Enhanced oscillator strength of interband transitions in coupled Ge/Si quantum dots

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We report a calculation of oscillator strength for the $\Delta_1-\Gamma_{25'}$ interband transition in two vertically coupled pyramidal Ge quantum dots embedded in Si. A six-band $\mathbf{k}\cdot\mathbf{p}$ formalism was used to study the $\Gamma_{25'}$ hole states, and a single-band approach was used to obtain the Δ_1 electron state interacting with the hole. The elastic strain due to the lattice mismatch between Ge and Si was included into the problem via the Bir–Pikus Hamiltonian. We find that when two dots are brought closely together, the oscillator strength may enlarge by a factor of about 2 as compared to the single-dot system. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2988469]

It is known that Ge/Si(001) quantum dots (QDs) represent heterostructures of the second type, in which charge carriers of different signs are separated by the heterointerface: holes are localized in Ge nanoclusters, and electrons are in the delocalized states of the conduction band of Si. However the tensile strain in the Si in the vicinity of the Ge island apex can cause a splitting of the sixfold-degenerate Δ valleys, thus yielding the localization of electrons near the Si/Ge boundary.[1–3] The localization energy of electron in a strain-induced potential well in a single Ge/Si QD is very small, ~7 meV.[4] This value enlarges vastly in multilayer Ge/Si structures with vertical stacking of Ge nanoclusters due to accumulation of strain energy from different dot layers.[3] The spatial separation of charge carriers involved in the interband absorption or radiative recombination process prevents the development of effective Ge-/Si-based optoelectronic devices. Recently it has been demonstrated that the vertically stacked Ge islands in Si show a strong photo- and electroluminescence at a wavelength of 1.5 μm up to room temperature.[5,6] Probably the enhanced emission from the coupled dots can be originated from the formation of collectivized or molecularlike electron and hole states in the structure, thus making the interband transitions in Ge/Si heterostuctures direct in a real space. In this work, we calculate the matrix elements for the $\Delta_1-\Gamma_{25'}$ interband transition involving the hole confined in two vertically stacked Ge QDs and the electron in the Si layer. We find that there is a distance between the dots at which the oscillator strength increases by a factor of about 2 as compared to the single QD.

We consider two *identical* vertically aligned pyramidal Ge nanoclusters with four {105}-oriented facets and a (001) base embedded into the Si matrix, as shown in Fig. 1. Each pyramid lies on a 4 ML Ge wetting layer. The nanoclusters are separated by a Si barrier of thickness t_{Si} measured from wetting layer to wetting layer. The pyramid base length l is 10 nm, and the pyramid aspect ratio h/l is fixed and equal to 0.1. The chosen geometrical parameters of the structure (the shape of Ge islands and their dimensions) correspond to a real situation often encountered in experiments. The typical size of computational cell (Ge wetting layers plus Ge islands plus Si environment) is $17.5\times17.5\times62.5$ nm^3 along x, y, and z axes, respectively. In order to check whether the calculation volume is large enough to give the proper (size-independent) result, we performed a numerical analysis also for different vertical sizes of computational domains ranging from 37.5 to 62.5 nm and found that the hole binding energy does not depend on the size of the supercell to within 1 meV of accuracy.

The finite element calculations of the three-dimensional (3D) spatial distribution of the strain components $\varepsilon_{\alpha\beta}$ were performed using the package COMSOL Multiphysics. The strain tensor elements are subsequently used as input to a strain-dependent Hamiltonian. The hole wave functions are calculated with a six-band $\mathbf{k}\cdot\mathbf{p}$ approximation (three valence bands and spin) based on the method of Bir and Pikus,[7] which includes spin-orbit and strain effects. A detailed analysis of single-particle hole orbitals in Ge/Si QD molecules can be found elsewhere.[8] Since the binding energies of holes are much larger than Coulomb interaction energies,[4] we neglect the influence of the Coulomb interaction between the electron and the hole on the hole states. The localization of the electron is due to, firstly, the inhomogeneous strain, which forms the confining potential for electrons in Si near the Ge pyramid apex and, secondly, the Coulomb attraction to the hole. The latter may dominate,[4] and, therefore, the electron-hole interaction should be taken into account while considering the electron states.

In the presence of strain, the 3D potential energy distribution of electrons in Δ_1 states with respect to the unstrained Si conduction band edge can be expressed as[9]

$$V_{\mathrm{strain}}(\mathbf{r}_e)=\Delta E_c x(\mathbf{r}_e)+\Xi_d\,\mathrm{Tr}[\varepsilon(\mathbf{r}_e)]+\Xi_u\varepsilon_{zz}(\mathbf{r}_e), \qquad (1)$$

where $\Delta E_c=0.34$ eV (Ref. 9) is the conduction-band offset between unstrained Si and Ge, $x(\mathbf{r}_e)=1$ on conditions that the

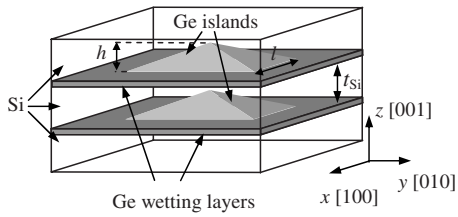

FIG. 1. Schematic picture of a Ge/Si double QD used for simulation.

