Donor-excited states and infrared-transition strengths in cylindrical GaAs-(Ga,Al)As quantum-well wires

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A variational calculation within the effective-mass approximation of the ground and lowest excited states of a donor impurity in a cylindrical GaAs-(Ga,Al)As quantum-well wire is presented. The corresponding impurity binding energies are calculated for various values of the GaAs-(Ga,Al)As quantum-wire radius and donor positions within the wire. The line strengths of transitions from the donor ground state to excited states of $2s$-like and $2p_z$-like symmetries are calculated as the donor position varies along the radial direction in the wire, for polarizations of the incident radiation perpendicular and parallel to the wire axis, respectively. Although the $1s \rightarrow 2s$ donor transition is forbidden in bulk materials, this transition is allowed for incident radiation polarized along the y radial direction of the wire with a quite considerable oscillator strength—comparable to the strength of the $1s \rightarrow 2p_z$ transition—for impurities away from the wire axis.

I. INTRODUCTION

The understanding of the properties of impurity energy levels in low-dimensional semiconductor structures is a subject of interest due to possible technological applications in electronic devices associated with these systems. In these nanostructures, besides Coulombic interactions, the impurities are affected by confining potentials which are more important as the dimension of the system is reduced. The electronic configuration for the impurity differs from that of the bulk, and depends on both the well size and the impurity position along the low-dimensional heterostructures. GaAs-(Ga,Al)As quantum wells (QW's) are the most investigated systems, and a number of studies[1–7] concerned with impurity-related properties have been reported in the literature. Greene and Bajaj[2] have calculated the binding and transition energies of the ground and first excited states for distinct wells and magnetic-field strengths in GaAs-(Ga,Al)As heterostructures.

On the other hand, experimental progress in determining the effects of confinement on shallow impurities in QW structures has also been made, mainly through distinct spectroscopic measurements. Jarosik *et al.*[4] have observed broad absorption lines which appear to be related to $1s$-$2p_{\pm}$ transitions at on-center hydrogenic donors in GaAs-$Ga_{0.75}Al_{0.25}As$ QW structures. Holtz *et al.*[5] have reported resonant Raman scattering and photoluminescence measurements which provided an accurate determination of the differences in energy between $1s$ and $2s$ acceptor states. Theoretical predictions from Fraizzoli and Pasquarello[6] and Fraizzoli, Bassani, and Buczko[7] are in good agreement with these experimental results. Far-infrared absorption spectrum of lightly doped GaAs-$Ga_{0.7}Al_{0.3}As$ superlattices were investigated by Helm *et al.*[8]

A considerable theoretical understanding of the properties of impurities in quantum-well wires (QWW's) has also been achieved.[9–12] However, although a high development of more sophisticated techniques in growing low-dimensional heterostructures has been observed in the last years, only a few impurity-related properties in QWW's have been experimentally reported. Hiruma *et al.*[13] have performed an optical characterization of GaAs quantum-wire microcrystals grown by organometallic vapor-phase epitaxy. Spectral features dominated by free-carrier to acceptor-impurity recombinations have appeared in the photoluminescence measurements, and were attributed to the presence of carbon acceptors in the wire microcrystal. A theoretical calculation by Oliveira, Porras-Montenegro, and Latgé[14] of the acceptor-related photoluminescence spectrum of a GaAs quantum-wire microcrystal was shown to be in good agreement with the experimental results.

The experimental work of Helm *et al.*[8] on far-infrared spectroscopy of confined donors in GaAs-(Ga,Al)As superlattices has motivated us to investigate the properties of excited states associated with confined donors in cylindrical GaAs-(Ga,Al)As QWW's. In this work, therefore, we present a calculation of the binding energies associated with the ground and some excited states of hydrogenic donors in cylindrical GaAs-(Ga,Al)As QWW's as functions of the wire radius and the impurity position along the wire cross section. The line strengths of transitions from the ground to excited states are also studied. In Sec. II we provide some theoretical background for the calculation of the donor-excited states within the effective-mass approximation. Theoretical results and transition line strengths are presented in Sec. III, and the work is summarized in Sec. IV.

