Scaling in the correlation energies of two-dimensional artificial atoms

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Abstract
We find an unexpected scaling in the correlation energy of artificial atoms, i.e., harmonically confined two-dimensional quantum dots. The scaling relation is found through extensive numerical examinations including Hartree–Fock, variational quantum Monte Carlo, density functional, and full configuration interaction calculations. We show that the correlation energy, i.e., the true ground-state total energy minus the Hartree–Fock total energy, follows a simple function of the Coulomb energy, confinement strength and number of electrons. We find an analytic expression for this function, as well as for the correlation energy per particle and for the ratio between the correlation and total energies. Our tests for independent diffusion Monte Carlo and coupled-cluster results for quantum dots—including open-shell data—confirm the generality of the scaling obtained. As the scaling also applies well to \( \gtrsim 100 \) electrons, our results give interesting prospects for the development of correlation functionals within density functional theory.

(Some figures may appear in colour only in the online journal)

1. Introduction

Artificial atoms—quantum dots (QD) [1, 2], i.e., nanoscopic semiconductor structures for which a set of electrons is confined—offer wider possibilities for engineering their properties than real atoms. The QD size, for example, can be changed from a few nanometers to hundreds of nanometers by modifying the experimental constraints. In turn, the degree of correlation of the electronic motion can be tuned by changing the system size, number of electrons, and type of the confining potential.

In the present paper, we focus on the standard, quasi-two-dimensional (quasi-2D), isotropic harmonic oscillator model of an artificial atom, given by the \( N \)-electron Hamiltonian

\[
H = \sum_{i=1}^{N} \left[ -\frac{\hbar^2}{2m^*} \nabla_i^2 + V_{\text{ext}}(r_i) \right] + \sum_{i<j}^N \frac{e^2}{4\pi\varepsilon|\mathbf{r}_i - \mathbf{r}_j|},
\]

where the external confinement in the \( xy \) plane is described by a parabolic potential,

\[
V_{\text{ext}}(r) = \frac{1}{2} m^* \omega^2 r^2.
\]

In spite of its simplicity, the model has been shown to predict very well the electronic properties of both vertical and lateral single quantum dots at the GaAs/AlGaAs interface [1, 3, 4]. The model can be essentially characterized by two parameters: the number of confined electrons \( N \) and the ratio between the Coulomb and oscillator energies, i.e.,

\[
\beta = \frac{E_{\text{Coul}}}{\hbar\omega} = \frac{e^2 m^{*1/2}}{4\pi\varepsilon\omega^{1/2} \hbar^{3/2}},
\]

where \( e \) is the electron charge, \( m^* \) its effective mass in the semiconductor material, and \( \varepsilon \) the dielectric constant. We point out that \( E_{\text{Coul}} \) is the total electron–electron interaction...
energy, i.e., the expectation value of the Coulomb interaction operator.

In a previous work [5], some of the present authors showed that the total energy of such an artificial atom with 20 \( \leq N \leq 90 \) electrons obeys the scaling relation dictated by the Thomas–Fermi (TF) theory

\[
E_{\text{gs}}(N, \beta) \approx N^{3/2} f_{\text{gs}}(z). 
\]

The variable \( z = N^{1/4} \beta \) combines in a particular way the numbers of \( N \) and the coupling constant \( \beta \). The function \( f_{\text{gs}} \) is universal in the sense that it depends only on \( z \), and not explicitly on the system parameters. This result is not very surprising, as the TF theory is known to predict well the total energies of ‘large’ electronic systems. In particular, in 2D the gradient corrections to the TF kinetic energy vanish to all orders [6], and, secondly, for the 2D Fermi gas in a harmonic potential the TF (non-interacting) kinetic energy is exact when using the exact density as the input [7]. It is remarkable, however, that the TF scaling for the total energy holds for a wide range of \( \beta \), from the strong-confinement limit (weak correlations, \( \beta \to 0 \)) to the weak-confinement limit (strong correlations, \( \beta \to \infty \); the so-called Wigner phase [8]).

In the present paper, we move a step further and examine whether a scaling \( \text{a la } \) Thomas–Fermi total energy, with different exponents, holds also for the correlation energy [9–11] \( E_c \), i.e., the difference between the total ground-state energy \( E_{\text{gs}} \) and the Hartree–Fock (HF) energy \( E_{\text{HF}} \):

\[
E_c = E_{\text{gs}} - E_{\text{HF}}. 
\]

We point out that within density functional theory (DFT) the HF energy in the above expression is commonly replaced by the total energy of an exact-exchange calculation. In practice, this is very close to the HF energy.

