Hydrogenic impurities in GaAs-(Ga,Al)As quantum dots

N. Porras-Montenegro and S. T. Pérez-Merchancano
Departamento de Física, Universidad del Valle, A. A. 25360, Cali, Colombia
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The ground-state energy and the binding energy of shallow hydrogenic impurities in spherical GaAs-(Ga,Al)As quantum dots have been calculated as functions of the radius of the dot. The binding energy has been calculated following a variational procedure within the effective-mass approximation. We have used a finite confining potential well with depth determined by the discontinuity of the band gap in the quantum dot and the cladding. Calculations were also performed for an infinite confining potential. For the infinite potential well we found that the impurity binding energy increases as the dot radius decreases whereas in the finite potential-well situation, the binding energy reaches a peak value as the dot radius decreases and then diminishes to a limiting value corresponding to the radius for which there are no bound states in the well. We found that the strong electronic confinement in these quantum dots reflects itself in the ground-state energy and in the impurity binding energies, which are higher than those found in GaAs-(Ga,Al)As quantum wells and quantum-well wires.

The development of experimental techniques such as chemical vapor deposition, liquid-phase epitaxy, and molecular-beam epitaxy have led to the fabrication of many quantum-well structures with dimensions comparable to the electronic de Broglie wavelength. Due to their small size these structures present some physical properties such as optical and electronic transport characteristics that are quite different from those of the bulk semiconductor constituents.[1,2] It is expected that these characteristics will be more pronounced as the electronic confinement is increased with the reduction of the dimensionality.

Structures produced by ultrathin-film growth are inherently two dimensional, and thus experimental and theoretical investigations have largely been devoted to heterostructures in which only the carrier momentum normal to the interfaces is quantized. Recent advances in microfabrication technology[3–5] have allowed the fabrication of structures with quantum confinement to one dimension ["quantum-well wires" (QWW's)] and have initiated intriguing investigations into one-dimensional physics. It is expected that the fabrication of semiconductor heterostructures with quantum confinement to zero dimensions ["quantum dots" (QD)] will show exotic electronic behavior. In this sense some authors have reported the fabrication of quantum dots and some electronic associated phenomena such as the observation of discrete electronic states in GaAs-(Ga,Al)As nanostructures[6] and the photoluminescence of overgrown GaAs-(Ga,Al)As quantum dots.[7]

The study of impurity states in these low-dimensional heterostructures is an important aspect to which many theoretical and experimental works have been devoted. Since Bastard's pioneering work[8,9] about the binding energy of a hydrogenic impurity within an infinite potential-well structure, using a variational approach, a lot of attention has been devoted to the study of impurity states in quantum wells (QW).[10–12] Results show an enhancement of the impurity binding energy with the decrease of the layer thickness. Also it has been found that the impurity's binding energy depends upon its location within the well.

Equally much theoretical work has been done to characterize the impurity states in QWW's. Bryant[13] studied the effect of changing the cross-sectional form of the QWW on the impurity's binding energy and found that, in the case of wires with the same cross-sectional area, the binding energies are nearly equal for the cylindrical and the rectangular QWW's provided that the rectangular form does not deviate too far from the square shape. Weber, Schulz, and Oliveira[14] calculated the impurity binding energy as a function of the impurity position and the density of impurity states in GaAs-(Ga,Al)As QWW's with different rectangular cross sections and for infinite well depths. Brown and Spector[15] calculated the impurity binding energies using infinite and finite cylindrical confining potentials for both axial and off-axis impurities. Porras-Montenegro, López-Gondar, and Oliveira[16] calculated the impurity binding energy as a function of the impurity position in the radial direction and the density of impurity states as a function of the binding energies for different well radii and for finite and infinite cylindrical GaAs-(Ga,Al)As QWW's. More recently, Latgé, Porras-Montenegro, and Oliveira[17] have calculated the infrared transitions between hydrogenic states in cylindrical GaAs-(Ga,Al)As QWW's. In this work we present results for both donor and acceptor impurity binding energies in spherical GaAs-(Ga,Al)As quantum dots as a function of the dot radius, using a variational procedure within the effective-mass approximation.