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II. THEORY

The Hamiltonian for a donor impurity in a cylindrical GaAs-(Ga,Al)As QWW with radius R may be written in the effective-mass approximation as

$$H=\frac{p^2}{2m^*}-\frac{e^2}{\varepsilon_0[|\rho-\rho_i|^2+z^2]^{1/2}}+V(\rho)\ , \tag{2.1}$$

where $V(\rho)$ is the confinement potential which is equal to V_0 when the impurity electron is outside the quantum wire and zero elsewhere, ε_0 is the dielectric constant which was assumed to be equal to the GaAs bulk value, and m^* is the electron effective mass. We have considered the Al concentration x equal to 0.3 which corresponds[15] to a potential barrier approximately equal to 224 meV. In our calculation, the impurity position ($\mathbf{r}_i=\boldsymbol{\rho}_i$) was chosen along the y axis, as is shown in Fig. 1.

We follow Brown and Spector,[9] who discussed GaAs-(Ga,Al)As QWW's for the cases of (i) an infinite potential well, (ii) a finite potential barrier with electron effective mass constant, and (iii) a finite potential barrier with electron effective mass changing. Boundary conditions used by Brown and Spector[9] require that the wave function and the radial particle current are continuous at the potential boundary. The main effect of considering the dependence of the effective mass on the distance from the axis of the wire is an enhancement in the binding energy over the constant-mass case for small ($R<50$ Å) wire radii. Also, Fraizzoli, Bassani, and Buczko,[7] Bastard *et al.*,[16] and Atanasov and Bassani[17] have considered the role of the dielectric mismatch in the calculation of impurity and exciton binding energies in QW's. For QWW's, one would expect that this effect would increase the binding energy as the quantum-wire radius decreases and as the donor approaches the barrier. In this work we choose to concentrate on a method for solving the donor problem (ground and some excited states) and in evaluating the infrared-transition line strengths from the donor ground state to some excited states. Therefore, we do not include effects of dielectric mismatch and dependence of the effective mass on the distance from the axis of the wire, and consider a model with finite-barrier potential with a constant electron effective mass and dielectric constant.

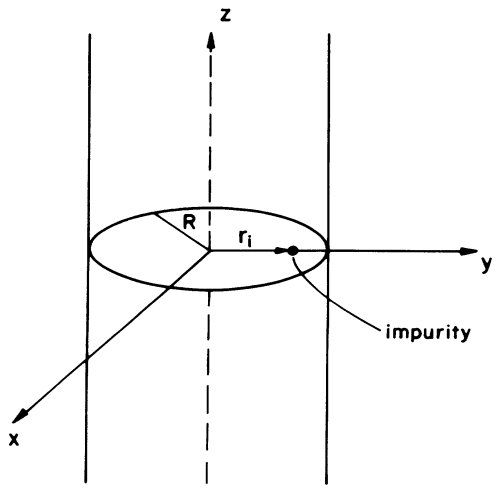

FIG. 1. Schematic picture of a cylindrical GaAs-(Ga,Al)As QWW with radius R and an impurity position along the y axis.

We adopted a variational approach within the effective-mass approximation. The trial wave functions in the variational scheme were labeled by their correspondent bulk hydrogenic limit, and taken as products[12] of hydrogenic functions with the ground-state wave function of the QWW, i.e.,

$$\Psi_{nl}(r,r_i)=N_{nl}\phi_R(\rho)\Gamma_{nl}(r,r_i,\{\lambda_{nl},\beta_{nl},\alpha_{nl}\})\ , \tag{2.2}$$

where $\phi_R(\rho)$ is the ground-state QWW solution[9] without the impurity potential, N_{nl} are the normalization constants, and Γ_{nl} are hydrogenic functions. The corresponding excited states were referred to as $2s$, $2p_{x,y,z}$, $3s$, and $3p_{x,y,z}$ ($n=1$, 2, and 3, with $l=1$ and 2), although they should not be identified with actual hydrogenic states since in general the above wave functions [Eq. (2.2)] are distorted by the barrier potential. $\{\lambda_{nl},\beta_{nl},\alpha_{nl}\}$ is the set of variational parameters of the hydrogenic wave functions. The origin of the coordinate system was chosen at the center of the wire well, and the impurity position is taken to vary within the y axis (cf. Fig. 1). In this coordinate system the distance from the impurity electron to the donor is $r=(|\rho-\rho_i|^2+z^2)^{1/2}$. The hydrogenic wave functions used in Eq. (2.2) are

