Self-consistent calculation of discrete and continuous states in spherical semiconductor quantum dots

G. Todorović
Faculty of Civil Engineering, University of Belgrade, Bulevar Revolucije 73, 11000 Belgrade, Yugoslavia

V. Milanović, Z. Ikonić, and D. Indjin
Faculty of Electrical Engineering, University of Belgrade, Bulevar Revolucije 73, 11000 Belgrade, Yugoslavia

(Received 17 July 1996; revised manuscript received 30 December 1996)

A self-consistent procedure for calculating the energy structure, wave functions, and charge distribution in spherically symmetric semiconductor quantum dots is presented that takes account of both bound and free-electron states. The Schrödinger and Poisson equations are solved iteratively while using the Morse-type parametrized potential to keep the charge neutrality in each iterative step. Numerical calculations performed for a GaAs-$Al_{0.3}Ga_{0.7}As$ based quantum dot indicate that under realistic doping conditions bound states account for most of the charge accumulated in the dot. However, the self-consistent potential very significantly modifies the free-state wave functions and hence the bound-free transition matrix elements.
[S0163-1829(97)01523-3]

I. INTRODUCTION

Semiconductor quantum dot structures have attracted considerable research attention, both theoretical and experimental. Analytical models based on the multiband effective mass approximation, have been developed for spherically symmetric dots,[1] and self-consistent studies of the accumulated charge have been done.[2] Yet, the free part of energy spectrum in these structures was not very thoroughly studied. In analogy to the more conventional quantum well structures one may expect that free-electronic states in the dot may also become very important in some phenomena or applications of quantum dots.[3] Here we present a self-consistent procedure for calculating the energy structure and charge distribution in quantum dots, taking account of both the bound and free states. Also, we explore the influence of the self-consistency on intraband bound-bound and bound-free optical transitions.

II. THEORETICAL CONSIDERATIONS

A. The effective mass Schrödinger equation for a quantum dot

Electronic states close to the conduction band extremum can be described by a single-electron one-band effective mass equation:

$$[E(-i\nabla)+U(\vec{r})]F(\vec{r})=EF(\vec{r}), \tag{1}$$

where $F(\vec{r})$ denotes the envelope function. In the case of GaAs and similar materials with the isotropic electron effective mass, Eq. (1) applied to a spherically symmetric quantum dot takes the form

$$\left[-\frac{\hbar^2}{2}\nabla\left(\frac{1}{m^*(r)}\nabla\right)+U(r)\right]R_e(r)Y_{\ell m}(\theta,\phi) = ER_e(r)Y_{\ell m}(\theta,\phi), \tag{2}$$

where $R(r)$ is the radial part of the envelope function, and $m^*(r)$ the radially dependent electron effective mass. In the structure considered it is given by

$$m^*(r)=\begin{cases} m_w, & r<R_0 \\ m_b, & r\geqslant R_0, \end{cases} \tag{3}$$

where R_0 is the dot radius and $m_w(m_b)$ are the effective masses in GaAs ($Al_xGa_{1-x}As$), i.e., the well and bulk materials. Introducing the new function $\phi_\ell(k,r)$ with $R_e(r)=r\phi_\ell(k,r)$, as usual in such problems, we recast Eq. (2) into

$$\frac{1}{r^2}\frac{d}{dr}\left[r^2\frac{1}{m^*(r)}\frac{d}{dr}\left(\frac{\phi_\ell(k,r)}{r}\right)\right]-\frac{\ell(\ell+1)}{r^3}\frac{\phi_\ell(k,r)}{m^*(r)} + \frac{2}{\hbar^2}[E-U(r)]\frac{\phi_\ell(k,r)}{r}=0 \tag{4}$$

with ℓ denoting the orbital quantum number, $U(r)$ the potential energy, and the constant k is related to energy, $E=(\hbar^2k^2/2m_b)$. The boundary conditions for the function $\phi_\ell(k,r)$, in the center of the structure is[4]

$$\lim_{r\to 0}(2\ell+1)!!\,r^{-\ell-1}\phi_\ell(k,r)=1. \tag{5}$$

The asymptotic form of the solution of Eq. (4) for large values of radii may be obtained by first writing its general solution as a linear combination of counterpropagating plane waves $f_\ell(\pm k,r)$ that satisfy the asymptotic boundary conditions[5]

