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Many-electron artificial atoms

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Artificial atoms, i.e., systems of excess electrons confined in semiconductor quantum dots, are studied by the unrestricted Hartree-Fock method. We consider a spherical quantum dot embedded in an insulating matrix and assume a confinement potential in a form of spherical potential well of radius R and depth V_0. The calculations have been performed for few- and many-electron artificial atoms with the number of electrons from 1 to 20. We have shown that bound many-electron states of atomlike properties are created in quantum dots if the values of R and V_0 are sufficiently large. The critical values of R and V_0 for the binding of N electrons in the quantum dots have been determined. We have found that the subsequent shells of the artificial atoms are filled by electrons according to the Hund rule. The characteristic behavior resulting from the full and half-filling of the shells is clearly visible in the dependence on the number of electrons of the calculated chemical potential, addition energy, and electric capacitance of the quantum dots. The present results have been compared with those of the classical Thomson model of atoms and applied to the quantum dots made of Si and GaAs. [S0163-1829(99)00320-3]

I. INTRODUCTION

Excess electrons introduced into semiconductor quantum dots (QD's) are subjected to an external potential, which confines electrons in all three dimensions.[1] A strength and range of the confinement potential are determined by a geometry of the nanostructure and/or an external voltage applied to the microelectrodes, i.e., can be changed intentionally, which gives us a unique opportunity of getting the electron energy spectra with the designed properties. For this reason, the system of excess electrons confined in the QD was called an artificial atom.[2] In the present paper, we consider a formation of artificial atoms in spherical QD's, which are grown by chemical methods as semiconductor nanocrystals of nearly spherical shape in an insulating matrix. The spherical QD's have been made of group-IV semiconductors[3] and III-V,[4] II-VI,[5] I-VII,[6] and IV-VI (Ref. 7) semiconducting compounds. The electron energy spectra of the QD's were studied by the far-infrared magneto-absorption,[8] capacitance spectroscopy,[9] and transport spectroscopy.[10] These experimental techniques allow us to study the atom-like properties of the excess electron systems confined in the QD's. Recently, the shell filling has been observed[11] for the artificial atoms in cylindrical QD's. A quantitative theoretical description of this effect has been given by the present authors.[12] In the present paper, we use a similar method to consider the shell filling in the artificial atoms of spherical symmetry.

A theoretical study of electrons in QD's was the subject of many papers.[13–30] The authors of these papers assumed the model confinement potential, which was either the infinitely deep rectangular potential well[13–15,26] or the harmonic-oscillator (parabolic) potential.[16–22,24,25,27–30] Both these potentials possess an infinite depth, which is in principle unphysical. Nevertheless, their application allowed the authors[17,19,27,28,30] to describe the main features of the confined electron systems, since the energy levels of experimental interest are located rather deeply in the corresponding potential well. Moreover, for quasi-two-dimensional cylindrical QD's, the harmonic confinement potential properly accounts for the nearly equal separations between the lowest-energy levels, although anharmonic corrections should also be included.[31] However, for both the harmonic and infinitely deep rectangular potential, a continuum-energy threshold does not exist; therefore, the corresponding Schrödinger equations possess exclusively bound-state solutions. This leads to the fundamental physical deficiency of these model potentials, namely, the binding and dissociation processes cannot be described with their use. Therefore, the theoretical models with these potentials put no limits on the number of electrons, which can be added to the QD. For the real QD — due to its finite nanoscale extensions in the three directions of space — the confinement potential has the finite depth and range. Therefore, the QD can be charged by the finite number of electrons. This number determines a quantum ''capacity'' of the QD. On the contrary to the classical electric capacitance of the QD, the quantum ''capacity'' of the QD can only be described with the use of the confinement potential of finite depth and range.

In order to solve the above-mentioned problems and obtain the finite binding energy and finite quantum ''capacity'' of the QD, we apply in the present paper the confinement potential in the form of the spherical potential well with the finite depth and range. The application of this confinement potential allows us to describe the characteristic properties of electrons confined in nearly spherical semiconductor nanocrystals. In particular, it leads to a clear physical interpretation of the binding of electrons in the QD and allows us to determine the quantum ''capacity'' of the QD. Therefore, the present theoretical model should properly, at least qualitatively, describe the basic properties of spherical QD's. Moreover, in the frame of this model we can construct a ''periodic table'' of artificial atoms. The present calculations for the many-electron artificial atoms have been performed with the help of the unrestricted Hartree-Fock method. We have studied the artificial atoms with the shells $1s$, $1p$, $1d$, and $2s$, which are defined by the one-electron states of the spherical

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potential well. A complete occupancy of these shells is achieved for $N=20$ electrons. As a result, we obtain the complete first ''period'' ($N=1,\ldots,18$) and the first two ''elements'' of the second ''period'' ($N=19$, 20).

