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Energy spectra of donors and acceptors in quantum-well structures: Effect of spatially dependent screening

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The energy spectra of shallow donors and acceptors in GaAs-$Ga_{1-x}Al_xAs$ quantum-well structures have been calculated. The binding energies of the impurities were obtained within a variational calculation in the effective-mass approximation. Calculations were performed for simple neutral and double singly ionized impurities as functions of the position of the impurity in a GaAs quantum well of infinite depth and for various slab thicknesses. The effect of the spatially dependent screening is modeled with a dielectric response of the form $\epsilon^{-1}(r)=\epsilon_0^{-1}+(1-\epsilon_0^{-1})e^{-r/a}$, with a screening parameter $a \approx 1.1$ a.u. characteristic of bulk GaAs. Results are compared with Bastard's theory, which is based on a constant-ϵ_0 screening, and it is found that spatially dependent screening effects are small for donors down to very thin slab thicknesses, but can be quite important for all acceptors in GaAs quantum wells over a large range of slab thicknesses. Calculated results with improved statistics are in quantitative agreement with experimental data on neutral donors and acceptors.

I. INTRODUCTION

Since the pioneering work of Esaki and Tsu[1] on synthesized semiconductor superlattices a great deal of work[2] has been devoted to the understanding of the unique nature of the electronic states associated with superlattices and heterostructures. Modern growth techniques such as molecular-beam epitaxy and metal-organic chemical vapor deposition have made possible the realization of high-quality systems consisting of alternating layers of two different semiconductors with controlled layer thicknesses and sharp interfaces between the layers. A superlattice that has received considerable attention[2–4] consists of alternating layers of $Ga_{1-x}Al_xAs$ ($x \lesssim 0.4$) and GaAs, with thicknesses varying from a few atomic layers to more than 500 Å. The band gap of $Ga_{1-x}Al_xAs$ is larger than that of GaAs, and both semiconductors are direct-gap, zinc-blende semiconductors with almost perfectly matched lattice parameters and band extrema at $\mathbf{k}=0$. Therefore the GaAs-$Ga_{1-x}Al_xAs$ heterostructure exhibits a position-dependent, forbidden gap at $\mathbf{k}=0$ between the valence and conduction bands which goes through sharp discontinuities at the interfaces between the layers. Depending on the Al content of $Ga_{1-x}Al_xAs$, its band gap can be blue-shifted by as much as 445 meV from the GaAs bulk value.[5] The band-gap discontinuity in the GaAs-$Ga_{1-x}Al_xAs$ heterostructure is, according to Dingle,[3] distributed 85% on the valence band and 15% on the conduction band. Recently, however, Miller *et al.*[6] proposed a 57—43 % split in order to explain better their optical data, whereas Wang *et al.*[7] found that a 62—38 % split would be more appropriate to explain charge-transfer measurements in p-type modulation-doped GaAs-$Ga_{1-x}Al_xAs$ heterostructures. Regardless of the exact value of the gap offset, both electrons and holes in a GaAs layer of the heterostructure are confined by a potential well arising from the larger gap and forbidden region of the neighboring $Ga_{1-x}Al_xAs$ layers. Because of the quantum size effects[3] of such potential distribution, the GaAs slab is referred to in the literature as a quantum well.

Because of the potential device applications to heterostructures,[8–10] the understanding of the nature of impurity states associated with quantum wells is a subject of considerable technical and scientific relevance. The study of these impurities was pioneered by Bastard[11] with a variational calculation of hydrogenic states confined within a quantum well with an infinite potential-energy barrier. He evaluated the binding energy of the impurity state as a function of layer thickness and of the impurity position within the layer. Because of the combination of the two potentials (Coulomb attraction and quantum well), there is an inhomogeneous broadening of the hydrogenic levels, which results in the formation of an inhomogeneous, localized "impurity band" in the semiconductor superlattice.

The original study of Bastard[11] was followed by several other calculations. Mailhiot *et al.*[12] and Greene and Bajaj[13] studied the spectra of donors in a GaAs slab symmetrically flanked by two semi-infinite $Ga_{1-x}Al_xAs$ crystals, as a function of both the potential barrier (or equivalently the concentration x) and the size of the quantum well. Greene and Bajaj[13] considered the impurity located at the center of the well, whereas Mailhiot *et al.*[12] studied both the on-center and the on-edge situations, and addressed as well the problem of effective-mass and dielectric-constant mismatches at the interfaces. Chaudhuri and Bajaj[14] considered the effect of nonparabolicity of the conduction band for an on-center donor in a finite quantum well. When finite conduction-band offsets are taken into account, the donor envelope wave function is allowed to penetrate into the $Ga_{1-x}Al_xAs$ bar-

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rier layer. Therefore the use of finite potential barriers has a considerable effect on the binding energies in the thin GaAs limit, but constitutes an essentially minor correction[15] to Bastard's[11] results for well sizes, L, larger than a_0^*, the impurity effective Bohr radius ($L > a_0^*$).

