Excitons in type-II quantum-dot systems: A comparison of the GaAs/AlAs and InAs/GaSb systems

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A calculation of the exciton binding energy (E_x) and oscillator strength for quantum dots in type-II semiconductor systems is presented. These structures consist of a spherical dot of one semiconductor embedded in a second semiconductor. As in the type-II exciton systems in quantum wells the electron is confined in one semiconductor and the hole is confined in the other due to band lineups in the two materials which make this arrangement energetically favorable. We have considered the systems (i) GaAs/AlAs where the electron is confined in the X state in the AlAs while the hole is confined in the GaAs dot (for a dot radius of less than 56 Å) and (ii) InAs/GaSb where the electron is confined in the InAs dot while the hole is confined in the GaSb (for a dot of radius less than 87 Å). While both of these systems are indirect in real space the GaAs/AlAs system is also indirect in k space. We compare E_x in the type-II dot to E_r, the binding energy of a bulk hydrogenic impurity in the bulk barrier. We find that for the case of infinite barriers $E_x < E_r$. For the finite barrier case with wave-function leakage into the dot, $E_x \gg E_r$ due to the effect of the overlapping electron and hole wave functions within the dot. The values of E_x for a type-II dot system are vastly larger than those for a type-II quantum-well system due to the extra correlation in the other two confined dimensions and for the GaAs/AlAs system the values are comparable to the type-I E_x values for $30 < a < 70$ Å where a, is the radius of the dot.

When two materials are joined to form a heterostructure, band lineups arising from energy considerations can result in a lower potential for both electrons and holes being created in one material (a type-I system) or a lower potential created for electrons in one material and for holes in the other material (a type-II system). Type-I systems can be considered to be direct in real space while type II are indirect, in analogy with band-structure terminology. The exciton binding energy E_x in a Type-II quantum well was calculated by Bastard *et al.*[1] He considered the system InAs/GaSb where the electron is confined in the InAs quantum well and the hole in the GaSb barrier which is bulklike. In this work the assumption was made of infinite barrier height or complete confinement. The calculated exciton binding energy was found to be 0.8^*R at vanishing well width, where R is the scaled hydrogenic Rydberg which is about 1 meV for this system, and to decrease from this value for increasing well width. This compares with an increase in the exciton binding energy at vanishing well width in a type-I exciton system of about 4^*R when infinite barriers are assumed. The exciton oscillator strength (OS) was not considered in this paper[1] but for type-I systems has been considered by Rorison and Herbert.[2] The enhanced E_x and OS for type-I systems have been exploited in a number of very important devices such as the quantum-well laser and modulator. Duggan and Ralph[3] considered the heavy-hole type-II exciton binding energy in a GaAs/AlAs multiple-quantum-well system including anisotropy of the electron and hole effective masses, again assuming infinite confining potentials. They obtained exciton binding energies of a similar magnitude in size to the type-I heavy-hole exciton in this system. Salmassi and Bauer[4] considered the same system and found that including finite barriers increased E_x by abut 10%.

Excitons in type-I wires have been considered by Banyai *et al.*[5] They find that, again assuming infinite confining barriers, the exciton binding energy is vastly increased in these structures compared with the three-dimensional (3D) and 2D cases. The case of excitons in quantum dots has been considered by Efros and Efros,[6] Brus,[7] and Hu, Lindberg, and Koch,[8] among others. More recently Kayanuma has considered a general variational wave function to deal with excitons in quantum wells, wires, and dots. To my knowledge no one has considered the type-II exciton in wire and dot systems. Now that it is possible to fabricate or grow semiconductor quantum wires[10] and dots[11] this calculation is of increasing relevance. We present calculations on the type-II dot systems GaAs/AlAs, which is type II for a dot radius of less than 56 Å and InAs/GaSb which is type II for a dot radius of less than 87 Å.