$$\Gamma_{1s}=\exp(-\lambda_{1s}r)\ , \tag{2.3a}$$

$$\Gamma_{2s}=(1-\beta_{2s}r)\exp(-\lambda_{2s}r)\ , \tag{2.3b}$$

$$\Gamma_{2p_x}=\rho\sin\varphi\exp(-\lambda_{2p_x}r)\ , \tag{2.3c}$$

$$\Gamma_{2p_y}=(\rho\cos\varphi-\rho_i)\exp(-\lambda_{2p_y}r)\ , \tag{2.3d}$$

$$\Gamma_{2p_z}=z\exp(-\lambda_{2p_z}r)\ , \tag{2.3e}$$

$$\Gamma_{3s}=(3-6\beta_{3s}r+2\alpha_{3s}^2r^2)\exp(-\lambda_{3s}r)\ , \tag{2.3f}$$

$$\Gamma_{3p_x}=(2-\beta_{3p_x}r)\rho\sin\varphi\exp(-\lambda_{3p_x}r)\ , \tag{2.3g}$$

$$\Gamma_{3p_y}=(2-\beta_{3p_y}r)(\rho\cos\varphi-\rho_i)\exp(-\lambda_{3p_y}r)\ , \tag{2.3h}$$

$$\Gamma_{3p_z}=(2-\beta_{3p_z}r)z\exp(-\lambda_{3p_z}r)\ , \tag{2.3i}$$

where φ is the angle between $\boldsymbol{\rho}$ and $\boldsymbol{\rho}_i$. We have determined the impurity ground and excited states via a variational procedure which involves minimizing $\langle\Psi_{nl}|H|\Psi_{nl}\rangle$ with respect to the variational parameters $\{\lambda_{nl},\beta_{nl},\alpha_{nl}\}$, with the requirement[18] that the hydrogenic functions Γ_{nl} form a set of orthogonal functions. This procedure leads to exact bulk hydrogenic results in the limit of large-radius QWW's. Of course, one should be aware that an appropriate calculation[19] within the effective-mass approximation would involve the diagonalization of the Hamiltonian matrix written on a basis of functions which also include excited states of the QWW without the impurity. As usual, the binding energies for all hydrogeniclike states are obtained by subtracting the corresponding minimized $\varepsilon_{nl}=\langle\Psi_{nl}|H|\Psi_{nl}\rangle$ from the first-conduction-subband energy obtained from the solu-

tion of the QWW without the impurity. In what follows, we present our results in reduced effective units of length and energy, which correspond to an effective Bohr radius $a_B = \hbar^2\varepsilon_0/m^*e^2$ and an effective Rydberg $R_B = m^*e^4/2\hbar^2\varepsilon_0^2$, respectively [for donors in GaAs-(Ga,Al)As QWW's, these units are $a_B \simeq 100$ Å and $R_B \simeq 5.7$ meV].

III. RESULTS AND DISCUSSION

The theoretical binding energies as functions of the GaAs-$Ga_{0.7}Al_{0.3}As$ QWW radius for the states $1s$, $2s$, $3s$, $2p_z$, and $3p_z$ are presented in Fig. 2 for on-center and on-edge donors. It is apparent that for on-center donors and very large QWW radii ($R \gg a_B$) the bound electrons no

