Asymmetry of single-particle hole states in a strained Ge/Si double quantum dot

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A six-band $\mathbf{k}\cdot\mathbf{p}$ formalism was used to study single-particle hole states of two vertically aligned pyramidal Ge quantum dots embedded in Si and separated by a distance t_{Si}. The elastic strain due to the lattice mismatch between Ge and Si was included into the problem via Bir-Pikus Hamiltonian. The three-dimensional spatial strain distribution was obtained by finite element method. We found that at small interdot separation ($t_{Si} < 3.5$ nm), when the quantum-mechanical coupling between the dots is significant, the molecule-type hole orbitals delocalized fairly over the two dots are formed. The ground (excited) states correspond to symmetric σ_S (antisymmetric σ_{AS}) linear combination of single-dot states. However the splitting of σ_S from σ_{AS} is not symmetric, the average hole binding energy decreases with decreasing interdot separation. Strain effects start to play the dominant role at larger t_{Si}. In this region hole wave functions are localized on different dots, showing symmetry breaking. The most interesting property of energy spectrum is the crossing of levels with different symmetry which occurs with changing t_{Si}. At $t_{Si} > 3.5$ nm, σ_{AS} becomes the ground state of the system, replacing σ_S.

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Semiconductor quantum dots (QDs) look promising candidates as the active region in device-oriented applications. To develop instruments capable of operating at room temperature, it is necessary to fabricate QDs whose size is on the order of 10 nm or smaller. The synthesis of arrays of vertically coupled small-size artificial atoms has become possible with the use of semiconductor nanostructures that become self-assembled in the course of the heteroepitaxial growth of materials with a large lattice mismatch. Examples of systems developing self-assembled islands are the Ge/Si and InAs/GaAs heteropairs. The role of the driving force that causes the formation of vertically coupled QDs in multilayer heterostructures is played by the elastic strains arising in the medium.[1] The perturbation of the stress fields from a completely formed nanocluster penetrates into the thin growing layer of the matrix material and forms the nucleation site for a new island at the next "floor." As a result, columns of QDs aligned in the growth direction are obtained. Thus, the nonuniform strain distribution in both QD and surrounding matrix is an essential feature of quantum-dot heterosystems. Elastic deformation can cause significant change in the electron energy spectrum because it modifies the volume and shape of the crystal unit cell leading to a shift of energy extremes of Brillouin zone and degeneracy release due to lowering symmetry.

Strained Ge/Si(001) layers 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.[2] The inhomogeneous stress distribution in the vicinity of heterointerfaces and the mutual influence of the elastic strain fields from the coupled QDs result in a lack of horizontal symmetry (inversion) plane. Therefore, the first and second QDs forming the artificial molecule can be strained in different ways, even if they have identical sizes, shapes, and element compositions. The asymmetry of strain distribution should strongly affect the formation of molecule-type orbitals in double-dot systems,[3,4] which are now considered as the building blocks of a quantum information processing.[5–8] The aim of our study is the theoretical analysis of the electronic configuration of single-particle hole states in a Ge/Si double quantum dot.

We considered 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; 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.[9] 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 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 supercell to within 1 meV of accuracy.

The finite element calculations of three-dimensional spatial distribution of strain components $\varepsilon_{\alpha\beta}$ were performed using the package COMSOL MULTIPHYSICS with the approach described in Ref. 10. The strain tensor elements were subsequently used as input to a strain-dependent Hamiltonian. The electronic structure was calculated with a six-band $\mathbf{k}\cdot\mathbf{p}$ ap-

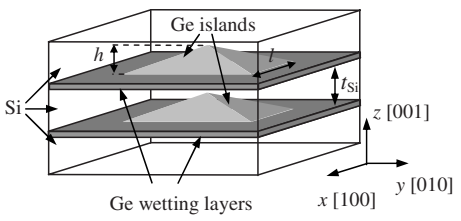

FIG. 1. Schematic picture of a Ge/Si double QD used for simulation of hole states.

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TABLE I. Material parameters used in calculations. Spin-orbit splittings Δ_0, deformation potentials a_v, b, and d (in eV, Ref. 17); Luttinger parameters (Ref. 18).