$$\lim_{r\to\infty}e^{\pm ikr}f_\ell(\pm k,r)=i^\ell, \tag{6}$$

in terms of which the solution of Eq. (4) reads

$$\phi_\ell(k,r)=C_1(k)f_\ell(k,r)+C_2(k)f_\ell(-k,r), \tag{7}$$

with

$$C_1(k)=\frac{i}{2k^{\ell+1}}\left(\frac{m_b}{m_w}\right)^{[(\ell+2)/2]} f_\ell(-k), \tag{8}$$

$$C_2(k)=-\frac{(-1)^\ell i}{2k^{\ell+1}}\left(\frac{m_b}{m_w}\right)^{-[(\ell+2)/2]} f_\ell(k), \tag{9}$$

and $f_\ell(\pm k)$ denotes the Jost function defined as[5]

$$f_\ell(\pm k)=\lim_{r\to 0}\frac{(\pm kr)^\ell f_\ell(\pm k,r)}{(2\ell-1)!!}. \tag{10}$$

The solution of Eq. (4) may thus be written as

$$\phi_\ell(k,r)=\frac{i}{2k^{\ell+1}\left(\frac{m_w}{m_b}\right)^{[(\ell+2)/2]}}[f_\ell(-k)f_\ell(k,r) -(-1)^\ell f_\ell(k)f_\ell(-k,r)]. \tag{11}$$

By comparing the solution of Eq. (4) for the constant effective mass case[6] in the limit $r\to\infty$,

$$\phi_\ell(k,r\to\infty)\to\frac{i^{\ell+1}e^{-i\delta_\ell(k)}}{2}[e^{-ikr}-(-1)^\ell e^{2i\delta_\ell(k)}e^{ikr}], \tag{12}$$

and Eq. (11) in the same limit

$$\phi_\ell(k,r\to\infty)=\frac{i^{\ell+1}}{2k^{\ell+1}\left(\frac{m_w}{m_b}\right)^{[(\ell+2)/2]}}[f_\ell(-k)e^{-ikr} -(-1)^\ell f_\ell(k)e^{ikr}], \tag{13}$$

we find that the ratio of Jost functions remains unchanged, i.e.,[5]

$$\frac{f_\ell(k)}{f_\ell(-k)}=e^{i2\delta_\ell(k)}, \tag{14}$$

so the asymptotic solution with the position-dependent mass (3) is of the same form as that with constant mass,

$$\phi_\ell(k,r\to\infty)\propto C(k)\sin\left[kr-\frac{\ell\pi}{2}+\delta_\ell(k)\right]. \tag{15}$$

The position-dependent effective mass is, however, reflected in the values of the phase-shift function.

B. Bound electron states

Value(s) k_0 in the lower half of the complex k plane in which the Jost function became zero correspond to bound states of the system.[6] From Eq. (11), then, it follows that the corresponding eigenfuntions asymptotically behave as

$$\phi_\ell(k_0,r\to\infty)=\frac{i^{\ell+1}}{2k^{\ell+1}\left(\frac{m_w}{m_b}\right)^{[(\ell+2)/2]}}f_\ell(-k_0)e^{-ik_0r} \propto e^{-|k_0|r}. \tag{16}$$

As suggested by Eq. (16) we substitute $\phi_\ell(k,r)=F_\ell(k,r)e^{-kr}$ in Eq. (4), and the differential equation for $F_\ell(k,r)$ inside the dot reads

$$\frac{d^2F_{1\ell}}{dr^2}-2k\frac{dF_{1\ell}}{dr}+\left[\left(1-\frac{m_w}{m_b}\right)k^2-\left(\frac{2m_w}{\hbar^2}U(r) +\frac{\ell(\ell+1)}{r^2}\right)\right]F_{1\ell}=0 \tag{17}$$

and in the bulk ($r>R_0$) it has the form

$$\frac{d^2F_{2\ell}}{dr^2}-2k\frac{dF_{2\ell}}{dr}-\left(\frac{2m_b}{\hbar^2}U(r)+\frac{\ell(\ell+1)}{r^2}\right)F_{2\ell}=0, \tag{18}$$

with the bound state energy $E=-(\hbar^2k^2/2m_b)$. The boundary conditions to be imposed to $F_{1\ell}(k,r)$ and $F_{2\ell}(k,r)$ follow from Eqs. (5) and (16) and from the continuity of radial function derivative at the heterointerface

$$\frac{1}{m_w}\frac{dR_{1\ell}}{dr}\bigg|_{r=R_{0-}}=\frac{1}{m_b}\frac{dR_{2\ell}}{dr}\bigg|_{r=R_{0+}}. \tag{19}$$