Photoluminescence spectra[16–18] of GaAs-$Ga_{1-x}Al_xAs$ heterostructures have revealed various impurity features which are much weaker than in bulk GaAs. They have been assigned variously to acceptors and donors. Shanabrook and Comas[18] reported donor binding energies ranging from 10 to 6 meV for GaAs well widths between 80 and 450 Å. These values are somewhat lower than but qualitatively well described by the theoretical calculations[11–13] of the *on-center* donors. The acceptor binding energies reported by Miller *et al.*[17] were quantitatively interpreted by Masselink *et al.*[19] as *on-center* acceptors within the effective-mass approximation, including the coupling of the top four valence bands and a short-range core potential for the acceptors. As discussed in this paper we believe that the observed spectra are not due exclusively to impurities at the *on-center* position, but to a more-or-less random distribution of positions throughout the thickness of the well.

In this contribution we study the effects of spatially dependent screening on the binding energies of simple neutral impurities and singly ionized double impurities as functions of the position of the impurity in a GaAs quantum well of infinite depth, and for various slab thicknesses. The inclusion of a spatially dependent screening is considered in Sec. II. Results and discussion are presented in Sec. III.

II. HYDROGENIC IMPURITIES IN A QUANTUM-WELL STRUCTURE

The Hamiltonian of a hydrogenic impurity in a single GaAs quantum well of infinite depth is, in the effective-mass approximation,

$$H=-\frac{\nabla^2}{2m^*}-\frac{Ze^2}{\epsilon(r)[\rho^2+(z-z_i)^2]^{1/2}}+V(z)\ , \qquad (2.1)$$

where $\rho=(x^2+y^2)^{1/2}$, the $z=0$ origin is chosen at the center of the well, z_i is the position of the impurity within the slab, $r=[\rho^2+(z-z_i)^2]^{1/2}$ is the distance from the carrier to the impurity site, $V(z)$ is the potential-energy barrier which confines the electron (hole) within the well of thickness L,

$$V(z)=\begin{cases}+\infty, & |z|>L/2\ ,\\ 0, & |z|<L/2\ ,\end{cases} \qquad (2.2)$$

and Z is the net charge of the hydrogenic impurity ($Z=1$ for the simple neutral impurities and $Z=2$ for the singly ionized, double impurities). The carrier effective mass and the GaAs spatially dependent dielectric screening are given by m^* and $\epsilon(r)$, respectively. The spatially dependent dielectric screening used in the calculation is that proposed by Hermanson,[20]

$$\epsilon^{-1}(r)=\epsilon_0^{-1}+(1-\epsilon_0^{-1})\exp(-r/a)\ , \qquad (2.3)$$

where ϵ_0 is the static dielectric constant and a is a screening parameter. Wang and Kittel[21] used this model dielectric function in the study of hyperfine splitting of muonium in Si and Ge; they chose a so that the Fourier transform of (2.3) fits the dielectric function of Walter and Cohen:[22] $\epsilon_0=11.47$ and $a=1.09$ a.u. for Si; $\epsilon_0=14.00$ and $a=1.15$ a.u. for Ge. In the case of GaAs the static dielectric constant[23] is $\epsilon_0=12.58$, and we took $a=1.1$ a.u. as the characteristic value for the screening parameter.

It should be noted that the dielectric function (2.3) is independent of both z_i, the location of the impurity, and L, the slab thickness. It possesses complete spherical symmetry and does not include any local-field effects. These neglected effects may have some importance for impurities near the edge of the slab and for all locations in extremely thin slabs.

As an exact solution of the Schrödinger equation for the Hamiltonian (2.1) is not possible, we have followed the variational approach of Bastard,[11] and have assumed a trial wave function of the form

$$\psi(\rho,z)=\begin{cases}N\cos(\pi z/L)\exp\{-\lambda^{-1}[\rho^2+(z-z_i)^2]^{1/2}\}, & |z|<L/2\ ,\\ 0, & |z|>L/2\ ,\end{cases} \qquad (2.4)$$

where λ is the variational parameter and N is a normalization factor. The trial impurity ground-state energy

$$\zeta(L,z_i)=\langle\psi|H|\psi\rangle/\langle\psi|\psi\rangle\ , \qquad (2.5)$$

is then minimized with respect to λ. All necessary integrals in (2.5) are performed analytically; only the minimization requires numerical handling.

