Development of polaron-transformed explicitly correlated full configuration interaction method for investigation of quantum-confined Stark effect in GaAs quantum dots

Christopher J. Blanton,1 Christopher Brenon,2 and Arindam Chakraborty1, a)

1) Department of Chemistry, Syracuse University, Syracuse, New York 13244, USA
2) East Syracuse Minoa High School, Syracuse, NY 13057

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The effect of external electric field on electron-hole correlation in GaAs quantum dots is investigated. The electron-hole Schrödinger equation in the presence of external electric field is solved using explicitly correlated full configuration interaction (XCFCI) method and accurate exciton binding energy and electron-hole recombination probability are obtained. The effect of the electric field was included in the 1-particle single component basis functions by performing variational polaron transformation. The quality of the wavefunction at small inter-particle distances was improved by using Gaussian-type geminal function that depended explicitly on the electron-hole separation distance. The parameters of the explicitly correlated function were determined variationally at each field strength. The scaling of total exciton energy, exciton binding energy, and electron-hole recombination probability with respect to the strength of the electric field was investigated.

It was found that a 500 kV/cm change in electric field reduces the binding energy and recombination probability by a factor of 2.6 and 166, respectively. The results show that the eh-recombination probability is affected much more strongly by the electric field than the exciton binding energy. Analysis using the polaron-transformed basis indicate that the exciton binding should asymptotically vanish in the limit of large field strength.

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I. INTRODUCTION

The influence of external electric field on optical properties of semiconductors has been studied extensively using both experimental and theoretical techniques. In bulk semiconductors the shift in the optical absorbing due to the external field is known as the Franz-Keldysh effect.1 In quantum wells and quantum dots, application of electric field has shown to modify the optical properties of nanosystems and is known as the quantum-confined Stark effect (QCSE).2,3 The application of the external field induces various modifications in the optical properties of the nanomaterial including, absorption coefficient, spectral weight of transitions, and change in λmax of the absorption spectra. In certain cases, the applied field can lead to exciton ionisation.4 The quantum-confined Stark effect has found application in the field of electro-absorption modulators,5 solar cells6 and the light-emitting devices.7 Recent experiments by Weiss et al. on semiconductor quantum dots have shown that the QCSE can also be enhanced by the presence of heterojunctions.8 In some cases, the QCSE can be induced chemically because of close proximity to ligands.5 The QCSE also plays a major role in electric field dependent photoc conductivity in CdS nanowires and nanobelts.9 Electric field has emerged as one of the tools to control and customize quantum dots as novel light sources. In a recent study, electric field was used in generation and control of polarization-entangled photons using GaAs quantum dots.10 It has been shown that the coupling between stacked quantum dots can be modified using electric field.11

The QCSE has been investigated using various theoretical techniques including perturbation theory,12–17 variational techniques,18–24 and configuration interaction method.25–35 In the present work, development of explicitly correlated full configuration interaction (XCFCI) method is presented for investigating effect of external electric field on quantum dots and wells. The XCFCI method is a variational method in which the conventional CI wavefunction is augmented by explicitly correlated Gaussian-type geminal functions.36 The inclusion of explicitly correlated function in the form of the wavefunction is important for the following two reasons. First, the addition of the geminal function increases the convergence of the FCI energy with respect to the size of the underlying 1-particle basis set.37 Second, inclusion of explicitly correlated function improves the form of the electron-hole wavefunction at small inter-particle distances which is important for accurate calculation of electron-hole recombination probability.38–40 The effect of explicitly correlated function on the convergence of CI energy has been investigated by Prendergast et al.37 and is directly re-
lated to accurate treatment of the Coulomb singularity in the Hamiltonian.\textsuperscript{37,41,42} Varganov et al. have demonstrated the applicability of geminal augmented multi-configuration self-consistent field wavefunction for many-electron systems.\textsuperscript{33} Elward et al. have also performed variational calculation using explicitly correlated wavefunction for treating electron-hole correlated in quantum dots.\textsuperscript{40,44}

One of the important features of the XCFCI method presented here is the inclusion of the external field in the ansatz of the wavefunction. This is achieved by defining a new set of field-dependent coordinates which are generated by performing variational polaron transformation\textsuperscript{45} and recasting the original Hamiltonian in terms of the field-dependent coordinates. The variational polaron transformation was introduced by Harris and Silbey for studying quantum dissipation phenomenon in the spin-boson system\textsuperscript{45} and is used in the present work because of the mathematical similarity between the spin-boson and the field-dependent electron-hole Hamiltonian.

The remainder of this article is organized as follows. The important features of the XCFCI method are summarized in Sec. II A, construction of the field dependent basis functions is presented in Sec. II B, the application of the XCFCI method using field-dependent basis is presented in Sec. III, and the conclusion are provided in Sec. IV.