In the frame of the classical mechanics, a ''periodic table'' of ''classical artificial atoms'' was obtained by Bedanov and Peeters,[32] who considered a system of classical charged particles confined in a two-dimensional parabolic potential. In this paper, we present the quantum-mechanical description of the ''periodic table'' formed by the artificial atoms. We provide solutions to the three-dimensional many-electron eigenvalue problems for the electrons confined in all the three dimensions.

The paper is organized as follows: the theoretical method is presented in Sec. II, the results and discussion are included in Sec. III, and the conclusions are in Sec. IV.

II. THEORY

We consider a system of excess electrons, which are confined in a single spherical QD embedded in an insulating matrix. The potential profile for this nanostructure is the following: the potential-well region inside the QD and the potential-barrier region in the surrounding matrix. We assume that the confinement potential for electrons has the form

$$V(\mathbf{r})=\begin{cases}-V_0 & \text{for } r<R,\\ 0 & \text{otherwise,}\end{cases} \tag{1}$$

where the potential-well depth $V_0>0$. The energy of the conduction-band minimum of the barrier material is set equal to zero and taken as the reference energy. Such a choice of the reference energy allows us to separate the discrete electron energy levels, which result from the size quantization, from the quasicontinuous conduction-band energy of the electrons in the barrier material. The Hamiltonian of the system of N excess electrons in the effective-mass approximation has the form

$$H=\sum_{i=1}^{N} h(\mathbf{r}_i)+\sum_{i=1}^{N}\sum_{j>i}^{N}\frac{2}{r_{ij}}, \tag{2}$$

where $\mathbf{r}_i$ is the position vector of the ith electron, r_{ij} is the electron-electron distance, and

$$h(\mathbf{r})=-\nabla^2+V(\mathbf{r}), \tag{3}$$

is the one-electron Hamiltonian. Throughout the present paper, the following units are used: donor rydberg $R_D=(m_e/m_{e0})Ry/\varepsilon_s^2$ is the unit of energy and donor Bohr radius $a_D=(m_{e0}/m_e)\varepsilon_s a_B$ is the unit of length, where the hydrogen rydberg $\mathrm{Ry}=13\,605.8$ meV, the hydrogen Bohr radius $a_B=0.0529$ nm, ε_s is the static dielectric constant, m_e is the electron conduction-band mass, and m_{e0} is the free-electron rest mass. We neglect the changes of the electron band mass and dielectric constant at the QD boundary.

The eigenvalue problem for Hamiltonian (2) has been solved by the unrestricted Hartree-Fock method. According to this approach, the N-electron wave function is taken on as the Slater determinant, which is constructed from the one-electron spin orbitals

$$\varphi_i(\xi_j)=\psi_{\mu s}(\mathbf{r}_j)\chi_s(\sigma_j). \tag{4}$$

Here, $i,j=1,\ldots,N$, $\xi=(\mathbf{r},\sigma)$, σ is the spin variable, $s=\pm 1/2$ is the spin quantum number, and $\mu=(klm)$, where l and m are the azimuthal and magnetic quantum numbers, respectively, and $k=n+1$, where n is the radial quantum number. In the Slater determinant, we take into account the following one-electron orbitals: $1s$, $1p$, $1d$, and $2s$, which are the solutions to the eigenvalue problem for Hamiltonian (3). The exact bound-state solutions to this eigenproblem are given by the spherical Bessel functions.[33] The present calculations have been performed with the help of the Roothan[34] modification of the Hartree-Fock approach. Accordingly, the one-electron orbitals have been proposed in forms of expansions in the Gaussian basis, i.e.,

$$\psi_{\mu s}(\mathbf{r})=\sum_{p_1p_2p_3q} c^{\mu s}_{p_1p_2p_3q}\, g_{p_1p_2p_3q}(\mathbf{r}), \tag{5}$$

where

$$g_{p_1p_2p_3q}(\mathbf{r})=x^{p_1}y^{p_2}z^{p_3}\exp(-\gamma_q r^2). \tag{6}$$

The parameters p_1, p_2, and p_3 take on values $p_i=0,1,2$, $c^{\mu s}_{p_1p_2p_3q}$ and γ_q ($q=1,\ldots,4$) are the variational parameters. Using the appropriate combinations of parameters p_i, we construct the s-, p-, and d-like solutions. The Hartree-Fock equations have been solved for each one-electron state with the quantum numbers $(klms)$ by the self-consistent iterative procedure with the minimization performed over the variational parameters $c^{\mu s}_{p_1p_2p_3q}$ and γ_q. Using these solutions, we have calculated the ground-state energy of the considered N-electron system.