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vector $\mathbf{r}_e$ points to the atom inside Ge islands (otherwise, $x(\mathbf{r}_e)=0$), and Ξ_d and Ξ_u are the deformation potentials. The electron wave functions $\psi_e(\mathbf{r}_e)$ are found by numericaly solving the 3D Schrödinger equation using the singe-band effective-mass approximation with the confining potential $U(\mathbf{r}_e)=V_{\text{strain}}(\mathbf{r}_e)+V_{eh}(\mathbf{r}_e)$, where

$$V_{eh}(\mathbf{r}_e)=\frac{e^2}{4\pi\epsilon}\int\frac{|\psi_h(\mathbf{r}_h)|^2 d\mathbf{r}_h}{|\mathbf{r}_h-\mathbf{r}_e|} \tag{2}$$

describes the electron-hole Coulomb interaction, $\psi_h(\mathbf{r}_h)$ is the hole wave function, ϵ is the dielectric constant, and $\mathbf{r}_{e(h)}$ denotes the electron (hole) position. We took the values of longitudinal and transversal effective masses at the Δ minimum in Si as m_z and m_{xy}, respectively; so $m_z=0.92m_0$ and $m_{xy}=0.19m_0$ (m_0 is the free electron mass).

In the dipole approximation, the oscillator strength of the transition between the valence and conduction bands is proportional to[10] $R=|\langle f|\mathbf{e}\cdot\mathbf{p}|i\rangle|^2$, where $|f\rangle$ and $|i\rangle$ represent the final and initial states, $\mathbf{e}$ is the direction of light polarization, and $\mathbf{p}$ is the momentum operator. The conduction-band (Δ_1) wave function is written as $|f\rangle=\psi_e(\mathbf{r}_e)u_c(\mathbf{r}_e)$, where $\psi_e(\mathbf{r}_e)$ is the electron envelop function and $u_c(\mathbf{r}_e)$ is the Bloch function at the bottom of the Δ_1 band. Correspondingly, in the valence band $|i\rangle=\psi_h(\mathbf{r}_h)u_v(\mathbf{r}_h)$, where $u_v(\mathbf{r}_h)$ is the Bloch functions at the top of the $\Gamma_{25'}$ band. Since the the periodic parts u_c and u_v vary rapidly over the characteristic length of variation of the envelop function ψ_e and ψ_h, the oscillator strength is reduced to evaluating the electron-hole wave-function overlap $M=\langle\psi_e|\psi_h\rangle$.[11] The result is $R\sim\|p\|^2\langle\psi_e|\psi_h\rangle^2$, where $\|p\|$ is the numerical version of the momentum matrix elements between Bloch functions. The hole states represent the six-component complex functions and are built from the heavy-hole (HH) states $|3/2,\pm 3/2\rangle$, the light-hole (LH) states $|3/2,\pm 1/2\rangle$, and the split-off (SO) states $|1/2,\pm 1/2\rangle$. Here $|J,J_z\rangle$ are the eigenstates of the effective angular momentum J and its projection J_z. The total hole envelop wave function, which is used for the calculation of matrix elements, is defined by $\psi_h=\sqrt{\Sigma_{J_z}\varphi_{J_z}^2}$. The sum extends over the four expectation values ($J_z=\pm 3/2$ for the HH and $\pm 1/2$ for the LH) of the $J=3/2$ multiplet and the two values $\pm 1/2$ for the SO $1/2$ state. The same approach is applied to find the matrix element for single QD, $M_{\text{SD}}=\langle\psi_e|\psi_h\rangle$.

As it has been shown in Ref. 8, coupled single-particle hole states of the Ge/Si double QD split into symmetric (σ_{S}) and antisymmetric (σ_{AS}) orbitals spread over two dots. Due to the competition of quantum mechanical tunneling and strain effects, the crossing of hole levels with different symmetries occurs with changing t_{Si}. At $t_{\text{Si}}\geqslant 4$ nm, σ_{AS} becomes the ground state of the system, replacing σ_{S}. Figure 2 shows the electron-hole wave-function overlap squared for σ_{S} and σ_{AS} hole states normalized on the squared matrix element for a single QD. The electron-hole overlap for the single QD with $l=10$ nm is calculated to be 32%. The factors $\langle\psi_e|\sigma_{\text{S}}\rangle^2$ and $\langle\psi_e|\sigma_{\text{AS}}\rangle^2$ enter into the definition of the oscillator strength of the $\sigma_{\text{S}}\rightarrow\psi_e$ and $\sigma_{\text{AS}}\rightarrow\psi_e$ interband transitions. It is clearly evident that there is a specific interdot separation ($t_{\text{Si}}\sim 3$ nm) at which the maximum oscillator strength can be obtained. For the $\sigma_{\text{S}}\rightarrow\psi_e$ transition, the oscillator strength increases by a factor of about 2 as compared to the single-dot system.

