Spin and valley control in single and double electrostatic silicene quantum dots

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We study quantum dots defined electrostatically within silicene. We determine the spin-valley structure of confined single- and two-electron systems, and quantify the effects of the intervalley scattering by the electron-electron interaction potential and the crystal edge. The double quantum dots are discussed in the context of the spatial symmetry of the extended orbitals. We determine the charge, spin and valley transition times induced by alternate electric fields. We show that the valley transition times can be changed within several orders of magnitude by the depth of the confinement potential. Also, the spin transition rates can be enhanced by orders of magnitude by the coupling of the bonding and antibonding orbitals mediated by the Rashba spin-orbit interaction.

I. INTRODUCTION

Quantum dots with electrostatic confinement defined in III-V materials are used for studies of the confined spin control [1, 2]. In graphene the spin coherence and relaxation times [56, 58] are long which should make this material attractive [8] for applications of confined spins in quantum computing [41]. However, the weakness of the spin-orbit interaction in graphene [9] excludes the spin manipulation by electric fields [10–13]. On the other hand, the graphene offers the valley [14, 15] instead of spin [16–18] for information processing. Nevertheless, the lack of the energy gap makes the purely confinement in pristine graphene excluded [19]. The gap can be opened in bilayer graphene [20–25] which solves the problem for electrostatic confinement [26–32]. However, the manipulation of the spin is still hampered by the weakness of the spin-orbit coupling [33].

An alternative material, in which both the spin and the valley [34] degrees of freedom could be controlled by electric fields, is the silicene [35, 36]. As in bilayer graphene, the perpendicular electric field opens the energy gap [37, 38] which allows for electrostatic confinement. Silicene is characterized by relatively strong intrinsic spin-orbit interaction [39, 41] with the coupling constant which by three orders of magnitude [36] exceeds the one for graphene. The fact inspires proposals for spin-active transport devices [42–47] defined in silicene.

Silicene was first grown on metals [48–52] and studies of the electronic properties require non-metallic substrates. Growth of silicene on AlN [53] and transition metal dichalcogenides (TMDCs) [35, 54, 55] was theoretically studied. A successful fabrication of a room-temperature field effect transistor was reported [56] for silicene on Al2O3. The Al2O3 substrate preserves the electron structure of free-standing silicene near the Dirac points [57]. Studies of silicene islands grown on graphite by van der Waals heteroepitaxy are also performed [58, 59].

In this work we determine the spin- and valley-structure of states electrostatically confined within silicene and consider manipulation of the spin and valley degrees of freedom using alternate electric fields of microwave or deep infrared frequency. The alternate electric fields were previously applied for states confined in carbon nanotubes [9, 60, 61]. The experiments [60, 61] resolve the spin and valley transitions using double quantum dots. Here, we consider one excess electron and an electron pair in single and double dots. We use the atomistic tight-binding approach that naturally accounts for the valley mixing effects of the crystal edge [92] and the intervalley scattering due to the short-range component of the Coulomb interaction [93, 94].

Previously, silicene flakes without the electrostatic confinement potential were studied [67, 70]. A type-I electrostatic quantum dot, that supports localization of conduction and valence band states was also discussed [71]. Here, we present a simpler set-up for electrostatic type-II quantum dots – that supports confinement of excess electrons in the conduction band only. We determine the intervalley scattering effects for the two-electron spectrum, and the tunnel coupling effects for the spatial symmetry of the wave functions in double quantum dots. We find that the spectrum for the double dots can be described in terms of separation of the spin-valley and spatial degrees of freedom. We demonstrate that both spin and valley transitions can be driven by AC electric fields and that the valley transition rates can be controlled in a large range by the tunable coupling to the edges of the flake. We also indicate that avoided crossings open by Rashba interaction between the bonding and antibonding states in double dots make the spin transition as fast as the spin conserving transitions.

II. THEORY

A. Single-electron Hamiltonian

We determine the single-electron eigenstates for an atomistic tight-binding Hamiltonian [41] which in the ab-
sence of the external magnetic field reads

\[
H_0 = -i \sum_{(k,l)\alpha} \hat{c}_{k\alpha} \sigma_{\alpha\beta} c_{l\beta} + i t_1 \sum_{(k,l)\alpha,\beta} \nu_{kl} \hat{c}_{k\alpha} \sigma_{\alpha\beta} c_{l\beta} + i t_2 \sum_{(k,l)\alpha,\beta} \mu_{kl} \hat{c}_{k\alpha} \sigma_{\alpha\beta} c_{l\beta} + i t_3 \sum_{(k,l)\alpha,\beta} F_z(r_k) \hat{c}_{k\alpha} \sigma_{\alpha\beta} c_{l\beta} + \sum_{k,\alpha} V_k c_{k\alpha} \hat{c}_{k\alpha} \]

with summations over the nearest-neighbor \(((k,l)\alpha)\), the next-nearest-neighbor \(((\langle k,l \rangle\alpha,\beta)\) ions, and the spins \((\alpha,\beta)\) of electrons localized at Si 3p_\perp orbitals. We use \(t_1 = 1.6\) eV \([36, 39, 41]\) for the nearest-neighbor hopping. The second term of Hamiltonian (1) is the intrinsic spin-orbit interaction \([36, 39, 41, 72]\) – the dominant spin-orbit coupling term for silicene. The intrinsic spin-orbit parameter is \(t_2 = 0.75\) meV \([39, 41]\), and \(\nu_{kl} = \pm 1\). The positive (negative) sign of \(\nu_{kl}\) is set for the counterclockwise (clockwise) next-nearest neighbor hopping via the common neighbor ion. The second line in Eq. (1) introduces the built-in Rashba spin-orbit interaction which results from the presence of a perpendicular electric field component for the buckled lattice, with \(t_1 = 1.6\) meV \([39, 41]\) and \(d_{kl} = r_{k}-r_{l},\) where \(r_{k} = (x_{k}, y_{k}, z_{k})\) indicates the position of the \(k\)-th ion, \(\mu_{kl} = \pm 1\), with plus for sublattice A and minus for sublattice B. The superscript \(z\) above the parentheses in the second and third line of Eq. (1) stands for the \(z\)-component of the vector operator defined by the cross product. The third line of Eq. (1) introduces the extrinsic Rashba interaction due to the external electric field perpendicular to the silicene plane, with \(t_3 = 0.589 \times 10^{-3}\) Å, which for \(F_z = 17\) meV/Å gives \(t_3 F_z = 10\) μeV \([39]\).

