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Article *in* Journal of Applied Physics · July 2013

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Citation: J. Appl. Phys. **114**, 043706 (2013); doi: 10.1063/1.4816099
View online: http://dx.doi.org/10.1063/1.4816099
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v114/i4
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The electronic properties of a two-electron multi-shell quantum dot-quantum well heterostructure

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(Received 11 June 2013; accepted 3 July 2013; published online 24 July 2013)

A detailed investigation of the electronic properties of a double electron in a core/shell/well/shell quantum dot heterostructure has been systematically studied for cases with and without an on-center donor impurity. For this purpose, the Poisson-Schrödinger equations have been solved self-consistently in the frame of the single band effective mass approximation and Hartree treatment. The variation of the binding energies of negatively charged donor impurity (D^-) have been examined for different core radii, shell thicknesses, and well widths. The results obtained have been presented comparatively as a function of layer thicknesses and probable physical reasons behind in their behavior have been discussed. © *2013 AIP Publishing LLC*.
[http://dx.doi.org/10.1063/1.4816099]

I. INTRODUCTION

Nowadays, the latest developments in fabrication techniques allow the production of semiconductor quantum dot nanocrystals (QDNCs) with layer by layer and different shapes.[1–7] These kinds of QDNCs have been the subject of intense research because of various potential applications, for example, in new-generation photovoltaic solar cells.[8–10] Therefore, a number of authors investigate both theoretically and experimentally the electronic and optical properties of QDNCs.[11–17] As is well known, if an impurity atom has excess one electron than host material that doped in, this impurity atom is called as a donor.[18] When a donor impurity is put in a QDNC and the radius of the QDNC with the impurity is large sufficient, the excess electron in the QDNC is bound to the impurity atom, and it is known as a neutral hydrogenic donor impurity (D^0) because it is very similar to a hydrogen atom.

The hydrogenic donor impurity problem is a very useful model in understanding of electronic and optical properties of quantum dot (QD) heterostructures. Pioneering studies on a donor impurity in a QD has been reported by Zhu *et al.* and Porras-Montenegro, and Perez-Merchancano.[19,20] Later, different studies related to an on-center or off-center donor impurity in a QD heterostructure have been published in the literature. In this context, Deng *et al.*[21] calculated the binding energies of shallow donors and acceptors in a spherical quantum dot for both a finite and an infinite high barrier using the variational approach, including the spatial variation of dielectric screening. Zhu and Chen[22] calculated the energy levels and binding energies of an off-center donor in a spherical quantum dot by a linear variational method. They showed that the alteration of the position of a single Coulomb center can largely change the single-electron spectrum in a quantum dot with a larger radius. Elabsy[23] investigated the effect of temperature on the binding energy of a donor impurity located at the center of a spherical semiconductor GaAs/AlGaAs quantum dot. Lien and Trinh[24] studied the binding energy of hydrogen impurities in spherical QDs with parabolic confinements and disc-like QDs with parabolic lateral confinements in an electric field. Peter[25] calculated the binding energy of shallow hydrogenic impurities in spherical QDs in the influence of pressure as a function of the dot size. There are a lot of paper with similar contents reported by many authors.[26–35]

The understanding of electronic and optical properties of a many-electron QDNC is rather important for device applications. If we consider a two-electron QDNC, its electronic and also optical properties are strictly dependent on the Coulomb interaction between the electrons. In a QDNC which contains neutral donor impurity, a second electron can also bind to the D^0 as a result of polarization if the radius becomes larger. In this case, the structure is called as a negatively charged donor impurity (D^- center). In such a structure, while the impurity pulls the energy levels down, the Coulomb interactions between the electrons shift the levels up. Therefore, the electronic structure of a D^- is rather complex, and the understanding of its electronic properties is very important. Zhu *et al.*[36] have examined binding energy of the D^- as a function of dot radius and barrier height in the frame of effective mass approximation using variational method. Szafran *et al.*[37] have been investigated ground and excited states' energy levels of D^- in a QD for spin-singlet and spin-triplet configurations as a function of QD radius and barrier depth. Xie[38–41] has calculated energy spectrum of a D^- center in a disc-shaped QD with parabolic confinement using the method of few-body physics. Also, he has investigated the magnetic field effects on the electronic properties of an off- and on-center D^- in a QD. In another study, Xie[42] has investigated the second bound state binding energy spectra of a D^- center QD system depending on magnetic field strength and QD radius by using the method of hyperspherical coordinates. Gu and Liang[43] have explored D^-