We perform extensive calculations for \( E_{\text{gs}} \) of QDs with \( 6 \leq N \leq 90 \) and find a unique scaling relation for \( E_c \). The fact that \( E_c \) scales is completely unexpected because, by definition, the electronic correlation is beyond mean-field properties [9, 10, 12–21]. In addition, the asymptotic behavior of the correlation potential accessible in DFT has a highly nontrivial scaling [22]. We find, as our second main result, that also the fraction of the total energy associated with \( E_c \) scales in a universal way.

The paper is organized as follows. In section 2 we briefly summarize the computational methods employed. In section 3, we present and discuss results from numerical calculations of \( E_c \). Finally, concluding remarks are given in section 4.

## 2. Numerical results

In order to validate our numerical values for \( E_c \), we perform extensive calculations by means of standard many-electron methods including HF, DFT in the local-density approximation (LDA) [23, 24], variational quantum Monte Carlo (VMC) [25–29], and full configuration interaction (FCI) methods [30]. We have used our own numerical codes with all the methods, but the HF results shown and some LDA results have been calculated with the OCTOPUS code [31].

For simplicity, we give the confinement strength \( \omega \) and energies in atomic units. These can be used to obtain the material-dependent effective atomic units; i.e., the effective Hartree energies and effective Bohr radii correspond to \( E_h^* = (m^* / m_0)(\epsilon / \epsilon_0)^2 E_h \) and \( a_0^* = (\epsilon / \epsilon_0)(m^* / m_0)a_0 \), respectively. Our choices for \( \omega \) used below represent typical experimental setups for GaAs quantum dots. For the GaAs case with \( m^* / m_0 = 0.067 \) and \( \epsilon / \epsilon_0 = 12.8 \), the effective atomic units correspond to \( E_h^* \approx 11.1277 \text{ meV} \) and \( a_0^* \approx 10.11 \text{ nm} \).

We find that both our VMC results (which are upper bounds for the total energy) and the DFT results correspond very well with FCI ones. This is demonstrated in figure 1 for the case \( N = 6 \) and \( \omega = 0.25 \text{ au} \) that corresponds to the most correlated case for our set of QDs (see figure 2). The FCI result is plotted as a function of the energy cutoff for the many-body configuration included in the basis [32], and it eventually converges to a value that is very close to the VMC and LDA ones. It is noteworthy that the HF ground-state energy (lacking the correlation) is higher, and thus the gray region in figure 1 corresponds to the correlation energy according to equation (5).

The FCI method becomes too expensive with the larger particle numbers considered. However, we can see from figure 1 that the VMC method is able to capture nearly all of the correlation energy for the case shown. Furthermore, this is the most correlated case that we study in this work. We stress that the correlation energy is only a few per cent (<6%) of the total energy, reaching the largest relative values for the smallest systems in the strong-coupling regime. This is visualized in figure 2, which shows the relative correlation energy \( \chi = |E_c / E_{\text{gs}}| \) as a function of the confinement strength. The gray area corresponds to the typical experimental regime.
when considering laterally or vertically confined GaAs QDs.

One can see that the correlation energy is largest at small particle number and with the weak confinements.

Our HF, LDA, and VMC results are summarized in table 1. All the systems considered are closed-shell QDs with ground-state angular momentum and spin quantum numbers \( L = S = 0 \). As the VMC calculations are expected to yield the exact ground-state energy with a good accuracy, we use those results in the following to compute the correlation energy as the difference from the HF energies. However, it is reassuring to see that the LDA energies are very close to the VMC results. The accuracy of the LDA for these systems is well expected due to the smooth potentials and densities, and it is in accordance with previous studies [4]. We notice that—for the parameters used in the calculations—the scaled variable \( z = N^{1/4} \beta \) takes values in the range \( 0 < z < 8 \) that covers the weak-coupling regime and a part of the strong-coupling regime; \( z \approx 1 \) is the point of transition between these regimes [33].