The impurity binding energy is finally given by

$$E(L,z_i)=(\hbar^2\pi^2/2m^*L^2)-\zeta(L,z_i)\ , \qquad (2.6)$$

where the first term corresponds to the energy of a free electron (hole) at the bottom (top) of the conduction (valence) band.

In the next section we present our results in reduced atomic units (a.u.*), which correspond to a length unit of an effective Bohr radius $a_0^*=\hbar^2\epsilon_0/m^*e^2$, and an energy unit of an effective Rydberg $R_0^*=m^*e^4/2\hbar^2\epsilon_0^2$. For GaAs these units are $a_0^*\approx100$ Å and $R_0^*\approx5.72$ meV for donors (electrons), and $a_0^*\approx22$ Å and $R_0^*\approx26$ meV for acceptors (holes).

III. RESULTS AND DISCUSSION

In Fig. 1(a) we display the effects of spatially dependent screening on the binding energy of the ground state, $E(L,z_i=0)$, of a simple neutral donor at the center of the GaAs quantum well as a function of the well width. We also show, in Fig. 1(b), $E(L,z_i)$ as a function of z_i for two well thicknesses, $L=25$ and 50 Å. It is apparent that, for

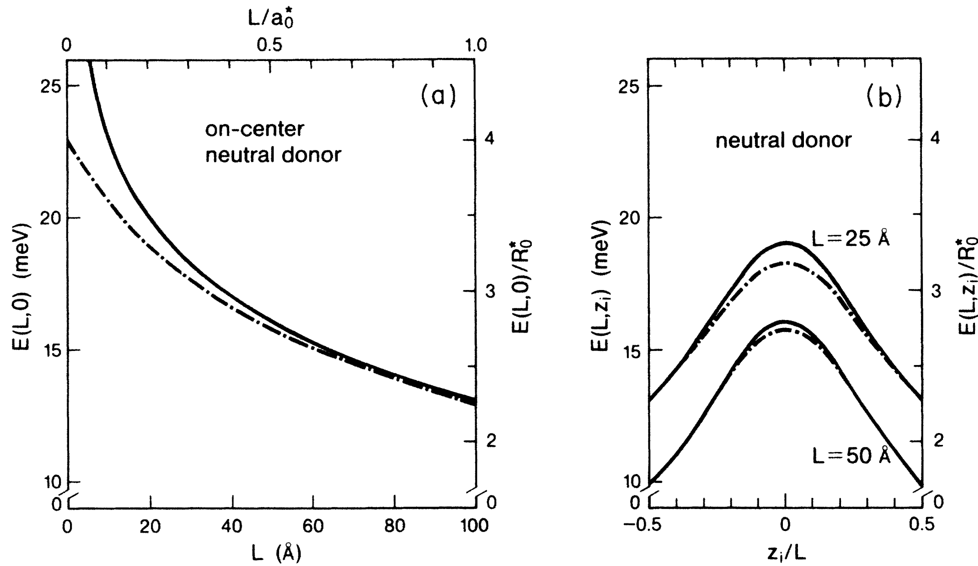

FIG. 1. Binding energy $E(L,z_i)$ for the ground state of a neutral donor as a function of (a) the GaAs quantum-well thickness, L, with the impurity at the center of the well; (b) the impurity position z_i within the quantum well for thicknesses $L=25$ and 50 Å. The dash-dotted curve is for constant $\epsilon=\epsilon_0$; the solid curve is for a spatially dependent $\epsilon=\epsilon(r)$ and leads to an on-center binding energy of $8.0R_0^*$ as L becomes vanishingly small.

very narrow GaAs quantum wells ($L\lesssim 25$ Å), the inclusion of spatially dependent screening, as opposed[11] to a dielectric constant ϵ_0, leads to a considerable increase of the binding energy of the on-center neutral donor. For $L\rightarrow 0$ and for the impurity at the on-center position [Fig. 1(a)], spatially dependent screening effects leads to an "effective Bohr radius" $\lambda\approx 0.26a_0^*$ and a binding energy $E(L,z_i=0)\approx 8R_0^*$, as compared to $\lambda=0.5a_0^*$ and $E(L,z_i=0)=4R_0^*$ for constant screening ϵ_0. In contrast, for $L=0.5a_0^*$ the effective Bohr radii are $\lambda=0.70a_0^*$ (for the $\epsilon=\epsilon_0$ case) and $\lambda=0.68a_0^*$ [for the $\epsilon=\epsilon(r)$ case], which leads to binding energies of $2.75R_0^*$ and $2.81R_0^*$, respectively. It is clear, therefore, that the effect of spatially dependent screening becomes less pronounced as the width of the GaAs slab increases, as the neutral-donor electron wave function becomes increasingly more spread out and, as a result, the importance of the spatial dependence of the dielectric response diminishes. A similar decrease in the importance of the effect occurs as the impurity ion approaches the boundary of the quantum well because of the increasing p-like character[24] of the wave function: spatially dependent screening effects are more