II. THEORY

A. Explicitly correlated full configuration interaction

The field dependent electron-hole Hamiltonian is defined as\textsuperscript{46,47}

\begin{equation}
H = -\frac{\hbar^2}{2m_e} \nabla_e^2 - \frac{\hbar^2}{2m_h} \nabla_h^2 + v^\text{ext}_e + v^\text{ext}_h \\
- \frac{1}{|\mathbf{r}_{eh}|} + |e|\mathbf{F} \cdot (\mathbf{r}_e - \mathbf{r}_h)
\end{equation}

where \(m_e\) is the mass of the electron, \(m_h\) is the mass of the hole, \(\epsilon\) is the dielectric constant, and \(\mathbf{F}\) is the external electric field. The external potential \(v^\text{ext}_e\) and \(v^\text{ext}_h\) represent the confining potential experienced by the quasi-particles. The form of the XCFCI wavefunction is defined as

\begin{equation}
\Psi_{\text{XCFCI}} = \hat{G} \sum_k c_k \Phi_k
\end{equation}

where \(c_k\) is the CI coefficient and \(\Phi_k\) are basis functions. The operator \(\hat{G}\) is known as the geminal operator and is an explicit function of \(\mathbf{r}_{eh}\) and is defined as

\begin{equation}
\hat{G} = \sum_{i=1}^{N_e} \sum_{j=1}^{N_h} \sum_{k=1}^{N_k} b_k e^{-\gamma_k r_{ij}},
\end{equation}

where \(N_e\) is the number of Gaussian functions included in the expansion, \(N_h\) and \(N_k\) are the number of electrons and holes, respectively. The parameters \(b_k\) and \(\gamma_k\) used in the definition of the geminal operator are obtained variationally. The construction of the basis functions used in the definition of XCFCI wavefunction in Eq. (2) will be discussed in Sec. II B. The XCFCI calculation is performed in two steps. In the first step, the parameters of geminal operator are obtained variationally by performing the following minimization

\begin{equation}
E[G_{\text{min}}] = \min_{b_k, \gamma_k} \frac{\langle G \Phi_0 | H | G \Phi_0 \rangle}{\langle G \Phi_0 | G \Phi_0 \rangle}.
\end{equation}

In the second step, the expansion coefficients \(\{c_k\}\) are obtained variationally and are defined by the following minimization procedure

\begin{equation}
E_{\text{XCFCI}} = \min_{\{c\}} \frac{\langle \Psi_{\text{XCFCI}} | H | \Psi_{\text{XCFCI}} \rangle}{\langle \Psi_{\text{XCFCI}} | \Psi_{\text{XCFCI}} \rangle}.
\end{equation}

The above equation can be rewritten as a FCI calculation of transformed operators

\begin{equation}
E_{\text{XCFCI}} = \min_{\{c\}} \frac{\langle \Psi_{\text{FCI}} | \tilde{H} | \Psi_{\text{FCI}} \rangle}{\langle \Psi_{\text{FCI}} | \Psi_{\text{FCI}} \rangle},
\end{equation}

where the transformed operators are defined as

\begin{align}
\tilde{H} & = G_{\text{min}}^\dagger H G_{\text{min}}, \\
\tilde{i} & = G_{\text{min}}^\dagger G_{\text{min}}.
\end{align}

The exact expression of the transformed operators in Eq. (7) and (8) and discussion relevant to their derivation has been presented earlier in Ref. 40 and 44 and is not repeated here. The \(E_{\text{XCFCI}}\) reduces to conventional FCI energy in the limit of geminal function equals to 1

\begin{equation}
E_{\text{FCI}} = \lim_{G \rightarrow 1} E_{\text{XCFCI}}
\end{equation}

We expect the \(E_{\text{XCFCI}}\) energy to be lower than the FCI energy for identical set of basis functions and earlier studies have shown this to be true.\textsuperscript{44}

After the successful completion of the XCFCI calculations, the field dependent exciton binding was calculated from the difference between the non-interacting and interacting ground state energies. Defining the non-interacting Hamiltonian as

\begin{equation}
H_0 = \lim_{\epsilon \rightarrow -1, \omega \rightarrow 0} H,
\end{equation}

the exciton binding energy is computed as

\begin{equation}
E_B[F] = E_{\text{XCFCI}} - E_0^{(0)},
\end{equation}

where \(E_0^{(0)}\) is defined in Eq. (12)

\begin{equation}
E_0^{(0)} = \min_{\Psi} \frac{\langle \Psi | H_0 | \Psi \rangle}{\langle \Psi | \Psi \rangle}.
\end{equation}
The field dependent electron-hole recombination probability is obtained from
the XCFCI wavefunction using the following expression\textsuperscript{10,44}
\[ P_{eh}|\mathbf{F}\rangle = \frac{\langle \Psi_{XCFCI}|\delta(\mathbf{r}_e - \mathbf{r}_h)|\Psi_{XCFCI}\rangle}{\langle \Psi_{XCFCI}|\Psi_{XCFCI}\rangle}. \] (13)

The exciton binding energy and the recombination probability are functionals of the applied external field and are indicated explicitly in Eq. (11) and (13), respectively.