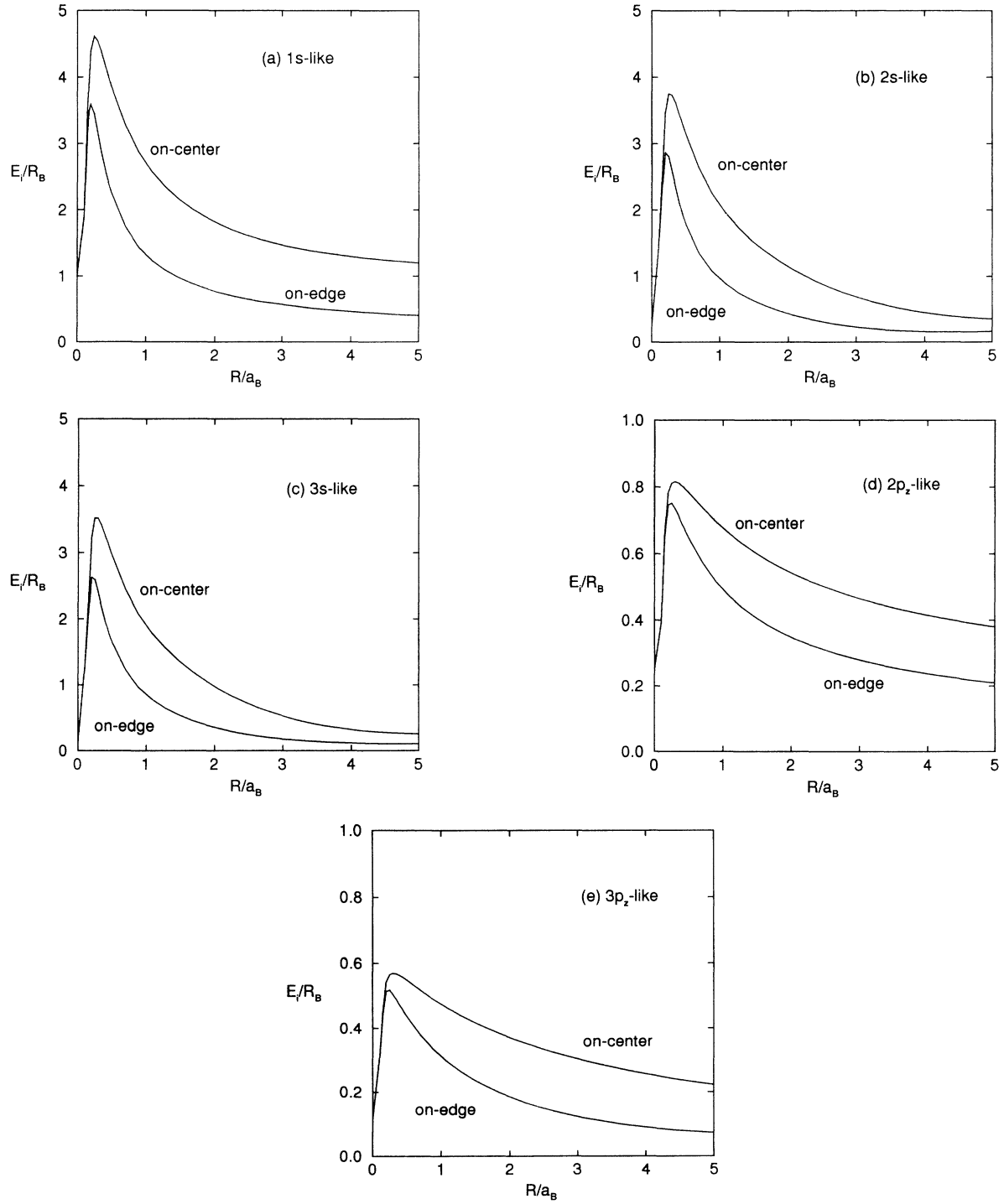

FIG. 2. Binding energies of an electron bound to a shallow-donor impurity at the center and the edge of a GaAs-$Ga_{0.7}Al_{0.3}As$ QWW as functions of the QWW radius (given in units of the effective Bohr radius $a_B \simeq 100$ Å): (a) $1s$-like, (b) $2s$-like, (c) $3s$-like, (d) $2p_z$-like, and (e) $3p_z$-like states. Impurity states are labeled by their bulk hydrogenic limits.

