



## Polarizability of few electron quantum dots: Extended coupled-cluster response approach

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### ABSTRACT

The dipole polarizability is studied for few electron quantum dots using extended coupled cluster linear response method (ECCLR). The polarizability of pure parabolic quantum dots depends only on the number of electrons and the parabolic confinement strength. We added impurity in a GAUSSIAN form to the external parabolic potential and used ECCLR to calculate polarizability. To examine the effect of external potential, we changed the parabolic confinement to square-well and repeated all calculations. We mainly looked at eigenvalue spectrum and charge density specially when the system shifts towards lower density regions.

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### 1. Introduction

Quantum dots (QD) are artificial atoms in which electrons are confined using an external potential [1–3]. Significant efforts have recently been put into understanding their electronic and optical properties. However less work is present as far as understanding the electric field response and calculating polarizability is concerned.

Among Few experimental works, Seufert [4,5] studied exciton polarizability by means of highly resolved photoluminescence spectroscopy while the permanent dipole moment in the lateral direction was found to be negligible.

Among theoreticians Holka et al. [6] used Coupled Cluster Singles and Doubles (CCSD) and Hartree–Fock (HF) methods to calculate the energy and numerically calculated polarizability of a 2-electron QD in presence of electric field, but their results were restricted to non-interacting Hamiltonian using parabolic confinement. In one of the early works of Kohn [8] examined interacting electron gas response to external magnetic field. His results and his formalism however was restricted to the case of short range  $e-e$  interactions. Sako and Dierksen [7] also calculated the polarizability of a Hydrogen anion and Helium atom in addition to 2-electron QD. They used configuration interaction (CI) to calculate the total energy and calculated the polarizability using the finite difference approximation and compared it with analytical values.

There are situations where analytical calculations fail to produce a correct answer, e.g., presence of impurity. Impurity happens

during the process of fabrication and its effect on electronic structure and properties of quantum dots has been studied by using different methods such as path integral Monte Carlo [9], spin-density-functional theory (SDFT) [10]. Pujari et al. [10] have observed that the impurity induces a localized magnetic moment that generates spin polarized configurations. The effect of a weak attractive on-center impurity on polarizability however has not been studied before specially when the system is moderately correlated. As we shall see, impurity can bring about significant changes in charge density and polarizability. Thorough understanding of the effect of impurity on the charge density and response of the system to the electric field by means of a powerful method is advised.

In this Letter we use ECCLR to calculate the polarizability of 2 and 6-electron QDs. We also obtain non-relaxed polarizability at HF level by using dipole integrals and HF orbital energies and relaxed polarizability by finite difference approximation (FF) using HF energies. The results of these calculations are compared with analytical value of the polarizability ( $\alpha_{xx} = N/\omega_x^2$ ) for the entire range of  $\omega$ . We know that scenario will be different if we add on-center attractive impurity in case of parabolic confinement, or if we change the external potential from parabolic to square-well.

The Letter is organized as follows: in Section 2 we present a brief description of ECCLR method with emphasis on ECC. In Section 3 details of the external potential of quantum dots and the computational details are presented. Section 4 provides results and discussions.

### 2. Theory

In this section we will briefly review the extended coupled cluster response approaches. Single-reference coupled-cluster (SRCC)

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[11–16] has been established as state of the art method for the calculation of electronic structure and properties of the atoms and molecules. First order properties can be obtained using expectation value approach or using analytic response approach. The initial response approach developed by Monkhorst [17] did not have  $(2n + 1)$  rule built in. Thus, the calculation of first order energy derivative required the first derivative of the wave function. This severely restricted the use of SRCC response approach for the calculation of properties, in particular when perturbation is geometric. This problem was eliminated with the help of Z-vector technique [18] by Bartlett and co-workers [19,20]. Thus, with the help of one extra set of perturbation independent amplitude energy derivatives can be calculated. However, this approach is very complicated to extend it for higher order properties. Jørgensen and Helgaker [21] independently developed apparently different response approach based on Lagrange variation. This approach can easily be extended for higher order properties [22,23]. Parallel to these developments, Pal et al. [24–26] proposed a fully stationary response approach. Choice of the energy functional is very important in this approach. Different functional like expectation value (XCC), unitary coupled cluster (UCC) and extended coupled cluster (ECC) exist in the literature. In the variational response approach the energy functional becomes nonterminating and needs to be truncated for the practical application. Pal and co-workers used the fixed number of cluster amplitudes in the energy functional as the truncation scheme. Bartlett and co-workers [27,28] implemented the CC approach for using XCC and UCC functionals for the energy calculations. However, the truncation scheme followed was on the basis of the perturbation order.