Before performing the many-electron calculations, we have checked that the Gaussian basis [Eqs. (5) and (6)] correctly reproduces the analytical solutions to the eigenvalue problem for one-electron Hamiltonian (3). The application of the equivalent Slater basis leads to a very good agreement of the results[35] with the exact eigenfunctions and eigenvalues for the one-electron problem. It is well known that the properly chosen Gaussian basis yields the same results as those obtained with the Slater basis. In the present work, we additionally performed the direct calculations with the Gaussian basis [Eqs. (5) and (6)] for the one-electron states in the spherical potential well and found that the present results reproduce the analytical results with a high accuracy.

In the Hartree-Fock approach, the electron-electron correlation is neglected. We have performed a detailed study[36] of the problem of correlation for two-electron artificial atoms in a spherical QD with a finite confinement potential.[36] It was shown[36] that the Hartree-Fock and exact results are almost the same for QD's of small and intermediate radius. Only for large QD's ($R>10a_D$), i.e., in the weak-confinement regime, we have obtained small deviations between both the results. In the frame of the present work, we have carried out an additional test of the reliability of the unrestricted Hartree-Fock method. We have considered the pair of electrons confined in a three-dimensional harmonic-oscillator potential of the form $V(r)=r^2/l^4$, where l is a characteristic length of the problem. After separating out the center-of-mass motion, the problem is reduced to the one-particle one-

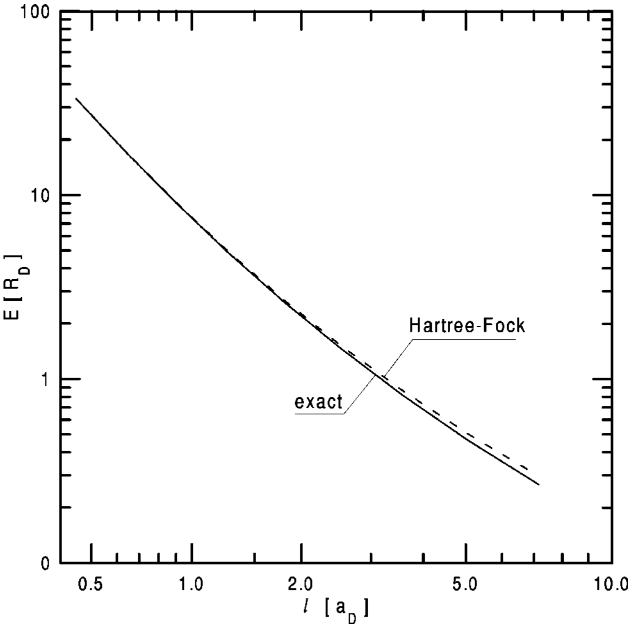

FIG. 1. Ground-state energy of two electrons confined in three-dimensional potential $V(r)=r^2/l^4$ calculated by the exact numerical method (solid curve) and unrestricted Hartree-Fock method (dashed curve). The donor rydberg R_D is the unit of energy and the donor Bohr radius a_D is the unit of length.

dimensional eigenvalue problem. For the ground state, we have solved this problem by a numerical shooting method, which yields exact results. A comparison of the exact results with the Hartree-Fock results obtained with the use of the procedure described in the context of Eqs. (4)–(6) is reported in Fig. 1. Figure 1 shows that only for large QD's, small differences between both the results are visible, which agrees with the results of our previous paper.[36] Based on the present and previous[36] study, we can state that the Hartree-Fock method provides the reliable results for the artificial atoms.

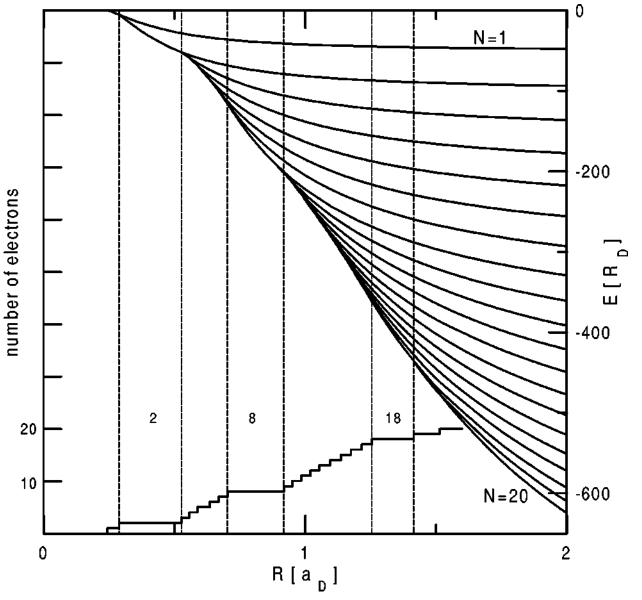

FIG. 2. Energy E of the ground state and number of electrons corresponding to the artificial atom of the lowest energy for $V_0 = 50R_D$ as functions of quantum-dot radius R. Vertical thin lines correspond to the filled shells. The donor rydberg R_D is the unit of energy, and the donor Bohr radius a_D is the unit of length.