longer interact with the wire boundary and behave as three-dimensional electrons in GaAs. Also, as the QWW radius diminishes, the binding energies, for any donor position in the wire, increase up to a maximum value as the donor wave function (for states with the above symmetries) becomes more compressed in the QWW, which leads to more binding. One should notice that the states labeled by $2p_z$ and $3p_z$ behave in the same qualitative way as $1s$, $2s$, and $3s$, as the electron wave function of a p_z-like state is essentially concentrated along the wire axis, and are affected in a similar manner by the confinement effects. For very small wire radii, the donor electrons leak out of the wire and behave as three-dimensional electrons in $Ga_{0.7}Al_{0.3}As$, and the exact bulk hydrogenic values are again recovered as expected. The behavior of the donor binding energies as the impurity position changes along the wire radial direction is shown in Fig. 3 for these same states and for $R=10$ nm. As the donor approaches the wire boundary, the binding energy decreases due to the repulsion of the donor-electron wave function by the barrier potential. This effect is clearly more pronounced for states labeled as s-like than for p_z-like states, as one would expect. Our results in Figs. 2 and 3 are very similar to the corresponding theoretical results[2,7] for donors in GaAs-(Ga,Al)As *quantum wells,* although the spread of the binding energies as the donor position varies is larger in QWW's than in QW's because of the larger confinement.

Figure 4 shows our theoretical results for the donor binding energies for the states labeled $2p_x$, $2p_y$, $3p_x$, and $3p_y$ in the case of varying GaAs-$Ga_{0.7}Al_{0.3}As$ QWW radii, and for on-center and on-edge donors. The np_x- and np_y-like states have electron densities essentially distributed around the xy plane, and are clearly degenerated (by symmetry) in the case of donors located at the wire axis, as a comparison between Figs. 4(a) and 4(c) (for $n=2$) and 4(b) and 4(d) (for $n=3$) shows. As the impurity position changes from $r_i=0$ (on-center donor) to $r_i=R$ (on-edge donor), the presence of the wire-barrier potential

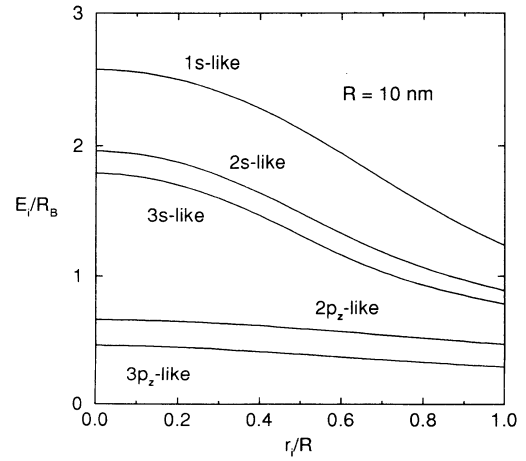

FIG. 3. Donor binding energies as functions of the impurity position for a GaAs-$Ga_{0.7}Al_{0.3}As$ QWW with a radius of 10 nm for different impurity states, labeled by their bulk hydrogenic limits.

breaks the symmetry between x and y and lifts these degeneracies. Independently of the donor position within the QWW, the behavior of these $2p_{x,y}$ and $3p_{x,y}$ states are very similar in the sense that for a QWW radius smaller than some particular value (which depends on the impurity position), they become unbound due to the rather large increase in the donor-electron kinetic energy as the electron wave function is compressed by the wire potential barrier [this occurs even for the $2p_y$-like on-edge case shown in Fig. 4(c), although this is not explicitly shown in the figure]. This unbound-donor behavior was also theoretically found for donors in *quantum wells* by Greene and Bajaj[2,20] and Fraizzoli, Bassani, and Buczko.[7] It would perhaps, be worthwhile to mention that the rather peculiar behavior, for a given QWW radius, of the donor binding energies with impurity positions of $2p_y$- and $3p_y$-like states is probably related to an interplay between the donor position, QWW radius, and espacial extension and nodes of the $2p_y$- and $3p_y$-like electron radial wave functions.

We have also calculated the line strengths for infrared transitions from the $1s$-like donor ground state to some excited states. The line strengths for these transitions were discussed by Greene and co-workers,[21,22] who considered x and y polarizations of the incident radiation for the case of QW's, and by Fraizzoli, Bassani, and Buczko.[7] Within the framework of the effective-mass approximation,[23] the momentum matrix elements of transitions from the impurity ground state (i) to excited states (f) may be expressed as

$$|\langle f|\nabla_j|i\rangle|^2=(m^*E_{fi}/\hbar^2)^2|\langle f|x_j|i\rangle|^2 , \tag{3.1}$$