The Hamiltonian of a single hydrogenic impurity in a spherical QD system is given by

$$H=\frac{p^2}{2m}-\frac{e^2}{\epsilon|\mathbf{r}-\mathbf{r}_0|}+V(\mathbf{r}) , \tag{1}$$

where m is the electronic effective mass, ϵ the dielectric

constant of the QD material, $V(\mathbf{r})$ the confining potential, and $\mathbf{r}_0$ is the impurity position, which we take at the center of the dot.

The eigenfunction of the Hamiltonian in the absence of the impurity for the ground state ($n=1$ and $l=0$) and for the infinite potential well is

$$\psi_{10}(\mathbf{r})=\frac{\sin(k_{10}r)}{(2\pi R)^{1/2}r}\ , \tag{2}$$

where $\mathbf{r}$ is (r,θ,ϕ). In order to satisfy the boundary conditions $\psi(r=R)=0$, the eigenenergies corresponding to Eqs. (1) and (2) are

$$E_{10k}=\frac{\hbar^2}{2m}k_{10}^2 \quad \text{with} \quad k_{10}=\frac{\pi}{R}\ , \tag{3}$$

where R is the dot radius.

Equation (2) is the wave function of a particle confined in an infinite spherical potential well. Inclusion of the impurity potential makes it necessary to use a variational approach to approximate the wave functions and eigenvalues implied by the Hamiltonian.

Taking into account the spherical confining geometry and the hydrogenic impurity potential, we use the trial wave function

$$\psi(\mathbf{r})=\begin{cases} N\dfrac{\sin(k_{10}r)}{r}e^{-\lambda r}, & r\le R \\ 0, & r\ge R \end{cases} \tag{4}$$

for the ground-state wave function, where N is the normalization constant of the wave function and λ is a variational parameter obtained by minimizing the binding energy.

The binding energy $E_b(R,\mathbf{r}_0)$ of the hydrogenic impurity is defined as the ground-state energy of the system without the impurity present, minus the impurity ground-state energy $\xi(R,\mathbf{r}_0)$, i.e.,

$$E_b(R,\mathbf{r}_0)=\frac{\hbar^2k_{10}^2}{2m}-\xi(R,\mathbf{r}_0)\ , \tag{5}$$

with $\xi(R,\mathbf{r}_0)=\langle T\rangle+\langle U\rangle$, where $\langle T\rangle$ and $\langle U\rangle$ are given by

$$\langle T\rangle=\frac{\hbar^2}{2m}(\lambda^2+k_{10}^2) \tag{6}$$

and

$$\langle U\rangle=-\frac{4\pi e^2N^2}{\epsilon}\int_0^R dr\frac{\sin^2(k_{10}r)}{r}e^{-2\lambda r}\ , \tag{7}$$

respectively, with

$$N^2=\frac{\lambda(\lambda^2+k_{10}^2)}{\pi(1-e^{-2\lambda R})}\ . \tag{8}$$

In the finite well model approximation the potential $V(\mathbf{r})$ in the Hamiltonian [Eq. (1)] will be taken as zero for $r\le R$ and v for $r\ge R$. The boundary conditions on the wave functions are that $\psi(\mathbf{r})$ and its first normal derivative are continuous at the potential boundary. The eigenfunctions of the Hamiltonian in the absence of the impurity are

$$\psi_{10}(\mathbf{r})=\begin{cases} N\dfrac{\sin(\zeta_{10}r)}{r}, & r\le R \\ N\dfrac{\sin(\zeta_{10}R)}{r}e^{\chi_{10}(R-r)}, & r\ge R \end{cases} \tag{9}$$

with

$$\zeta_{10}=\left[\frac{2mE_{10}}{\hbar^2}\right]^{1/2} \quad \text{and} \quad \chi_{10}=\left[\frac{2m(V-E_{10})}{\hbar^2}\right]^{1/2}\ , \tag{10}$$

where E_{10} is the energy of the ground state given by the transcendental equation

$$-\zeta_{10}=\chi_{10}\tan(\zeta_{10}R)\ , \tag{11}$$

which can be written as

$$-\left[\frac{V}{E}-1\right]^{1/2}=\tan(\zeta_{10}R)\ , \tag{12}$$

and from which it can be found that the smallest radius for the existence of a bound state is given by