These read

$$F_{1\ell}(k,r=0)=0,\quad F_{1\ell}(k,r=R_{0-})=F_{2\ell}(k,r=R_{0+}),$$

$$F_{2\ell}(k,r\to\infty)=1,\quad \frac{dF_{2\ell}}{dr}\bigg|_{r\to\infty}=0 \tag{20}$$

and

$$\frac{dF_{1\ell}(k,r=R_{0-})}{dr}=\frac{m_w}{m_b}\frac{dF_{2\ell}(k,r=R_{0+})}{dr}+\left(1-\frac{m_w}{m_b}\right) \times\left(k+\frac{1}{R_0}\right)F_{2\ell}(k,r=R_{0+}). \tag{21}$$

The normalization constants $C_\ell(k)$ of the eigenfunctions $\phi_\ell(k,r)$ are to be determined from the unity-norm condition.

C. Free-electron states

The free- (continuous) electron spectrum is characterized by positive energies $E=(\hbar^2k^2/2m_b)$, measured from the conduction band edge deep in the bulk. From the asymptotic expression (15) it follows that $\phi_\ell(k,r)$ for large values of r can be written as

$$\phi_\ell(k,r)=\mathrm{Im}\{e^{i[kr-(\ell\pi/2)+\delta_\ell(k)]}F_\ell(k,r)\}. \tag{22}$$

Substituting it in Eq. (4) we find that for $r>R_0$,

$$\frac{d^2F_{2\ell}}{dr^2}+2ik\frac{dF_{2\ell}}{dr}-\left(\frac{2m_b}{\hbar^2}U(r)+\frac{\ell(\ell+1)}{r^2}\right)F_{2\ell}=0, \tag{23}$$

and for $r<R_0$,

$$\frac{d^2F_{1\ell}}{dr^2}+2ik\frac{dF_{1\ell}}{dr}+\left[\left(\frac{m_w}{m_b}-1\right)k^2-\left(\frac{2m_w}{\hbar^2}U(r)+\frac{\ell(\ell+1)}{r^2}\right)\right]F_{1\ell}=0. \tag{24}$$

The boundary condition for $F_{1\ell}$ and $F_{2\ell}$ is found using similar arguments as in the case of bound states, and reads

$$F_{2\ell}(k,r\to\infty)=1,\left.\frac{dF_{2\ell}}{dr}\right|_{r\to\infty}=0,$$

$$F_{1\ell}(k,r=R_{0-})=F_{2\ell}(k,r=R_{0+}), \tag{25}$$

and

$$\frac{dF_{1\ell}(k,r=R_{0-})}{dr}=\frac{m_w}{m_b}\frac{dF_{2\ell}(k,r=R_{0+})}{dr}+\left(\frac{m_w}{m_b}-1\right)\times\left(ik-\frac{1}{R_0}\right)F_{2\ell}(k,r=R_{0+}). \tag{26}$$

To complete the construction of radial eigenfunctions one should find the phase shifts $\delta_\ell(k)$. Using Eq. (4) we find

$$\mathrm{Im}\{e^{i[kr-(\ell\pi/2)+\delta_\ell(k)]}F_{1\ell}(k,r=0)\}=0, \tag{27}$$

wherefrom

$$\tan[\delta_\ell(k)]=-\frac{\mathrm{Im}\{F_{1\ell}(k,r=0)\}}{\mathrm{Re}\{F_{1\ell}(k,r=0)\}}. \tag{28}$$

D. Electron density

The normalized bound state wave functions, upon taking the modulus squared, and multiplying by the Fermi-Dirac distribution function $f_{\mathrm{FD}}E_i=[e^{[(E_i-E_F)/k_BT]}+1]^{-1}$ are directly applicable for calculating the electron density on bound states, via

$$n_b(r)=\frac{1}{2\pi}\sum_{\ell=0}^{\ell_{\max}}(2\ell+1)\sum_{i=1}^{N_b(\ell)}|R_{i,\ell}(r)|^2f_{\mathrm{FD}}(E_{i,l}), \tag{29}$$

where $N_b(\ell)$ denotes the number of bound states for any particular value of ℓ and the factor $2(2\ell+1)$ accounts for the spin and magnetic quantum number degeneracy, and the functions are taken to be normalized as $\int_0^\infty R_\ell^2r^2dr=1$.