	Δ_0	a_v	b	d	γ_1	γ_2	γ_3
Si	0.04	2.46	−2.1	−4.8	4.28	0.34	1.45
Ge	0.30	1.25	−2.9	−5.3	13.38	4.24	5.69

proximation (three valence bands and spin), based on the method of Bir and Pikus,[11] which includes spin-orbit and strain effects. The contribution of electronic bands was neglected because of its small value (~0.5%) according to Ref. 12. The $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consists of four components,

$$H = H_{vv} + H_{so} + H_{\text{strain}} + U, \tag{1}$$

where H_{vv} contains terms which depend on wave vector $\mathbf{k}$, H_{so} describes spin-orbit interaction, H_{strain} is the strain-dependent contribution, and U accounts for the average valence-band offset at the Ge/Si heterojunction. In the basis of Bloch amplitudes, H_{vv} is given by Eq. (46) of the original work of Dresselhaus, Kip, and Kittel.[13] H_{strain} is the same as H_{vv} where there is the standard change in notation $k_{xy} \rightarrow \varepsilon_{xy}$ (Ref. 11). The spin-orbit Hamiltonian H_{so} is described by Eq. (12) in Ref. 14. $U=0$ in Si and $U=0.54$ eV in Ge.[15,16] Material parameters used in calculations are reported in Table I.

From the uncertainty principle, it follows that the electron binding energy in the ground state of coupled quantum dots is always higher than the electron energy in a single QD. Therefore, this kind of a stable state is called the bonding state. It usually corresponds to the symmetric combination of atomic wave functions (σ_S). Conversely, the antibonding state is formed by the antisymmetric orbital (σ_{AS}) which corresponds to a lower electron energy level. For nonidentical QDs, the states σ_S and σ_{AS} split with an energy difference $\Delta_{\text{SAS}}=\sqrt{\Delta_T^2+\delta^2}$ which consists of the energy difference between the isolated levels, δ, and the quantum-mechanical coupling energy, Δ_T. This is the conventional quantum-mechanical approach used to describe the formation of molecular orbitals in a system of tunnel-coupled QDs.[19] It should be mentioned that there is no inversion center in the double quantum dots under consideration. So the hole eigenstates can be neither symmetric nor antisymmetric in the full sense. Nevertheless we will follow the usual practice calling the σ_S and σ_{AS} states by symmetric and antisymmetric, correspondingly, although these names are not really justified.

Calculated hole binding energies in symmetric and antisymmetric states of Ge/Si double QD are presented in Fig. 2(a) as a function of dot separation t_{Si}. As distinct from the anticipated behavior following from the conventional quantum-mechanical model, the single-particle states in a Ge/Si double QD have some specific features. First, the splitting of σ_S from σ_{AS} is not symmetric: the average energy $E_{\text{SAS}}=(E_{\sigma_S}+E_{\sigma_{\text{AS}}})/2$ decreases with decreasing interdot separation, implying an asymmetry of single-particle hole states. Recent calculations[3] show similar behavior for electronic states of InAs/GaAs quantum-dot molecules. Second,

