Scaling of Coulomb and exchange-correlation effects with quantum dot size

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The role of many-body effects in quantum dots is of both academic and practical interest. We study the electron-electron interaction within a simplified spherical quantum dot using the local spin-density approximation. We experiment with a variety of confining potentials (triangular, harmonic, square well, etc.) and with a varying number of electrons ($N=2$ to 20). We carry out a detailed study of the scaling behavior of the ''Hubbard U'' potential, which is a measure of the capacitive energy, with quantum dot size R ($U\sim 1/R^{\beta}$). We find that the scaling exponent β is $\approx 1/2$ for harmonic confinement and equal to 1 for the square-well confinement. The dependence of the scaling exponents on the confining potential and the number of electrons N is elucidated. We also examine the relative importance of Coulomb, exchange, and correlation terms in the Hubbard U potential and find that correlation plays a relatively more important role at a larger size. We provide a partial explanation for the value of the exponent in the Appendix.

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I. INTRODUCTION

Quantum dots (QD) are structures in which charge carriers are essentially trapped in a three-dimensional potential. They are also known as ''artificial atoms''[1,2] and consist of a 10^3–10^6 atoms, with system sizes in the range of 1–10 nm. They are of fundamental and technical interest for next generation electronic devices. An important goal of today's technological drive towards smaller and smaller devices is to fabricate the so-called single-electron transistor which can be operated at room temperature.[3] They may also form the basis of new generations of lasers. The emission in quantum dot lasers originates from the recombination of excitonic complexes, so it is important to study and understand the quantum dot's internal electronic structure.[4]

Theoretical studies of a QD involve multielectron effects. Review of earlier work in this direction maybe found in the recent work of Jiang *et al.*[5] and Ranjan *et al.*[6] In this work, we investigate the behavior of the direct Coulomb and the exchange correlation with the shape of the confinement potential and size of the quantum dot. Before we discuss the motivation behind this work, we define a few relevent terms. The ''Hubbard U'' potential is composed of a number of terms

$$U=U_c+U_{xc} \tag{1.1}$$

and

$$U_{xc}=U_x+U_{corr}, \tag{1.2}$$

where U_c is the direct Coulomb term, U_x is the exchange, and U_{corr} is the correlation term. Note

$$U=\frac{2}{N(N-1)}U(N), \tag{1.3}$$

where N is the number of electrons and $U(N)$ could be $U_c(N)$, $U_x(N)$, or $U_{corr}(N)$. $U(N)$ is the effective potential energy of N electron system while U is the ''pair'' interaction. In the local-density approximation (LDA), all three terms given in Eqs. (1.1) and (1.2) are present, whereas in certain other approximations, namely, Hartree-Fock and Harbola-Sahni (HS),[7] the correlation term is absent.

We experiment with various shapes of the confinement potential. The model external potential we have chosen is

$$V_{ext}(r)=\begin{cases}(V_0/R^k)r^k-V_0, & r\leqslant R\\ 0, & r>R.\end{cases} \tag{1.4}$$

Here, V_0 is the height of the potential. This can be given by the conduction-band offset (valence-band offset) between the QD and the surrounding layer for the electron (hole). R is the radius of the QD ($2R$ is the width of the potential) and k assumes positive integral values from 1 to say a very large number. Changing the value of k results in the change of the shape of the potential. In particular, $k=1$ is quasitriangular, $k=2$ is quasiharmonic confinement, and $k\geqslant 10$ is quasisquare-well confinement. As $k\rightarrow\infty$, the potential becomes square well. The capacitive energy is defined as

$$\frac{e^2}{C(N)}=E(N+1)-2E(N)+E(N-1), \tag{1.5}$$

where $E(N)$ is the total ground-state energy of N-electron QD. The capacitive energy essentially tracks the Hubbard U potential except for the peaks. The Hubbard U potential is found to be equal to the difference between the ionization potential and electron affinity, i.e.,

$$\frac{e^2}{C(N)}=I(N)-A(N). \tag{1.6}$$

In chemistry, this difference is known as the chemical hardness. More the chemical hardness, the more stable an atom or molecule is.