### 3. Scaling of the correlation energy

Here, we first assume that the correlation energy has a particular scaling with respect to \( N \). Thus, we suggest an ansatz of the form

\[
E_c(\omega) = N^\alpha f_c(N^{1/4} \beta). \tag{6}
\]

Next, we fit our data for \( N \) and \( \beta \) to the ansatz by varying the scaling exponent \( \alpha \). We minimized the normalized root mean square deviation of the data as a function of \( \alpha \). The minimum deviation is obtained with \( \alpha \approx 3/4 \), and thus we set \( \alpha \) to this value. For the function \( f_c \), a two-parameter fit of the form \( \alpha z^\gamma \) leads to

\[
\frac{E_c}{\omega N^{3/4}} = -0.0668 z^{1.51}. \tag{7}
\]

The scaled correlation energies are shown in figure 3 along with the function \( f_c(z) \). It can be seen that the quality of the analytical expression obtained for \( f_c \) is very good over the whole range of \( z \), showing a mean deviation of around 5% for \( E_c \). We point out that equation (7) can be straightforwardly rewritten in such a way that the dependence of \( E_c \) on the system parameters (say, \( N \) and \( \omega \)) becomes explicit:

\[
E_c(N, \omega) = -0.0668 N^{1.275} \omega^{0.245}, \tag{8}
\]

where \( E_c \) is given in au. From this expression, we can trivially obtain an expression also for the correlation energy per particle \( E_c/N \).

Next we confirm the applicability of the scaling relation obtained, by comparing the scaling function \( f_c \)
in equation (7) against independent calculations for small and medium-size QDs, including open-shell systems. In particular, we consider diffusion Monte Carlo (DMC) [34] calculations for \(N = 2, \ldots, 13\) and coupled-cluster singles–doubles (CCSD) calculations [35] for \(N = 2, 6, 12\) compared against the scaling function \(f_c\).

Another interesting quantity to consider for the scaling is \(\chi\) as defined above, i.e., the relative fraction of the correlation energy with respect to the total energy. It can be shown that \(\chi\) also follows a similar scaling relation as a function of the parameter \(z\). We use equation (7) for \(E_c\) and for the total ground-state energy we use the result

\[
\frac{E_{\text{gs}}}{\hbar \alpha N^{3/2}} = \frac{2}{3} + \frac{0.698}{1 + 2.149} z^{4/3} + 1.5 z^{4/3} + 2.175 z^{2/3}. \tag{9}
\]

obtained in [5]. It has been found that the last expression performs better for systems with large \(N\), in particular for \(N > 20\). For the fraction \(\chi\), we obtain

\[
\chi(z) N^{3/4} = f_\chi(z) = \frac{p(z)}{q(z)}, \tag{10}
\]

where

\[
p(z) = 0.200 z^{1.513} + 0.431 z^{1.846} + 0.301 z^{2.180} + 0.436 z^{2.513}. \tag{11}
\]

and

\[
q(z) = 2 + 4.298 z^{1/3} + 3z^{2/3} + 6.444 z + 4.5z^{4/3} + 6.525 z^{5/3}. \tag{12}
\]

The computed values of \(\chi\), labeled according to \(N\), are shown in figure 4 together with the analytic expression obtained. It can be seen that equation (10) works remarkably well for systems with \(N > 20\). Let us stress that for the weak-confinement regime \((z \gg 1)\) equation (10) predicts an almost linear dependence of \(\chi N^{3/4}\) on the parameter \(z\), leading to \(\chi \sim N^{-0.54} e^{-0.42}z\).

In the extreme low-density limit, the system approaches the so-called Wigner phase [8]. The mean-field methods break the rotational symmetry in this case, and the electrons localize in space [36]. In the many-body treatment, the localization can be seen in the conditional densities [29]. Here, our results are not in that limit, and analysis of this limit is left for future studies.

Finally, we note that for real (three-dimensional) atoms, the TF theory predicts for the total energy the following dependence [38]:

\[
E_{\text{gs}}(N, Z) \approx N^{7/3} f_\text{gs}(N/Z), \tag{13}
\]

where \(Z\) is the nuclear charge. The correlation energy also seems to show a scaling \(a\) to TF with

\[
E_c(N, Z) \approx N^\alpha f_c(N/Z). \tag{14}
\]

The coefficient \(\alpha\) is near 4/3 [39–48]. We stress, however, that real atoms, unlike artificial ones, are always in the weak-correlation regime.

4. Concluding remarks

In summary, we have performed extensive numerical calculations for semiconductor quantum dots and found an unexpected universal scaling relation for the correlation energy, equation (7), which resembles the scaling coming from Thomas–Fermi theories. A universal scaling relation for the fraction of the total energy associated with the correlations was also obtained (equation (10)). Such an expression provides information on the degree of correlation of the system and the accuracy of the HF estimation, even without any calculations. The material parameters (effective mass, dielectric constant) are contained in the scaling variable \(z\). Our result has direct implications for the design of new correlation functionals for DFT calculations [37, 49], and may also supplement the recently formulated DFT for strictly correlated electrons [50] and related approaches.
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