The expression for free-electron density, analogous to Eq. (29), depends on the type of free wave function normalization. Here we use the ''box'' normalization, i.e., take the structure to be embedded in a spherical box (infinite potential), its radius R_{inf} being very large. The continuous spectrum in such a case becomes quasicontinuous, i.e., formally very dense discrete. With the effective mass dependence given by Eq. (3) the unnormalized free spectrum wave functions $\phi_\ell(k,r)$ satisfy[7]

$$2k\int_0^{R_{\mathrm{inf}}}\phi_\ell^2dr=\left[\frac{\partial\phi_\ell}{\partial k}\frac{\partial\phi_\ell}{\partial r}-\phi_\ell\frac{\partial^2\phi_\ell}{\partial r\,\partial k}\right]_{R_{\mathrm{inf}}}. \tag{30}$$

From Eq. (15) $\phi_\ell(k,R_{\mathrm{inf}}\to\infty)\propto\sin[kR_{\mathrm{inf}}-(\ell\pi/2)+\delta_\ell(k)]$ and it follows from the above expression that

$$\int_0^{R_{\mathrm{inf}}\to\infty}\phi_\ell^2dr=\frac{1}{2k}\left[k\left(R_{\mathrm{inf}}+\frac{\partial\delta_\ell}{\partial\kappa}\right)-\frac{1}{2}\sin2\left(kR_{\mathrm{inf}}-\frac{\ell\pi}{2}+\delta_\ell(k)\right)\right]_{R_{\mathrm{inf}}\to\infty}, \tag{31}$$

i.e.,

$$\int_0^{R_{\mathrm{inf}}\to\infty}\phi_\ell^2dr=\frac{C_\ell^2(k)}{2}\left[R_{\mathrm{inf}}+\frac{\partial\delta_\ell}{\partial\kappa}\right]_{R_{\mathrm{inf}}\to\infty}, \tag{32}$$

because $\phi_\ell(k,R_{\mathrm{inf}})=0$. For the quasicontinuous spectrum we have[8]

$$\frac{dk}{\pi}=\frac{1}{R_{\mathrm{inf}}+\dfrac{\partial\delta_\ell(k)}{\partial k}},\quad C_\ell^2(k)=\frac{2\,dk}{\pi} \tag{33}$$

and, using Eq. (29), the free-electron density is given by

$$n_c(r)=\frac{1}{\pi^2}\sum_{\ell=0}^{\ell_{\max}}(2\ell+1)\int_0^\infty|R_{\ell k}^{(n)}(r)|^2f_{\mathrm{FD}}\left(\frac{\hbar^2k^2}{2m_b}\right)dk. \tag{34}$$

The bulk is taken to be uniformly doped with donors, and degree of their ionization being

$$n_d(r)=\frac{N_d}{1+2e^{\{[E_{Fb}-E_{Db}-U(r)]/k_BT\}}}, \tag{35}$$

where N_d, E_{Fb}, and E_{Db} denote the donors density, bulk Fermi level, and donor ionization energy. Equation (35) accounts for the influence of the local potential $U(r)$ on the degree of ionization.

E. Poisson equation

The electrostatic potential is found by solving the Poisson equation,

$$\nabla[\epsilon(r)\nabla\varphi(r)]=-\rho, \tag{36}$$

where ρ denotes the charge density and $\varepsilon(r)$ the dielectric permittivity which is also position dependent: $\varepsilon(r)=\varepsilon_1$ for $r<R_0$ and $\varepsilon(r)=\varepsilon_2$ for $r>R_0$. Due to the spherical symmetry of structure Eq. (36) thus becomes

$$\frac{1}{r^2}\frac{d}{dr}\left[\epsilon(r)r^2\frac{d\varphi}{dr}\right]=-\rho, \tag{37}$$

with $\varphi(r)$ satisfying the boundary conditions $\varphi(0)=0,d\varphi/dr|_{r=0}=0$, the first of which is simply the choice of reference. Integrating (37) then delivers

$$\varphi(r)=\frac{1}{r}\int_0^r\frac{v(v-r)\rho(v)}{\varepsilon(v)}dv. \tag{38}$$

Since $\rho(r)=-e[n_b(r)+n_c(r)-n_d(r)]$, the ionized donors density is

$$n_d(r)=\begin{cases}0, & r<R_0\\ \dfrac{N_d}{1+2e^{[E_{Fb}-E_D(r)]/k_BT}}, & r>R_0\end{cases}. \tag{39}$$

The potential inside the dot ($r<R_0$) is

$$\varphi(r)=\frac{e}{\varepsilon_1}\left[\int_0^r v[n_b(v)+n_c(v)]dv - \frac{1}{r}\int_0^r v^2[n_b(v)+n_c(v)]dv\right] \tag{40}$$

and outside ($r>R_0$)

$$\varphi(r)=\frac{e}{\varepsilon_1}\left[\int_0^{R_0} v[n_b(v)+n_c(v)]dv-\frac{1}{r}\int_0^{R_0} v^2[n_b(v)+n_c(v)]dv\right]+\frac{e}{\varepsilon_2}\left[\int_{R_0}^r v[n_b(v)+n_c(v)-n_d(v)]dv - \frac{1}{r}\int_{R_0}^r v^2[n_b(v)+n_c(v)-n_d(v)]dv\right] \tag{41}$$

and the potential energy is $U(r)=-e\varphi(r)-\Delta E_c\cdot\theta(R_0-r)$.