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centres' quantum dot in a Gaussian confining potential. They obtained the energy spectrum and showed that the property of the ground and low-lying states of the quantum dot is rather sensitive to the strength of the confinement potential. Garcia *et al.*[44] have investigated the dependencies of the neutral and negative donor binding energies and their ratios on the base radius in the pyramid, lens, and disk shaped QDs. Later, a number of studies related to electronic properties of D^- centers have been reported by many authors for different physical conditions.[45–50]

Developments in growth technology have resulted in manufacturing of multi-layered spherical quantum dots (MSQD).[51,52] Recently, the electronic and optical properties of multilayered spherical quantum dot under some different physical effects are investigated by many authors.[16,17,53–58] It is seen from these studies that the electronic and optical properties and impurity binding energies are very sensitive to the layer thicknesses. In all of these studies, a single electron and/or neutral donor impurity have been taken into consideration. The electronic properties of a double electron and/or a D^- in a MSQD have not been investigated yet.

The objective of this study is to investigate the electronic properties of a double electron MSQD for cases with and without a donor impurity in the frame of the single band effective mass approximation. For this purpose, the Poisson-Schrödinger equations have been solved full numerically and self-consistently. The results have been presented comparatively for cases with and without the impurity.

The rest of the paper is organized as follow: In Sec. II, we describe our model and its theory. In "Results and Discussion" section, we present our calculation results, and their probable physical reasons are discussed. In the last section, a brief conclusion is introduced.

II. MODEL AND THEORY

In this study, we consider a multi-shell spherical CdSe/ZnS/CdSe/ZnS (i.e., core/shell/well/shell) quantum dot nanocrystal. The CdSe core material with radius R_1 is coated with ZnS shell, which has a wider band gap than that of the CdSe. The shell thickness is $T_s = R_2 - R_1$. This structure is further coated with CdSe for well region with a width of $T_w = R_3 - R_2$. Finally, whole structure is covered with ZnS shell material in order to constitute a confinement region in the well layer. The potential profile of this structure is shown in Figure 1. In this structure, the donor impurity is on-center of the core region.

Here, we consider interacting two electrons in a core/shell/well/shell QD. In the effective mass approximation and BenDaniel-Duke boundary conditions, for a spherically symmetric quantum dot, the single-particle Schrödinger equation is given by

$$\left[-\frac{\hbar^2}{2}\vec{\nabla}_r\left(\frac{1}{m_e^*(r)}\vec{\nabla}_r\right) - e\phi_{sc} - \frac{Ze^2}{\kappa(r)r} + V_e(r)\right]R_{n\ell}(r) = \varepsilon_{n\ell}R_{n\ell}(r). \quad (1)$$

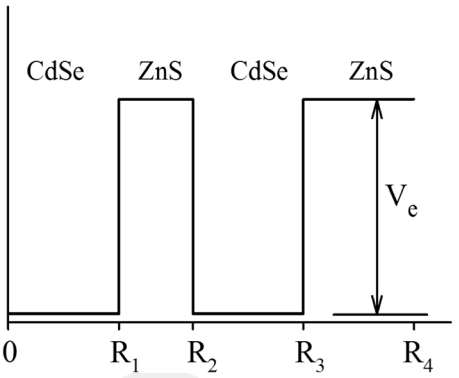

FIG. 1. Schematic representation of the potential profile of multi-shell CdSe/ZnS quantum dot heterostructure.