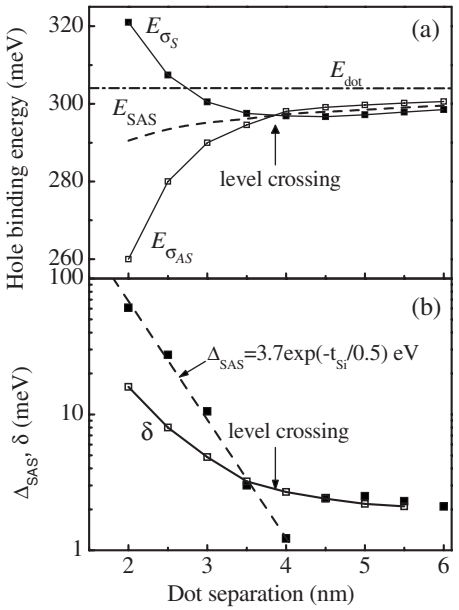

FIG. 2. (a) Evolution of hole binding energy in symmetric (σ_S) and antisymmetric (σ_{AS}) states as a function of the distance between QDs. As reference, also the ground-state energy of *isolated* single dot E_{dot} is shown. All energies are counted with respect to the valence-band edge in bulk Si. The dashed line is the average of E_{σ_S} and $E_{\sigma_{\text{AS}}}$. The σ_S and σ_{AS} cross at about 3.9 nm, as indicated by the arrow. (b) The energy difference between symmetrical and antisymmetrical states $\Delta_{\text{SAS}}=|E_{\sigma_S}-E_{\sigma_{\text{AS}}}|$ (filled squares) and between the isolated levels δ (empty squares). The dashed line corresponds to $\Delta_{\text{SAS}}=3.7\ \exp(-t_{\text{Si}}/0.5)$ eV.

the change in t_{Si} causes crossing between the energy levels corresponding to σ_S and σ_{AS} states. As a result, at $t_{\text{Si}}>3.5$ nm, the σ_{AS} orbital becomes the ground state of the system. The splittings of energy between σ_S and σ_{AS} and between the isolated levels (δ) are plotted in Fig. 2(b) on a logarithmic scale.[20] Two different regions are evident in Fig. 2(b). At $t_{\text{Si}}<4$ nm, $\Delta_{\text{SAS}}>\delta$ and decays exponentially. It can be fitted as $\Delta_{\text{SAS}}=3.7\ \exp(-t_{\text{Si}}/0.5)$ eV. We define this region as the strong-coupling region. Note the decay length of 0.5 nm is shorter than the decay length ~1 nm of Δ_{SAS} found for electrons in InAs/GaAs double dots[4] due to larger carrier effective mass in Ge/Si heterosystem. At longer distances the energy splitting $\Delta_{\text{SAS}}\sim\delta$ and becomes a weak function of the interdot separation varying approximately as t_{Si}^{-1}. In this weak-coupling region, Δ_{SAS} originates from asymmetry of strain distribution.

The symmetry breaking is also demonstrated in Fig. 3, where we plot the $|y\rangle$ component of the envelope wave function for the ground and first excited states. In the strong-coupling region, $\Delta_T>\delta$. Therefore, both σ_S and σ_{AS} states are extended quasiequally in both dots with the symmetric (bonding) state being the ground state. At larger distances an asymmetry of the molecular orbitals appears. The ground (excited) states are more localized on the bottom (top) dot. Finally, at $t_{\text{Si}}>3.5$ nm, the level structure becomes inverted: the asymmetric wave function turns out to be the ground state.

The anomalous behavior at large t_{Si} can be understood by

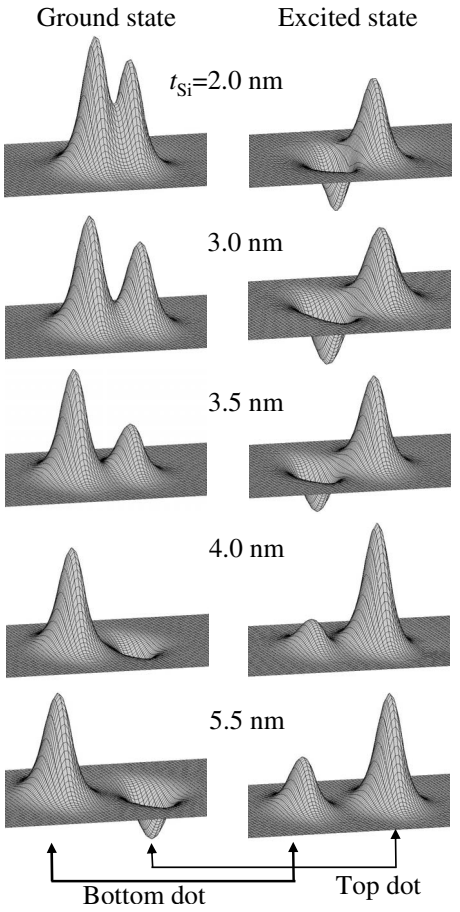

FIG. 3. Spatial distributions of the ground-state (left panel) and excited-state (right panel) hole wave functions in the (yz) plane, where the z axis goes though the vertical symmetry axis of the pyramids. Here we show only real part of the $|y\rangle$ component calculated in a basis of Bloch functions.