F. Charge neutrality of the structure

The global charge neutrality of the structure implies that

$$\int_0^\infty [n_b(r)+n_c(r)-n_d(r)]r^2dr=0. \tag{42}$$

Substituting the expressions for charge densities in Eq. (42) and using some analytical properties of free-state wave functions (see the Appendix), we find

$$\frac{1}{\pi^2}\sum_{\ell=0}^{\infty}(2\ell+1)\left[\frac{\hbar^2}{4m_bk_BT}\int_0^\infty\frac{\delta_\ell(k)kdk}{\cosh^2\left(\dfrac{\hbar^2k^2-2m_bE_{Fb}}{2m_Bk_BT}\right)} - -\frac{\delta_\ell(0)}{1+e^{-E_{Fb}/k_BT}}\right]+\frac{1}{2\pi}\sum_{\ell=0}^{\ell_{\max}}(2\ell+1)\sum_{i=1}^{N_b(\ell)}f_{\mathrm{FD}}(E_i) - \int_0^{R\to\infty}[n_{cb}-n_d(r)]r^2dr=0, \tag{43}$$

where n_{cb} in the last integral denotes the electron density in the bulk.

G. The parametric self-consistent procedure

Within the self-consistent procedure the Schrödinger and Poisson equations are solved iteratively. Somewhat different from the conventional self-consistent procedure that deals only with bound states, we have employed the parametrization of Hartree potential[8,9] within each iterative step, which enables one to enforce the charge neutrality condition and contributes to the numerical stability of the procedure. In effect, inside the dot the potential is obtained by direct numerical integration of the Schrödinger equation, and in the bulk region the potential is written as a Morse-type function with three parameters

$$\varphi(r-R_0)=\frac{\varphi_0}{1-\beta}[e^{-\lambda(r-R_0)}-\beta e^{-2\lambda(r-R_0)}], \tag{44}$$

as is customarily done in such a case.[8,9] Here φ_0 represents the potential of the heterointerface ($r=R_0$), while λ and β are related through the conservation of dielectric displacement at the interface:

$$\beta=\frac{\varepsilon_2\lambda\varphi_0+\varepsilon_1\left.\dfrac{d\varphi}{dr}\right|_{r=R_{0-}}}{2\varepsilon_2\lambda\varphi_0+\varepsilon_1\left.\dfrac{d\varphi}{dr}\right|_{r=R_{0-}}}. \tag{45}$$

Therefore, a single free parameter (λ) remains to fit the Morse potential in order to get the charge neutrality, Eq. (43). As usual, a suitably chosen convergence factor f was introduced to make the procedure stable,[10] i.e., the input potential in the next iteration was taken as weighted average of the input and output potentials of the previous iteration,

$$U_{\mathrm{in}}^{(n+1)}=U_{\mathrm{in}}^{(n)}+f(U_{\mathrm{out}}^{(n)}-U_{\mathrm{in}}^{(n)}). \tag{46}$$

The value of $f=0.7$ was taken in our calculations.

H. Intraband transition matrix elements

Having found the wave functions of bound and free states in a quantum dot, it is straightforward to calculate the matrix elements of bound-bound and bound-free optical transitions. Given the spherical symmetry of the system these are polarization insensitive. For bound-free transitions (quantum dot ''ionization'') the dipole matrix element[11] is given by

$$M(n\ell,E\ell')=\int_0^\infty R_{n,\ell}r^3R_{E,\ell'}dr, \tag{47}$$

where n denotes the principal quantum number of bound, and E the energy of the free state. The same expression, upon substitution $E\to n'$, holds for bound-bound transitions. In either case the orbital quantum numbers ℓ and ℓ' satisfy the selection rule $\ell-\ell'=\pm1$.