CALCULATIONS

Exciton binding energy

In Figs. 1(a) and 1(b) we show the band lineups in the systems GaAs/AlAs and InAs/GaSb, respectively. The radial part of Hamiltonian for the exciton in this spherical type-II system is

$$H(r_c,r_f)=H(r_c)+H(r_f)-V(r_c)+V(r_f)-1/(\varepsilon(r_c-r_f)) , \tag{1a}$$

where the first term is the kinetic energy of the confined

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particle—the hole in the GaAs/AlAs system and the electron in the InAs/GaSb system, the second is the kinetic energy of the free particle—the electron in the GaAs/AlAs system and the hole in the InAs/GaSb system, the third and fourth terms are the potentials for the confined and free particle, respectively, and the fifth is the Coulomb energy. (Throughout this paper in all equations we use atomic units taking $\hbar=1$, $e=1$, and $m_0=1$.) If we neglect the Coulomb term the Hamiltonian separates into

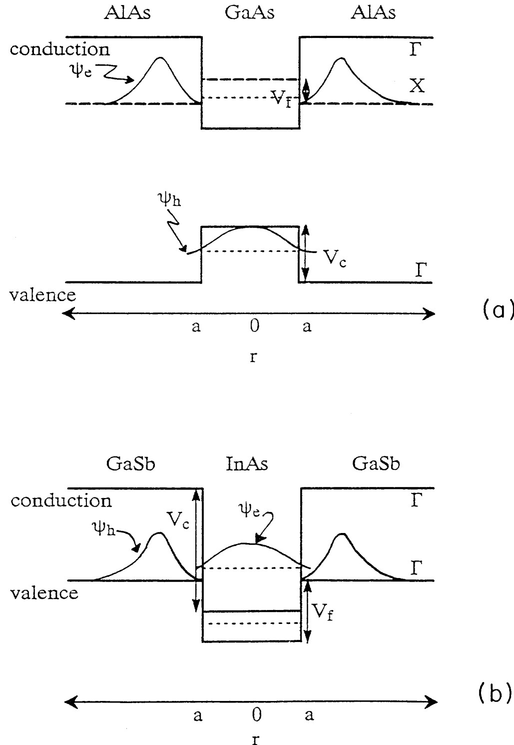

FIG. 1. (a) A cross section through a quantum dot in the GaAs/AlAs system showing the potential E as a function of real space r is shown. Both the Γ and X conduction bands and the Γ valence bands in the two materials are shown. A band-offset ratio of 67/33 (Γ conduction band/Γ valence band) was taken, which results in an effective potential barrier for the confined hole of $V_c=0.526$ eV and an effective barrier for the free electron of $V_f=0.278$ eV. A schematic view of the electron and hole wave functions is also shown. The system becomes type-I when $a>56$ Å when the confined state in the Γ conduction band in the GaAs falls below the X conduction band in the AlAs system. (b) A cross section through a quantum dot in the InAs/GaSb system showing the potential E as a function of real space r is shown. The Γ conduction and valence bands in the two materials and the band lineups are shown. The InAs valence band lies 0.15 eV below the GaSb valence band but provided that the confined level in the InAs dot is above the GaSb valence band the system will be type II. The system is determined to be type II for dot sizes of less than 87 Å and to become semimetallic for larger dot radii. The resulting effective potential barrier for the confined electron is $V_c=0.820$ eV and the effective barrier for the free hole is $V_f=0.510$ eV. A schematic view of the electron and hole wave functions is also shown.

$$H(r_c)=-1/(2m_c^*)[1/r_c^2(\partial/\partial r_c)r_c^2\partial/\partial r_c)-V(r_c)\ , \quad (1b)$$

where $V(r_c)=\text{const}$ for $r_c<a$ and $V(r_c)=0$ for $r_c>a$, where a is the radius of the dot and

$$H(r_f)=-1/(2m_f^*)[1/r_f^2(\partial/\partial r_f)r_f^2\partial/\partial r_f]+V(r_f)\ , \quad (1c)$$

where $V(r_f)=\text{const}$ for $r_f<a$ and $V(r_f)=0$ for $r_f>a$. We therefore propose taking a separable wave function as the solution to Eq. 1(a) of the form

$$\Psi(r_c,r_f)=\Psi(r_c)\Phi(r_f)\ , \quad (2)$$

where $\Psi(r_c)$ is the solution to (1b) and $\Phi(r_f)$ is a variational wave function which models the buildup of the free-particle wave function near the dot.