### B. Electrostatic quantum dot potential

The last term of Hamiltonian (1) introduces the electrostatic potential that should open the energy gap and define the confinement potential. In the calculations for a single quantum dot we model the potential as due to the setup that is depicted in Fig. (a). The silicene layer is sandwiched within a dielectric that separates two gates. The top gate contains a circular protrusion of radius \(R_p\). Solution of the Laplace equation for the system of Fig. (a) for \(V_b = -3.8\) V and \(V_t = 79.5\) V on the sublattices is presented in Fig. (b). The vertical electric field introduces potential difference between the ions at the bottom (A, black line in Fig. (b)) and top (B, red line in Fig. (b)) sublattice, and the difference opens the energy gap \([37, 38, 67, 71]\). The potential applied to the top gate is attractive for electrons and its protrusion that is closer to silicene layer induces the dip of the confinement potential for the conduction-band electrons on both sublattices A and B [Fig. (b)]. For the geometry assumed in Fig. (b) the potential has a form of a Gaussian of nearly equal depth / width on both sublattices. Based on this finding for \(V_k\) in Eq. (1) we set

\[
V_k = V(r_k) = \begin{cases} 
V_A(r_k) = -w - we^{-r_k^2/R_p^2} & \text{for } k \text{ in A} \\
V_B(r_k) = -w - we^{-r_k^2/R_p^2} & \text{for } k \text{ in B}
\end{cases}
\]

with \(R_p = 10\) nm, equal to the radius of the protrusion.

For the single quantum dot we consider a regular hexagonal flake with an armchair edge and side length of 22 nm [Fig. 2(b,c)]. The positions of the ions of the A sublattice \(r_k^A = k_1 a_1 + k_2 a_2\) are generated with the crystal lattice vectors \(a_1 = a \left( \frac{1}{2}, \frac{\sqrt{3}}{2}, 0 \right)\) and \(a_2 = a \left( 0, 0, 0 \right)\), where \(a = 3.89\) Å is the silicene lattice constant, and \(k_1, k_2\) are integers. The B sublattice ions are generated by \(r_k^B = r_k^A + (0, d, \delta)\), with the in-plane nearest neighbor distance \(d = 2.25\) Å and the vertical shift of the sublattices \(\delta = 0.46\) Å.

### C. Electron confinement

The energy levels calculated with the tight-binding Hamiltonian are displayed as a function of the potential.
FIG. 2. (a) Energy levels as a function of the potential depth $w$. The colorscale indicates the localization of the energy levels in the quantum dot calculated as the integral of the charge density within the radius of $1.1 R_p$ from the dot center. The results are calculated in the absence of the external magnetic field. The conduction (valence) band states are found for $E > 0$ ($E < 0$). (b) The $K'$ spin-down ground-state electron density on the A (b) and B (c) sublattices for $w = 200$ meV. In the continuum approximation the angular momentum quantum number is 0 on the A sublattice and -1 on the B sublattice.

depth $w$ in Fig. 2(a). The colorscale in Fig. 2(a) shows the localization of the eigenstates within the range of $1.1 R_p$ from the center of the electrostatic potential. The localized states are found only in the conduction-band side ($E > 0$) of the spectrum. The states of the valence band are localized outside of the dot. The energy gap rapidly increases with $w$. Namely, for $w = 100$ meV, 200 meV and 300 meV the gap is 170 meV, 290 meV and 380 meV, respectively.

The wave function of the spin-down ground state is given for $w = 200$ meV in Fig. 2(b,c). The electron density of the localized conduction band states is nearly entirely localized at the sublattice A.

D. Magnetic field

The external magnetic field is introduced by the Peierls phase via modification of the hopping terms $c_{k\sigma}^\dagger c_{k\sigma'} \rightarrow c_{k\sigma}^\dagger c_{k\sigma'} e^{i\mathbf{r}_k \cdot \mathbf{A}}$ with the vector potential $\mathbf{A}$. For the perpendicular magnetic field $\mathbf{B} = (0, 0, B)$ we use the symmetric gauge $\mathbf{A} = (-Bx/2, By/2, 0)$. With the Peierls phase included the energy operator changes to $H_0 \rightarrow H_A$, and the Hamiltonian is completed by the spin Zeeman term

$$H_B = H_b + \frac{1}{2} g \mu_B B \sum_{k, \alpha} \sigma_z c_{k\alpha}^\dagger c_{k\alpha},$$

where $\mu_B$ is the Bohr magneton $g = 2$ and is the Landé factor.

E. Continuum approximation

When the electron system gets localized inside the quantum dot, the intervalley mixing by the edge becomes negligible. Then, the valley index becomes a good quantum number. In order to identify the valley we perform calculations using the continuum approximation [73] of the tight-binding Hamiltonian [1] that keeps track of the diagonal intrinsic spin-orbit interaction. For the identification of the quantum numbers we neglect the Rashba coupling that, although crucial for spin manipulation, produces only a slight modification to the energy levels and majority-spin eigenfunctions. For the wave function with two components, each corresponding to a single sublattice $\psi = (\psi_A, \psi_B)$, the continuum Hamiltonian reads [73]

$$H_\eta = \hbar V_F (k_x \tau_x - \eta k_y \tau_y) + V(r) r_z + \frac{g \mu_B B}{2} \sigma_z - \eta \tau_z \sigma_3 \sqrt{3} t_x,$$

where the valley index is $\eta = 1$ for the $K$ valley and $\eta = -1$ for the $K'$ valley, and $\tau_x$, $\tau_y$ and $\tau_z$ are the Pauli matrices in the sublattice space, $\mathbf{k} = -i \nabla + \frac{e}{\hbar} \mathbf{A}$, and $V_F = \frac{3\hbar t_x}{2\eta}$ is the Fermi velocity. For the circular potential $V(r)$, the $H_\eta$ Hamiltonian eigenstates $\Psi_\eta$ are also eigenstates of the valley-orbital angular momentum operator of form $J_z = L_z \left( \begin{array}{c} 1 \\ 0 \end{array} \right) + \eta \frac{\hbar}{2} \tau_z$, where $L_z = -i \hbar \frac{\partial}{\partial \phi}$ stands for the operator of the orbital angular momentum.