Here, first term is kinetic energy term of the electron, $\hbar$ is the reduced Planck constant, $m_e^*(r)$ is the position-dependent effective mass of the electron, e is the unit electronic charge, ϕ_{sc} is the self-consistent Hartree potential between the electrons, the third term is the Coulomb interaction between electron and donor impurity, Z is charge of the impurity, $V_e(r)$ is the position dependent confining potential of the electron, $\kappa(r)$ is the position dependent dielectric constant, $\varepsilon_{n\ell}$ is the single particle energy eigenvalue, and $R_{n\ell}(r)$ is the radial wave function of the electron. Here, if $Z = 1$, there is a hydrogenic donor impurity on the center of the QD and the structure is called as D^- center. However, if $Z = 0$, there is no impurity and the structure is called as double-electron QD. Mathematical expression of the confinement potential is

$$V_e(r) = \begin{cases} 0, & r \le R_1 \text{ and } R_2 \le r \le R_3 \\ V_e, & R_1 < r < R_2 \text{ and } r > R_3 \end{cases}, \quad (2)$$

where V_e is the conduction band offset between CdSe and ZnS.

The self-consistent electrostatic potentials caused by the electrons are calculated by means of the Poisson equation. The Poisson equation can be written as

$$\vec{\nabla}\kappa(r)\vec{\nabla}\phi_{sc} = \frac{e}{\varepsilon_0}\rho_e(r), \quad (3)$$

where ρ_e is the density of the electron and ε_0 is the dielectric permittivity of the vacuum. This equation contains the image potential contributions due to the surface polarization at the interfaces.[27] The electron density is

$$\rho_e(r) = \frac{1}{4\pi}\sum_{\ell=0}^{p} 2(2\ell+1)\sum_{n=1}^{np}|R_{n,\ell}^{elec}(r)|^2 + \frac{1}{4\pi}q|R_{nq,\ell q}^{elec}(r)|^2, \quad (4)$$

Here, $2(2\ell + 1)$ is the spin and magnetic degeneracies; p and np are the angular momentum quantum number and the principle quantum number of the fully occupied states, respectively; q is the number of remaining electrons in the last state; nq and ℓq are the principle quantum number and angular momentum quantum number of the last state, respectively.

In common application of the Hartree approximation, all electrons charge densities in the system considered have

been used in the Poisson equation and this potential is substituted into the Schrödinger equation. In this application, the self-interaction correction and exchange-correlation effects are not been taken into account, and so these corrections must be done for more realistic results. In this study, we have considered the Hartree approximation, a slightly different from the traditional manner. In the calculations, we use the charge density of one of the electron and the other one moves in a mean potential created by this electron. Since we limit ourselves to just ground state, 1s, our utilization is rather reasonable. As a result, the charge density will be

$$\rho_e(r) = \frac{1}{4\pi} q |R^{elec}_{nq,\ell q}(r)|^2. \tag{5}$$

We assume that our structure is spherically symmetric. The calculations are performed depending on the full numeric self-consistent solution of the Poisson-Schrödinger equations (1), (3), and (5), in the Hartree approximation. We employ matrix diagonalization technique for the determination of single particle energies. For this purpose, the Hamiltonian operator is discretized on a uniform radial mesh in one dimension (1D) using the finite difference technique, and hence, Eq. (1) is reduced to a matrix eigenvalue equation. Here, the width Δr between two mesh points is chosen as 0.001. Eigenvalues and eigenvectors of these matrix equations are determined by the EISPACK subroutine.

Total energy of the two-electron QD is

$$E_{tot} = \sum_{i=1}^{2} \varepsilon_{n\ell}(i) - \frac{e}{4\pi\varepsilon_0\kappa(r)} \int R_{n\ell}\phi_{sc}R_{n\ell}r^2 dr. \tag{6}$$

It should be noted that, in our calculation scheme explained above, since the Coulomb interaction between the electrons is counted two times, one of them is subtracted in total energy calculation. The binding energy of D^- is defined as

$$E_b(D^-) = E_0 + E(D^0) - E(D^-), \tag{7}$$

where E_0 is the ground state energy of a single electron without the impurity in the QD, $E(D^0)$ and $E(D^-)$ are the lowest energy state of the neutral donor and negatively charged donor center, respectively.