III. NUMERICAL RESULTS AND DISCUSSION

Numerical calculations were performed for a GaAs dot with the radius $R_0=10$ nm embedded in $Al_{0.3}Ga_{0.7}As$ bulk. Based on data in Ref. 12 the following parameters were used in calculation: the effective masses in the dot and bulk materials $m_w=0.067$ and $m_b=0.092$ (in free-electron mass units), and the dielectric permittivities $\varepsilon_1=13.18$ and $\varepsilon_2=12.24$, respectively, the donor binding energy $E_{Db}=8.21$ meV and conduction band offset $\Delta E_c=227.9$ meV. The Fermi level for a given donor doping level was calculated from Eqs. (A3) and (35). All the calculations were done for the temperature $T=300$ K, and the donor density in the bulk was taken in the range $10^{14}-10^{15}$ cm^{-3}, i.e., not far from unintentional doping levels in these materials, the

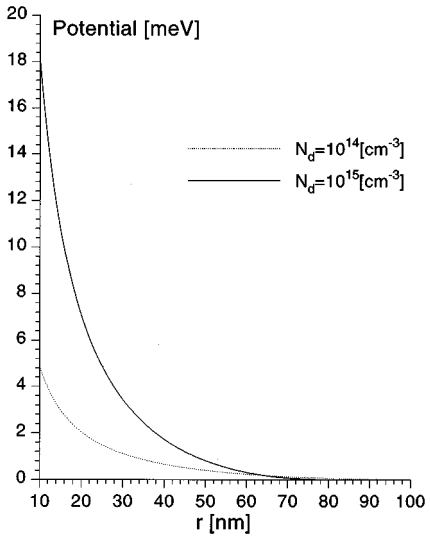

FIG. 1. Self-consistent Hartree potential in the bulk region of $R=10$ nm GaAs quantum dot structure, at two different donor doping levels of the bulk.

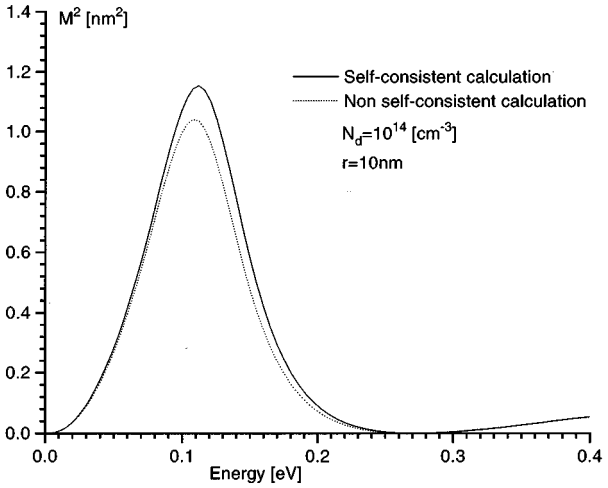

FIG. 2. Dipole matrix element squared for the bound-free transition $(n=0,\ell=1)\rightarrow(\text{continuum},\ell=0)$ in $R=10$ nm GaAs/$Al_{0.3}Ga_{0.7}As$ quantum dot, calculated with or without the self-consistent effects, with the donor doping level $N_d=10^{14}$ cm^{-3}.

Fermi level being -0.229 eV and -0.169 eV in two limit cases, measured from the conduction band edge deep inside the bulk.

To find the energy spectrum Eqs. (17)–(18) for discrete and Eqs. (23)–(24) for free states were numerically integrated (from the origin to $r=90$ nm), and the charge density was calculated according to Eqs. (29) and (34). The numerical procedure employed was found to be very stable and of a high accuracy.[13] Within the continuous spectrum the energy range included in calculations extended to $E_{\max}=130$ meV above the bulk conduction band edge. This was sufficient to cover all significantly populated states while keeping the nonparabolicity low (the largest k vector were $<1/20$ of the distance to the Brillouin zone boundaries).

There are five bound states in this dot. The non-self-consistent calculation gives their energies (the notation is $E_{n,\ell}$) as $E_{0,0}=-0.189$ eV, $E_{1,0}=-0.080$ eV, $E_{0,1}=-0.148$ eV, $E_{0,2}=-0.098$ eV, and $E_{0,3}=-0.040$ eV. The corresponding values obtained by the self-consistent calculation are -0.181 eV, -0.078 eV, -0.142 eV, $-.092$ eV, and -0.034 eV at $N_d=10^{14}$ cm^{-3} and -0.162 eV, -0.052 eV, -0.124 eV, -0.075 eV, and -0.018 eV at $N_d=10^{15}$ cm^{-3}. Effects of self-consistency are thus quite significant. However, they are here due mostly to bound, not free, states space charge.