We take an average of the effective mass for the electron in the X state according to $1/m_f^*=\frac{1}{3}(1/m_\parallel^* + 2/m_\perp^*)$ where we taken $m_\perp^*$ and $m_\parallel^*$ to be $0.19m_0$ and $1.56\ m_0$, respectively,[12] and use the $k=0$ value for the hole mass which is taken to be that of the heavy hole $m_c^*=0.34$.[12] We neglect the mixing of the light and heavy holes and the resulting band splitting and warping, considering this as something to be included at a later date. The confining potentials are shown in Figs. 1(a) and 1(b) and can be estimated from the data given in Ref. 12. Image charges are neglected and ε is taken to be the average of the two materials.[12] The values used in these calculations are given in the caption of Figs. 1(a) and 1(b). Before we consider this system, however, we can study the model system with infinite confining potentials.

Infinite barriers

With the appropriate choice of variational wave function the binding energy becomes analytical. For this case the wave functions are completely resident in their appropriate semiconductors. The wave function and corresponding energies for the confined particle are[13]

$$\Psi_{n=1,l=0,m=0}(r_c) = \sqrt{(2/a^3)}j_0(\alpha_{10}r/a)/j_1(\alpha_{10})Y_{l=0,m=0}(\Omega)\ , \quad (3a)$$

with corresponding binding energy

$$E=\alpha_{10}^2/(2m_c^*a^2)\ , \quad (3b)$$

where j_0 is the zeroth-order spherical Bessel function which has its first zero at $r=a$, $\alpha_{10}=\pi$ and $Y_{l=0,m=0}(\Omega)$ is the corresponding spherical harmonic. The exact form of the wave function of the unconfined particle has not been evaluated in detail. It is free-particle-like everywhere except in the region $r<a$, where it is forced to zero. The energy of this state will be zero since it is unconfined.

The confined particle will not be altered by the Coulomb interaction since E(confinement) $\gg E$(Coulomb) for infinite barriers. The unconfined or free particle will be affected by the Coulomb interaction and its wave function will peak near the dot and will be forced to vanish at $r=a$ for infinite barriers. For infinite barriers we propose a variation wave function of the form[14]

$$\Phi(r_f)=(r_f-a)\exp[-(r_f-a)b/2]/(2a^2/b^3+12a/b^4+24/b^5) \quad \text{for } r_f>a ,$$
$$\Phi(r_f)=0 \quad \text{for } r_f<a , \tag{4}$$

where b is the variational parameter which is $b=2/a_0$, where a_0 is the effective orbital size. The solution to the Hamiltonian acting upon these wave functions is

$$E_x(b)=\{1/(2m_f^*)[a^2/(2b)+a/b^2+2/b^3]-\pi(2a/b^3+6/b^4)/\varepsilon\}/(2a^2/b^3+12a/b^4+24/b^5) , \tag{5}$$

where the first term is the kinetic energy of the unbound particle, which increases with decreasing b showing that it is unfavorable to localize this free particle. The second term is the Coulomb term, which is negative in sign and increases for large localization of the free particle. The minimization of this energy function with respect to the variational parameter b gives the binding energy and the wave function [through b in Eq. (4)], which can be evaluated numerically. E_x for the system considered assuming infinite barriers is shown in Figs. 2 and 3.