The motivations behind the study of the Hubbard U potential and its constituent terms are both of academic as well as of practical interest. It has been reported that the Coulomb repulsion is heavily attenuated for multiple charge states of the impurity in semiconductor.[8,9] It may even assume negative values.[10,11] In this paper, we study the electron-electron interaction within a simplified spherical quantum dot using

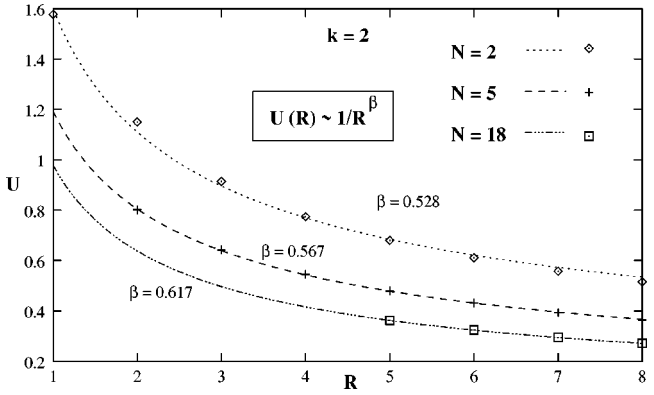

FIG. 1. The effective electron-electron interaction energy U, is depicted as a function of size R of a QD for three different values of the total number of electrons N and for $k=2$ (quasiharmonic confinement). The interaction energy $U\sim 1/R^{\beta}$ with the exponent $\beta=0.528$, 0.567, and 0.617 for $N=2$, 5, and 18, respectively. Notice the weak dependence of β on N.

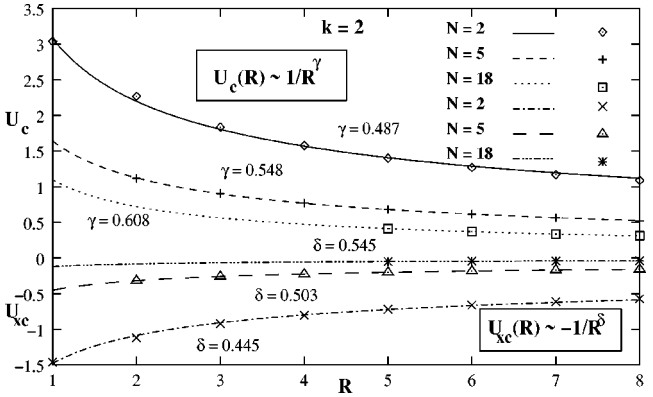

FIG. 2. The individual terms in U, Coulomb U_c, and exchange correlation U_{xc} are depicted as a function of size R of a QD for three different values of the total number of electrons, N and for $k=2$ (quasiharmonic confinement). The values of N and k are the same as in the figure. The Coulomb term scales as $U_c\sim 1/R^{\gamma}$. The exchange correlation scales as $U_{xc}\sim 1/R^{\delta}$. Note that the exponents increase slightly with N.

the local spin-density approximation (LSDA). We experiment with a variety of confining potentials (triangular, harmonic, square well, etc.) for the electrons [see Eq. (1.4)]. We carry out a detailed study of the scaling behavior of the Hubbard U potential, which is a measure of the capacitive energy, with quantum dot size R. Our main thrust is to study the size dependence of the Coulomb and exchange correlation for two distinct and extensively used shapes of the potential, namely, harmonic ($k=2$) and square well ($k\geqslant 10$). We also compare our LSDA results with LDA and the HS scheme and state the results.

II. RESULTS

In our calculations, we have treated exchange-correlation effect in its Gunnarsson-Lundqvist[12] parametrized form. Our calculations have been performed in atomic units. Unless otherwise stated, all the results are based on LSDA calculations. The plot of the capacitive energy as a function of the electron number N exhibits shell filling effects. We shall not dwell on them since it is not the focus of the current paper except to mention that partial filling of shells is observed in certain cases. *This implies that the spherical quantum dot maybe electronically unstable and may undergo distortion. This is suggestive of a Jahn-Teller–like effect in quantum dots.* The scaling exponents reported in this section have been extracted using the Levenburg-Marquardt procedure. We investigate how U and the individual terms comprising it [see Eq. (1.1)] scale with R for different shapes of the confinement.