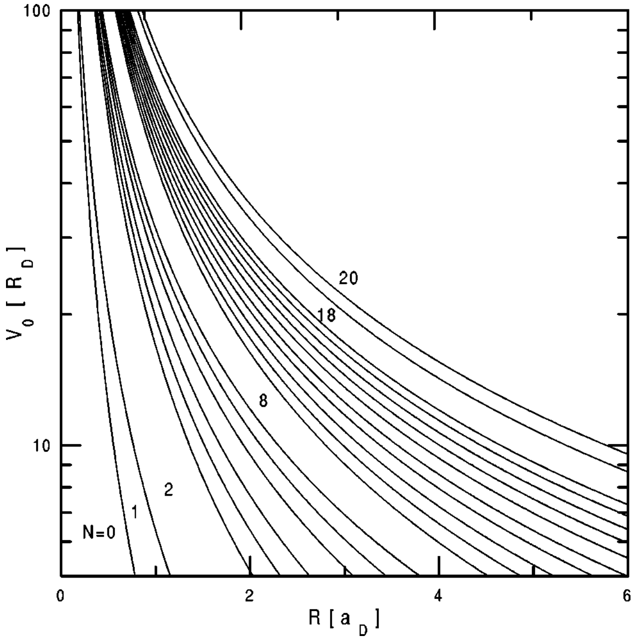

FIG. 3. ''Phase diagram'' showing the formation of N-electron artificial atoms. The system of N electrons confined in the quantum dot possesses the minimum ground-state energy if the quantum dot is characterized by the parameters R and V_0 from the region, which is labeled by N and limited by the corresponding curves.

III. RESULTS AND DISCUSSION

The results of the Hartree-Fock calculations are reported in Fig. 2. The right upper part of Fig. 2 displays the ground-state energy of N-electron artificial atoms for $N=1,\ldots,20$ and the left lower part — the numbers of electrons corre-

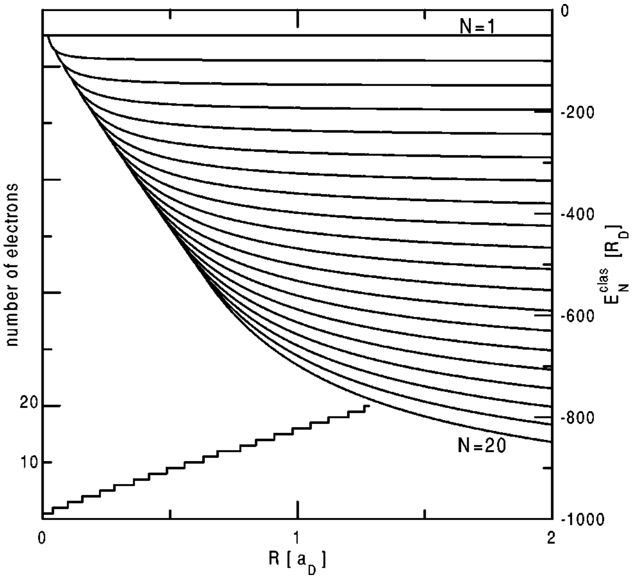

FIG. 4. Classical potential energy $E_N^{\rm clas}$ and number of electrons corresponding to the most stable configuration of interacting electrons subjected to the confinement potential with $V_0=50R_D$ as a function of radius R.