$$\Psi_\eta = \left( \begin{array}{c} f_A(r) \exp(i\phi) \\ f_B(r) \exp(i(l + \eta)\phi) \end{array} \right)$$

where $l$ is the orbital quantum number, and the eigenequation for the radial functions reads

$$(V^A(r) - \eta 3\sqrt{3} t_x \sigma_z + \frac{g \mu_B B \sigma_z}{2}) f_A$$

$$+ V_F \left[ -\eta \frac{i\hbar}{r} (l + \eta)f_B - i \hbar f'_B - \eta \frac{iBr}{2} f_B \right] = E f_A, \quad (6)$$

$$(V^B(r) + \eta 3\sqrt{3} t_x \sigma_z + \frac{g \mu_B B \sigma_z}{2}) f_B$$

$$+ V_F \left[ \eta \frac{i\hbar}{r} f_A - i \hbar f'_A + \eta \frac{iBr}{2} f_A \right] = E f_B. \quad (7)$$

The system of eigenequations [77] is solved with a finite difference method.

F. Electron pair

In graphene flakes, the Coulomb interaction of the excess electrons can be strong enough to generate an extra electron and hole pair [75]. The present electrostatic
quantum dot attracts the electrons but repulses the holes and the confinement potential does not support localized hole states of the valence band [Fig. 2(a)]. The cost of generation of the electron-hole pair is of the order of the energy gap, e.g. 290 meV for \( w = 200 \text{ meV} \). Even for the small size of the present dot – the electron-electron interaction energy is eight times smaller than 20 meV (for \( w = 200 \text{ meV} \)). For a pair of confined conduction band electrons the generation of extra electron-hole pair will not decrease the electron-electron repulsion, since the extra electron is added to the dot, and the hole needs to stay outside. For that reasons below we fix the number of conduction band electrons [74] to two and consider the Hamiltonian

\[
H_{2e} = \sum_i d^\dagger_i d_i E_i + \frac{1}{2} \sum_{ijkl} d^\dagger_i d^\dagger_j d_k d_l V_{ijkl},
\]

where \( d^\dagger_i \) is the electron creation operator for the single-electron energy level \( E_i \) and the Coulomb matrix elements read

\[
V_{ijkl} = \kappa \langle \psi_i (r_1) \psi_j (r_2) \rangle \frac{1}{|\mathbf{r}_{12}|} |\psi_k (r_1) \psi_l (r_2)\rangle,
\]

where \( \kappa = e^2 / (4\pi \varepsilon_0) \) and we use the Al\(_2\)O\(_3\) dielectric constant \( \varepsilon_0 = 9.1 \). We integrate the Coulomb elements for the single-electron wave functions \( \psi \) spanned by the Si atomic orbitals 3\( p_z \),

\[
\psi_i (r_1) = \sum_{k,\sigma} C_{k,\sigma_i}^i p_{z_k}^\sigma (r_1),
\]

where the summation over spin accounts for the effects of the non-spin-diagonal Rashba interaction. Although the spins are nearly polarized perpendicular to the silicene plane by the spin-diagonal intrinsic-spin-orbit interaction, the trace contribution of the minority spins allows for the spin transitions. The Coulomb matrix element reads

\[
V_{ijkl} = \kappa \langle \psi_i (r_1) \psi_j (r_2) \rangle \frac{1}{|\mathbf{r}_{12}|} |\psi_k (r_1) \psi_l (r_2)\rangle = \frac{e^2}{|\mathbf{r}_{12}|} \sum_{a,\sigma_a; b,\sigma_b; c,\sigma_c; \delta d,\delta_d} |C_{a,\sigma_a}^i C_{b,\sigma_b}^j C_{c,\sigma_c}^k C_{d,\delta_d}^l \delta_{\sigma_a,\sigma_b} \delta_{\sigma_c,\delta_d} \rangle \langle \psi_a (r_1) \psi_b (r_2) \rangle \frac{1}{|\mathbf{r}_{12}|} |\psi_c (r_1) \psi_d (r_2)\rangle.
\]

For the Coulomb integral we apply the two-center approximation [74]

\[
\langle \psi_a (r_1) \psi_b (r_2) \rangle \frac{1}{|\mathbf{r}_{12}|} |\psi_c (r_1) \psi_d (r_2)\rangle = \frac{1}{|\mathbf{r}_{ab}|} |\psi_a (r_1) \psi_b (r_2)\rangle = N_z (1 - \frac{2Z}{r}) \exp (-Zr / 3),
\]

where \( N \) is the normalization constant and \( Z \) is the effective screened Si nucleus charge as seen by 3\( p_z \) electrons. The single-center integral can then be calculated analytically and is equal to \( I_{a=b} = \frac{3577}{46090} N_z Z \). The Slater screening rules for 3\( p \) electrons produce \( Z = 4.15 \), then \( I_1 = 8.76 \text{ eV} \).