To understand the scaling behavior, in Fig. 1, we plot the Hubbard U potential with size R of the QD, for the number of electrons, $N=2,5$, and 18 and $k=2$ (quasiharmonic confinement). Hubbard U potential is the average electron-electron interaction energy in a many-electron quantum dot. This figure reveals that U scales as

$$U\sim\frac{1}{R^{\beta}}, \tag{2.1}$$

where the exponent $\beta=0.53$, 0.57, and 0.62 for $N=2$, 5, and 18, respectively.

Figure 2 shows how the individual terms in U scale with size. The external confining potential is quasiharmonic ($k=2$). The Coulomb term drops with size while the exchange correlation plays a compensating role. In other words, the roles of Coulomb and exchange correlation are antithetical. For small size, U_{xc} becomes increasingly attractive while the Coulomb term becomes increasingly repulsive. For $N=2$, $U_c\simeq 2|U_{xc}|$ which is expected. As the electron occupancy N increases, the magnitudes of U_{xc} and U_c become smaller. However, the compensating nature of the two terms is not affected. A Levenburg-Marquardt least-squares fit reveals that

$$U_c\sim\frac{1}{R^{\gamma}}, \tag{2.2}$$

where the exponent $\gamma=0.49$, 0.55, and 0.61 for $N=2$, 5, and 18, respectively. U_{xc} follows a similar scaling behavior, i.e., $U_{xc}\sim -1/R^{\delta}$ with the exponent values $\delta=0.45$, 0.50, and 0.55 for $N=2$, 5, and 18, respectively. Both the exponents γ and δ are $\approx 1/2$ for quasiharmonic confinement.

Now for completeness of our study, we would like to investigate the scaling behavior for other shapes of the confinement, for example, quasisquare ($k=10$) and quasitriangular ($k=1$), etc. These results are displayed in Table I. The effect of the confining potential on the scaling exponent β is dramatic, whereas for $k=2$, $\beta\sim 1/2$, as $k\to\infty$, $\beta\to 1$. This latter result maybe understood in classical terms ($C\sim R_{eff}$). Interestingly for the quasitriangular confinement ($k=1$), the scaling exponent is $\approx 1/3$. In the Appendix, we attempt to explain why U_c scales as $1/\sqrt{R}$ for quasiharmonic confinement and as $1/R$ for quasisquare confinement. We note that with increasing N, β increases. This is due to the fact that with increasing N, the effective potential becomes more flat as the effective size of the quantum dot increases.

TABLE I. The "Hubbard U" potential is found to scale as $\sim 1/R^{\beta}$. Similarly, the Coulomb and exchange-correlation energy scale as $\sim 1/R^{\gamma}$ and $\sim 1/R^{\delta}$, respectively. The exponents β, γ, and δ depend on the shape of the potential, indexed by k [see Eq. (1.5)]. This table lists the values of β, γ, and δ for different values of k and for $N=2$, 5. Note that the exponents increase from 1/3 to 1 as k increases from 1 to 10.

N	k	β	γ	δ
	1	0.386	0.355	0.325
	2	0.528	0.487	0.445
2	4	0.721	0.660	0.601
	8	0.862	0.784	0.710
	10	0.894	0.812	0.735
	1	0.393	0.381	0.351
	2	0.567	0.548	0.503
5	4	0.746	0.718	0.653
	8	0.884	0.846	0.762
	10	0.917	0.877	0.787

In other words because of the spatial extension of the effective potential, electrons find more space to move and hence Coulomb interaction becomes weaker. With these results we see that the Hubbard U potential actually depends on a number of variables, namely, the shape of the confinement, the number of electrons, and also on size of the quantum dot. We find the following relationship between the exponent β and and the confining parameter k:

$$\beta(k) = 0.92(1 - 0.78\exp[-0.27k]). \tag{2.3}$$

In Table I, we also display the exponents γ and δ for the Coulomb and exchange correlation. This establishes scaling laws for U_c and U_{xc}. Further, we also investigate how the Hubbard U potential depends on the number of electrons. We know that classically there should not be any N dependence in U. However, we do find a weak-N dependence. Assuming that the exponent β in the scaling of U has a N dependence, so that

$$\beta(N) \sim N^{\eta} \tag{2.4}$$

then

$$\ln\beta \sim \eta \ln N. \tag{2.5}$$

A logarithmic plot of β as a function of N gives the value of η between 0.05 and 0.08 which is very small. This result has a phenomenological implication. In experiments on nearly spherical quantum dots, one may now safely assume that U—the difference in the electron affinity and the ionization potential, is essentially unchanged with the shell filling.