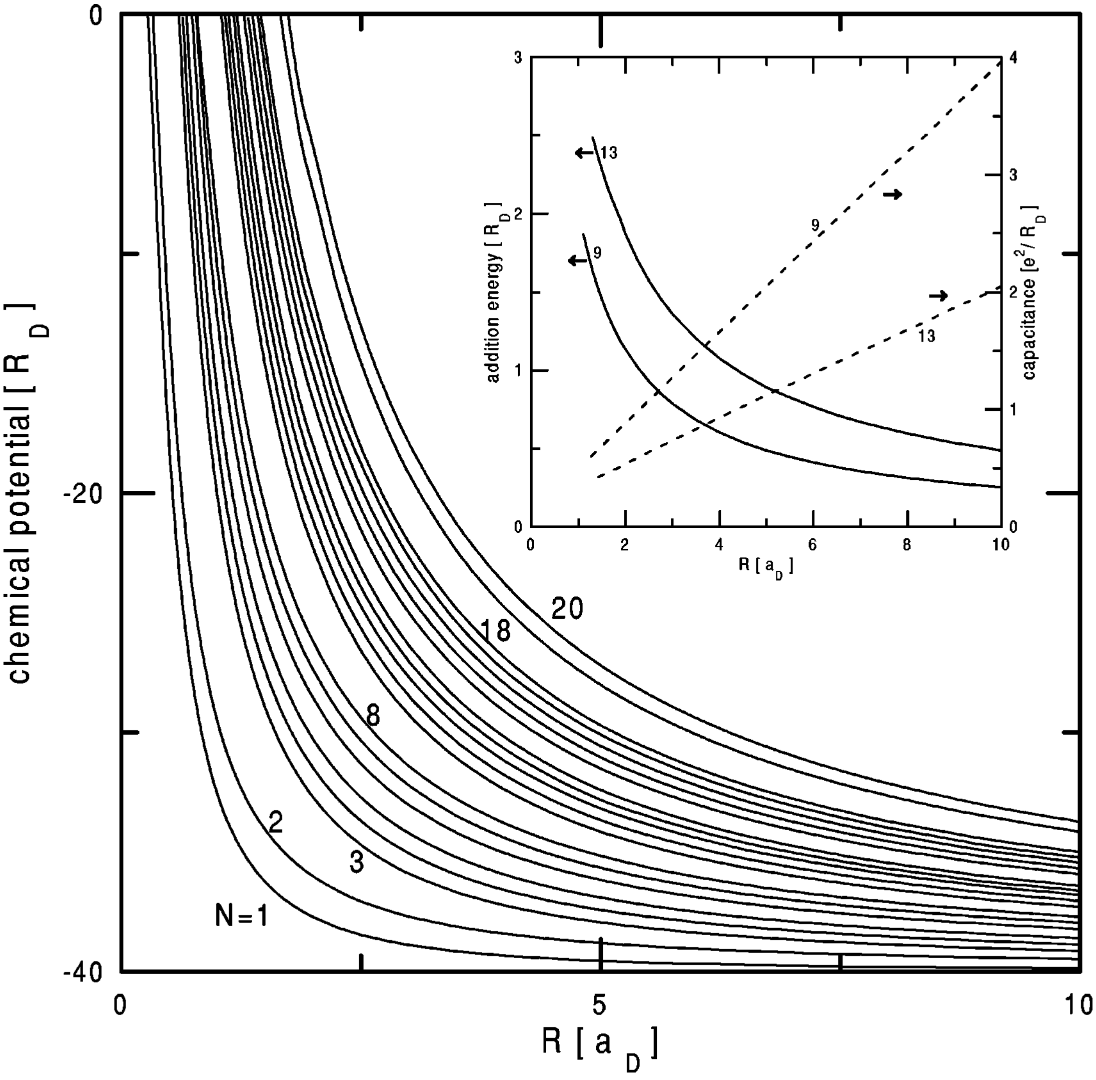

FIG. 5. Chemical potential of N-electron artificial atoms for $V_0 = 40R_D$ as a function of the quantum-dot radius R. Inset: Addition energy (solid curves) and electric capacitance (dashed curves) of quantum dots with $V_0 = 40R_D$ consisting of 9 and 13 electrons as functions of R.

sponding to the stable configuration. The N-electron system confined in the QD forms a bound state if the following condition of binding is fulfilled:

$$E_N < E_{N-1}, \tag{7}$$

where E_N and E_{N-1} are the ground-state energies of the artificial atoms consisting of N and $N-1$ electrons, respectively. For the assumed reference energy, the corresponding condition of binding of the one-electron state has the form: $E_1 < 0$. Applying these conditions of binding to the results of Fig. 2 we see that the artificial atoms with the increasing number of electrons become stable if the radius of the QD is sufficiently large, which is in agreement with the classical constant-interaction model. The effect of binding of N-electron artificial atoms is clearly visible in the left lower part of Fig. 2, that shows the number of electrons corresponding to the stable atomlike states. The calculations performed for several values of the potential-well depth V_0 lead to the conclusion that the critical value of the QD radius for the binding of the N-electron artificial atom decreases with increasing V_0. The present results allow us to determine the quantum "capacity" of the QD. This means that for the given values of the parameters of the QD (R and V_0) we can estimate the number of electrons, which can be added to the QD forming the bound state. The Hartree-Fock method provides the upper bounds to the ground-state energy; therefore, the estimated critical values of R and V_0 for the binding of N-electron artificial atoms are also the upper bounds on the quantum "capacity" of the QD. We have constructed a "phase diagram" (Fig. 3), which shows the regions of stability of N-electron artificial atoms on the (R, V_0) plane. The curves drawn in Fig. 3 correspond to the upper limits on the parameters of the QD, above which the system of N electrons confined in the QD forms a bound state. For example, the first region in Fig. 3 ($N=0$) corresponds to no electrons bound in the QD, the second ($N=1$) — one electron bound, etc.