where x_j is x, y, or z, and E_{fi} is the difference in energy between the initial and final states involved in the impurity transition. The line strengths of these infrared transitions are therefore proportional to the square of the dipole matrix elements between the initial and final states. In Fig. 5, we present the square of the dipole matrix element between the $1s$ and $2s$ states for radiation polarized in the y direction, $|\langle 2s|y|1s\rangle|^2$, and between the $1s$ and $2p_z$ states in the case of z polarization, $|\langle 2p_z|z|1s\rangle|^2$, for all impurity positions within a GaAs-$Ga_{0.7}Al_{0.3}As$ QWW of radius $R=10$ nm. Because of the symmetry of the Hamiltonian, the strength of the y-polarized radiation between $1s$ and $2s$ states is zero for on-center donors. Away from the center, the $|\langle 2s|y|1s\rangle|^2$ matrix elements increase but remain smaller than that of the $1s\rightarrow 2p_z$ transition. Our results for $|\langle 2s|y|1s\rangle|^2$ and $|\langle 2p_z|z|1s\rangle|^2$ are quite similar to those obtained by Greene and Lane[21] and Fraizzoli, Bassani, and Buczko[7] for GaAs-(Ga,Al)As *quantum wells,* although no maximum is observed in the $|\langle 2s|y|1s\rangle|^2$ QWW line strength, in contrast to the result by Fraizzoli, Bassani, and Buczko[7] for $L=400$-Å QW's.

IV. CONCLUSIONS

In this work we have presented a variational calculation within the effective-mass approximation for the states of a donor impurity in a cylindrical GaAs-(Ga,Al)As QWW. The binding energies of the ground