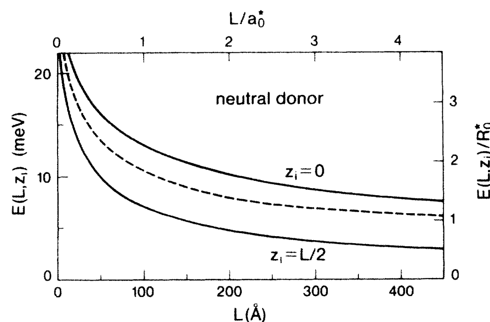

FIG. 2. Binding energy $E(L,z_i)$ versus quantum-well thickness, L, for a neutral donor at the center ($z_i=0$) or at the boundary ($z_i=L/2$) of the well. Results are obtained with a spatially dependent screening. The dashed line indicates the center of gravity of the impurity band.

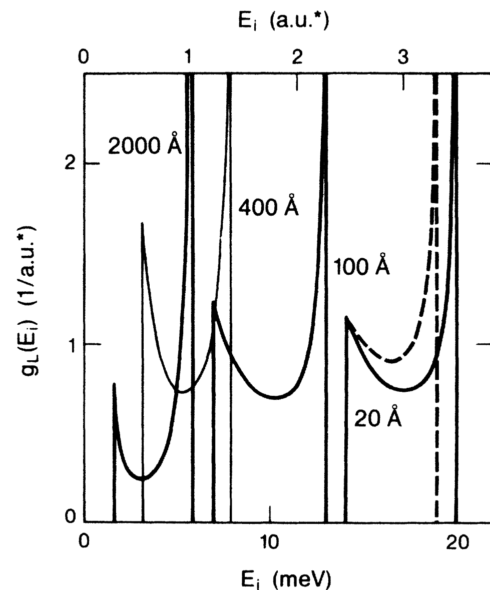

FIG. 3. Density of states $g_L(E_i)$ in reduced atomic units as a function of the neutral-donor binding energy $E_i=E(L,z_i)$ for various quantum-well thicknesses. Solid curves are obtained with a spatially dependent screening $\epsilon=\epsilon(r)$. Shown for comparison are also the results (dashed curve) calculated with a constant $\epsilon=\epsilon_0$ screening for $L=20$ Å.

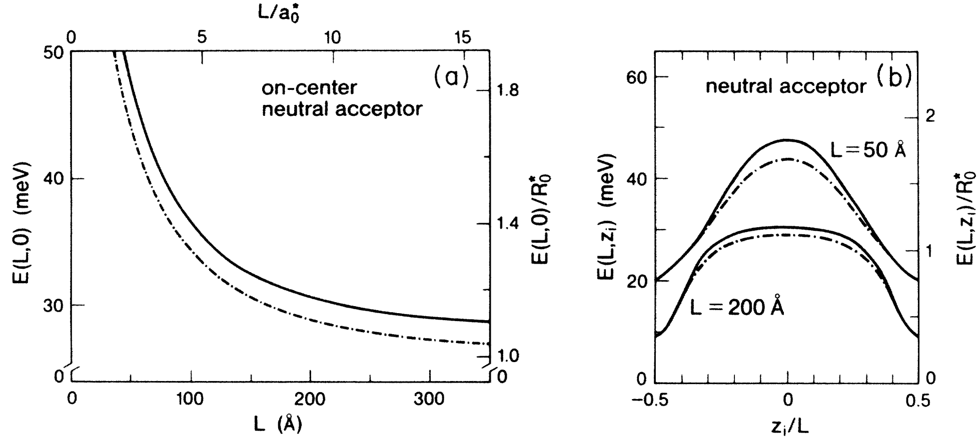

FIG. 4. Binding energy $E(L,z_i)$ for the ground state of a neutral acceptor as a function of (a) the GaAs quantum-well thickness, L, with the impurity at the center of the well; (b) the impurity position z_i within the quantum well for thicknesses $L=50$ and 200 Å. The dash-dotted curve is for constant $\epsilon=\epsilon_0$; the solid curve is for a spatially dependent $\epsilon=\epsilon(r)$.