Finite barriers

If we consider real systems the barrier is always only a few hundred meV (0.526 eV for the confined hole and 0.278 eV for the free electron in the GaAs/AlAs system and 0.820 eV for the confined electron and 0.510 eV for the free hole in the InAs/GaSb system) leading to penetration of the confined wave function out of the dot and of the free particle into the dot. This should greatly increase the Coulomb energy. This penetration can be represented as an exponential decay into the other semiconductor according to[15]

$$\Psi(r_c)=N\sin[\sqrt{(2m_c^*E)}r_c]/r_c \quad r_c<a , \tag{6a}$$

$$\Psi(r_c)=MN\exp\{-\sqrt{[2m_c^*(V-E)]}r_c\}/r_c \quad r_c>a . \tag{6b}$$

By matching $\Psi(r_c)$ and $1/m_c^*\partial\Psi(r_c)/\partial r_c$ at $r_c=a$ the energy can be determined and the parameters M and N (matching and normalization constants) determined.[14] In this model we neglect the effect of the Coulomb interaction on the hole wave function since E(confinement) dominates the physics of the confined particle. The effect of the Coulomb interaction on the confined particle will be to shift its maximum from $r=0$ to some finite value of r where this shift will be a function of V_c, the Coulomb energy, and m_c^*.

The free particle's wave function can be written as

$$\Phi(r_f)=MN\exp[k_b(r_f-a)/2] \quad r_f<a , \tag{7a}$$

$$\Phi(r_f)=N(r_f-a+z_0)\exp[-b(r_f-a)/2] \quad r_f>a . \tag{7b}$$

By matching $\Phi(r_f)$ and $1/m_f^*\partial\Phi(r_f)/\partial r_f$ at $r=a$ and by normalization we can relate the parameters N,M,z_0 in terms of b and k_b. Since the energy should depend much

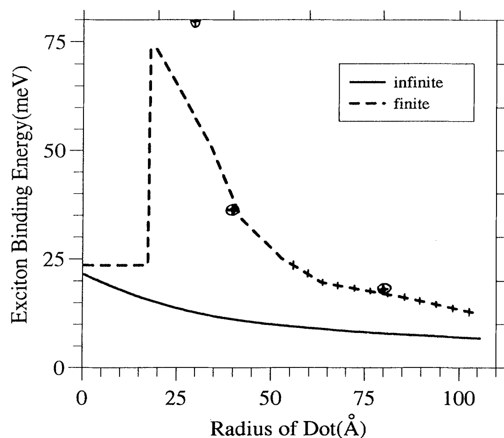

FIG. 2. The exciton binding energy E_x in meV is shown as a function of the radius of the quantum dot in Å for the system GaAs/AlAs. The full line shows E_x assuming infinite barriers and no wave-function leakage while the dashed line shows E_x taking the barriers to be those shown in Fig. 1(a). These values of E_x should be compared to the binding energy of a hydrogenic impurity in bulk E_r which is 36.3 meV and the bulk exciton binding energy which is 23.6 meV. The system becomes type I for $a>56$ Å, and so the finite and infinite curves are not valid for $a>56$ Å which is indicated by the lines being crossed. Three values of E_x for a type-I system are shown as dots for comparison.

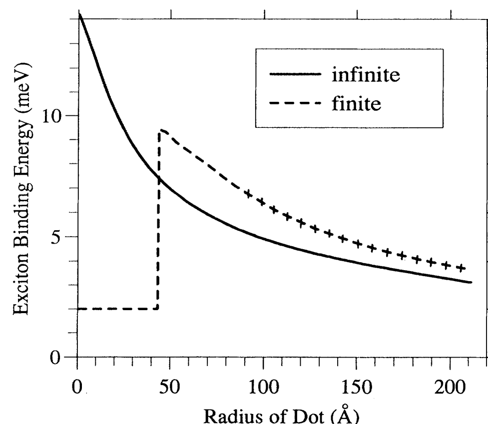

FIG. 3. The exciton binding energy E_x in meV is shown as a function of the radius of the quantum dot in Å for the system InAs/GaSb. The full line shows E_x assuming infinite barriers and no wave-function leakage, while the dashed line shows E_x taking the barriers to be those shown in Fig. 1(b). These values of E_x should be compared to the binding energy of a hydrogenic impurity in bulk E_r which is 19.9 meV and the bulk exciton binding energy which is 2.06 meV. The system becomes semimetallic for $a>58$ Å, and so the finite and infinite curves are not valid for $a>87$ Å which is indicated by the lines being crossed. A comparison can be made with the value of E_x for a type-I system of 60 meV for a radius of 30 Å.

less on k_b than on b we have fixed k_b at the value $k_b = 2\sqrt{(2m_f^* V)}$ and use b as a simple variational parameter.