III. RESULTS AND DISCUSSION

The atomic units have been used throughout the calculations, where $\hbar = m_0 = e = 1$. The material parameters are $m^*_{CdSe} = 0.13m_0$ and $m^*_{ZnS} = 0.28m_0$, $\kappa_{CdSe} = 9.3$, and $\kappa_{ZnS} = 8.1$.[59] The electron confinement potential, $V_e = 1.05\,\text{eV}$.[60] The effective Bohr radius and the effective Rydberg energy are determined as $a_0 = 37.84\,\text{Å}$ and $R_y = 20.44\,\text{meV}$, respectively.

In the calculation results, we observe that when the core radii are comparable with well widths ($R_1 \cong T_w$) or $R_1 < T_w$, the finding probability becomes larger in the well region, accordance with the principle of minimum energy. In contrast, when the core radius is larger than the well width, the finding probability in the core region becomes more. Figure 2 shows the finding probability of the electrons depending on the layer widths. Top and bottom panels correspond to $T_s = 0.2\,a_0$ and $T_s = 0.4\,a_0$, respectively. As seen from the figure, although it can be mentioned the finding probability, even a little, in the core region for $R_1 = 0.4$ and $T_s = 0.2\,a_0$ in case of $Z = 1$, it becomes approximately zero for $T_s = 0.4\,a_0$. Similarly, as the finding probability of $Z = 0$ case exists in the well region for $R_1 = 0.8$ and $T_s = 0.2\,a_0$, it vanishes for $T_s = 0.4\,a_0$ at the same core radius as can be seen from bottom panel of the figure. That is, the shell thickness has also a