Now we come to the next important point, what is the role of exchange correlation in deciding the overall scaling of U? We have checked the scaling of U_c and U_x with size of the quantum dot for $k=2$ (harmonic confinement) and for $N=2$, 5, 18 within the HS scheme. We find that both Coulomb and exchange interaction scale as in LDA/LSDA with the exponents almost the same in both cases. This shows that the correlation plays a key role in deciding the overall scaling of U with the shape of the confinement and size of the quantum dot.

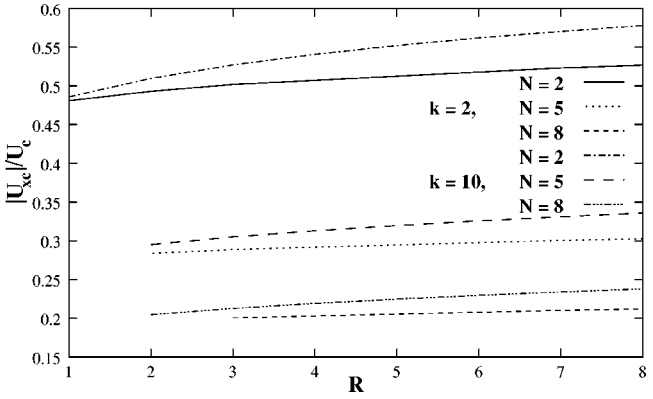

FIG. 3. The ratio of exchange correlation U_{xc} to the Coulomb U_c energy is plotted as a function of the quantum dot size R for $k=2$ and 10 and total number of electrons $N=2$, 5, and 8. The ratio is nearly constant for $k=2$ and this is consistent with the figure. For quasisquare confinement ($k=10$), however, there is noticeable increase with dot size R.

The exchange correlation becomes important when the size is large. The correlation energy is relatively more sensitive to the size and shape of the potential. What is shown in Fig. 3 is the ratio of absolute value of exchange correlation energy to the Coulomb energy with the size of the QD. One can extract the following facts: For large N (density), the Coulomb interaction dominates and hence the ratio $|U_{xc}|/U_c$ is small. It becomes large for a small number of electrons and varies over a wide range; thus showing that the exchange correlation becomes more pronounced. We also find that the exchange correlation is large for large k. The simplest reason one might think of is that as the shape becomes quasisquare well, the spatial extension of the potential is more and hence electrons find more space to move. This attenuates the Coulomb interaction as the density becomes small. In other words, correlation effects are largest in the low-density limit. As the system size becomes larger (or the electron density spreads out more), a more accurate analysis would require the use of asymptotically correct exchange-correlation potential in place of LSDA. The latter is not accurate in the outer regions of a system.

The above results make it clear that correlation plays the key role in deciding the scaling of U. To emphasize this point, we compare the two distinct methodologies, namely, the LDA/LSDA and the HS schemes for the ratio $|U_{xc}|/U_c$ (LDA/LSDA) and $|U_x|/U_c$ (HS) with size R. We find that in case of HS, the ratio is constant, in particular, it is 0.5 for $N=2$. The exchange correlation is always larger for a given N, R, and k in case of LDA/LSDA than the near exact exchange in case of HS. We have also checked the behavior of exchange-only LDA which is also known as Dirac exchange-only LDA and it turns out to be smaller for a given N, R, and k in LDA than in HS. This is because in HS scheme, the exchange energy is calculated exactly. Interestingly, the ratio

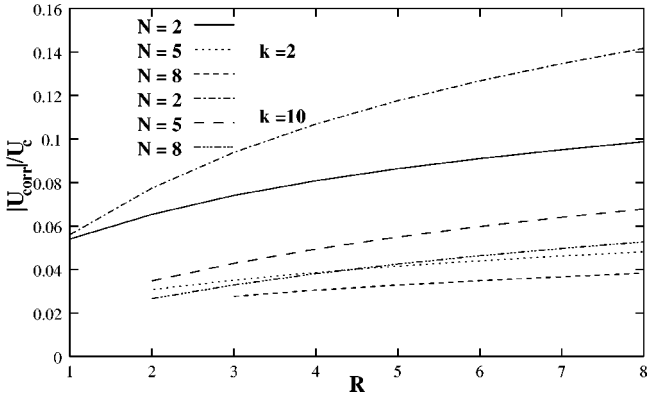

FIG. 4. The ratio of correlation energy U_{corr} to the Coulomb energy U_c is plotted as a function of the quantum dot size R for $k=2$ and 10 and total number of electrons $N=2$, 5, and 8. The correlation is more effective for few-electron quantum dots. It increases with both R and k.