Using these wave functions in Eq. (1) we minimize the energy as before by varying b to evaluate E_x and the wave functions. E_x is measured relative to the initial free state E_i^f which has energy equal to zero, while the energy of the confined state E^c is assumed not to be altered by correlation with the free particle since E(confinement) $\gg E$(Coulomb) and therefore E_x is calculated according to $E_x = E_f - E_i = (E_f^f + E^c) - (E^c + E_i^f) = E_f^f$, where E_f^f is the calculated final-state energy of the free electron. The binding energy so calculated is presented for the GaAs/AlAs system in Fig. 2 and for the InAs/GaSb system in Fig. 3. Comparison is also made with the infinite barrier case. The wave functions for the system near maximum E_x, which corresponds to a dot radius of 15.85 Å for the GaAs/AlAs system and 44.9 Å for the InAs/GaSb system, are plotted in Figs. 3 and 4, respectively.

Exciton oscillator strength

The radiative recombination rate of both band-to-band and excitonic transitions is directly related to its oscillator strength, which is proportional to[2]

$$R = |\langle f|\mathbf{p}|i\rangle|^2/N , \quad (8)$$

where $|f\rangle$ and $|i\rangle$ represent the final and initial states, respectively, $\mathbf{p}$ is the momentum operator derived from the $\mathbf{A}\cdot\mathbf{p}$ electron-photon coupling, and N is the number of dots in the crystal. We follow the usual approximations and take $\mathbf{p}$ to operate on the Bloch components of the wave functions when R can be reduced to evaluating the electron-hole wave-function overlap. The result is

$$R \sim \|p\|^2 \langle \Phi(r_f)|\Psi(r_c)\rangle^2 \Omega , \quad (9)$$

where $\|p\|$ is the numerical version of the momentum matrix elements between Bloch functions and Ω is the dot atomic volume.

For type-II systems the overlap factor will be small since it depends on the overlap of the exponential tails into and out of the well. Within this level of approximation the exciton recombination in type-II systems with infinite barriers is zero. For finite barriers the exponential tails contribute and as the dot gets smaller, pushing the ground state upwards, this exponential contribution increases sharply until the "confined" particle becomes unbound when the overlap should increase strongly.

As mentioned previously the GaAs/AlAs system is indirect in k space as well as real space. This complicates the analysis significantly, but the effect will be to reduce the oscillator strength (OS) further. We make the assumption that the Bloch states and therefore the Bloch matrix elements are not altered significantly between the materials and incorporate the reduction in k-space overlap into a phenomenological parameter α. For quantum wells a value of α of 0.4 has been evaluated from consideration of experimental results.[16] The wave-function overlap squared factor in Eq. (9) is shown for the GaAs/AlAs and InAs/GaSb systems in Figs. 6 and 7, respectively.