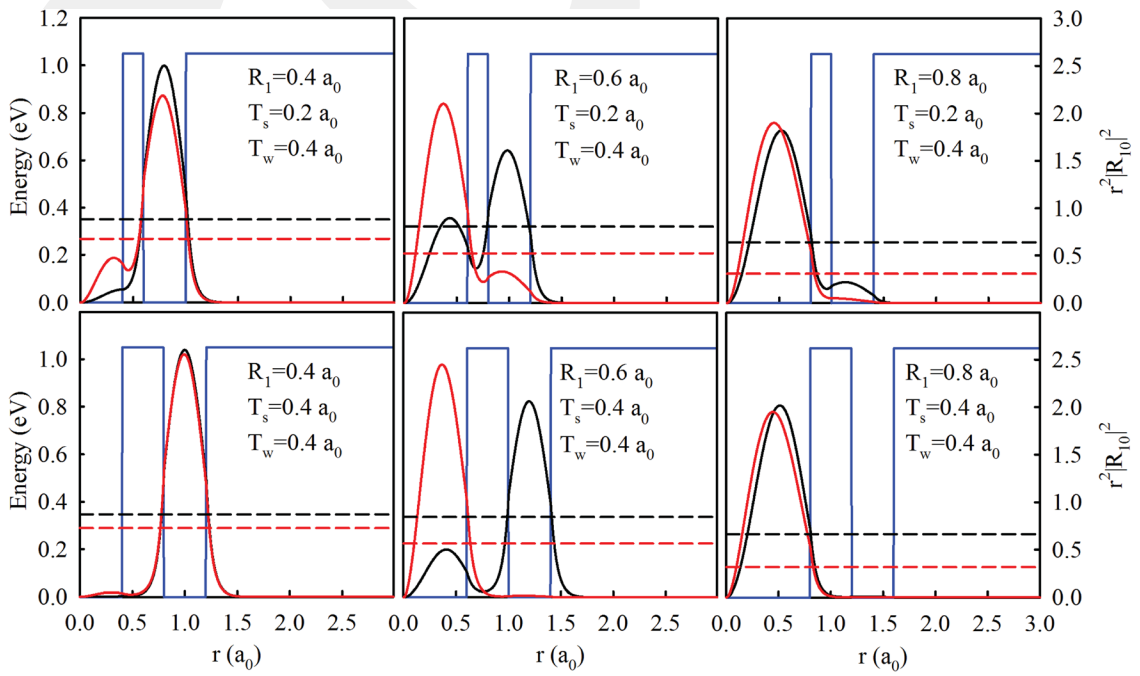

FIG. 2. Probability densities of the electrons as a function of the radius for various core radii (R_1) and well widths (T_w). The top panel for $T_s = 0.2\,a_0$ and the bottom panel for $T_s = 0.4\,a_0$. Black and red lines correspond to $Z = 0$ and $Z = 1$ cases, respectively. The dashed lines show the single particle energy levels.

great influence on the electronic properties of the multi-shell quantum dot structures.

Figures 3(a)–3(d) show the total energy of double electron and negatively charged donor impurity in a MSQD heterostructure, comparatively, as a function of core radius, R_1, for different well widths and shell thicknesses. It should be noted that the $Z=0$ corresponds to the double-electron cases and the $Z=1$ corresponds to negatively charged donor center. As can be seen from the figure, all total energies exhibit a decrease behavior with increasing R_1. These decreasing tendencies are more evident for $T_w = 0.2\,a_0$ in all cases. The total energies become independent from T_w for $R_1 \geq 1.2\,a_0$ for $Z=0$ cases, and so they have same values with further increasing of R_1 after this certain values. Similar behaviors are observed for $Z=1$ cases except $R_1 > 0.8\,a_0$. These results can be explained as follows: In small core radii, the electrons are more energetic, and hence, the changes of well region size affect the energies drastically. When the R_1 reaches the certain values mentioned above, the electrons are almost completely confined to the core region and are not influenced by the well region. It is noted that if the T_w is comparable with the R_1, finding probability of the electron in the well region becomes much more. In contrast to this, if the $R_1 > T_w$, in this case, the finding probability of the electron in the core region is much more. As the repulsive Coulomb interaction between the electrons raises the single particle energy, the attractive Coulomb potential of the impurity pulls this energy down a little. Therefore, the total energy of double-electron MSQD is greater than that of the negatively charged donor impurity in all cases. More interesting result is that there are the abrupt decreases on the energies at around the certain R_1 values. In case of $Z=0$, while a sudden decreasing is observed only at $T_w = 0.8\,a_0$ for $T_s = 0.2\,a_0$, there is no slump in $Z=1$ case, as seen in Figs. 3(a) and 3(b). These sudden decreases are much more evident at $T_s = 0.4\,a_0$ when $T_w > 0.2\,a_0$ for both $Z=0$ and $Z=1$ cases as can be seen from bottom panel of the figure. These changes show that the energy levels are affected from the shell thickness, between the core and well regions.

Figures 4(a) and 4(b) show the binding energies of the D^- center as a function of the core radius for different well widths at two various T_s values specified on the figures. In both figures, the binding energies exhibit rather different characters when almost $R_1 < 1.2\,a_0$. When we look at the top panel, we see that the binding energies are decreasing first with increasing R_1. This decrease is smaller for small T_w values and becomes more with increasing T_w. In addition, the decreasing binding energies reach to larger R_1 values at large well widths. After the binding energies reaches to minimal values, they increase until $R_1 = 1.2\,a_0$. The binding energies have same energy values and exhibit a typical single core/shell QD binding energy behavior for $R_1 \geq 1.2\,a_0$. If we focus on the bottom panel of Fig. 4, we observe that these changes in the binding energies are more complicated for $T_s = 0.4\,a_0$. In addition, the binding energies are a bit larger when compared with the top panel. The physical reason of these variations can be explained as follows: When $T_s = 0.2\,a_0$, the electron(s) can tunnel easily between the core and well regions. Hence, the electron(s) confined in the core (well) region is affected more from the well (core) region. In addition, the repulsive Coulomb interactions are not changed drastically due to thin shell thickness. We conclude that the structure behaves a somewhat single core/shell QD structure. As a result, the binding energies exhibit a smooth changes for $T_s = 0.2\,a_0$. In case of $T_s = 0.4\,a_0$, the tunneling is a bit harder between the core and well regions. Therefore, the electron(s) may be more confine in one of the regions. The repulsive Coulomb interactions between the