The Hamiltonian [3] is diagonalized with the configuration interaction approach in the basis of up to \( \sim 3000 \) two-electron Slater determinants constructed from the lowest-energy eigenfunctions of the single-electron Hamiltonian (3).
G. Driven transitions

We study the charge, spin and valley dynamics for the system subject to an in-plane AC electric field. The time-dependent Hamiltonian for the field oriented along the $y$ axis reads

$$H_t = H_B + H' = H_B + eF_{ac} \sum_{k,\alpha} y_k \sin(2\pi vt) c_{\alpha k}^{\dagger} c_{\alpha k}. \quad (12)$$

The amplitude and the frequency of the AC electric field are denoted by $F_{ac}$ and $\nu$, respectively. For integration of the time dependent Schrödinger equation $i\hbar \frac{\partial \Psi}{\partial t} = H \Psi$, we use the eigenstates of the stationary Hamiltonian ($H_B \psi_n = E_n \psi_n$),

$$\psi = \sum_n A_n(t) \exp(-\frac{i E_n t}{\hbar}) \psi_n,$$

which gives the following system of equations

$$i\hbar \frac{dA_n(t)}{dt} = \sum_n A_n(t) eF_{ac} \sin(2\pi vt) y_{kn} e^{-i \frac{E_n - E_k}{\hbar} t}, \quad (14)$$

with the dipole matrix elements $y_{kn} = \langle \Psi_k | y | \Psi_n \rangle$.

For two electrons the system of equations for description of the time-dependence is formally identical, only with $E_n$ standing for the two-electron eigenenergies and the matrix elements $y_{kn}$ calculated for the two-electron wave functions. In the calculations we set the stationary ground-state in the initial condition and study the system dynamics for the AC pulse duration of 3.74 ns.

III. A SINGLE QUANTUM DOT

A. Single-electron states

The splitting of the energy levels in Fig. 3(a) at large $w$ is due to the intrinsic spin-orbit coupling. Each of the energy levels is two-fold degenerate. The structure of the low-energy spectrum for the conduction band states localized in the dot is displayed in Fig. 3. Figure 3(a) shows the results of the atomistic tight-binding calculations with the color of the lines showing the spin-up and spin-down states. In Fig. 3(b) we plotted, for comparison, the results of the continuum approach. In Fig. 3(b) the levels are labelled by the valley index $K$ or $K'$, the spin $u$ or $d$ in the subscript for the spin-up and spin-down states, and the integer shows the angular momentum quantum number on the A sublattice. For comparison Fig. 3(c) indicates the results without the spin-orbit coupling, when at $B = 0$ each energy level is degenerate with respect to the valley and the spin. The intrinsic spin-orbit coupling introduces the valley-spin interaction that splits the fourfold degenerate states to spin-valley doublets. The states of opposite valleys produces electrical currents of opposite orientations. The lower-energy (higher) doublets correspond to parallel (antiparallel) orientation of the orbital and spin magnetic moments.

B. The electron pair

Near the ground-state each of the two electrons occupies the states of the two lowest-energy single-electron doublets. In consequence, the two-electron ground-state is nearly sixfold ($\binom{5}{3}$) degenerate. The spectrum in the absence of the spin-orbit interaction is displayed in Fig. 4(a) with the dominant contributions to the two-electron wave function given in the Figure near the energy levels. At $B = 0$, one finds a ground-state triplet, and an excited state doublet that is next followed by a singlet.

Let us discuss the structure of the energy levels in Fig. 4(a). For the spin-down polarized ground-state $K_d K_d$ the interaction integral $I = \langle \psi | K_d K_d \psi \rangle$ with the antisymmetrized wave functions is $I = \langle K_d(1) K_d(2) | K_d(1) K_d(2) \rangle - \langle K_d(2) K_d(1) | K_d(1) K_d(2) \rangle = E_C - E_X$, where $E_C$ and $E_X$ are the Coulomb and exchange integrals, respectively. The exchange integral is non-zero due to intervalley scattering induced by the short-range component of the electron-electron interaction potential. The same result is obtained for the other spin-polarized state of the ground-state triplet $K_u K_u$.

For the $K'$-valley-polarized state $K'_d K'_d$ the interaction integral is $I_2 = E_C \mp (K'_d(1) K'_d(2) | K'_d(1) K'_d(2) \rangle - \langle K'_d(2) K'_d(1) | K'_d(1) K'_d(2) \rangle = E_C \mp 0$. Here, the exchange integral vanishes due to the spin mismatch of the wave functions for both the first and the second electron in the integral. The same result is obtained for the $K$-valley-polarized state $K_d K_u$ state. Therefore, at $B = 0$ the valley-polarized states $K'_d K'_u$ and $K_d K_u$ form a degenerate doublet at the energy $E_x$ above the ground-state energy level [see Fig. 4(a)].

In Fig. 4(a) there are two energy levels which are neither spin nor valley polarized $\binom{5}{2} (K'_d K_u \pm K'_u K_d)/2!$. For these states the interaction integral is $I_3 = C \mp E_X$. Hence, the lower-energy state enters the ground-state triplet, and the other is the singlet at the energy of $2E_X$ above the ground state.

The role of the short-range component of the Coulomb interaction for the intervalley exchange can be illustrated by comparison of Fig. 4(a) and Fig. 4(b). In Fig. 4(b) we plotted the spectrum that is obtained for the Coulomb potential $1/r_{12}$ replaced by a function $V(r_{12}) = \min(1/r_{12}, a/a)$, where $a$ is the silicene lattice constant. This potential removes the shortest-range maximum of the Coulomb interaction. In Fig. 4(b) we notice that the splitting of the energy levels is reduced nearly 10 times. The modified Coulomb interaction is used only in Fig. 4(b).
Figure 4(c) shows the results with the spin-orbit interactions included. The states that were not polarized neither in spin nor in valley that in Fig. 4(a) were split only by a small value of $2\Delta SO$ now differ in the energy much more, i.e., by twice the single-electron spin-orbit splitting ($2\Delta SO$).

In the ground-state of Fig. 4(c) the configuration $K_d'K_u$ with both electrons in the states of the lowest single-electron doublet [cf. Fig. 3(a, b)] is dominant. The contribution of the excited doublet with interchanged valley indices $K_u'K_d$ is only $\approx 4\%$. This contribution corresponds to both electrons in the higher-energy single-electron doublet. Since the contribution is small, the intervalley exchange energy is negligible.