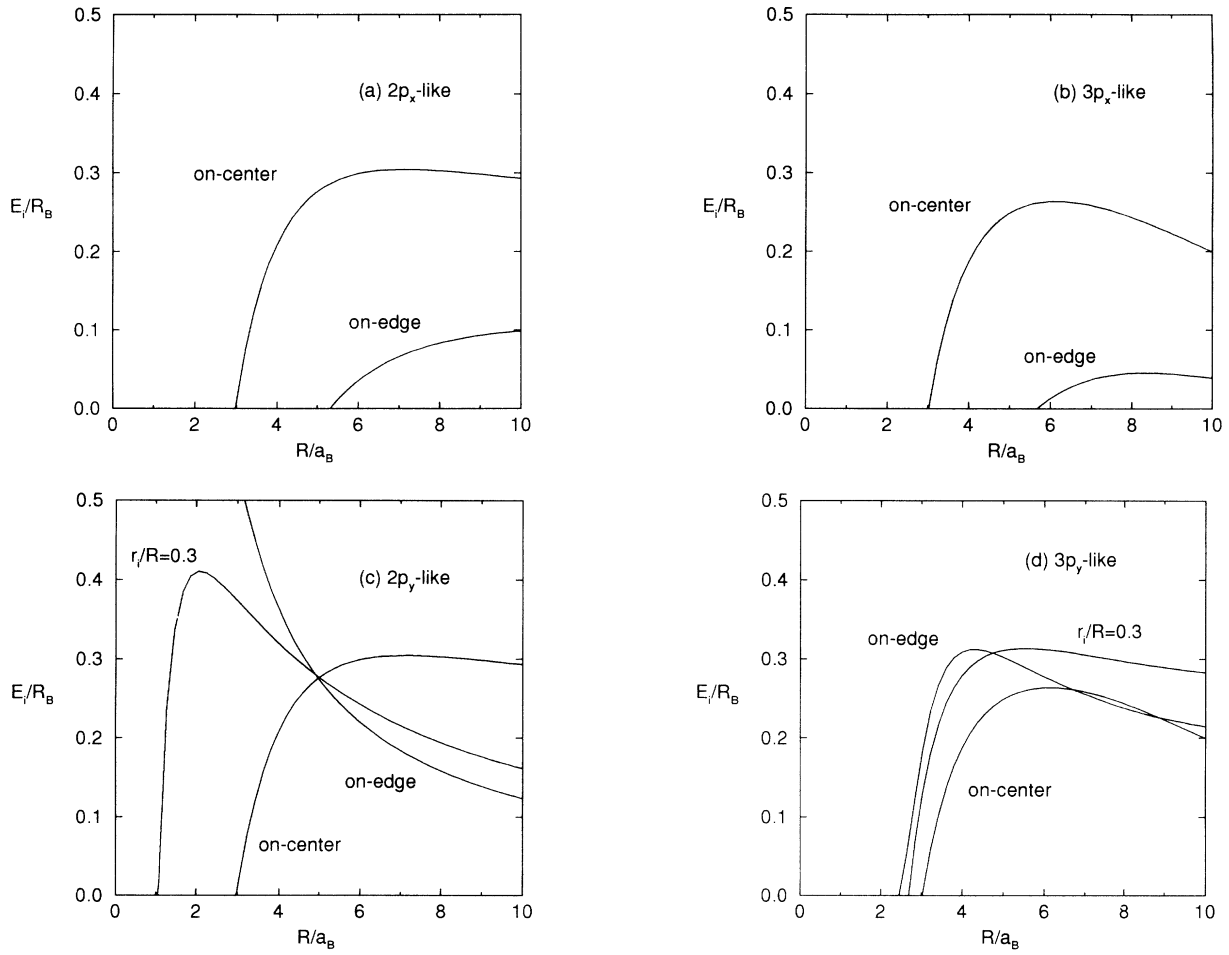

FIG. 4. Binding energies of an electron bound to on-center and on-edge shallow-donor impurities in a GaAs-$Ga_{0.7}Al_{0.3}As$ QWW as a function of the QWW radii (given in units of the effective Bohr radius $a_B \simeq 100$ Å): (a) $2p_x$-like, (b) $3p_x$-like, (c) $2p_y$-like, and (d) $3p_y$-like states.

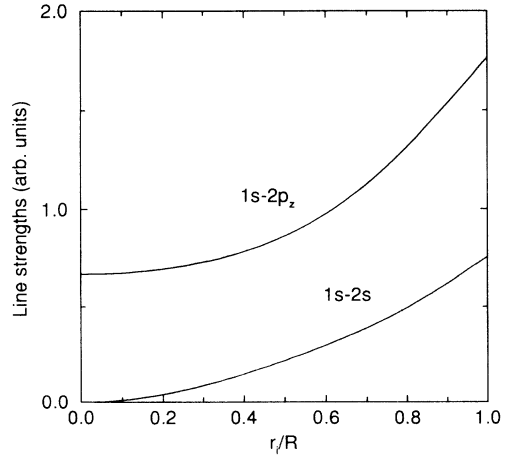

FIG. 5. Line strengths $|\langle 1s|y|2s\rangle|^2$ for the $1s \rightarrow 2s$ transition (with y-polarized radiation), and $|\langle 1s|z|2p_z\rangle|^2$ for the $1s \rightarrow 2p_z$ transition (with z-polarized radiation) as functions of the donor position along the radial direction, for a GaAs-$Ga_{0.7}Al_{0.3}As$ QWW radius $R = 10$ nm.

and lowest excited states—labeled for convenience by their bulk hydrogenic limits—were calculated for various values of the GaAs-(Ga,Al)As QWW radius and impurity positions within the quantum wire. Our results are very similar to those obtained in previous theoretical calculations[2,7] for impurity states in GaAs-(Ga,Al)As QW's, although one should take into account that confinement effects are larger in quantum wires than in QW's and therefore the spread of the donor binding energies as the donor position varies is larger in QWW's than in QW's of comparative dimensions. The line strengths of transitions from the donor ground state to excited states of $2s$- and $2p_z$-like symmetries were calculated for an $R = 100$ Å GaAs-(Ga,Al)As QWW as the donor position varies along the radial direction in the wire, for polarizations of the incident radiation perpendicular and parallel to the QWW axis, respectively. Although the $1s \rightarrow 2s$ donor transition is forbidden in bulk materials, this transition is allowed with a quite considerable oscillator strength—comparable to the strength of the $1s \rightarrow 2p_z$ transition—for impurities away from the wire axis in a GaAs-(Ga,Al)As QWW. Experiments in selectively donor-doped GaAs-(Ga,Al)As QWW's using far-infrared

spectroscopy[8] should be performed in order to investigate the detailed properties associated with impurity transitions.

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