The $N(R)$ dependence displayed in the left lower part of Fig. 2 exhibits a staircaselike structure with "stairs" of small and large "width" (measured as a corresponding change of the QD radius). The "stairs" of small "width" correspond to the filling by electrons of the states belonging to the same shell. For the spherical QD's, these are the shells $1s$, $1p$, $1d$, and $2s$ filled by 2, 6, 10, and 2 electrons, respectively. The ''stairs'' of large ''width'' correspond to the change of the shell, i.e., the formation of the next shell requires a larger increase of the QD radius. Moreover, the results of the present calculations for the half-filled $1p$ and $1d$ shells show that the Hund rule is fulfilled. The electrons with the same spin fill the spin orbitals of the $1p$ and $1d$ shells up to the half, which corresponds to the artificial atom with the maximum value of the total spin. In order to increase the number of electrons above 5 and 13 in the $1p$ and $1d$ shells, respectively, we have to put an additional electron with the opposite spin, which requires a slightly larger increase of the QD radius than for the states with the same spin. A close inspection of the staircaselike picture in Fig. 2 allows us to recognize this slight increase of the ''width'' of stairs. In summary, the results of Figs. 2 and 3 show how the ''periodic table'' is built from the artificial atoms in the case of the three-dimensional finite confinement potential.

We mention that the ''periodic table'' was proposed for classical charged particles[32] in the frame of the classical mechanics and Hund's rule was obtained by Koskinen *et al.*[28] and Steffens *et al.*[30] with the use of the density-functional theory. The authors[28,30,32] considered the two-dimensional QD's with the parabolic confinement.

The filling of the shells of artificial atoms by adding the electrons to the QD can be treated in the frame of classical electrostatics as charging the capacitor of the capacitance C. According to the classical electrostatics, the energy $\Delta E^{\mathrm{clas}} = e^2/C$ is required to add one electron to the QD. In order to obtain a deeper physical insight into the process of formation of artificial atoms, we have studied the corresponding classical problem of charging the sphere. Recently, several authors[37–39] considered the similar problem, which is equivalent to the classical Thomson model of atom. In the Thomson problem, the charge carriers are strictly constrained to the surface of the sphere of radius R. In the present work, we have considered the system of N charge carriers with equal charges that are subjected to an external field with the potential given by Eq. (1). This classical model provides an exact analog to the quantum-mechanical model presented in Sec. II. We were looking for the lowest potential energy of the system of N charge carriers using the Monte Carlo minimization technique. Figure 4 demonstrates that — according to the classical model — the energies required to add one electron to the QD are nearly the same; however, the small differences between these energies for different N are also visible. For the classical N-electron system confined in the three-dimensional finite potential — on the contrary to the two-dimensional case[32] — no shell-filling effect takes place.

The overall properties of the N-electron energies are similar in both the quantum-mechanical (Fig. 2) and classical (Fig. 4) description. The lowest N-electron potential energy calculated in the present work by the classical electrostatics approach can be written down in the form

$$E_N^{\mathrm{clas}} = -NV_0 + U_N/R, \tag{8}$$

where U_N is the parameter dependent on the number of electrons. The second term in Eq. (8) is the classical charging energy. We have checked that the N dependence of this term very well agrees with the fitted form.[39]

The quantum-mechanical problem of N electrons constrained to move on the surface of the sphere of constant radius was solved in the one-electron basis.[40] We note that the present model is more realistic: the electrons are allowed to be spread over the volume of the potential-well sphere and can penetrate the barrier region outside the sphere. For the electrons strictly confined to the surface of the sphere no clear shell-filling effect was obtained.[40]