In the above range of doping densities the Fermi level is well below the conduction band edge, i.e., the population of free states is quite low. The charge density in the dot region almost entirely originates from bound states, and the same holds true for the Hartree potential. It is only at very much larger doping densities, of the order of 10^{17} cm^{-3} or more, that free-state space charge and Hartree potential would become non-negligable in respect to bound states contributions. However, quantum dot structures with such doping do not seem to be of interest at present. The calculated self-consistent Hartree potential for two values of doping, is given in Fig. 1, indicating the increasing importance of the self-consistent calculation as the doping increases. The depletion region extends from the dot boundary to deep inside the bulk.

While the free-state space charge may not have large effects on the electronic structure, except for very large doping levels, the wave functions of (mostly unpopulated) free states may be substantially affected by the self-consistent Hartree potential of accumulated bound states charge. This is reflected in the values of bound-free intraband optical transition matrix elements, describing the absorption related to quantum dot ''ionization,'' to use the analogy with atomic physics. Such transitions have been considered in Ref. 14, with the space charge effects neglected. In Figs. 2 and 3 we give the dipole matrix elements squared vs free-state energy depedence for bound-free $(n=0,\ell=1)\rightarrow(E,\ell'=0)$ transition, calculated via Eq. (47) self-consistently or non-self-consistently. The accumulated space charge tends to increase the matrix elements squared, taken at the peak absorption energy, by a modest 11% at the doping density of 10^{14} cm^{-3}, or a very significant 65% at 10^{15} cm^{-3}. It is mostly the sensitivity of wave functions in continuum which brings about the importance of self-consistency. The bound-bound

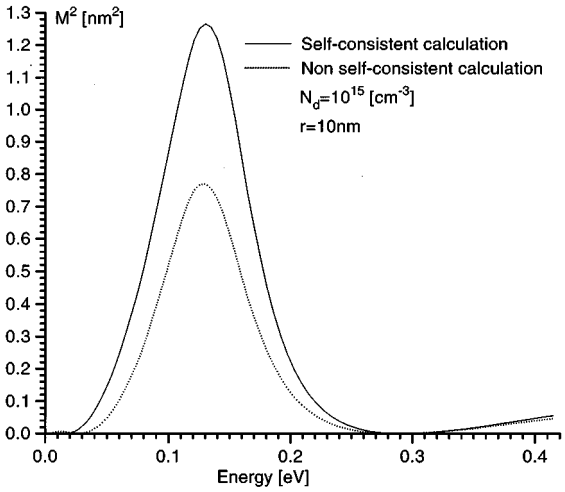

FIG. 3. Same as in Fig. 2, but for $N_d=10^{15}$ cm^{-3}.

transition matrix elements squared, calculated via the two approaches at 10^{15} cm^{-3}, differ by less than 5%. This is because bound states wave functions are essentially determined by the built-in confining potential and are not very sensitive to the details of mild self-consistent corrections, Fig. 1. On the other hand, free-state wave functions come out to be sensitive, apparently because the Hartree potential has a rather long range, just like the character of the wave functions themselves.

IV. CONCLUSION

The self-consistent procedure for calculating the electronic structure and charge density in semiconductor quantum dots, taking into account both the discrete and continuous parts of energy spectrum is presented. Numerical calculations performed for a GaAs-$Al_{0.3}Ga_{0.7}As$ based quantum dot show that only bound states significantly contribute to the total charge in the dot region, unless the doping level of bulk is quite large ($>10^{17}$ cm^{-3}). However, the free-state wave functions are considerably affected by the Hartree potential of the accumulated charge. This shows in the values of bound-free optical transition matrix elements, calculated self-consistently or non-self-consistently, the difference between the two approaches becomming prominent at doping levels $10^{14}-10^{15}$ cm^{-3}. Calculating the bound-free intraband absorption in most structures is thus likely to require the self-consistant procedure.

APPENDIX

In a homogeneous, uniformly doped bulk semiconductor the free-electron density is given by

$$n_{cb}(r)=\frac{1}{\pi^2}\sum_{\ell=0}^{\infty}(2\ell+1)\int_0^{\infty}j_\ell^2(kr)f_{\rm FD}(k)k^2dk, \quad \text{(A1)}$$

where $j_\ell(kr)$ are the spherical Bessel functions, i.e., the regular solution of Eq. (4). Using the property of spherical Bessel functions[15]

$$\sum_{\ell=0}^{\infty}(2\ell+1)j_\ell^2(kr)=1, \quad \text{(A2)}$$

we find from (A1) that the electron density in bulk is

$$n_{cb}=\frac{1}{\pi^2}\int_0^{\infty}f_{\rm FD}(k)k^2dk. \quad \text{(A3)}$$

Using Eq. (A3) in Eq. (34) we get

$$n_c(r)=n_{cb}+\frac{1}{\pi^2}\int_0^{\infty}\{[R_\ell^{(n)}(r)]^2-j_\ell^2(kr)\}f_{\rm FD}(k)k^2dk. \quad \text{(A4)}$$