considering effects of strain on the band structure. From our calculations we observed that the hydrostatic strain $\varepsilon_h=\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}$ (not shown here) resides entirely inside the dots and is approximately the same for the double-dot structures and the single dot. Unlike the hydrostatic component, the biaxial strain $\varepsilon_b=\varepsilon_{zz}-0.5(\varepsilon_{xx}+\varepsilon_{yy})$ inside QDs is decreased when the two dots are brought closely together (Fig. 4). Furthermore, the biaxial strain field is different on both geometrically identical dots due to the lack of inversion symmetry with respect to the medium plane between the dots. The top Ge island proves to be less strained. The hole state in Ge QD is built mainly from valence-band states, namely heavy-hole (HH) states $|\frac{3}{2},\pm\frac{3}{2}\rangle$, the light-hole (LH) states $|\frac{3}{2},\pm\frac{1}{2}\rangle$, and the split-off (SO) states $|\frac{1}{2},\pm\frac{1}{2}\rangle$. Here $|J,J_z\rangle$ are the eigenstates of the effective angular momentum J and its projection J_z. In a bulk material, when no strain is present, spin-orbit effects raise degenerate HH and LH bands with respect to SO band. Biaxial strain lifts the degeneracy of the heavy- and light-hole bands at Γ point, and mixes light-hole and spin-split-off bands. For biaxial strain along [001] orientation, the model-solid theory[21] predicts the following strain-induced energy shifts with respect to the average valence-band energy E_{av}:

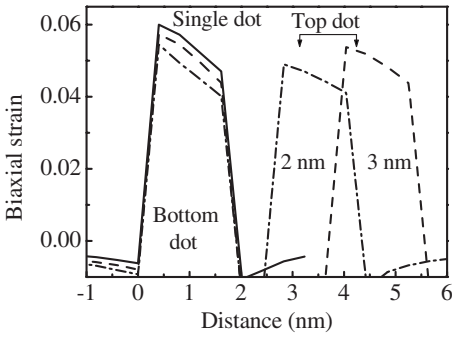

FIG. 4. Biaxial component of the strain in single dot (solid line) and double-dot structures for two selected interdot distances of 2 nm (dashed-dotted line) and 3 nm (dashed line) as a function of the position from the bottom boundary of the bottom wetting layer along the z axis through the pyramid apex.

$$\Delta E_{HH} = |b|\varepsilon_b, \tag{2}$$

$$\Delta E_{LH} = -\Delta E_{HH} - 0.5[(\Delta_0 - \Delta E_{HH}) - \sqrt{(\Delta_0 - \Delta E_{HH})^2 + 8(\Delta E_{HH})^2}], \tag{3}$$

$$\Delta E_{SO} = -\Delta E_{HH} - 0.5[(\Delta_0 - \Delta E_{HH}) + \sqrt{(\Delta_0 - \Delta E_{HH})^2 + 8(\Delta E_{HH})^2}]. \tag{4}$$

Under the action of strain, the split HH band remains a pure $|\frac{3}{2},\frac{3}{2}\rangle$ state, while LH and SO bands become mixtures of $|\frac{3}{2},\frac{1}{2}\rangle$ and $|\frac{1}{2},\frac{1}{2}\rangle$.