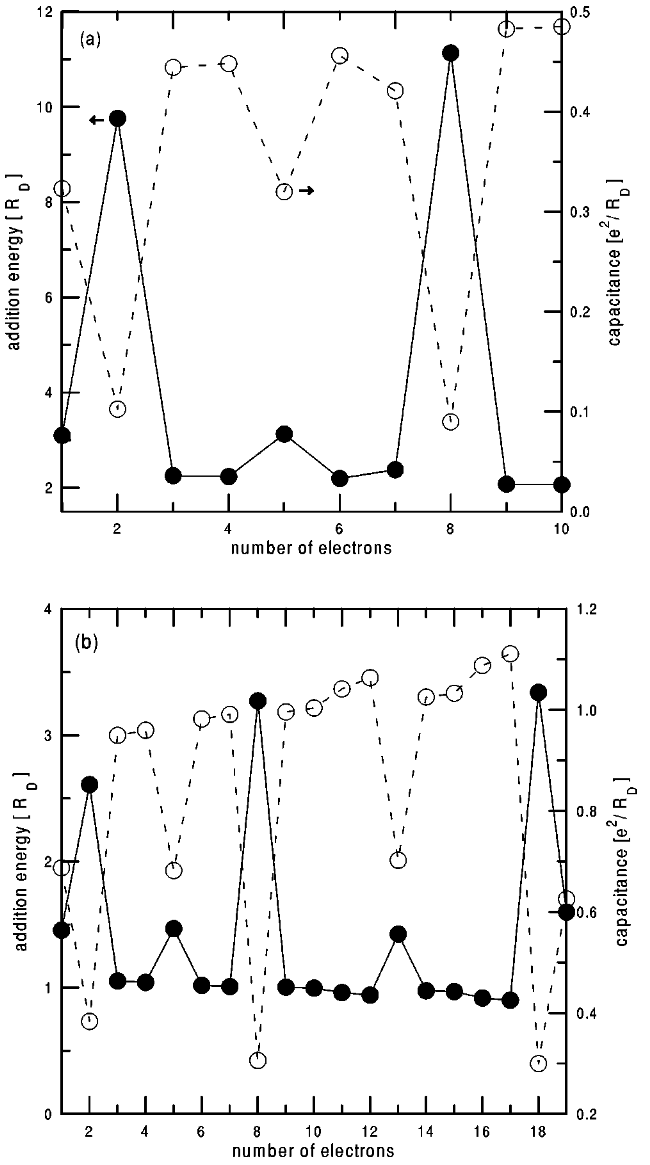

FIG. 6. Addition energy (solid circles) and electric capacitance (open circles) of the quantum dots with (a) $R = a_D$ and $V_0 = 50R_D$, and (b) $R = 2.5a_D$ and $V_0 = 100R_D$, as functions of the number of electrons. Thin lines are drawn as guides for the eye. In case (a), only 11 electrons are bound.

The characteristic behavior connected with the shell filling is also visible if we calculate the chemical potentials for the artificial atoms (Fig. 5). The chemical potential of N-electron system is defined as $\mu_N = E_N - E_{N-1}$ and interpreted as the energy needed to increase the number of electrons from $N-1$ to N. The N-electron artificial atom is stable if the corresponding chemical potential μ_N is negative, which means that the amount of energy $|\mu_N|$ is released when this artificial atom is created. The nearly equidistant curves in Fig. 5 show the filling of the empty one-electron states in the unfilled shell. Two types of de-

TABLE I. Quantum ''capacity'' of the quantum dots made of the material listed in the first column. The depth V_0 of the potential well (in eV) is listed in the second column and the next columns yield the values of QD radii (in nm), for which the dot is filled by the maximum number of electrons given in the first row. The highest occupied orbital is given in the parentheses.

Material	V_0	$1(1s^1)$	$2(1s^2)$	$5(1p^3)$	$8(1p^6)$	$13(1d^5)$	$18(1d^{10})$	$20(2s^2)$
Si/SiO_2	3.1	0.33	0.39	0.75	0.87	1.26	1.44	1.66
Porous-Si	1.7	0.47	0.56	1.14	1.36	2.2	2.43	2.86
$GaAs/Al_{0.2}Ga_{0.8}As$	0.21	2.75	3.30	6.80	8.20	12.7	14.6	17.3

viations from this behavior are visible in Fig. 5. The larger increase of the chemical potential corresponds to the complete filling of shells $1s$, $1p$, and $1d$ by 2, 6, and 10 electrons, respectively. The smaller increase of μ_N for $N=5$ and 13, i.e., for the half-filled $1p$ and $1d$ shells, is a signature of the Hund rule.