$$R=\left[\frac{\pi^2\hbar^2}{8mV}\right]^{1/2}\ . \tag{13}$$

In a similar way as in the infinite well situation, inclusion of the impurity potential in the Hamiltonian forces use of the variational approach. Then the trial wave function for the ground state with the impurity present is taken as

$$\psi(\mathbf{r})=\begin{cases} N\dfrac{\sin(\zeta_{10}r)}{r}e^{-\lambda r}, & r\le R \\ N\dfrac{\sin(\zeta_{10}R)}{r}e^{\chi_{10}(R-r)}e^{-\lambda r}, & r\ge R \end{cases} \tag{14}$$

with

$$N^{-2}=4\pi(A+B)\ , \tag{15}$$

where

$$A=\frac{1-e^{-2\lambda R}}{4\lambda}-\frac{\lambda+e^{-2\lambda R}[-\lambda\cos(2\zeta_{10}R)+\zeta_{10}\sin(2\zeta_{10}R)]}{4(\lambda^2+\zeta_{10}^2)}\ , \tag{16}$$

and

$$B=\frac{\sin^2(\zeta_{10}R)e^{-2\lambda R}}{2(\chi_{10}+\lambda)}\ , \tag{17}$$

and λ is a variational parameter.

Defining the binding energy E_b as before we have

$$E_b(R,\mathbf{r}_0)=\frac{\hbar^2\zeta_{10}^2}{2m}-\xi(R,\mathbf{r}_0)\ . \tag{18}$$

Calculating $\xi(R,\mathbf{r}_0)=\langle T\rangle+\langle U\rangle$, we have

$$\langle T\rangle=-\frac{4\pi\hbar^2N^2}{2m}\left[(\lambda^2-\zeta_{10}^2)A-\frac{\lambda\zeta_{10}}{2(\lambda^2+\zeta_{10})}C+(\chi_{10}+\lambda)^2B\right], \tag{19}$$

where C is given by

$$C=\zeta_{10}-e^{-2\lambda R}[\lambda\sin(2\zeta_{10}R)+\zeta_{10}\cos(2\zeta_{10}R)]\ , \tag{20}$$

and for the potential energy

$$\langle U\rangle=-\frac{4\pi e^2N^2}{\epsilon}[D+\sin^2(\zeta_{10}R)e^{2\chi_{10}R}F]+2\pi N^2V\frac{\sin^2(\zeta_{10}R)}{(\chi_{10}+\lambda)}e^{-2\lambda}\ , \tag{21}$$

where

$$D=\int_0^R dr\frac{\sin^2(\zeta_{10}r)}{r}e^{-2\lambda R} \tag{22}$$

and

$$F=\int_R^\infty dr\frac{e^{-2(\chi_{10}+\lambda)r}}{r}\ . \tag{23}$$

Our results are presented in reduced atomic units (a.u.), which correspond to a length unit of an effective Bohr radius $a^*=\hbar^2\epsilon/m^*e^2$, and an energy unit of an effective Rydberg $R_0^*=m^*e^4/2\hbar^2\epsilon^2$. For GaAs-(Ga,Al)As QD's these units are $a^*\cong100$ Å and $R_0^*\cong5.72$ meV for donors (electrons) and $a_0^*\cong22$ Å and $R_0^*\cong26$ meV for acceptors (holes). In our calculations we have assumed a spherical effective mass for both donors and acceptors. Also, we have ignored variations in the effective mass and dielectric constant and considered the values of GaAs throughout the QD heterostructure.

We assume that the band-gap discontinuity[18,19] in a GaAs-$Ga_{1-x}Al_xAs$ QD heterostructure is distributed about 40% on the valence band and 60% on the conduction band with the total band-gap difference ΔE_g between GaAs and $Ga_{1-x}Al_xAs$ given as a function of the Al concentration $x<0.45$ as[20] ΔE_g (eV) $=1.247x$.