important for s-like states, which are more concentrated near the impurity ion. The results of Fig. 1 are in overall agreement with a recent calculation of screening effects by Csavinszky and Elabsy.[25] As already mentioned, for *very thin* quantum-well structures, effects of finite conduction-band offsets, nonparabolicity of the conduction band, and effective mass and dielectric mismatches at the interfaces between the GaAs slab and the neighboring $Ga_{1-x}Al_xAs$ layers, not considered in this work, are also of considerable importance.[12–15] For narrow superlattices tunneling between potential wells should also be considered.[26,27]

The binding energy as a function of the GaAs well thickness is presented in Fig. 2 for a simple neutral donor located at the center ($z_i=0$) or at the boundary ($z_i=L/2$) of the well. We also display there the variation with well thickness of the center of gravity of the "impurity band." The concept of an impurity band for quantum-well impurities, first proposed by Bastard,[11] is a consequence of confinement effects, which leads to inhomogeneous broadening of the impurity levels: this is not a true impurity band, in the sense that all electronic states are here atomiclike, isolated, and belonging to a single impurity in a single quantum well. If the quantum well is not too thin, one may treat the impurity position z_i as a continuous random variable and, provided that there is no intentional doping, define a density of impurity states[11] per unit binding energy, $g_L(E_i)$, as

$$g_L(E_i)=[2/L]\,|\,\partial E_i/\partial z_i\,|^{-1}\,,$$
$$E_i=E(L,z_i),\ z_i\geqq 0\,. \tag{3.1}$$

The density of impurity states for the case of the simple neutral donor is shown in Fig. 3 for various layer thicknesses. It should be noticed that $\partial E_i/\partial z_i$ vanishes for the impurity at the on-center position, which leads to an infinite value of $g_L(E_i)$ for $E_i=E_i^{\max}=E(L,z_i=0)$. There is also a second peak in $g_L(E_i)$ for the impurity at the on-edge position, i.e., for $E_i=E_i^{\min}=E(L,z_i=\pm L/2)$, for *all* values of the GaAs slab thickness.[28] For increasing thickness, $L\gg a_0^*$, the strength of the $E_i^{\min}$ peak diminishes, whereas the strength of the $E_i^{\max}$ singularity is enhanced, leading to a center of gravity of the impurity band which converges to the $E_i^{\max}$ value (which becomes the impurity binding energy in bulk GaAs).

Our results for the center of gravity of the impurity band for the simple neutral donor as a function of well thickness (Fig. 2) compare well with the photoluminescence data of Shanabrook and Comas,[18] who reported donor binding energies in the range 10–6 meV for GaAs quantum wells of width between 80 and 450 Å. It should be mentioned that Shanabrook and Comas[18] argue that

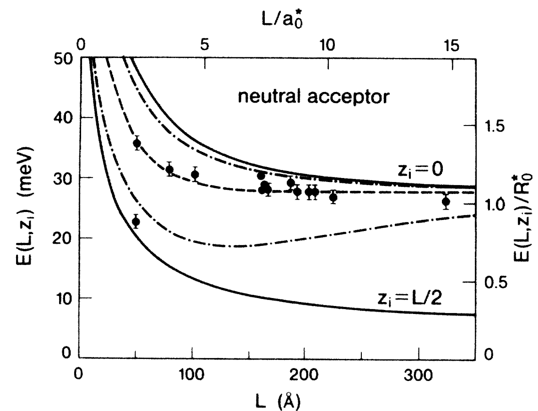

FIG. 5. Binding energy $E(L,z_i)$ versus quantum-well thickness, L, for a neutral acceptor at the center ($z_i=0$) or at the boundary ($z_i=L/2$) of the well. Results are obtained with a spatially dependent screening. The dashed line indicates the center of gravity of the impurity band. The two dash-dotted curves indicate the energies for which the integrated density of states reaches the values 0.25 and 0.75. Experimental results (●) are those of Miller *et al.* (Ref. 17).