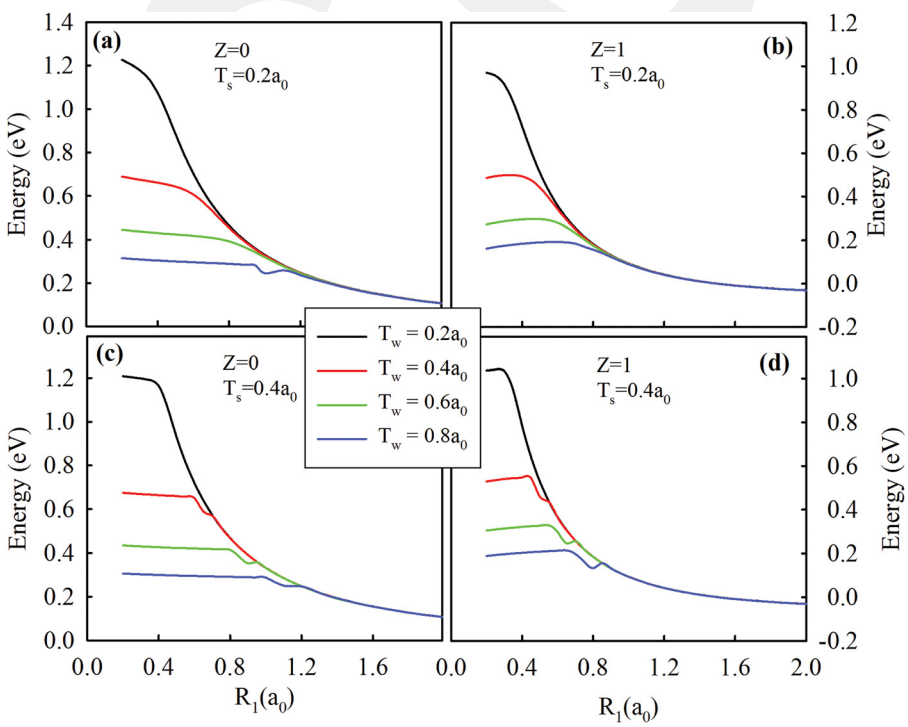

FIG. 3. Variation of total energy as a function of core radius for various well widths (T_w) (a) for $Z=0$ and $T_s = 0.2\,a_0$, (b) for $Z=1$ and $T_s = 0.2\,a_0$, (c) for $Z=0$ and $T_s = 0.4\,a_0$, and (d) for $Z=1$ and $T_s = 0.4\,a_0$.