In the four states in the center of the spectrum in Fig. 4(c) one of the electrons occupies a single-electron state of the lower doublet, and the other a state of the higher doublet. These states at $B = 0$ form two doublets. The energy of the spin-polarized doublet is lower by the intervalley exchange $E_X$ as in Fig. 4(a).

C. Transitions in the alternate electric field: the single-electron

For the system in the alternate electric field the transition times are proportional to the inverse of the dipole matrix elements which are displayed by lines in Fig. 5(a) as functions of the potential depth $w$. The results are calculated for the $K_d'$ ground state as the initial state. In Fig. 5(b) the spin-flip (valley-flip) transition is obtained for $K_u'$ (Fig. 6(b)) as the final state. The matrix element for the simultaneous flip of both the valley and the spin $K_d' \rightarrow K_u$ is too small to fit in the Figure. We find that the matrix element for the valley flip vanishes for large $w$. The armchair edge of the flake is responsible for the intervalley coupling (22). For large $w$ the confined states are entirely localized within the dot (Fig. 2), and the intervalley flip by the electric field is no longer possible.

In Fig. 5(b) we translate the matrix elements to the transition times as obtained for the amplitude of the electric field of $F_{AC} = 2$ kV/cm. For shallow confinement ($w \approx 150$ meV) the valley flips are very fast, of the order of 100 ps. For $w = 400$ meV the valley flip times are as large as 20 $\mu$s. On the other hand the spin-flip times are about 1 to 2 ns and weakly depend on the potential depth. Note, that the spin flip time slightly increases at large $w$, which might be counterintuitive since the electric field and thus the extrinsic contribution to the Rashba coupling is enhanced for larger $w$. However, the orbital extent of the wave functions decreases with growing $w$, and the latter effect is dominant for the values of the dipole matrix elements.

Figure 6 shows the results of the time-dependent calculation as function of the AC frequency $\nu$ for $F_{AC} = 2$ kV/cm with values of $w$. The magnetic field is set to 1 T, the system is started in the $K_u'$ ground state and the simulation lasts 3.74 ns. The plots in Fig. 6 show the maximal square of the projection of the time dependent wave function on the stationary states. For both the spin transition to $K_u'$ and the valley transition to $K_d$ we list the times upon which occupation of the final state exceeds 10% and 50%. For $w = 250$ meV [Fig. 6(c)] the valley transition time is already much longer than time covered by the simulation.
FIG. 5. (a) Solid lines show the transition matrix elements \( y_{ij} = \langle \phi_j | \gamma_i | \phi_i \rangle \) for a single excess electron confined in the quantum dot. For the initial wave function \( \phi_i \) we take the ground state \( K_u' \) wave function at 1 T. The spin-flipping matrix element is calculated for the \( \phi_j \) set as \( K_u' \). The valley transition matrix element is obtained for \( \phi_j \) set as \( K_d' \) energy level — the highest energy level for the quadruple of lowest-energy conduction band states in Fig. 3(b). The points correspond to the results obtained for two confined electrons. The results are obtained for the ground-state set as \( \phi_i \) [see the spectrum Fig. 4(c)]. The valley (spin) flipping transition obtained for \( \phi_j \) identified with the first [black line in Fig. 4(c)] [second [blue line in Fig. 4(c)] energy level wave function. The matrix elements for both spin and valley flips are to small to enter the plot. (b) Same as (a) only for the transition times obtained for the AC electric field amplitude of \( F_{AC} = 2kV/cm \), as \( 0.103/y_{ij} [\mu m \times ns] \).

D. Transitions in the AC electric field: the electron pair

The dot symbols in Fig. 5 indicate the transition matrix elements and transition times for the electron pair. The results were calculated for the ground-state of Fig. 4(c) to \( K_u'K_u' \) or \( K_d'K_d' \) excited states, where the listed spin-valley configurations stand for the two-electron Slater determinants. The dominant contributions to the ground state wave function is \( \psi = (K_u'K_u + cK_d'K_d)/N \), where \( N \) is the normalization constant and \( c \) is small \( (|c|^2 << 1) \). The transition to the \( K_u'K_u' \) state is identified with the valley transition since it involves the valley flip \( K_u'K_u \rightarrow K_u'K_u \) in the dominant term of the ground-state wave function, or \( K_u'K_u \rightarrow K_u'K_u \) in the smaller term. For the similar reason the transition from the ground state to \( K_d'K_d \) can be identified with the spin flip.

Figure 5 shows that the spin-flip times as calculated for the electron pair are very close to the results for the single-electron. The two-electron results for the valley flip follow the single-electron trend, but for large \( w \), when the system is separated from the edge, the short range component of the electron-electron enhances the valley transition rates.

The results for the time-resolved simulation of the driven transitions in the two-electron quantum dot are presented in Fig. 6 for the ground state in the initial condition [cf. Fig. 4(c)]. The transitions to spin-polarized excited state \( K_u'K_d \) and \( K_d'K_u \) both involve the spin flip of one of the electrons in the ground state. At resonant energies it takes the driven wave function about 0.3 ns (1 ns) to reach about 10% (50%) admixture of the spin-polarized state. The valley flips which are very fast (several ps) for shallow confinement potential, dissappear for larger \( w \).
IV. DOUBLE QUANTUM DOT

A. Single electron in a symmetric pair of quantum dots

For description of the double quantum dot we consider a generalization of the model confinement of Eq. (2)

\[
V_A^t(r) = w + \min \{ V^t(r), V^u(r) \},
\]

\[
V_B^t(r) = -w + \min \{ V^t(r), V^u(r) \},
\]

with

\[
V^t(r) = -w_l \exp \left( -\frac{(x + l/2)^2 + y^2}{R_p^2} \right),
\]

\[
V^u(r) = -w_u \exp \left( -\frac{(x - l/2)^2 + y^2}{R_p^2} \right),
\]

where \(w_l\) and \(w_u\) are the potential depths of the quantum dot below and above the \(y = 0\) axis, respectively, and \(l\) is the distance between the dot centers. This form of potential for \(w_l = w_u\) allows one to obtain the single dot potential both at \(l = 0\) and in the limit of large \(l\). For the study of the double dot the vertical edge of the flake was extended to 46.3 nm (see Fig. 8).