In Fig. 1 we present the energy of the ground state without the impurity potential in a spherical GaAs-$Ga_{1-x}Al_xAs$ QD as a function of the dot radius for the conduction band (a) and for the valence band (b) for different Al concentrations. In both cases the energy increases as the radius decreases and approximates a limiting value corresponding to the height of the well when the radius is close to the value given by Eq. (13). Also, it is seen that the energy goes to zero in the bulk limit for large radii.

In Fig. 2 we present the binding energy in a spherical GaAs-$Ga_{1-x}Al_xAs$ QD as a function of the dot radius for hydrogenic donor (a) and acceptor (b) impurities, and for different Al concentrations. The binding energy increases as the radius decreases, reaches a maximum, and then diminishes to a limiting value corresponding to a particular radius of the well, the limiting radius at which it is possible to find the fundamental energy level as shown in Fig. 1. The smallest radii for which it is possible to obtain the donor binding energy are $0.20a^*$, $0.25a^*$, and $0.35a^*$ for Al concentrations of 0.45, 0.30, and 0.15, respectively, and for the acceptor binding energy are $0.53a^*$, $0.65a^*$, and $0.93a^*$ with the same Al concentrations. Also, it is observed that the binding energy goes to $1R_0^*$ in the bulk limit for the large dot radius.

In Fig. 3 we present the binding energy of a donor impurity in a spherical GaAs-(Ga,Al)As quantum dot as a function of the radius for the infinite potential model (a), and for a finite barrier corresponding to Al concentration $x=0.30$ (b). With the same Al concentration we present the binding energy of a donor impurity in a cylindrical QWW (Ref. 17) of radius R (c), and in a QW of size $L_z=R$ of the same material[11] (d). As observed in this figure the quantum confinement produces evident changes in the binding energy of shallow impurities as the dimensionality of the structure is diminished. Whereas the maximum of the binding energy is about $2.5R_0^*$ and $4.6R_0^*$ for GaAs-$Ga_{1-0.3}Al_{0.3}As$ QW's and

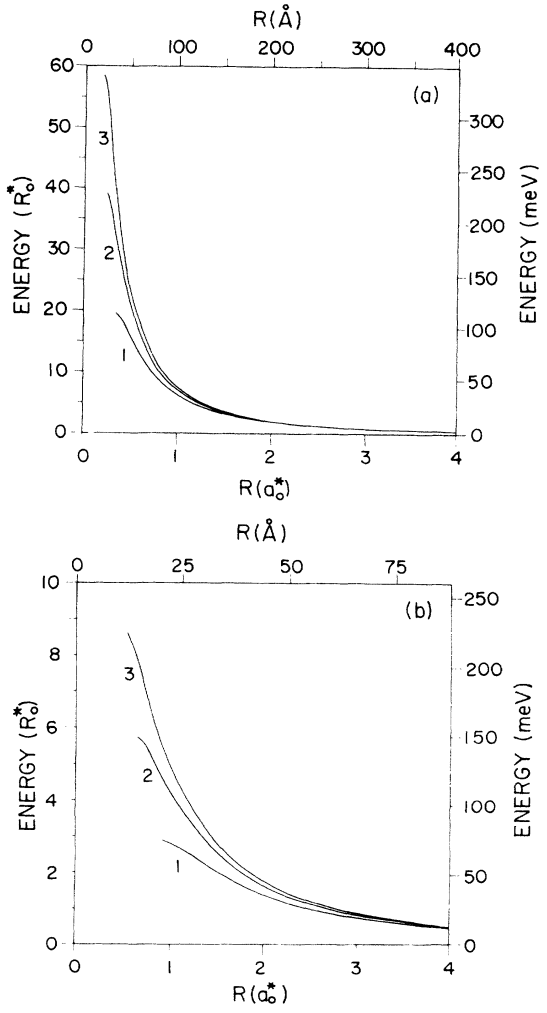

FIG. 1. Energy of the ground state in a spherical GaAs-$Ga_{1-x}Al_xAs$ QD as a function of the dot radius for the conduction band (a) and for the valence band (b), for different Al concentrations: 1 for $x=0.15$, 2 for $x=0.30$, and 3 for $x=0.45$.