To analyze the hole state composition, we resort to the Luttinger-Kohn representation of the $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian. The basis of Bloch functions can be transformed into the Luttinger-Kohn basis using the set of functions given by Eq. (V.11) of the article by Luttinger and Kohn.[22] Results of the wave function expansion in the Luttinger-Kohn basis for σ_S and σ_{AS} states are shown in Fig. 5. The contribution of the states with $J_z=\pm\frac{3}{2}$ (the heavy-hole states) is predominant and amounts to about 85–90 %. In Ge regions, biaxial strain is positive (Fig. 4), and the heavy-hole band is shifted upward [see Eq. (2)]. Partial strain relaxation upon dot stacking in the coupled dot system causes downward shift of the HH states. This is why the energy of the bonding state E_{σ_S} is

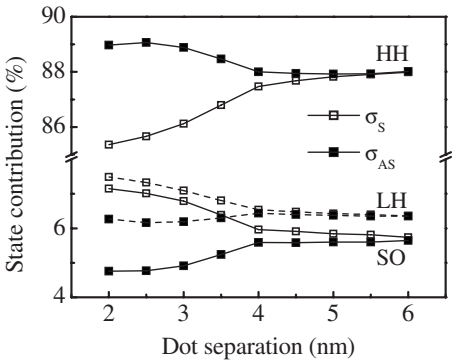

FIG. 5. Contribution of the HH, LH, and SO states for σ_S and σ_{AS} hole orbitals.

reduced below the value of the single dot in the weak-coupling region, where $\delta > \Delta_T$. The shorter interdot distance the less strain. Therefore the average energy E_{SAS} decreases with decreasing t_{Si}.

The strain asymmetry between the dots leads to splitting of the energy levels for the top and bottom QDs, δ [Fig. 2(b)]. The bottom dot is more strained and, hence, the respective energy level is deeper than that for the top dot. As a result, in the weak-coupling region, the ground hole state (σ_S or σ_{AS}, depending on t_{Si}) is localized on the bottom QD, while the excited state is mostly confined on the top one.

Now we discuss the crossing of levels with different symmetry observed in Fig. 2. In the strong-coupling region, the position of energy levels is determined by the interdot quantum-mechanical coupling and, hence, E_{σ_S} lies above $E_{\sigma_{\mathrm{AS}}}$, being the ground-state energy level at $t_{\mathrm{Si}} < 4$ nm. In the weak-coupling region, strain-induced energy shifts dominate over tunneling splitting. As we can see in Fig. 5, σ_{AS} state contains larger contribution of HH component than σ_S and smaller contribution of LH and SO. Therefore, σ_{AS} shifts to the bottom of the potential well, eventually replacing σ_S and causing level crossing. The question is why the $|\frac{1}{2}\rangle$ states make smaller contribution to σ_{AS}. Figure 6 demonstrates that HH state provides the main part in the dot center, while the tails of the hole wave function outside the dot are determined by LO and SO components. Since for asymmetric state the wave function changes the sign, there should be a surface near the middle of the barrier where the probability to find the hole is exactly equal to zero. On the contrary, for symmetric state this probability is finite at any distance. This clarifies deficiency of $|\frac{1}{2}\rangle$ states for σ_{AS}.

To conclude, we investigated the single-particle hole states in the double quantum dots made of two identical, vertically stacked, Ge/Si nanoclusters. As consequence of inhomogeneous strains, the symmetry of states is breaking. The splitting of bonding state, σ_S, from the antibonding one, σ_{AS}, is not symmetric; the average hole binding energy decreases with decreasing interdot separation. The change in interdot separation t_{Si} causes crossing between the energy levels corresponding to σ_S and σ_{AS} orbitals. As a result, at $t_{\mathrm{Si}} \gtrsim 4$ nm, the antibonding state σ_{AS} becomes the ground state of the system, replacing the σ_S state.

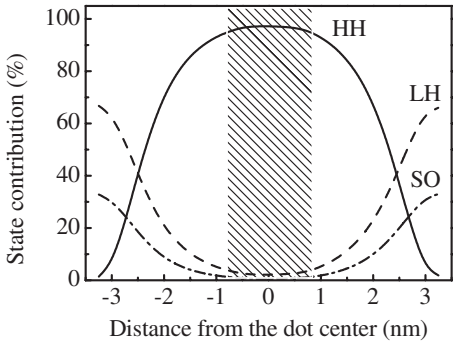

FIG. 6. Calculated spatial distribution of the relative contribution of HH, LH, and SO components in the wave function of the hole ground state for a single Ge/Si dot. The profiles are plotted along the vertical symmetry axis z of a pyramid. The shaded region corresponds to Ge pyramid and wetting layer.

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