The initial implementation of Pal and coworkers was based on the expectation-value (XCC) functional [29–31]. The XCC functional, however, is a non-terminating series and the stationary equations resulting from the functional when truncated to a fixed power in cluster amplitudes contain disconnected terms. The extended coupled cluster functional proposed by Arponen and coworkers [32–34] was the alternative functional. It uses an exponential parametrization of both left and right vectors and is based on a double-similarity transformation of the Hamiltonian using the excitation and de-excitation operators, respectively. Linearization of the left cluster amplitudes in ECC functional leads to SRCC equations. In this sense ECC functional has the ability to sum up higher order terms compared to the SRCC normal functional (NCC). Pal and co-workers used this functional to developed response approach for the calculation of molecular properties. Using this functional Pal and co-workers successfully calculated electric [29–31,35–38] and magnetic properties [39] of the molecules. The form of the functional looks like

$$E = \langle \phi_0 | \exp(\Sigma)(H \exp(T))_L | \phi_0 \rangle_{DL} \quad (1)$$

There,  $L$  denotes Linked functional and  $DL$  means double linking. The  $T$  operator to the right of  $H$  must be connected to the Hamiltonian and the operator  $\Sigma$  to the left of  $H$  must either be connected to  $H$  or to two different  $T$  amplitudes. This restriction on the functional makes sure that the series is always linked and terminates naturally. It can be seen from the functional that we have doubled the number of cluster amplitudes and hence doubled the number of equations in ECC compared to usual CC method. Differentiation of the energy functional with respect to the  $T$  amplitudes gives equation for  $\Sigma$  and differentiation with respect to  $\Sigma$  gives equation for  $T$  amplitudes.

It can be seen that the functional, though naturally terminating, terminates at quite higher order. For the practical applications we truncate the series. There are two ways of truncation. One based on the perturbation order while other based on the number of cluster amplitudes present in the functional. We followed the latter using the cubic truncation. The other truncation that is generally fol-

lowed is the coupled cluster singles and doubles (CCSD) approximation. Thus, with in CCSD we have total of three cluster amplitudes present in the energy functional. The cluster operators truncated up to two-body excitations is what termed as ECCSD-cubic approximation.  $\Sigma$  and  $T$  amplitudes are obtained by the stationarity of the functional equations

$$\frac{\delta E}{\delta T^0} = 0; \quad \frac{\delta E}{\delta \Sigma^0} = 0 \quad (2)$$

At the stationary point, functional form is simplified using the stationary conditions of the cluster amplitudes i.e., Eq. (2). The derivative cluster amplitudes are obtained by making the derivative energy functional stationary w.r.t. to the ground state Sigma and  $T$  amplitudes. The derivative energy functional involves the ground state sigma and  $T$  amplitudes along with the derivative of Sigma and  $T$  amplitude. Thus, the equations

$$\frac{\delta E^{(1)}}{\delta t^{(0)}} = 0; \quad \frac{\delta E^{(1)}}{\delta \sigma^{(0)}} = 0 \quad (3)$$

provide us with the equation for  $\Sigma^{(1)}$  and  $T^{(1)}$  amplitudes. Since, the energy and all derivative energy functionals are double linked all the properties calculated are size extensive. Using these derivative cluster amplitudes we can obtain properties up to third order.

### 3. Computational details

The material of the dot is assumed to be GaAs. We use effective mass approximation with an effective mass  $m^* = 0.067 m_e$ , where  $m_e$  is the mass of an electron, and dielectric constant  $\epsilon = 12.9$ . The units of length and energy are scaled to effective atomic units. The effective Bohr radius is  $a_B^* = \hbar^2(4\pi\epsilon\epsilon_0)/m^*e^2$  and effective Hartree is  $Ha^* = 2 * Ry = m^*e^4/2\hbar^2(4\pi\epsilon\epsilon_0)^2$ . For GaAs  $a_B^* = 9.8$  nm and  $Ry^* = 6$  meV. All the results presented in this Letter are in effective atomic units.

We use real space grid technique to solve HF and obtain the orbital energies. Our technique initiates the self-consistency with one of the several hundred guesses of charge density in search for minimum energy (for more details refer to Ref. [10]), which assures the detection of actual ground state of the system.

To study the effect of electron correlation we have used ECCLR method which has cubic truncation of cluster amplitudes. Though it is truncated in cubic approximation, the left amplitudes (which are de-excitation amplitude) in this functional are fully exponential.

We also calculated polarizability (considering relaxation effects) numerically by finite difference approximation method using HF energies [40]

$$\alpha_{\text{rel}} = \left[ \frac{\partial^2 E(F)}{\partial F^2} \right]_{F=0} \approx - \frac{E(+F) + E(-F) - 2E(0)}{F^2} \quad (4)$$

where the field strength  $F$  has been chosen to be 0.001 au. To study the effect of orbital relaxation, the results are compared with polarizability calculated using the non-relaxed HF wavefunctions and orbital energies:

$$\alpha_{\text{non-rel}} = \sum_i^{\text{occ}} \sum_a^{\text{vir}} \frac{\mu_{ai}^* \mu_{ai}}{e_a - e_i} \quad (5)$$

where  $\mu_{ai}$  are the dipole integrals and  $e_i$  and  $e_a$  are the occupied and virtual HF orbital energies.

#### 3.1. Harmonic oscillator confinement

Polarizability is the relative tendency of charge distribution to be distorted from its normal shape by an external electric field

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