The energy spectrum for a symmetric pair of dots \(w_l = w_u\) is displayed in Fig. 8(a). For the symmetric pair of dots the eigenstates can be labelled by the parity operator eigenvalues. For both the valleys the continuum Hamiltonian including the Rashba interaction \([73]\) for a point-symmetric potentials commutes with the parity operator \(P_4\)

\[
P_4 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix},
\]

with the two first (last) components that correspond to the spin-up (spin-down) wave functions, the subscripts denote the sublattice. In the definition of \(P_4\), the scalar parity operator \(P\) is used that inverts the argument of a scalar wave function \(Pf(x) = f(-x)\).

In Fig. 8(a) the energy levels are labelled by the valley index \(K\) or \(K'\) and the subscript \(e\) or \(o\) for even and odd \(P_4\) eigenstates that correspond to the eigenvalue +1 or −1, respectively. The spin of the levels is marked by the color of the lines.

In Fig. 8(d-g) we plot the ground-state electron densities as obtained with the atomistic approach for the spin-up (d,e) and spin-down (f,g) components on sublattice A (d,f) and B (e,g) for \(B = 0.5\) T and \(l = 18\) nm. The ground-state is the lowest odd \(P_4\)-parity \(K'\) spin-down energy level. The spin-down A sublattice is dominant [Fig. 8(f)] as for the single-dot ground state and the spin-up components are very small [Fig. 8(d,e)], which results from the essential weakness of the Rashba interaction in silicene. The spin-down A and the spin-down B [Fig. 8(d,g)] components vanish at the origin, which results from the spatial antisymmetry of these components with respect to the point inversion \(P\). The two other wave function components are even eigenfunctions of the scalar parity operator \(P\).

The energy spectrum of Fig. 8 at large \(l\) is a degenerate version of the single-dot spectrum for \(l = 0\). Let us discuss the energy levels starting from \(l = 30\) nm and going to lower \(l\) values. The splitting of the energy levels at finite \(l\) results from activation of the tunnel coupling between the dots. Since the A sublattice component of the wave functions is dominant, one can attribute the bonding character to: spin-down odd \(P_4\) parity states and spin-up even \(P_4\) parity states. The corresponding energy levels fall in energy when \(l\) is reduced below 30 nm. The remaining states: the spin-down even-parity and the spin-up odd-parity energy levels are antibonding and increase when \(l\) is reduced below 30 nm.
As a consequence of the tunnel coupling, the energy levels of opposite bonding-antibonding character change their order near \( l = 16 \text{ nm} \) – the region enlarged in Fig. 8(b). The energy levels which correspond to the same valley but opposite spin enter into an avoided crossing that is open by the Rashba spin-orbit coupling, which mixes the spins of the eigenstates within the range of the avoided crossing.

The avoided crossing of energy levels appears at a narrow range of \( l \) but it leaves a much wider signature on the transition matrix elements. They are displayed in Fig. 8(c) for the spin-down \( K'_o \) ground state set as the initial state of the transition. The valley and spatial parity selection rules allow the transition only to \( K'_o \) states [cf. Fig. 8(c)]. Near the avoided crossing of Fig. 8(b), the spin-flipping transition increases by two orders of the magnitude [Fig. 8(c)]. The result indicates that one can arrange for very fast spin flips using double dot potentials.

**B. Single electron: asymmetric double dots**

The ideal symmetry assumed for Fig. 8 can hardly be achieved experimentally. For that reason we considered also asymmetric systems. In Fig. 9(a) we plotted the energy levels as functions of the interdot distance for the dot defined at the lower part of the flake \( y < 0 \) made deeper by 6 meV (\( w_l = 1.03 w_u \)). The colors in Fig. 9(a) indicate the localization of the charge density. For \( l < 15 \text{ nm} \) the electron density is distributed nearly equally at both sides of the \( y = 0 \) line which indicates a strong tunnel coupling between the dots. For \( l > 15 \text{ nm} \) the states exhibit a distinctly stronger localization in one of the dots. The avoided crossing open by the Rashba interaction discussed above is still observed [Fig. 9(b)]. At the center of the avoided crossing not only the spins are exchanged, but the charge density is equally distributed between the dots.

Fig. 9(c) shows the dipole matrix elements for the transitions from the ground-state. For a non-symmetric system a second spin-flipping transition appears. For
the symmetric system the transition to the higher-energy \( K'_d \) state was forbidden by the spatial symmetry: the higher-energy \( K'_d \) spin-up state in the symmetric quantum dot [Fig. 8(a)] is \( P_3 \)-odd, similarly as the ground-state. Moreover, for the symmetric system the transition to the spin-down even-parity state constantly grows with \( l \) [Fig. 8(c)]. For an asymmetric system the matrix element for the transition from the \( K'_d \) ground-state to the first excited state of the same spin-valley is a non-monotonic function of the interdot distance [Fig. 9(c)] and decreases at large \( l \) since the ground and the excited state end localized in different dots and the overlap between the wave functions vanishes.

The simulation of driven transitions for \( F_{AC} = 200 \) \( \text{V/cm} \) is displayed in Fig. 10. Note that we reduced \( F_{AC} \) ten times with respect to Figs. 6 and Fig. 7. The ground state at \( B = 0.5 \text{T} \), i.e. the \( K' \) spin-down oriented, mostly confined in the lower dot \((y < 0)\), is applied for the initial condition. The occupation of any state in the \( K \) valley reaches at most \( \approx 0.2\% \) during the 3.74 ns that is covered by the computation. Figure 10(a) was calculated at \( l = 18 \) nm – near the center of avoided crossing in Fig.
FIG. 11. (a) Energy spectrum for a single electron in an asymmetric double dot system, with $w_{l} = 1.1w_{u}$ and $w_{u} = 200\text{ meV}$, for $B = 0.5\text{ T}$ as a function of the interdot distance $l$. The color of the line indicates the localization in the lower dot (blue) or upper dot (red). (b) Transition matrix elements from the $K'_{d}$ ground state to the excited states. The color of the lines indicates the localization of the excited state.