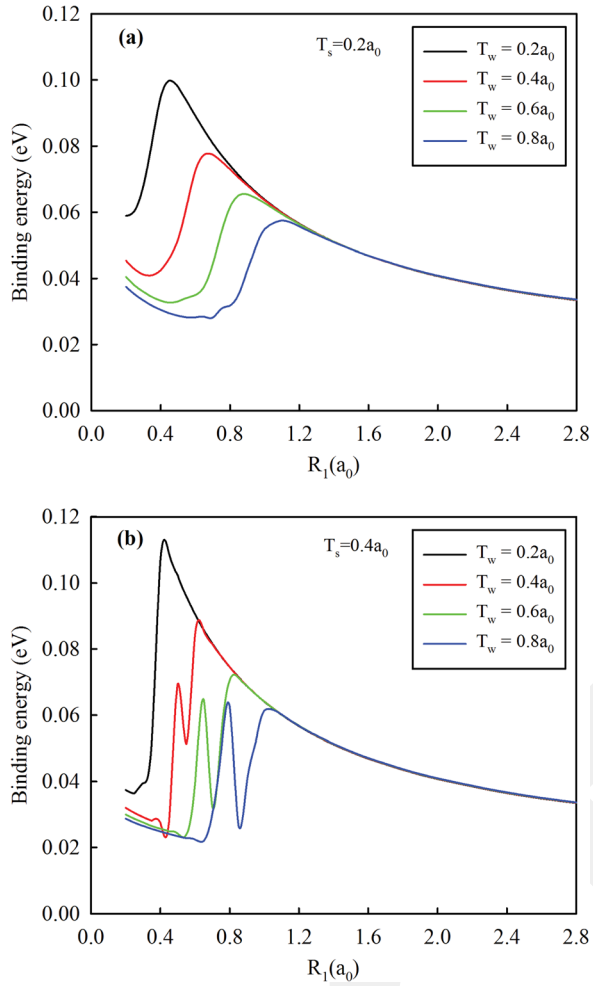

FIG. 4. The binding energy of the D^- as a function of core radius for different well widths (T_w) (a) for $T_s = 0.2\,a_0$ MSQD and (b) for $T_s = 0.4\,a_0$.

electrons are also dramatically influenced from the shell thickness, and hence, the binding energies display rather different character.

In Fig. 5, we have plotted the variation of the binding energies with shell thickness. As observed from the figure, while the binding energy decreases with increasing shell thickness for $R_1 = 0.2\,a_0$, it increases a little for $R_1 = 0.6\,a_0$. These behaviors can be explained as follows: When $R_1 = 0.2\,a_0$, the finding probability of the electron(s) expands to core and well regions at thin shell layers and the impurity effect is perceived more by the electron(s) and so the binding energy is also higher. When the shell thickness becomes more, the finding probability is to be maximum in the well region because the tunneling probability to core region decreases. Therefore, influence of the impurity on the electrons diminishes. This causes the lower binding energies. When $R_1 = 0.6\,a_0$, the finding probability is maximum in the core region because well layer is very small in comparison with core one. Therefore, the impurity effect is larger and hence the binding energy is larger. When the shell thickness increases, the tunneling probability to well region diminishes and the electron(s) are almost completely confined in the core region. In this case, the binding energy grows a bit more up with increasing shell thickness.

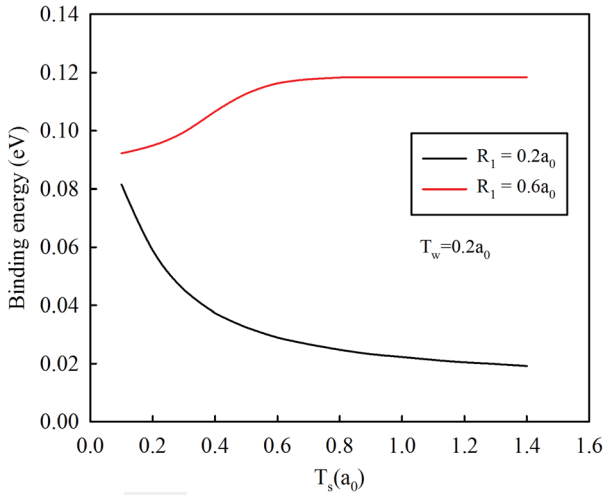

FIG. 5. The binding energy of the D^- as a function of shell thickness for different core radii (R_1). The well width, $T_w = 0.2\,a_0$.