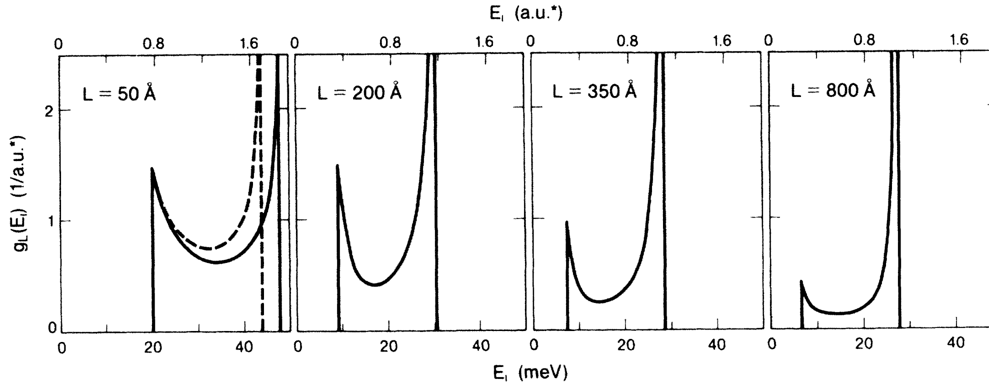

FIG. 6. Density of states $g_L(E_i)$ in reduced atomic units as a function of the neutral-donor binding energy $E_i = E(L,z_i)$ for various quantum-well thicknesses. Solid curves are obtained with a spatially dependent screening $\epsilon = \epsilon(r)$. Shown for comparison are also the results (dashed curve) calculated with a constant $\epsilon = \epsilon_0$ screening for $L = 50$ Å.

they tried to incorporate donor atoms into the structure at particular locations in the wells by means of a 50 Å wide donor spike, incorporated either at the on-center or at the on-edge position. For the on-edge case they claim that the donors were either diffusing of segregating over distances of at least 50 Å during crystal growth. We believe that the good agreement between their reported data and the center of gravity of our theoretical results indicate that their data correspond to donor impurities more-or-less randomly distributed throughout the quantum wells.

The spatially dependent screening effects on the binding energies of simple neutral acceptors at the on-center position as a function of slab thickness is shown in Fig. 4(a). There is a substantial increase in the acceptor binding energy with respect to the ϵ_0 constant-screening theory over a large range of thicknesses, with increases ranging from ~4 meV for $L = 50$ Å to ~2 meV for $L = 350$ Å. In Fig. 4(b) we display the binding energies of neutral simple acceptors as functions of z_i, the impurity position, for $L = 50$ and 200 Å. The importance of the spatially dependent screening effects diminishes as the impurity approaches the edge of the GaAs quantum well and the hole wave function increases its p-like character.

Theoretical results for the simple neutral donor are compared with the photoluminescence measurements of Miller *et al.*[17] in Fig. 5. The density of impurity states

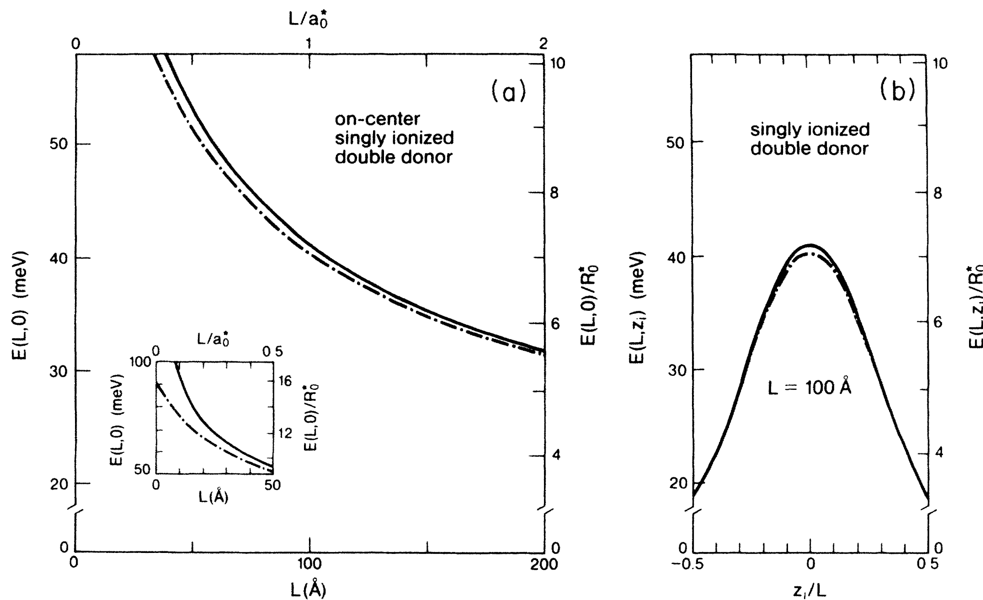

FIG. 7. Binding energy $E(L,z_i)$ for the ground state of a singly ionized, double donor as a function of (a) the GaAs quantum-well thickness, L, with the impurity position at the center of the well; (b) the impurity position z_i within the quantum well for a thickness $L = 100$ Å. The dash-dotted curve is for constant $\epsilon = \epsilon_0$; the solid curve is for a spatially dependent $\epsilon = \epsilon(r)$.

