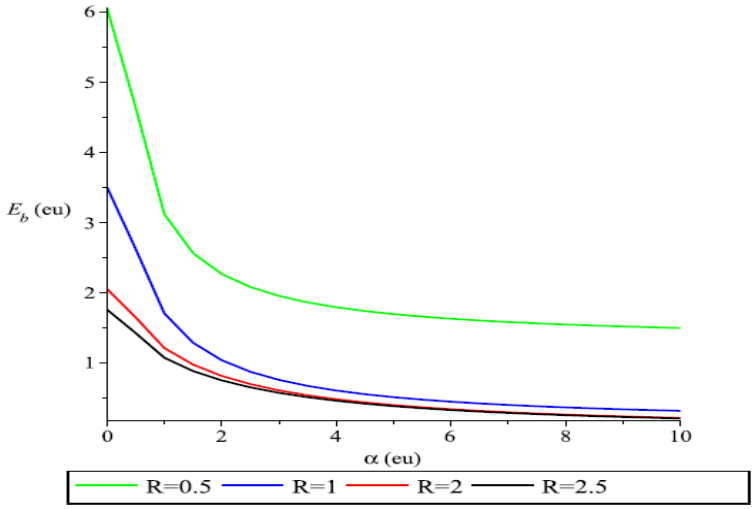

Fig 1. Exciton binding energy in GaAs/ Ga_{1-x} Al_x As versus field parameter α for different quantum dot radii

This behavior is consistent with the fact that a strong laser light is responsible for the electron-hole Coulomb interaction. The field is high enough that the charge carriers move in a very rapid oscillatory ("trembling") motion, and this causes the Coulomb potential to be spread out over a long distance α_0, which in turn weakens the electron-hole correlation captured by $\varphi_{eh} = \exp(-\lambda r_{eh})$. The resulting decrease in binding energy is hence the same as the variation. Most interestingly, the decay rate is highly related to the dot radius, as the resulting curves show a clear contrast between geometric confinement and the external laser drive. In the strong confinement regime ($R = 0.5\ a^*$, top curve), the binding energy is only gradually reduced from its peak value and is still large (1.93 R^*) even at $\alpha_0 = 10$. Small dots have a tendency to compress the electron and hole wavefunctions and in general the carriers are close together so the perturbation brought by α_0 is small compared with the strong Coulomb attraction and the exciton is not strongly ionized by the field. In the weak confinement regime ($R = 2.5\ a^*$, bottom curve) however, the binding energy is low (1.6 R^*) and is driven down to zero as α_0 increases. The exciton is loosely bound with a pronounced hydrogenic character, the laser displacement of α_0 is very close to the exciton Bohr radius and the dressed potential $Vc(\alpha_0)$ is heavily screened and the electron-hole correlation is very weak. The best variational parameter λ is very small and the exciton approaches dissociation at high field intensities. The shapes of the dots are also slightly different in the middle configuration ($R = 1.0\ a^*$ and $R = 2.0\ a^*$) and the curves are of a transitional character between the two limits. It is interesting to note that the presented results are very different from the ones reported by Ouadgui et al. For the infinite-confinement case the E_β and Vc curves are symmetric, which is not the case in finite confinement (see Figures 1 and 3).

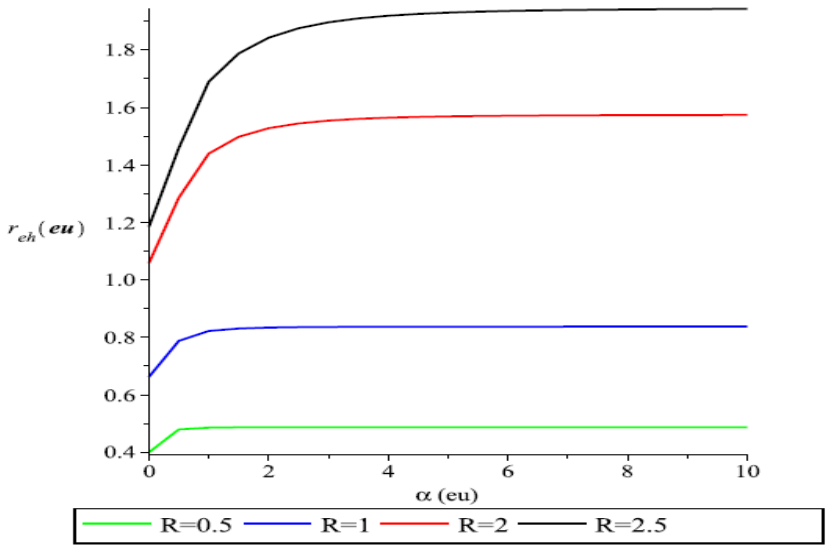

Fig 2. Changes in the exciton's spatial extension (r_{eh}) versus the field parameter (α) for different dot radii in GaAs/ Ga_{1-x} Al_x As for x=0.3.