Figures 6(a) and 6(b) show the D^- binding energies as a function of the well width, T_w, for different core radii and shell thicknesses, top panel for $T_s = 0.2\,a_0$ and bottom panel for $T_s = 0.4\,a_0$. When we compare the top and bottom panels with each other, although the general trends of the binding

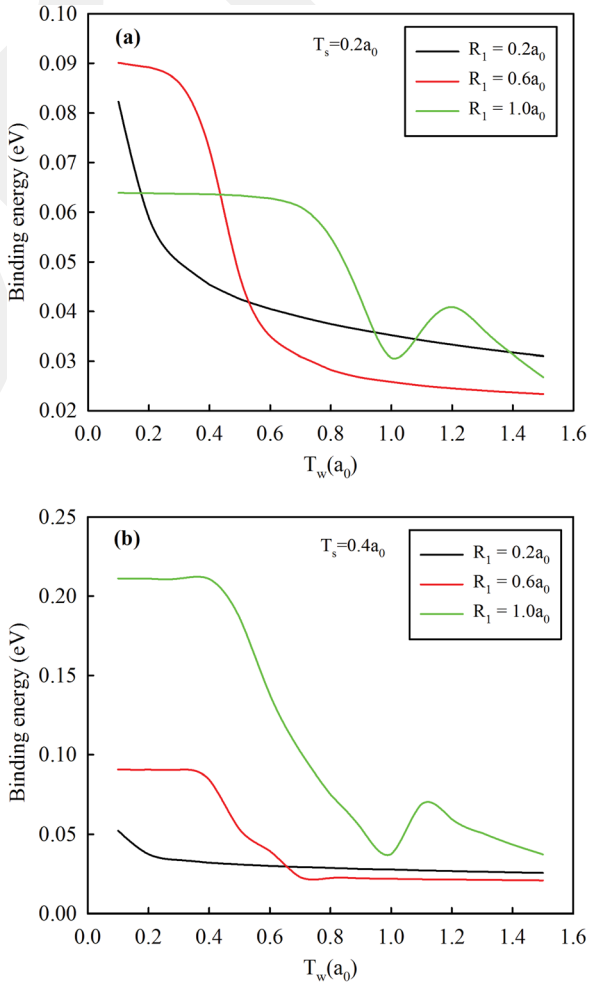

FIG. 6. The binding energy of the D^- as a function of well width for different core radii (R_1) specified on the figures. (a) $T_s = 0.2\,a_0$ and (b) $T_s = 0.4\,a_0$.

energies are similar for same parameters, we see that the binding energies in the top panel have lower values except of $R_1 = 0.2\,a_0$. Because, the electrons can tunnel easily between the core and well regions, and at the same time, the repulsive Coulomb interaction between the electrons becomes more dominant in $T_s = 0.2\,a_0$ case. The binding energies for $R_1 = 0.2\,a_0$ are decreasing with increasing well width because the electrons become localized in the well region and the influence of the impurity reduces at larger well widths. In case of $R_1 = 0.6\,a_0$, the binding energies remain almost fixed first until certain well widths and decreasing with further increasing of the well layer. These behaviors associated with the electrons are forced to be confined in the core region at smaller well widths, and until a certain well width, the electrons do not feel sufficiently in the well region. After this certain well width, the electrons tunneled to the well region are influenced lesser. Similar variations are observed for $R_1 = 1.0\,a_0$ cases. In this case, as distinct from the previous core radii, there are observed a shoulder about $T_w = 1.2\,a_0$.

IV. CONCLUSION

We have carried out a detail investigation of the electronic properties of double-electron MSQD heterostructure for cases with and without a hydrogenic donor impurity. In this context, the single particle and total energies, probability densities, and binding energies of the D^- have been determined as a function of layer thicknesses. In the calculations, the Poisson-Schrödinger equations have been solved self-consistently in frame of the effective mass approximation. We have found that the electronic properties are drastically influenced from the layer thicknesses. We hope that this study will contribute to the understanding of the electronic properties of double electron MSQD for cases with and without the donor impurity.

ACKNOWLEDGMENTS

This work is a part of the Ph.D. thesis preparing by R. Aydin at Physics Department of Selcuk University.

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