RESULTS AND DISCUSSION

It is useful to compare the results displayed in Fig. 2 with the case of a hydrogenic impurity in the bulk system which has a binding energy of $E_r = m_f^*/(2\varepsilon^2)$ (Ref. 14) with a corresponding $1S$ wave function $\Phi(r_f) = 1/\sqrt{(\pi a^3)}\exp(-r_f/a_0)$ and orbital size $a_0 = \varepsilon/m_f^*$. Since the confined particle is restricted to the dot, as the radius of the dot tends to zero the physics approaches that of a hydrogenic impurity in a bulk semiconductor. In the case of the hydrogenic impurity, however, the free-particle wave function is able to maximize its correlation by peaking at the position of the impurity in a scaled $1S$ orbital as given above. In the type-II systems the free particle is excluded from the dot (at least in the infinite barrier case) and so its wave function most resembles a $2P$-like wave function which vanishes at the center of the dot. This type of wave function will have a smaller orbital size because there is no $1S$ orbit with its two carriers to repel the carrier in the $2P$ orbital and so may be expected to have larger binding energy than the usual $2P$ wave function, but it is still expected to be less than the $1S$ state. For infinite barrier systems E_x will always be less than E_r and the OS will be zero for the case of infinite barriers and no wave-function leakage.

For finite barrier systems there will be wave-function leakage which will enhance the Coulomb interaction allowing the exciton binding energy to increase above that of the infinite barrier case for the same radius of dot. However, the type of wave-function leakage will also affect E_x. If the wave-function leakage arises from the confined particle leaking out of the dot and overlapping with the free-particle wave function, this will cause an admixture of a system in which one particle is fixed and the other particle correlates with it in a 3D environment—thus it should approach the E_r value of a $1S$ state given above. If the wave-function leakage arises from tunneling of the free particle into the dot this results in wave-function overlap of this wave function with the strongly peaked wave function of the confined particle producing a large enhancement of both E_x and OS. A similar effect is observed in the case of a hydrogenic impurity confined in a quantum well[17] where the wave function of the correlating particle is forced to be much more peaked about the impurity by the confinement which increases E_r. As the dot size increases in the finite barrier case, wave-function leakage from the confined state and from the free state decreases and the finite and infinite barrier cases approach each other. The OS for finite barrier cases arises from wave-function leakage and its magnitude will also reflect whether it is leakage into or out of the dot. Strong wave-function leakage into the well will result in a strong contribution to the OS since this tunneling wave function will overlap with a strongly peaked confined wave function.

In the GaAs/AlAs case the masses of both of the particles are reasonably heavy. The large effective mass of the confined particle allows it to remain bound for a small ra-

dius of the dot while the large mass of the correlating particle results in a small effective orbit and large E_x. Figure 4 shows that there is contribution from the wave function of the confined particle tunneling out of the dot and the correlating particle tunneling into the dot which is very effective in increasing both E_x and OS over the infinite barrier case. There is significant wave-function overlap at $r=0$ for the case of small dot radii in the GaAs/AlAs system as can be seen in Fig. 4 for a radius of 15.85 Å. The GaAs/AlAs system is also indirect in both real and k space. The InAs/GaSb system has a light confined particle which means that the confined level very quickly moves out of the confining potential as the size of the dot is decreased. This occurs before the correlating particle has felt the effects of the Coulomb attraction strongly enough to tunnel into the dot sufficiently. Therefore any increase in E_x and OS over the infinite barrier case is due to wave-function leakage of the confined particle out of the dot. The "effective" size of the dot is thus larger than its physical size, i.e., for the case shown in Fig. 5 for a radius of 44.9 Å the effective size is about 80 Å. Therefore E_x reflects two competing effects—the enhancement of the Coulomb interaction caused by wave-function overlap and the reduction in confinement and subsequent spreading out of the wave function. The wave-function overlap at $r=0$ is virtually negligible for all dot radii in the InAs/GaSb system. As can be seen in Figs. 6 and 7, this contribution to tunneling is not very effective in increasing E_x or OS. By consideration of these two systems we propose that the most favorable system in which to maximize the exciton binding would be one in which the wave-function tunneling came chiefly from the free correlating particle tunneling into the dot and overlapping with a strongly confined and strongly peaked wave function.