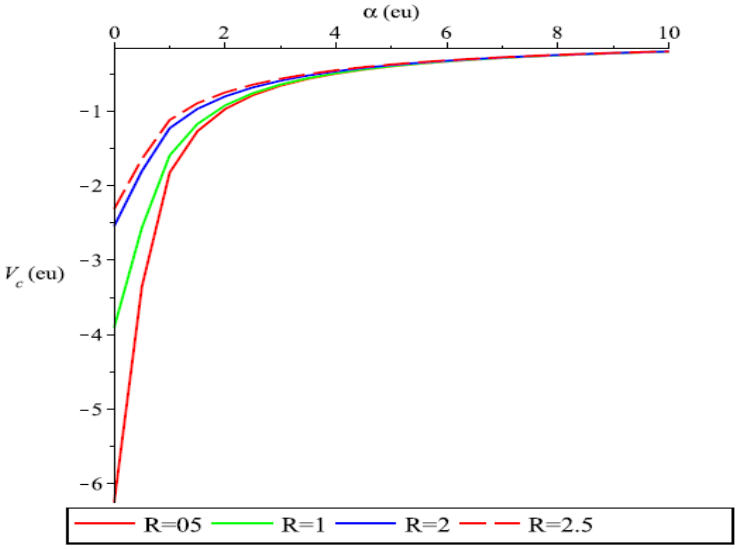

Fig 3. Exciton dressed potential against field parameter (α) in a GaAs quantum dot for different dot radii (e.u.)

In the absence of the laser field ($\alpha = 0$), the calculated binding energy for a weakly confined dot with ($R = 2\ a^*$), This value is reasonably close to the result $E_\beta \approx 2.1$, which is in good agreement with the value $E_\beta \approx 2.5$ found in [15], although the difference should not be described as exact agreement. The finite value of E_b indicates that the exciton remains bound at zero field. As the laser-dressing parameter increases, however, the electron–hole interaction becomes progressively weaker, in qualitative agreement with previous studies on bulk excitons [10] and quantum-well excitons [13]. The present calculations alone are not sufficient to establish a photoluminescence redshift, since this would require an explicit evaluation of the optical transition energy. Nevertheless, the laser-induced modification of the excitonic states may influence the absorption edge and could therefore be relevant to tunable optoelectronic devices based on quantum-dot structures. As shown in Fig. 2, the mean electron–hole separation reh increases with both the quantum-dot radius and the laser-dressing parameter. The increase is most pronounced at low values of α, after which r_{eh} gradually approaches a saturation regime. This trend reflects the progressive weakening of the spatial correlation between the electron and the hole and resembles the behavior previously observed in bulk excitonic systems. The separate contributions of the kinetic energy T and the dressed Coulomb potential Vc were also examined. Figure 3 shows that the magnitude of the Coulomb contribution depends on both R and α, while Vc approaches zero as α becomes very large. Figure 4 indicates that the kinetic energy decreases mainly in the low-field region and then varies only weakly once the laser-dressing parameter exceeds approximately unity.

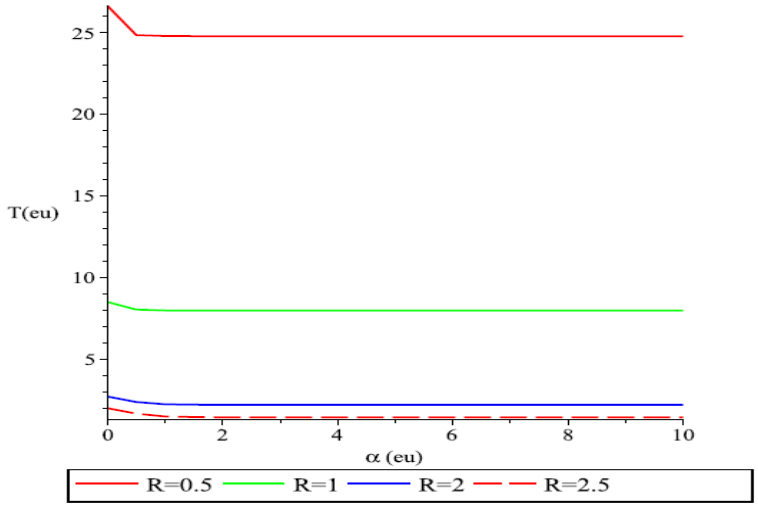

Fig 4. Kinetic energy variation with laser field for various quantum dot radii

Finally, Figures 5 and 6 show how electron and hole confinement potentials change when the fields are strongly constrained ($R = 0.5\ a^*$). Both potentials rise linearly up to $\alpha = 2$, after which the growth of V is negligible, saturating to near 0.32 for the electron and 0.185 for the hole.