$|U_x|/U_c$ in both HS and LDA does not change with size and remains constant for all value of N. For large N, it goes down as well.

To see further how important a role correlation plays, in Fig. 4, we plot the ratio $|U_{corr}|/U_c$ against the size R. We find that the ratio is always large for a given N and for large k. This means that for a fixed density of electrons, as we increase k, we effectively decrease the density. This makes the correlation more pronounced. As the dot size increases, the correlation becomes significantly important. We may understand this by looking at the scaling of U_c and U_{xc} for both $k=2$ and 10 (see Table I). The Coulomb part decreases at a faster rate as compared to the correlation, effectively making the ratio large for large k. This is because for $k=2$, the electronic charge density is confined towards the center of the QD, whereas for $k=10$ it is more spread out towards the surface.

Finally, in Fig. 5, we plot the ratio U_{corr}/U_x against the size R. Once again we find that the ratio is large for a given N and for large k and varies over a wide range. In this case also as the dot size increases, the correlation becomes more pronounced. It is interesting to note that for $N=5$, the exchange energy dominates more and hence for both $k=2$ and $k=10$, the correlation to exchange ratio for $N=5$ is smaller than that for $N=8$. We also worked out the relative difference in exchange-correlation energy calculated using the LDA and LSDA as a function of the number of electrons. This we show in the inset of Fig. 5. The striking feature is the increase of the relative difference where there are unpaired electrons in the dot. That is, when the shell is half filled, for the number of electrons $N=5$, 13, 19, 27, and for quasiharmonic confinement. The relative difference decreases from a maximum of 5% for $N=5$ to 1% for $N=27$ as a function of N. Interestingly, for quasisquare confinement, the relative difference is also found at $N=1$ and is as large as 25%, whereas for quasiharmonic confinement, it is almost zero. For those values of N other than the half-filled shell, the relative difference is less than 1%. This we believe maybe due to the numerical inaccuracy. This shows that the LSDA is a better approximation to the many-body exchange energy than the LDA, particularly when the shell is half filled, giving rise to nonzero total-spin angular momentum. We close the discussion with the statement that the exchange correlation does play an important role in deciding the scaling of the Hubbard U potential. However, it does not play a dramatic role and leads to negative U as predicted by Katayama-Yoshida and Zunger in 1985.[11] It would perhaps be interesting to examine many-body effects for an impurity in a quantum dot.

III. CONCLUSION

We reiterate that as the system size becomes larger (or the electron density spreads out more), a more accurate analysis would require the use of asymptotically correct exchange-correlation potential in place of LSDA. The latter is not accurate in the outer regions of a system. Shell filling effects which indicate the possibility of a Jahn-Teller–like effect in quantum dots have been observed. These will be examined further. The scaling of U, U_c, and U_{xc} strongly depends on the shape of the confining potential. Exchange correlation does play an important role in the scaling of Hubbard U potential but it does not show any anomalous behavior as to outweigh the Coulomb interaction and make the overall U negative. In particular, correlation is more sensitive to the