It is interesting to compare the type-I (Refs. 18 and 19) and type-II exciton systems in the dot. The discussion by Bryant[19] on correlation effects and quantum confinement shows very clearly that because a dot is confined in three dimensions, resulting in discrete confined states, correlation is only possible through interaction with higher states which move away from the ground state with increasing confinement. This makes the correlation of the electron and hole (both composed of these confined states) to form the exciton unfavorable. However, the Coulomb interaction does increase in the dot since the separation between the particles decreases. For small dots the kinetic energy of the system is that of the noninteracting ground state of the electron and hole while the interaction that reduced the total energy of the system is the Coulomb energy. It is this interaction energy lowering that we call the E_x in the dot. We can compare the E_x calculated by Brus[7] and Xia[18] for spherical dots of infinite confining potentials with the values of E_x we have calculated for the type-II system. These values are shown in Figs. 2 and 3 for comparison. We would expect the E_x

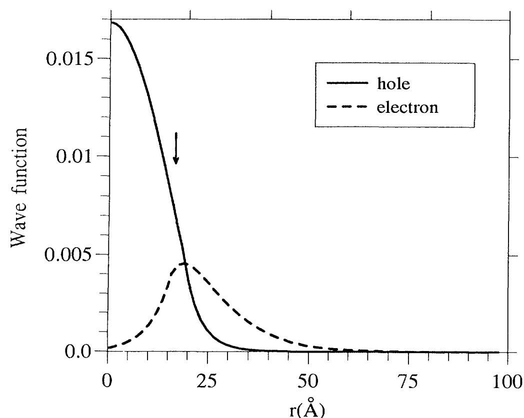

FIG. 4. The wave function of the electron (dashed line) and hole (full line) in atomic units as a function of the distance r from the center of the dot is shown for the GaAs/AlAs system for a dot of a radius of $r=15.85$ Å.

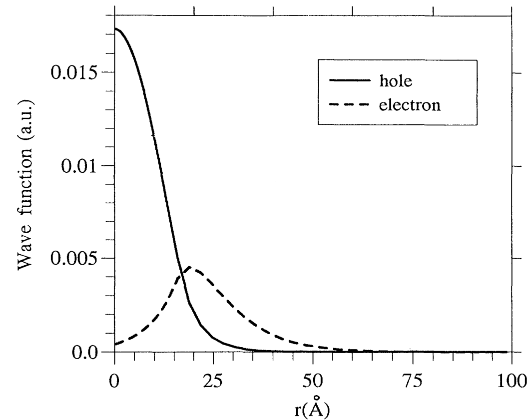

FIG. 5. The wave function of the electron (dashed line) and hole (full line) in atomic units as a function of the distance r from the center of the dot is shown for the GaAs/AlAs system for a dot of a radius of $r=44.9$ Å.

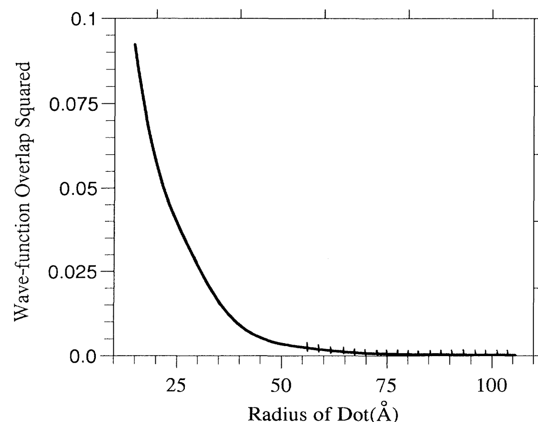

FIG. 6. The wave-function overlap squared $\langle \Phi(r_f)|\Psi(r_c)\rangle^2$ in atomic units as a function of the radius of the quantum dot in Å for the GaAs/AlAs system. This factor enters into the definition of the oscillator strength as given in Eq. (9) and will be the dominating factor in this equation. The system becomes type I when $a > 56$ Å, so the results shown for the type-II system for this region are shown crossed.