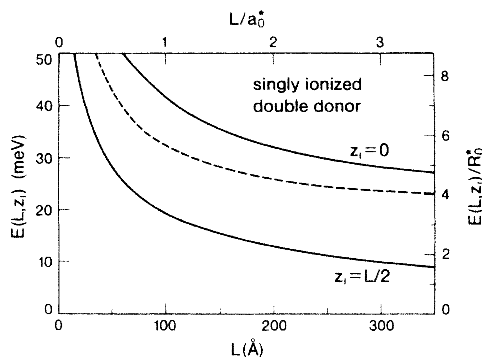

FIG. 8. Binding energy $E(L,z_i)$ versus quantum-well thickness, L, for a singly ionized, double donor at the center ($z_i=0$) or at the boundary ($z_i=L/2$) of the well. Results are obtained with a spatially dependent screening. The dashed line indicates the center of gravity of the impurity band.

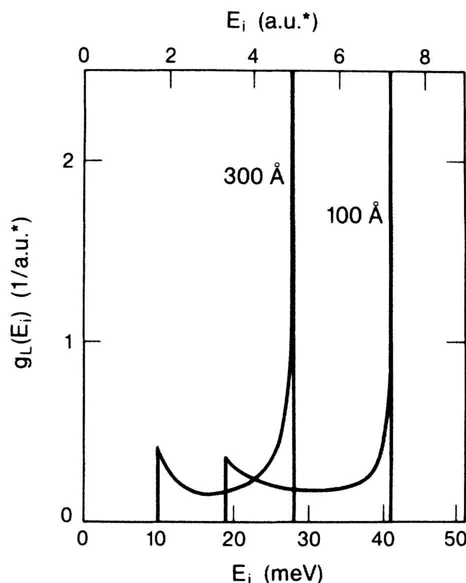

FIG. 9. Density of states $g_L(E_i)$ in reduced atomic units as a function of the singly ionized, double donor binding energy $E_i=E(L,z_i)$ for quantum-well thicknesses $L=100$ and 300 Å. Results are obtained with a spatially dependent screening.

(shown in Fig. 6 for various slab thicknesses) allows one to evaluate the energy of the center of gravity of the impurity band (dashed line in Fig. 5) as well as the energies for which the integrated density of impurity states reaches the values 0.25 and 0.75 (dash-dotted curves in Fig. 5). A clear picture of the relative strength of the two singularities, $E_i^{\min}$ and $E_i^{\max}$, emerges. For $L=50$ Å, the on-edge peak has considerable strength, in good agreement with the additional shoulder observed in photoluminescence data.[17] It is quite apparent from Fig. 5 that there is very good agreement between the theoretical results for the center of gravity of the impurity band and the experimental measurements. Our results indicate that proper consideration of spatially dependent screening effects, as well as the detailed shape of the density of impurity states are essential for the quantitative understanding of the experimental results for simple neutral acceptors in GaAs-$Ga_{1-x}Al_xAs$ quantum-well structures.

Finally, theoretical results for singly ionized, double donors and acceptors in GaAs quantum wells are shown in Figs. 7–12. It is found that spatially dependent screening effects are small for singly ionized, double donors