At the frequency $h\nu$ below 14 meV a Rabi spin-flip to the $K'$ state localized in the upper dot ($K'_{u}(y > 0)$) is observed with the half transition time of 0.79 ns. At $h\nu \simeq 7\text{ meV}$ transitions to as many as three states occur. The transitions are driven to the energy levels $K'_{u}(y > 0)$ and $K'_{u}(y < 0)$ that enter into the avoided crossing [Fig. 9b]. The fastest is the transition to $K'_{d}$ state — that amounts in the electron charge hopping from the lower to upper dot with conserved spin and valley. The spin-flipping transition to $K'_{u}(y < 0)$ leaves the charge in the lower dot and lasts a few times longer. Anyway, the spin-flip time is of the order of the charge transition thanks to the proximity of the avoided crossing open by the Rasba interaction that mixes the spins of the states involved.

The third transition for this energy is the one to $K'_{u}(y > 0)$ for $h\nu$ which is half the direct Rabi transition. This two-photon transition is enhanced by the overlap with the proximity of the transition to $K'_{u}(y < 0)$. The electron first flips its spin passing from the ground state to $K'_{u}(y < 0)$ and next jump to the other dot to $K'_{u}(y > 0)$. The peaks at lower energy in Fig. 10 are the two-photon and three-photon transitions to the discussed states.

Figure 10(b) was taken for $l = 24\text{ nm}$, far from the avoided crossing of Fig. 11. The magnetic field of 0.5 T and the amplitude of the AC field of 2 kV/cm. The results present the maximal occupation probability for the simulation time of 3.74 ns. The system is initially in the $K'_{d}$ ground state localized mostly in the lower quantum dot (the deeper one). We give the time required for a given excited state to appear with a 50% contribution to the time dependent wave function. Results for $l = 18\text{ nm (a)}$ and $l = 24\text{ nm (b)}$. 

FIG. 12. Transitions driven by AC electric fields for asymmetric double dot of Fig. 11 the magnetic field of 0.5 T and the amplitude of the AC field of 2 kV/cm. The results present the maximal occupation probability for the simulation time of 3.74 ns. The system is initially in the $K'_{d}$ ground state localized mostly in the lower quantum dot (the deeper one). We give the time required for a given excited state to appear with a 50% contribution to the time dependent wave function. Results for $l = 18\text{ nm (a)}$ and $l = 24\text{ nm (b)}$. 

The first transition to the $K_{d}(y < 0)$, 0.31 ns peak at lower energy in Fig. 10(b) was taken for $l = 24\text{ nm}$, far from the avoided crossing of Fig. 11. The half-time for the charge hopping to $K'_{d}(y > 0)$ states is only 3 ps. The spin-flip occurs faster with the charge hopping to the other dot ($K'_{u}(y > 0)$, 1.6 ns) than the spin-flip within
FIG. 13. Energy spectrum for two electrons confined in double quantum dots. In (a) and (b) an asymmetric system with $w_1 = 1.03w_0$ is considered (cf. the single-electron results in Fig. 9). The color of the line shows the $z$ component of the two-electron wave function in the $b/2$ units. In (a) the interdot distance of $l = 18$ nm was applied. The energy levels that shift in pairs are split by the interdot exchange energy that is due to tunnel coupling. For comparison in (b) $l = 24$ nm was taken and the splitting can no longer be resolved. In (c) a symmetric system with $w_1 = w_0$ is taken with $l = 18$ nm. In (a) the dominant contributions to the two-electron wave functions in the valley-spin space are given. Here, the left (right) term of each product is attributed to electron label 1 (2).

FIG. 14. Simulation for $l = 18$ nm and $B = 1$ T and the weekly asymmetric quantum dots of Fig. 13(a). The initial state is the spin-valley polarized state with both electrons in $K'$ energy level. Maximal occupation probabilities for the AC electric field applied for 3.7 ns and $F = 2$ kV/cm are given by the solid lines. The two values in ns give the time needed to increase the occupation of the eigenstate above 10% and 50%.

the dot $[K'_y(y < 0)]$. The change of the order of spin flip times with and without charge hopping is consistent with the result of Fig. 14(c) for the matrix elements of transitions to $K'_u$ states.

As the last case for the single-electron in a double dot we consider a stronger asymmetry of the potentials in Fig. 11 with $w_1 = 1.1w_0 = 0.22$ eV. For this potential difference the avoided crossing [Fig. 9(b), Fig. 9(b)] no longer occurs [Fig. 11(a)]. However, the spin-flipping matrix elements [Fig. 11(b)] still possess a local maximum for $l$ between 15 and 18 nm.

The driven transitions are displayed in Fig. 12 for $l = 18$ nm (a) and $l = 24$ nm (b). Here, the amplitude was increased back to $F_{AC} = 2$ kV/cm. The spin-flip is about three times faster for the transition to the other dot (to $K'_y(y > 0)$) than the intradot transition (to $K'_u(y < 0)$), and the interdot distance $l$ changes the transition times by a factor of $\simeq 3$. For both $l$ considered in Fig. 12 we observe fast charge hopping transitions (red lines) with the single-photon Rabi resonance above $h\nu_R = 15$ meV and a series of $n$-photon transitions for the energies of $\nu = n\nu_R/n$.

C. The electron pair

Figures 13(a) and 13(c) show the spectrum for the intradot distance of $l = 18$ nm for asymmetric and symmetric double dots, respectively. For the electron pair the asymmetry of the double dot potential is of limited importance for the charge distribution, since the Coulomb interaction keeps the carriers localized in separate dots, so the spectra of Fig. 13(a) and Fig. 13(c) are qualitatively identical. The separation of electrons makes the intervalley exchange discussed for a single dot in Fig. 4 negligible.