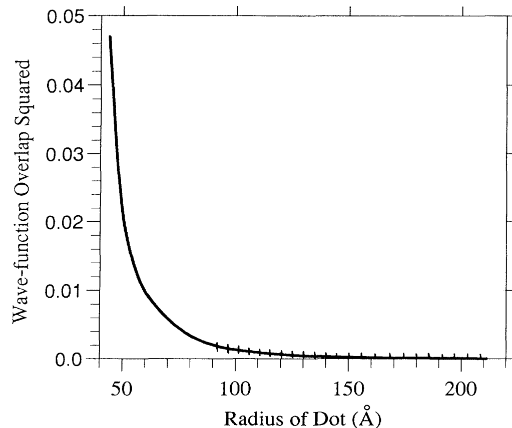

FIG. 7. The wave-function overlap squared $\langle \Phi(r_f)|\Psi(r_c)\rangle^2$ in atomic units as a function of the radius of the quantum dot in Å for the InAs/GaSb system. This factor enters into the definition of the oscillator strength as given in Eq. (9) and will be the dominating factor in this equation. The system becomes semimetallic when $a > 87$ Å, so the results shown for the type-II system for this region are shown crossed.

of the type-I systems to be larger than the E_x of the type-II systems but find in the GaAs/AlAs system that they are comparable for $30 < a < 70$ Å. This reflects the fact that the electron is in the X valley in the AlAs with its heavier m_c^* and also shows that the effect of the correlation of the "free" electron compensates for its relatively weak overlap with the hole wave function (compared to the type-I case). It is interesting to note that the plot of E_x as a function of dot radii for the type-II GaAs/AlAs system with finite barriers follows the form of the dependence of the E_x of the type-I GaAs/AlAs system showing that there is a contribution of type-I exciton behavior arising from the contribution of the electron and hole wave functions within the dot.

At some radius of the dot, however, the confined particle will become unbound[15]—at this point our model breaks down, although we can qualitatively describe the behavior. If the confined particle moves out of its potential and becomes free it is able to move away from the dot and form a free bulk exciton in the barrier material. For the GaAs/AlAs system this would result in a bulk exciton with an E_x of 23.6 meV in the AlAs and an overlap factor $\langle \Phi(r_f)|\Psi(r_c)\rangle^2 = 8(m_f^* m_c^*)^{3/2}/(m_f^* + m_c^*)^3$ of 0.88 for the electron in the X valley in the AlAs. For the InAs/GaSb system this would result in an E_x of 19.9 meV and an overlap factor of 0.26. If there is significant wave-function leakage into the dot there may exist a metastable state with the confined particle essentially free and the correlating particle in a $1S$-$2P$ hybridized state with significant wave function at $r=0$. It may be energetically quite favorable for this state to localized at this "defect" in the bulk barrier material due to the energy lowering from localization. Localization allows the correlating particle to have maximum correlation and the barrier system results in a bulk localized hydrogenic impurity energy E_r given earlier.

In conclusion we have calculated the exciton binding energy and envelope wave-function overlap required for the calculation of the oscillator strength in a type-II quantum dot for the GaAs/AlAs and InAs/GaSb systems. E_x for these type-II dot systems are very large compared to the 3D free-exciton binding energies showing *both* the effect of localizing one of the particles allowing the other particle of the exciton to fully correlate with it *and* the effect of confinement on the wave function within the dot. The E_x values for the type-II dot are also significantly larger than the exciton binding energies calculated for quantum-well systems[1] showing that the correlation in the extra two dimensions possible in the quantum dot enhances the exciton binding energy very greatly. The optical properties of type-II excitons are very different from that of type-I excitons or localized excitons[2] due to the electron and hole being indirect in real space. The envelope wave-function overlap will dominate the physics. This will be small but is expected to increase very strongly as the wave functions tunnel into the other semiconductor and to increase drastically when one of the particles moves out of its confining potential and becomes free to correlate with the other particle in a type-I situation. This great change in the overlap factor, which enters squared into the absorption of spontaneous emission, could be exploited by a number of physical perturbations to change from one condition to another. It should be emphasized that this is a zero-temperature theory and to be exploited temperature broadening would have to be considered.

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