The second term in (A4) is the ''excess'' density that appears due to the heterojunction. In Eq. (32) for the free-electron density it is convenient to use $R_\ell^*(r)=(1/k)R_\ell^{(n)}(r)$ instead of the unnormalized radial function $R_\ell^{(n)}(r)$. Prior to substituting (A4) into (42) the integrals $\int_0^{R\to\infty}(R_\ell^*)^2r^2dr$ and $\int_0^{R\to\infty}j_\ell^2(kr)r^2dr$ are evaluated, using Eq. (32)

$$\int_0^{R\to\infty}(R_\ell^*)^2r^2dr=\frac{1}{2k^3}\left[k\left(R+\frac{d\delta_\ell(k)}{dk}\right)-\frac{1}{2}\sin 2\left(kR-\frac{\ell\pi}{2}+\delta_\ell(k)\right)\right], \quad \text{(A5)}$$

$$\int_0^{R\to\infty}j_\ell^2(kr)r^2dr=\frac{1}{2k^3}\left[kR-\frac{1}{2}\sin 2\left(kR-\frac{\ell\pi}{2}\right)\right], \quad \text{(A6)}$$

wherefrom

$$\int_0^{R\to\infty}[(R_\ell^*)^2-j_\ell^2(kr)]r^2dr=\frac{1}{2k^2}\frac{d\delta_\ell(k)}{dk}+\frac{1}{4k^3}\left[\sin 2\left(kR-\frac{\ell\pi}{2}\right)-\sin 2\left(kR-\frac{\ell\pi}{2}+\delta_\ell(k)\right)\right]. \quad \text{(A7)}$$

and, following the same lines as described in Ref. 8, we find

$$\int_0^{R\to\infty}n_c(r)r^2dr=\int_0^{R\to\infty}n_{cb}r^2dr+\frac{1}{\pi^2}\sum_{\ell=0}^{\infty}(2\ell+1)\int_0^{\infty}f_{\rm FD}\frac{\partial\delta_\ell(k)}{\partial k}dk. \quad \text{(A8)}$$

From Eq. (41) the bound electron density may be written as

$$\int_0^{R\to\infty}n_b(r)r^2dr=\frac{1}{2\pi}\sum_{\ell=0}^{\ell_{\max}}(2\ell+1)\sum_{i=1}^{N_b(\ell)}f_{\rm FD}(E_i)\int_0^{R\to\infty}|R_{i,\ell}(r)|^2r^2dr=\frac{1}{2\pi}\sum_{\ell=0}^{\ell_{\max}}(2\ell+1)\sum_{i=1}^{N_b(\ell)}f_{\rm FD}(E_i). \quad \text{(A9)}$$

Substituting Eqs. (A8) and (A9) into Eq. (42) the charge neutrality equation (43) is derived.

[1] P. C. Sersel and K. J. Vahala, Phys. Rev. B **42**, 2690 (1990).

[2] A. Kumar, S. Laux, and F. Stern, Phys. Rev. B **42**, 5166 (1990).

[3] M. Tadić and Z. Ikonić, Phys. Rev. B, **52**, 8266 (1995).

[4] R. Newton, J. Math. Phys. **1**, 319 (1960).

[5] M. Golberger and K. Watson, *Collision Theory* (Wiley, New York, 1964).

[6] C. Joachain, *Quantum Collision Theory* (North-Holland, Oxford, 1975).

[7] J. Friedel, Philos. Mag. **43**, 153 (1952).

[8] G. Baraff and J. Appelbaum, Phys. Rev. B **5**, 475 (1972).
[9] D. Ehlers and D. Mills, Phys. Rev. B **34**, 3939 (1986).
[10] F. Stern, J. Comput. Phys. **6**, 56 (1970).
[11] V. A. Fock, *Fundamentals of Quantum Mechanics* (Mir, Moscow, 1982).
[12] S. Adachi, J. Appl. Phys. **58,** 1 (1985).
[13] D. Indjin, G. Todorović, V. Milanović, and Z. Ikonić, Comput. Phys. Commun. **90**, 87 (1995).
[14] R. Buczko and F. Bassani, Phys. Rev. B **54**, 2667 (1996).
[15] *Handbook of Math Functions*, edited by M. Abramowitz and I. Stegun (Dover, New York, 1972).