For $B = 0$ we observe three groups of states. In the lowest (the highest) group including 4 levels both the electrons occupy the states of the ground-state (excited
state) doublet $K'_{d}$ or $K_{u}$ ($K_{d}$ or $K'_{u}$) for each of the dots [Fig. 3(b)]. In the central group of 8 energy levels an electron in one dot occupies a state of the ground-state doublet ($K'_{d}$ or $K_{u}$) and an electron in the other dot occupies the excited state ($K_{d}$ or $K_{u}$).

If one neglects the intervalley coupling – by the edge, and the contribution of both spins to the wave function – due to the weak Rashba interaction, the wave functions for the electron pair in separate dots can be interpreted in terms of separable products of spin-valley ($\psi_{sv}$) and spatial wave functions ($\psi_{sp}$), i.e., $\Psi(1, 2) = \psi_{sp}(1, 2)\psi_{sv}(1, 2)$. Both the spatial and spin-valley parts have a definite and opposite symmetry with respect to the electron interchange.

For carriers localized in separate dots and four accessible spin-valley single-electron states ($K_{d}, K_{u}, K'_{d}, K'_{u}$) the two-electron spin-valley $\psi_{sv}$ wave functions takes one of the 16 forms which are explicitly given close to the energy levels in Fig. 13(a). The normalization is skipped in the formulae given in the Figure, and in the products of single-electron spin-valley terms the first term corresponds to the first electron and the other to the second, i.e. $K_{d}K_{d}$ stands for $K_{d}(1)K_{d}(2)$, etc. In Fig. 13(a) one finds ten symmetric and six antisymmetric spin-valley wave functions with respect to the electron interchange. Four of the ten symmetric functions are spin-valley polarized ($K_{d}(1)K'_{d}(2)$ etc.). The twelve remaining states which are not spin-valley polarized appear in both symmetric and antisymmetric forms and the corresponding energy levels shift in pairs as functions of the magnetic field [Fig. 13(a)]. The spatial part of the wave function can be in the crudest approximation expressed as $\psi_{sp}(1, 2) = \frac{1}{\sqrt{2}} (\phi_{1}(r_{1})\phi_{2}(r_{2}) \pm \phi_{2}(r_{1})\phi_{1}(r_{2}))$, where $\phi_{1}$ and $\phi_{2}$ are the single-electron orbitals. For the antisymmetric $\psi_{sv}$ the orbital part needs to be symmetric. For the symmetric spatial function both the electrons can occupy the same e.g. the bonding single-electron orbital, hence the redshift of the antisymmetric spin-valley states with respect to the symmetric ones in Fig. 13(a). The splitting of the energy levels that shift is pairs is due to the tunnel coupling between the dots and is known from the quantum dots in 3D materials [77]. Fig. 13(b) provides the result for the interdot distance increased to $l = 24$ nm. Here, the splitting of the symmetric and antisymmetric states can no longer be resolved since the tunnel coupling between the dots is already quenched. In the absence of the valley degree of freedom, the splitting appears between symmetric and antisymmetric spin states and is referred to as the "spin exchange energy" [77].

Figure 14 shows the results of the simulation for the transitions driven at 1T from the spin-valley polarized ground-state $K'_{d}(1)K'_{u}(2)$. The asymmetric dots of Fig. 13(a) are taken, with the magnetic field of 1T, the $F_{AC}$ amplitude of 2kV/cm, and the duration of the AC pulse of 3.74 ns. We do not observe effective ($> 10\%$) transitions from the $K'_{d}(1)K'_{u}(2)$ ground state to the remaining three-states of the lowest-energy group of energy levels of Fig. 13(a), since these transitions require both the spin and the valley flip for one of the electrons. For the central group of energy levels relatively fast transitions are observed to $K'_{d}(1)K'_{u}(2)+K'_{u}(1)K'_{d}(2)$ and $K_{d}(1)K'_{d}(2) + K'_{u}(1)K_{d}(2)$ states. These spin-valley states are symmetric with respect to the electron interchange and require either the spin or the valley flip in the ground-state $K'_{d}(1)K'_{d}(2)$ wave function. Note, that the transitions driven by the $F_{AC}(y_{1} + y_{2})\sin(\omega_{T}t)$ perturbation would be strictly forbidden between the exactly separable states of opposite symmetry of the spatial part of the wave function.

For the highest group of the energy levels one observes only the transitions to the $K'_{d}K'_{u}$ energy level. This transition requires the spin flip of both the electrons in the ground state $K'_{d}K'_{d}$. The corresponding transition peak is very small at the nominally resonant energy of about $\hbar\nu = 15$ meV, but a high transition peak is found at half the resonant energy. The peak overlaps with the transition to $K'_{d}(1)K_{u}(2) + K'_{u}(1)K_{d}(2)$ state. Hence, the transition to $K'_{d}K'_{u}$ at the energy if $\approx 7$ meV has an indirect two-photon character and occurs by sequential flips of the two spins, one after the other.

V. SUMMARY AND CONCLUSIONS

We studied the spin and valley properties of electrons confined in single and double electrostatic quantum dots defined within the silicene using atomistic tight binding and exact diagonalization approach for both stationary Hamiltonian eigenstates and the spin-valley dynamics of the system driven by microwave or deep infrared field.

The effects of the intervalley exchange interaction and the interdot tunnel coupling on the spectra confined in single and double quantum dots were explained. We determined the transition rates involving charge hopping, spin flipping and valley switching. We found that the valley-transition times can be changed by orders of magnitude by tuning the coupling of the confined system to the crystal edge by the electrostatic confinement potential. With the control of the electrostatic confinement one can both produce very fast valley flips or remove the orbital-valley coupling. The intervalley scattering due to the electron-electron interaction increases the valley transitions times to a significant extent only when the coupling to the edge is removed. The spin-transition times depends on the strength electrostatic confinement is weaker. Ultrafast spin transitions in the double dots due to the coupling of the bonding and antibonding orbitals by the Rashba interaction were demonstrated.
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