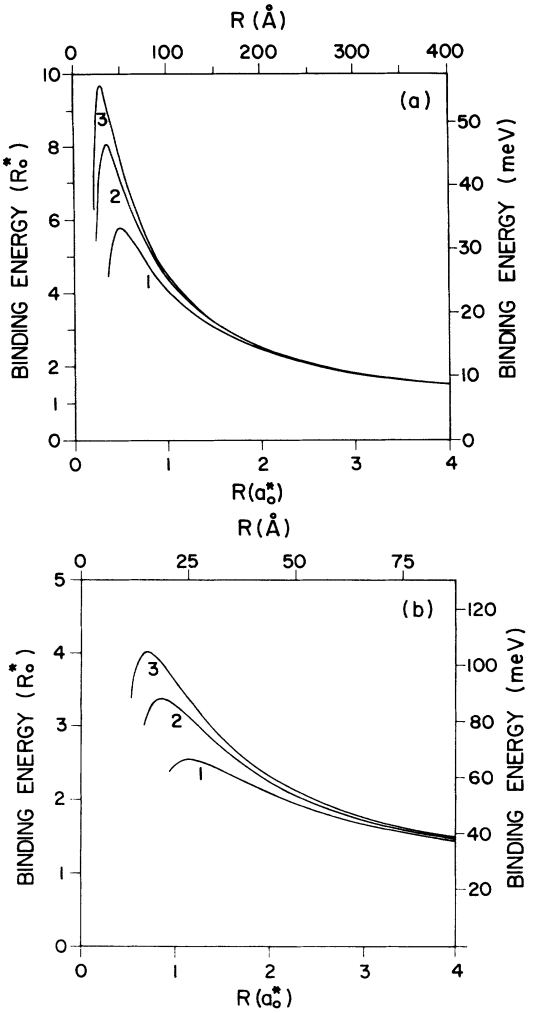

FIG. 2. Binding energy in a spherical GaAs-$Ga_{1-x}Al_xAs$ QD as a function of the dot radius for the hydrogenic donor (a) and acceptor impurities (b), and for different Al concentrations: 1 for $x=0.15$, 2 for $x=0.30$, and 3 for $x=0.45$.

QWW's, respectively, it reaches $\cong 8.0R_0^*$ in a spherical QD of the same material.

Summing up, we have calculated the ground-state energy and the binding energies for on-center shallow donor and acceptor impurities in spherical GaAs-(Ga,Al)As QD's following a variational procedure within the effective-mass approximation. For the infinite potential well we found that the binding energy increases as the dot radius decreases, whereas in the finite potential well the binding energy reaches a peak value as the dot radius decreases and then diminishes to a limiting value corresponding to a value of the radius for which there is no bound states in the well. As expected, we found that the strong electronic confinement in these QD structures reflects itself in the ground-state energy and in the impurity binding energies, which are considerably higher than those found in cylindrical GaAs-$Ga_{1-x}Al_xAs$ QWW's.

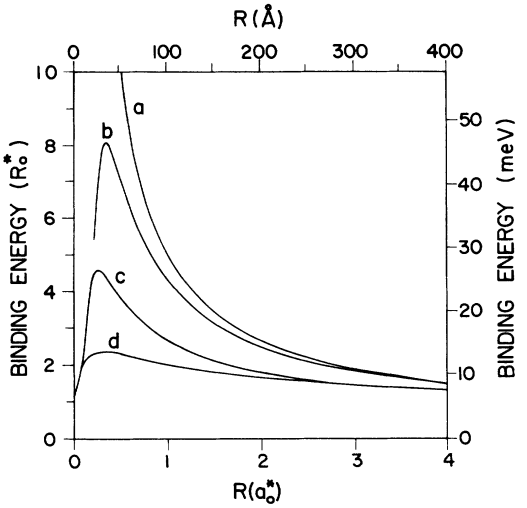

FIG. 3. Binding energy of a donor impurity in a spherical GaAs-(Ga,Al)As quantum dot as a function of the radius for the infinite potential model, **a**, and with a potential barrier corresponding to an Al concentration $x=0.30$, **b**. With the same Al concentration the binding energy of a donor impurity in a cylindrical QWW of radius R, **c** (taken from Ref. 17), and in a QW of size $L_z=R$, **d** (taken from Ref. 11).

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