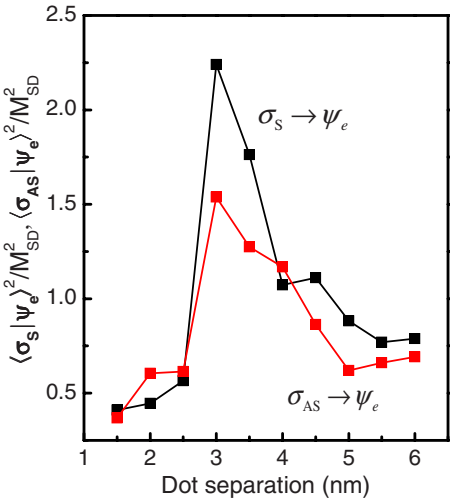

FIG. 2. (Color online) The electron-hole wave-function overlap squared for the σ_{S} (black) and σ_{AS} (red) hole states. The data are expressed in unit M_{SD}^2, where $M_{\text{SD}}=0.32$ is the matrix element for a single QD.

The physics of such behavior can be understood by considering the envelop electron and hole wave functions. Figure 3 shows the spatial distributions of the HH wave function component and the electron wave function for single and double QDs along the vertical symmetry axis of the structure (the z axis). The electron wave function ψ_e is mostly confined in Si and, in this region of t_{Si}, has a predominant form of a single peak. Due to the opposite parity of the electron ground state and the σ_{AS} hole orbital, the matrix element for the $\sigma_{\text{AS}}\rightarrow\psi_e$ transition is basically smaller than that for the $\sigma_{\text{S}}\rightarrow\psi_e$. In a single QD, the electron is localized in Si near the Ge island apex, where Si is more tensile strained, in

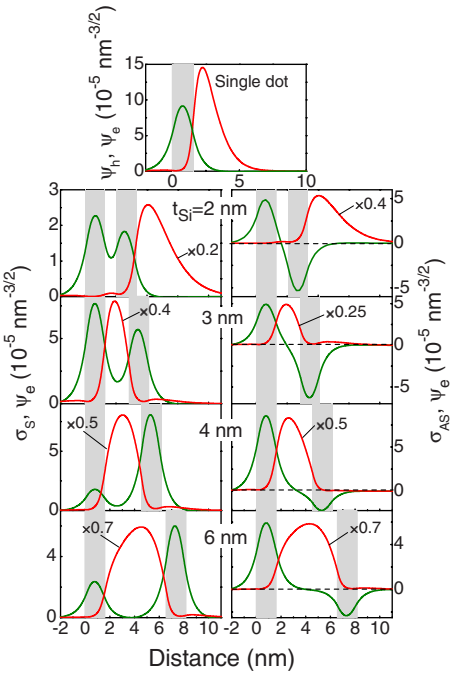

FIG. 3. (Color online) Spatial distribution of the hole (green) and electron (red) wave functions along the vertical symmetry axis of the dots (the z axis) for a single-dot and three selected double-dot structures. As a typical example of the hole states, we depict the HH component calculated using a 6×6 $\mathbf{k}\cdot\mathbf{p}$ model. The shaded regions correspond to Ge pyramids and wetting layers. The origin of the horizontal coordinates corresponds to the lower boundary of the lowest Ge wetting layer.

agreement with the previously published results.[4] A similar behavior is observed for the two dots separated by a shot distance. Two closely spaced QDs are "seen" by the electron as a single but thicker QD. Therefore, the electron-hole wave-function overlap at $t_{Si}<3$ nm is reduced below the value of the original single dot (see Fig. 2). At $t_{Si}\sim 3$ nm, the electron resides between two dots in a region where the probability to find the hole is finite due to quantum mechanical coupling between the dots. This makes the interband transition partially direct in the real space, thereby increasing the oscillator strength. When the two dots become separated by a more long distance, the coupling between the electron confined between the dots and the hole localized on the two long-distance dots is reduced again. The result of the electron-hole wave-function overlap includes two competing effects. On one hand, large interdot distance reduces the coupling between the electron confined between the dots and the hole spread over the two dots. On the other hand, the strain between the dots is also reduced, leading to a lower tunneling barrier for both electrons and holes, thus increasing coupling. The local shoulder of $\langle\psi_e|\sigma_S\rangle^2(t_{Si})$ dependence at $t_{Si}=4$ nm is a consequence of this competition.

Finally, one can expect that at $t_{Si}\rightarrow\infty$, the electron wave function will peak near both dots due to the attraction to the hole, and the wave-function overlap will be close to the value of the single dot. Probably, this is the reason for the oscillator strength's tendency to increase, observed in Fig. 2 at $t_{Si}>5.5$ nm.

In summary, we have calculated the electron-hole wave-function overlap required for the determination of the oscillator strength in type-II single and double Ge/Si QDs. An enhancement of the oscillator strength of the $\Delta_1-\Gamma_{25'}$ interband transition occurs in double Ge/Si QDs, depending on the interdot separation t_{Si}. Simulations show at least a twofold increase in the squared electron-hole wave-function overlap at $t_{Si}=3$ nm as compared to the single-dot structure. The results of this work have implications for the development of efficient Si-based optoelectronic devices.

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