B. Construction of field dependent basis set

One of the key features of the electron-hole Hamiltonian used in the present work is the presence of the field-dependent term in Eq. (1). Since the convergence of the CI expansion depends on the quality of the underlying 1-particle basis, it is desirable to construct and use efficient single particle basis sets. In the present work, we have developed field-dependent basis functions and the details of the derivation are presented as following. Starting with the expression of $H_0$ in Eq. (10), the zeroth-order Hamiltonian is expressed as a sum of non-interacting electronic and hole Hamiltonians
\[ H_0 = H_0^e + H_0^h, \] (14)
where the expression for the single-component non-interacting Hamiltonian is given as
\[ H_0^e = T_e + v_e^{\text{ext}} + |e|\mathbf{F} \cdot \mathbf{r}_e \] (15)
\[ H_0^h = T_h + v_h^{\text{ext}} - |e|\mathbf{F} \cdot \mathbf{r}_h. \] (16)

As seen from the above equation, the coupling between the external field and the quasiparticle coordinates is linear. The above Hamiltonian shares mathematical similarity with the spin-boson Hamiltonian that has been used extensively in quantum dissipative systems.\textsuperscript{48} In the present method, we perform analogous transformation which is defined by the follow equations
\[ \mathbf{q}_e = \mathbf{r}_e + \lambda_e \mathbf{F} \] (17)
\[ \mathbf{q}_h = \mathbf{r}_h - \lambda_h \mathbf{F}. \] (18)

Similar to the polaron transformation in the spin-boson system, the coordinates of the quasiparticle experience a shift due to the presence of the external field.\textsuperscript{38} Using the method of variational polaron transformation by Harris and Silbey,\textsuperscript{45} the shift parameter $\lambda$ is determined variationally. The field-dependent electronic basis functions are obtained by first constructing the Hamiltonian matrix using Gaussian-type orbitals (GTO) and then diagonalizing the resulting matrix
\[ H_0^e \Phi^e_i = \epsilon_i^e(\lambda_e) \Phi^e_i \] (19)
\[ H_0^h \Phi^h_j = \epsilon_j^h(\lambda_h) \Phi^h_j \] (20)
The value of the shift parameter is obtained variationally by minimizing the trace
\[ \min_{\lambda} \sum_i \epsilon_i^e \implies \lambda_e. \] (21)
The $\lambda_h$ is also obtained by a similar procedure. The electron-hole basis functions for the FCI calculations are constructed by taking a direct product between the set of electronic and hole single-component basis sets
\[ \{ \Phi_k \} = \{ \Phi^e_i \} \otimes \{ \Phi^h_j \}. \] (22)
The procedure described above is a general method that is independent of the exact form of the external potential. However if the external potential is of quadratic form, the field dependent zeroth-order single-component Hamiltonian has an uncomplicated mathematical form and additional simplification can be achieved.

III. RESULTS AND DISCUSSION

The electron-hole Hamiltonian in Eq. (1) has been used extensively for studying optical rectification\textsuperscript{15,17,46,47,49} effect in GaAs quantum dots and all the system specific parameters were obtained from previous calculations on the GaAs system.\textsuperscript{10,47} The parabolic confinement potential has found widespread applications\textsuperscript{12,15,20,23,50–61} in the study of quantum dots and was used in the present work to approximate the external potential term in the Hamiltonian. All the parameters that are needed for complete description of the electron-hole Hamiltonian used in the calculations are presented in Table I. Following earlier work on the effect of electric field on non-linear optical properties of GaAs quantum dots,\textsuperscript{10,47} the external electric field was aligned along the z-axis and the field strength was varied from zero to 500 kV/cm. Similar to the spin-boson Hamiltonian, the polaron transform resulted in shifted harmonic oscillators.\textsuperscript{38} The eigenvalues and eigenfunctions of the $H_0$ were obtained analytically, and the lowest ten eigenstates of the shifted harmonic oscillator Hamiltonian were used in the construction of the 1-particle basis. The direct product between the electronic and the hole basis sets was performed to generate the electron-hole basis for the FCI calculations. The geminal minimization was performed using a set of

| Parameter       | Value   |
|-----------------|---------|
| $m_e$           | 0.067m_0 |
| $m_h$           | 0.090m_0 |
| $k_e$           | $9.048 \times 10^{-7}$ a.u. |
| $k_h$           | $1.122 \times 10^{-6}$ a.u. |
| $\epsilon$     | 13.1e_0  |
three \( \{ b_k, \gamma_k \} \) parameters at each field strength, and the optimized values are presented in Table II. The total exciton energy for the field-free case was found to be 269.45 meV. The total exciton energy of the system as a function of the field strength is presented in Fig. 1. It is seen that the total energy decreases with increasing field strength. Earlier studies on this system indicate that the exciton energy is a quadratic function of the applied field.\(^{62,63}\)