The addition energy ΔE and electric capacitance C of the QD are the quantities of experimental interest. We have calculated both these quantities as follows: $\Delta E=\mu_{N+1}-\mu_N$ and $C=e^2/\Delta E$. The inset of Fig. 5 reports the present results for ΔE and C as functions of the QD radius for $N=9$ and 13 electrons. Figures 6(a) and 6(b) show the dependence of ΔE and C on the number of electrons for the parameters corresponding to the QD's made of a porous Si [Fig. 6(a)] and Si/SiO_2 [Fig. 6(b)] nanostructure. Similarly as in Fig. 5, the high jumps of the addition energy are associated with the filling of the next shell if the previous shell is fully occupied and the lower jumps — the filling of the one-electron orbital with the opposite spin if the shell is half-filled by electrons of the same spin. For the parameters of Fig. 6(a), the maximum number of electrons that can be added to the QD is eleven. The inset of Fig. 5 demonstrates that the electric capacitance of the QD is a linear function of R and the addition energy varies like $1/R$. However, we note that although both these functions resemble those for the corresponding classical quantities, the coefficients of the proportionality depend on the number of electrons, which expresses the quantum nature of the addition energy and electric capacitance.

IV. CONCLUSIONS

The results of the present paper allow us to determine the important characteristics of the spherical QD's. If the parameters of the QD, i.e., potential-well depth V_0 and radius R, are given, we can determine the maximum number of electrons that can be added to the QD filling all the available quantum states. In such a way, we find the quantum ''capacity'' of the QD. On the other hand, if the number N of electrons is given, we can determine the parameters of that QD, in which the bound N-electron state can be created. Both these characteristics should be helpful in planning the experiments with the artificial atoms formed in QD's. The present results can be applied to the spherical QD's prepared by chemical methods[3–7] and to the nanocrystals formed in a porous Si.[41] We note that the confinement potential of the finite depth should apply to InAs/GaAs self-assembled QD's (Ref. 42) as well as to gate-controlled QD's.[43] However, in the last case the shape of the potential has to be modified.[12]

As an example of an application of the present theoretical treatment, we list in Table I the results, which can be helpful in experiments with spherical QD's made of Si nanocrystals in SiO_2 matrix and GaAs in $Al_{0.2}Ga_{0.8}As$ matrix. The atom-like states were found in resonant-tunneling measurements in spherical Si nanocrystals.[3] The $GaAs/Al_xGa_{1-x}As$ quantum dots usually possess the cylindrical shape.[9] The spherical QD's made of GaAs were prepared in an organic solvent.[4] It was argued[41] that the theoretical model using the effective-mass approximation and the finite confinement potential fairly well describes the electronic properties of a porous Si. Therefore, we quote in Table I the results for the QD's formed from the nanocrystals in the porous Si.

Let us briefly discuss the reliability of the present results when applied to the real QD's. In the present paper we have neglected the change of the electron band mass and dielectric constant at the QD boundary. The influence of spatially dependent electron band mass can be taken into account in the frame of the BenDaniel-Duke model.[44] Using this model, we have performed[35] the calculations for the one-electron states in spherical QD made of the $GaAs/Al_{0.2}Ga_{0.8}As$ nanostructure. The results[35] show that the effect of the spatially variable electron band mass can be neglected for $R>\sim a_D$, i.e., in the intermediate and weak-confinement regime, which is caused by the small penetration of electrons into the barrier region. In the present study, we have found that this penetration is also very small for many-electron systems in the QD's of intermediate and large size. Therefore, we expect that this effect will be negligibly small for the N-electron artificial atoms.

The change of the dielectric constant at the QD boundary gives rise to an induced electric polarization.[13,15] In the case of the Si/SiO_2 nanostructure, the nanocrystal with the larger dielectric constant (Si) is embedded in the dielectric medium (SiO_2) with the smaller dielectric constant. Then, the induced electric polarization repulses the electrons from the QD boundary and pushes them towards the dot center.[13,15] This leads to a larger localization of electrons inside the QD. If the dielectric constants of both the materials only slightly differ between themselves, which is the case of the $GaAs/Al_xGa_{1-x}As$ nanostructure with small content of Al, the effect of the induced electric polarization is negligible.

In summary, we have shown that the many-electron artificial atoms are stable if the range and strength of the confinement potential are sufficiently large. The results of the present paper allow us to determine the critical values of the parameters of the QD's, in which the artificial atoms consisting of up to 20 electrons are formed. The fundamental qualitative properties of the artificial atoms are the same as those of the natural atoms. For the spherical confinement potential, the atomic shells $1s$, $1p$, $1d$, and $2s$ are filled in order. The shell-filling effect and Hund rule are clearly expressed by the calculated chemical potential, addition energy, and electric capacitance of the QD's. In the classical limit, the artificial atoms exhibit the properties similar to those being characteristic for the Thomson model of atom.

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