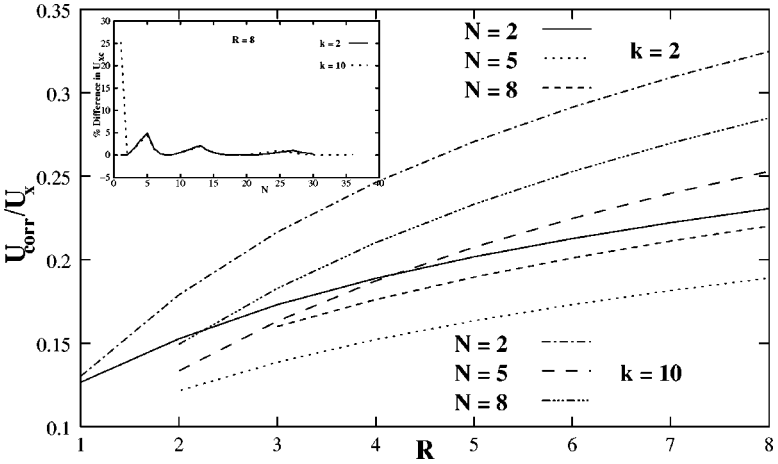

FIG. 5. The ratio of correlation to the exchange energy is plotted as a function of the quantum dot size R for $k=2$ and 10 and total number of electrons $N=2$, 5, and 8. The correlation energy is more pronounced for a small number of electrons in a quantum dot. It increases with both R and k. Interestingly, for $N=5$, the exchange energy dominates more and hence for both $k=2$ and $k=10$, the correlation to exchange ratio for $N=5$ is smaller than that for $N=8$. The inset shows the percentage difference in the exchange energy calculated using the LDA and LSDA, i.e., $[(U_{xc})_{LSDA}-(U_{xc})_{LDA}]\times 100/(U_{xc})_{LSDA}$.

quantum dot size and the shape of the confinement. It has long been understood that electron correlation is of critical importance in atoms, molecules, and solids and it should also be so in quantum dots. This stands justified through our calculations.

ACKNOWLEDGMENTS

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APPENDIX

One can understand the scaling behavior of the Coulomb term U_c on the basis of following simple argument. This argument resembles first-order perturbation-theory analysis.

Harmonic confinement $(k=2)$. In the first-order perturbation-theory sense, the Kohn-Sham orbital maybe taken to be a harmonic-oscillator type. The ground-state wave function and density are given by

$$\psi(r)=\left(\frac{m^*\omega}{\pi}\right)^{3/4} e^{-m^*\omega r^2/2}, \tag{A1}$$

$$\rho(r)=\left(\frac{m^*\omega}{\pi}\right)^{3/2} e^{-m^*\omega r^2}. \tag{A2}$$

Now from the relation

$$\frac{1}{2}m^*\omega^2 r^2=\frac{V_0 r^2}{R^2},$$

$$\omega=\sqrt{\frac{2V_0}{m^*R^2}}. \tag{A3}$$

The electron-electron interaction energy is given by

$$U_c=\frac{1}{2}\int\int \frac{\rho(r)\rho(r')}{|\vec{r}-\vec{r}'|}d\vec{r}d\vec{r}' \tag{A4}$$

$$\frac{1}{2}\left(\frac{m^*\omega}{\pi}\right)^3\int\int \frac{e^{-m^*\omega r^2}e^{-m^*\omega r'^2}}{|\vec{r}-\vec{r}'|}r^2 dr d\Omega r'^2 dr' d\Omega', \tag{A5}$$

where $d\Omega=\sin\theta d\theta d\phi$. We now make substitutions and introduce new dimensionless variables ζ and ζ' in place of r and r' such that

$$\zeta^2=m^*\omega r^2, \tag{A6}$$

$$\left(\frac{1}{m^*\omega}\right)^{3/2}\zeta^2 d\zeta=r^2 dr, \tag{A7}$$

$$U_c=\frac{1}{2}\left(\frac{m^*\omega}{\pi}\right)^3\left(\frac{1}{m^*\omega}\right)^3 (m^*\omega)^{1/2} \times\int\int \frac{e^{-\zeta^2}e^{-\zeta'^2}}{|\vec{\zeta}-\vec{\zeta}'|}\zeta^2 d\zeta d\Omega \zeta'^2 d\zeta' d\Omega'. \tag{A8}$$

Using Eq. (A3), we can write

$$U_c=\frac{1}{2\pi^3}\left(\sqrt{\frac{2m^*V_0}{R^2}}\right)^{1/2}\Gamma, \tag{A9}$$

that is,

$$U_c\sim\frac{1}{\sqrt{R}}, \tag{A10}$$

where Γ is the value of the integral and is a constant, given by

$$\Gamma=\int e^{-\zeta^2}\zeta^2 d\zeta d\Omega\int_0^\infty\int_0^\pi\int_0^{2\pi} \times\frac{e^{-\zeta'^2}\zeta'^2 d\zeta'\sin\theta' d\theta' d\phi'}{(\zeta^2+\zeta'^2-2\zeta\zeta'\cos\theta')^{1/2}}. \tag{A11}$$