To investigate the scaling of the total exciton energy with respect to the field strength, we have performed least-square fit of the calculated values with a second order polynomial and the results are presented in Fig. 1. The results from these calculations confirm that the quadratic scaling of the exciton energy as a function of the field strength.

The exciton binding energy was calculated using Eq. (11) and was found to be 28.52 meV for the field-free case. The effect of the external field on the exciton binding energy was investigated by calculating the relative binding energy which is defined by the following equation

\[
\tilde{E}_B = \frac{E_B[F]}{E_B[F = 0]}. \tag{23}
\]

It is seen from Fig. 2 that the exciton binding energy decreases with increasing field strength. As the field strength is increased from 0 to 500 kV/cm, the exciton binding energy decreases by a factor of 2.6. In addition to calculation of binding energy, the effect of the field on electron-hole recombination probability was also investigated. Analogous to the relative binding energy, the relative recombination probability is defined as

\[
\tilde{P}_{eh} = \frac{P_{eh}[F]}{P_{eh}[F = 0]}, \tag{24}
\]

and is presented in the Fig. 2. It is seen that the there is a sharp decrease in the recombination probability with increasing field strength and the recombination probability at 500 kV/cm is lower than the field-free case by a factor of 166. One of the key results from this study is that exciton binding energy and eh-recombination probability are affected differently by the external electric field. It is seen that the exciton binding energy and eh-recombination probability follow different scaling with respect to field strength.

The polaron transformation also provides insight into the effect electric field on the exciton binding energy in the limit of high field strengths. Starting with the transformation defined in Eq. (17), the electron-hole Coulomb interaction in the transformed coordinate can be expressed as

\[
\frac{1}{|r_e - r_h|} = \frac{1}{\| (q_e - q_h) - (\lambda_e + \lambda_h)F \|} = v_{eh}(q). \tag{25}
\]

It is seen in the above equation that the above expression will be dominated by the field-dependent term in the limit of high field strength. A direct consequence of the above condition is that in the limit of high field strengths, we expect the exciton binding energy to be small

\[
H(q) \approx H_0(q) \Rightarrow E_B \approx 0 \quad \text{for} \; 1 \ll |F| < \infty. \tag{26}
\]

It is important to note that the above conclusion is independent of the choice of the external potential.

**IV. CONCLUSION**

The effect of external electric field on exciton binding energy and electron-hole recombination probability was computed using explicitly correlated full configuration interaction method. Field-dependent basis functions were used in the calculations and a variational polaron transformation scheme was developed for construction of
field-dependent basis functions. It was found that both exciton binding energy and electron-hole recombination probability decrease with increasing field strength. One interesting conclusion from this study is that the binding energy and recombination probability follow different scaling with respect to the external electric field. For the range of field strengths studied, the recombination probability and exciton binding energy decrease by a factor of 166 and 2.6, respectively. These results give important insight into the application of electric field for manipulating excitons in quantum dots.

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| $F_z$ (kV/cm) | 0    | 100  | 200  | 300  | 400  | 500  |
|-------------|------|------|------|------|------|------|
| $b_1$       | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |
| $\gamma_1$  | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| $b_2$       | $1.40 \times 10^{-4}$ | $9.99 \times 10^{-1}$ | $4.99 \times 10^{-2}$ | $5.78 \times 10^{-3}$ | $1.00 \times 10^{-2}$ | $5.59 \times 10^{-3}$ |
| $\gamma_2$  | $2.29 \times 10^{-4}$ | $4.60 \times 10^{-6}$ | $1.11 \times 10^{-2}$ | $1.11$ | $1.11$ | $1.11$ |
| $b_3$       | $4.35 \times 10^{-2}$ | $1.08 \times 10^{-1}$ | $8.90 \times 10^{-2}$ | $1.67 \times 10^{-2}$ | $2.00 \times 10^{-2}$ | $1.58 \times 10^{-2}$ |
| $\gamma_3$  | $1.13 \times 10^{-2}$ | $1.00 \times 10^{-2}$ | $3.01 \times 10^{-3}$ | $1.11 \times 10^{-1}$ | $1.01 \times 10^{-1}$ | $1.02 \times 10^{-1}$ |

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