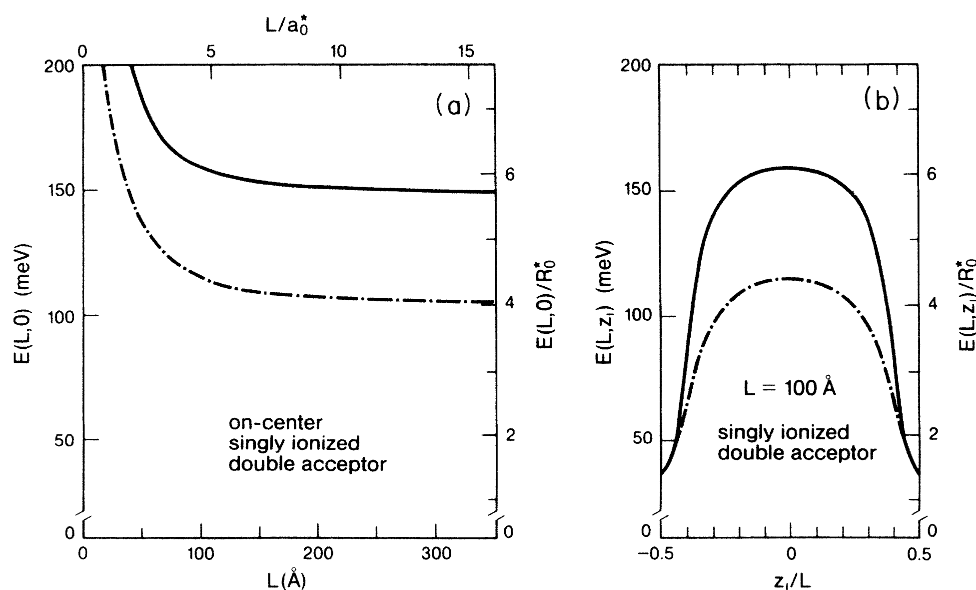

FIG. 10. Binding energy $E(L,z_i)$ for the ground state of a singly ionized, double acceptor as a function of (a) the GaAs quantum-well thickness, L, with the impurity position at the center of the well; (b) the impurity position z_i within the quantum well for a thickness $L=100$ Å. The dash-dotted curve is for constant $\epsilon=\epsilon_0$; the solid curve is for a spatially dependent $\epsilon=\epsilon(r)$.

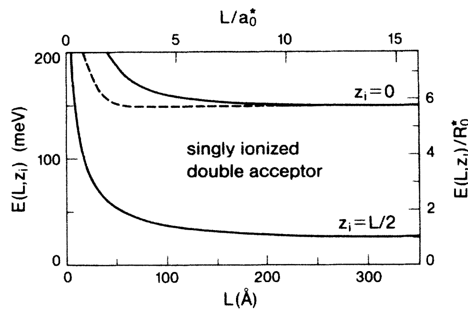

FIG. 11. Binding energy $E(L,z_i)$ versus quantum-well thickness, L, for a singly ionized, double acceptor at the center ($z_i=0$) or at the boundary ($z_i=L/2$) of the well. Results are obtained with a spatially dependent screening. The dashed line indicates the center of gravity of the impurity band.

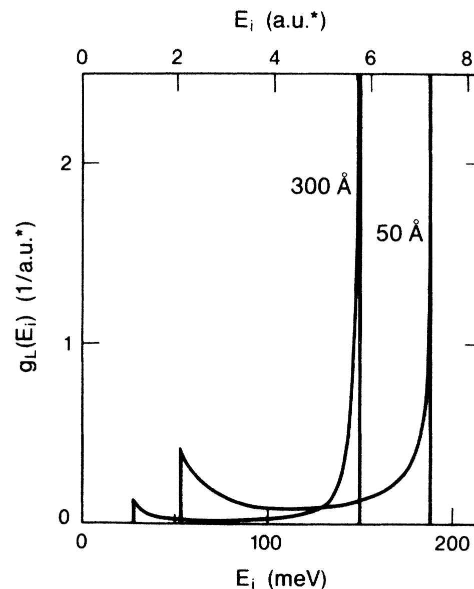

FIG. 12. Density of states $g_L(E_i)$ in reduced atomic units as a function of the singly ionized, double acceptor binding energy $E_i=E(L,z_i)$ for quantum-well thicknesses $L=50$ and 300 Å. Results are obtained with a spatially dependent screening.

down to very thin layer thicknesses ($L\sim 50$ Å), but can be quite significant for singly ionized, double acceptors. It should be mentioned that double donors and double acceptors should be common in GaAs quantum-well structures, a consequence of antisite defects in which an As (Ga) atom occupies a Ga (As) site and produces a double donor (double acceptor).

In conclusion we would like to point out two features of the experiments as they relate to the present calculations. (1) As seen in Fig. 5 the experimentally measured energy levels, which have an error of ± 1 meV, seem to be scattered throughout the impurity band, and not necessarily concentrated at the top, the *on-center* location. If at all, they seem to follow the center of gravity of the band. Therefore this comparison seems to indicate a *random distribution of impurities* in the slab. Previous comparisons with experiment, assuming only an *on-center* location, should thus be viewed with caution. (2) It is important to realize that the binding energies obtained for the singly ionized double donors (Fig. 8) are very close to those of the simple neutral acceptors (Fig. 5). The assignment of observed lines to neutral acceptors should therefore be made with extreme caution based on the overall donor-acceptor characteristics of the sample, since energetics alone do lead to ambiguities and, as speculated above, single ionized double impurities caused by antisite defects, could be fairly abundant.

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[28]Notice that Fig. 4 of Bastard's paper (Ref. 11) displays the results for $[2E(L,0)\,|\,dz_i/dE_i\,|\,/L]$, which do *not* correspond, as claimed, to the density of impurity states $g_L(E_i)$. In addition it is straightforward to show that this function, as well as $g_L(E_i)$, exhibit a double-peaked structure at $E_i=E_i(\text{max})$ and at $E_i=E_i(\text{min})$ for *all* values of the GaAs layer thickness.