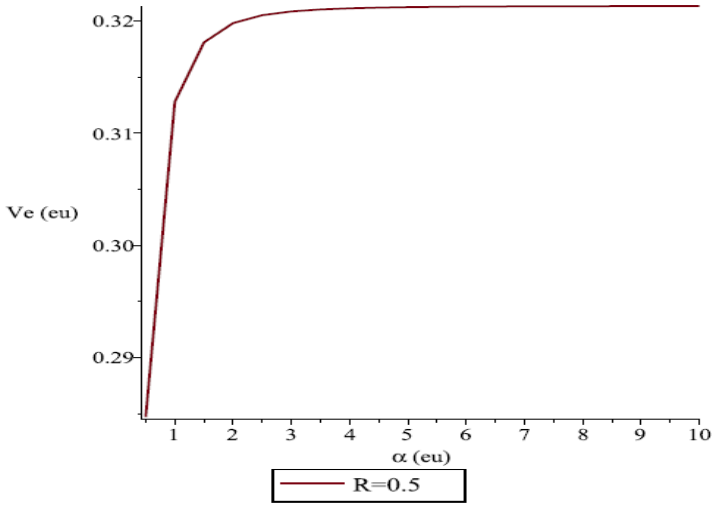

Fig 5. Potential confinement variation V_e for R=0.5 with laser field parameter α

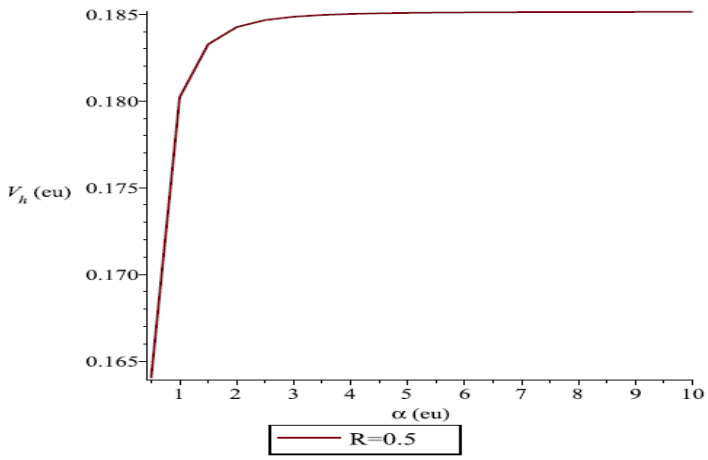

Fig 6. Potential confinement variation V_h for R=0.5 with laser field parameter α

5. Applications in Optoelectronics

The ability to tune exciton binding energies continuously with an external laser opens up several opportunities for device engineering, including terahertz modulators and optical switches. An intense laser pulse can be used to drive the binding energy down far enough to cause rapid exciton dissociation, changing the absorption regime from the one of exciton to one of free conductive charge carriers in the shorter timescale. Wavelength-tunable nanolasers. Because the laser field transforms the overall energy of the electronic state, the emission behavior of a dot-based device can be adjusted post-fabrication. Enhanced photovoltaics. Lowering the binding energy in quantum dot solar cells lowers the energy required for charge separation and thus reduces recombination losses.

6. Conclusion:

In this study, we used a variational method combined with a non-perturbative laser-dressed potential to evaluate the exciton binding energy in finite GaAs/GaAlAs quantum dots. The results show that increasing the laser-dressing

parameter weakens the electron–hole Coulomb attraction and consequently reduces the exciton binding energy. The magnitude of this effect strongly depends on the dot radius. In small quantum dots, strong geometric confinement maintains a large overlap between the electron and hole wavefunctions, allowing the exciton to remain comparatively well bound. In larger dots, the weaker confinement makes the excitonic state more sensitive to the applied laser field and favors its progressive dissociation. These findings suggest that intense THz radiation could provide a useful means of controlling excitonic states in quantum-dot-based optical modulators, tunable semiconductor lasers, and other optoelectronic devices. Further calculations of optical transition energies and experimental validation are required before practical device applications can be confirmed.

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