Square-well confinement $(k\geqslant 10)$. Once again, in a first-order perturbation-theory approximation, the Kohn-Sham orbitals are square-well–type wave function. The ground-state wave function and density are given as

$$\psi=\frac{A\sin k_{in}r}{k_{in}r}, \tag{A12}$$

$$\rho=\frac{|A|^2\sin^2 k_{in}r}{k_{in}^2 r^2}, \tag{A13}$$

where

$$A=\frac{k_{in}}{\sqrt{4\pi}}\left[\frac{R}{2}\left(1-\frac{\sin(2k_{in}R)}{2k_{in}R}\right)+\frac{1-\cos(2k_{in}R)}{4k_{out}}\right]^{-1/2}, \tag{A14}$$

$$k_{in}=\sqrt{2m^*E},\qquad k_{out}=\sqrt{2m^*(V_0-E)}. \tag{A15}$$

Now from the definition of electron-electron interaction energy [Eq. (A4)],

$$U_c=\frac{|A|^4}{2}\int\int\frac{\sin^2(k_{in}r)\sin^2(k_{in}r')}{(k_{in}r)^2(k_{in}r')^2|\vec{r}-\vec{r}'|}d\vec{r}d\vec{r}'=\frac{|A|^4}{2}\int\int\frac{\sin^2(k_{in}r)\sin^2(k_{in}r')r^2 dr d\Omega r'^2 dr'\sin\theta' d\theta' d\phi'}{(k_{in}r)^2(k_{in}r')^2(r^2+r'^2-2rr'\cos\theta')^{1/2}}, \tag{A16}$$

where $d\Omega = \sin\theta d\theta d\phi$. Now we introduce new dimensionless variables ξ and ξ' such that

$$k_{in} r = \xi, \qquad k_{in} r' = \xi'. \tag{A17}$$

Thus,

$$U_c = \frac{|A|^4}{2} \frac{1}{k_{in}^5} \int \int \frac{\sin^2\xi \, d\xi d\Omega \sin^2\xi' \, d\xi' \sin\theta' d\theta' d\phi'}{(\xi^2 + \xi'^2 - 2\xi\xi'\cos\theta')^{1/2}}$$

$$= \frac{|A|^4}{2} \frac{2\pi}{k_{in}^5} \int \sin^2\xi d\xi d\Omega \int \sin^2\xi' d\xi' I(\xi), \tag{A18}$$

where the angle integral

$$I(\xi) = \int_0^\pi \frac{\sin\theta' d\theta'}{(\xi^2 + \xi'^2 - 2\xi\xi'\cos\theta')^{1/2}} = \begin{cases} \left(\frac{2}{\xi}\right), & \xi > \xi' \\ \left(\frac{2}{\xi'}\right), & \xi < \xi'. \end{cases}$$

Hence, the electron-electron interaction energy becomes

$$U_c = |A|^4 \frac{\pi}{k_{in}^5} 8\pi \int_0^\infty \sin^2\xi \, d\xi \left[\frac{1}{\xi} \int_0^\xi \sin^2\xi' \, d\xi' + \int_\xi^\infty \frac{\sin^2\xi'}{\xi'} d\xi' \right]$$

$$= \frac{8\pi^2 |A|^4}{k_{in}^5} C, \tag{A19}$$

where C is a constant and is the value of the integral. Now from Eqs. (A14), (A15), and (A19), we have

$$U_c = \frac{C}{2\sqrt{2m^*E}} \left[\frac{R}{2} \left(1 - \frac{\sin 2\sqrt{2m^*E}\, R}{2\sqrt{2m^*E}\, R} \right) + \frac{(1 - \cos 2\sqrt{2m^*E}\, R)}{4\sqrt{2m^*(V_0 - E)}} \right]^{-2}. \tag{A20}$$

Assuming $E \sim 1/R^2$ and also $V_0 \gg E$, one can write

$$U_c = \frac{C_1 R}{[C_2 R + C_3/\sqrt{V_0}]^2}, \tag{A21}$$

where C_1, C_2, and C_3 are constants. For V_0 large enough, the last term in the denominator of the above equation can be neglected. Therefore,

$$U_c \sim \frac{1}{R}. \tag{A22}$$

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