# Electrical spin manipulation in double SrTiO<sub>3</sub>/LaAlO<sub>3</sub> quantum dots

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The spin dynamics in two electron double quantum dots embedded in two dimensional electron gas at the interface between  $SrTiO_3$  and  $LaAlO_3$  is studied by an exact numerical solution of the timedependent Schrödinger equation, in the context of the electric dipole spin resonance experiment. Based on the three band model of 3d-electrons localized at Ti ions on the square lattice we analyze in details the singlet-triplet transition induced by the AC electric field, in the magnetic field range close to the avoided crossing which appears as a result of the spin-orbit coupling. Our calculations show that for symmetric double quantum dots the single photon spin-flip transitions is prohibited due to the parity symmetry and the transition can occur only by the higher order two-photon processes. For a weakly asymmetric system, when the first order singlet-triplet transitions are released due to the parity symmetry breaking, the spin-flip transition has a character of the Rabi oscillations for a low electric field amplitude. As the amplitude is increased the frequency of the transition is blueshifted (redshifted) for the magnetic field below (above) the single-triplet avoided crossing. Interestingly, for a sufficiently high magnetic field and high AC field amplitude the electric field drives the system across the avoided crossing inducing the spin-flip by the Landau-Zener-Stueckelberg-Majorana transitions with 100% spin flip probability for a slow sweep. Finally, the optimization of the geometrical parameters of the system with respect to the time of spin-flip of its fidelity is also presented.

# I. INTRODUCTION

The ability to control the spin of electrons confined in quantum dots (QDs) has attracted growing interest in recent years, as it lies at the heart of developments towards a scalable spin-based quantum computer [1, 2]. The effiency of spin manipulation is entirely contingent upon the properties of the material platform, which dictate the strength of the spin-orbit coupling (SOC) [3–5] or the hyperfine interaction with the nuclear spin bath. The latter is a source of spin decoherence [6] limiting the usability of a specific material platform in accordance with one of the Loss di Vincenzo criteria [7], whereby the quantum system must remain coherent for times much longer than the duration of elementary logic gates. For this reason, silicon-based QDs have been intensively studied over the last years due to their prolonged spin coherence time [8].

With respect to that, another promising but still unexplored platform for QD-based spin qubits technology are transition metal oxide heterostructures, such as  $SrTiO_3/LaAlO_3$  (LAO/STO) interfaces [9–13]. The electronic properties of the two-dimensional electron gas (2DEG) formed at the LAO/STO interface is determined by the 3*d*-orbitals [14–18], implying that direct and indirect decoherence stemming from interaction with the nuclear bath can be significantly mitigated, as it is proportional to the square of the wave function at the nuclei position [6] - this value diminishes to zero for 3delectrons. This property, combined with the strong spinorbit interaction [19, 20], high mobility [21], and susceptibility to electrostatic gates at a level comparable to semiconducting materials [22–24], initiates the currently ongoing research on LAO/STO-based quantum dots as potential spin-qubits with inherent scalability of 2D systems. The first experimental realization of electrostatically defined LAO/STO QDs has been already reported, yielding a Coulomb blockade diamond characteristic for a well-defined confinement [22, 23].

The demonstration of electrical spin manipulation in these systems constitutes the next milestone in spinqubit realization based on the transition metal oxide heterostructures. Significant progress in this field has been recently achieved in electrostatically defined semiconducting QDs [25], where the microwave magnetic field, previously used to control spin dynamics [26, 27], has been replaced by the ac gate voltage [2, 28–30]. In this technique, called electric dipole spin resonance (EDSR), the microwave potential applied to one of the gates generates a time-dependent potential which, due to the momentum-dependent spin-orbit interaction [3– 5], hybridizes the spin-up and spin-down states, leading to Rabi oscillations between the Zeeman-split electronic states [26]. For two electrons, the SOC hybridization induces level repulsion between the spin singlet and triplet states, which is often used to determine the strength and direction of the effective SOC field [31]. Recently, EDSR technique applied to double quantum dots (DQDs) allowed for a demonstration of Pauli spin blockade [2, 32]. In this regime, the system initially set to the spin triplet state  $T_{-}(1, 1)$ , with tunneling from the left to the right QDs prohibited by selection rules, transmits to the spin singlet state (0, 2), which is possible due to the SOC generated by the ac field applied to one of the gates [33].

Although spin manipulation in LAO/STO QDs has not yet been demonstrated experimentally, theoretical studies allow us to gain a first insight into the spin dynamics in these multiorbital systems. In our recent paper [34], we have discussed in detail the manipulation of electron spin in a single QD in the context of EDSR experiment. We have shown that for a single electron, the spin-flip in the ground state has the character of a Rabi resonance, while for two electrons, the singlet-triplet transition is forbidden by parity symmetry. The latter one is possible via a second-order, two-photon process, which exhibits a two-state Rabi character for low ac field amplitude.

Here we extend our study to the case of two-electrons in double quantum dots. Based on the time dependent scheme we simulate the singlet-triplet transition induced by the AC electric field. We consider the magnetic field range in the nearest of the avoided crossing which appears as result of the SO coupling. The calculated transition between singlet-triplet states, its characteristics, i.e. duration time and fidelity, are discussed with respect to the system asymmetry which is needed to induce the single photon transition by the parity symmetry breaking, as well as the coupling strength between the dots. Finally, the full symmetric system is also analyzed with respect to the higher order singlet-triplet transitions.

The manuscript is organized as follows: in Sec. II we present a theoretical model of double quantum dots in 2DEG at the (001) LAO/STO interface as well as the scheme used for time-dependent two electron calculations. Sec. III contains the analysis of electronic spectrum of a two-electron double quantum dots as well as results of time dependent simulations (EDSR), finally summary and conclusions are included in Sec. IV.

# II. THEORETICAL MODEL

### A. The single-electron problem

We consider 2DEG in the quantum well created at the (001) LAO/STO interface [12, 35]. The system is characterized by a significant atomic SO coupling and a strong Rashba interaction due to the asymmetry of the vertical electric field at the interface [14–16]. We use the real space representation [34] of the tight-binding Hamiltonian [14] spanned by 3d orbitals of Ti ions that are arranged in a square lattice. The Hamiltonian of the system

is given by

$$\hat{H} = \sum_{\mu,\nu} \hat{C}^{\dagger}_{\mu,\nu} (\hat{H}^{0} + \hat{H}_{SO} + \hat{H}_{B}) \hat{C}_{\mu,\nu} +$$
(1)  
$$\sum_{\mu,\nu} \hat{C}^{\dagger}_{\mu+1,\nu} \hat{H}^{x} \hat{C}_{\mu,\nu} + \sum_{\mu,\nu} \hat{C}^{\dagger}_{\mu,\nu+1} \hat{H}^{y} \hat{C}_{\mu,\nu} +$$
$$\sum_{\mu,\nu} \hat{C}^{\dagger}_{\mu+1,\nu-1} \hat{H}_{mix} \hat{C}_{\mu,\nu} -$$
$$\sum_{\mu,\nu} \hat{C}^{\dagger}_{\mu+1,\nu+1} \hat{H}_{mix} \hat{C}_{\mu,\nu} + h.c.,$$

where  $\hat{C}_{\mu,\nu} = (\hat{c}^{\dagger}_{\mu,\nu,xy}, \hat{c}^{\dagger}_{\mu,\nu,xy}, \hat{c}^{\dagger}_{\mu,\nu,xz}, \hat{c}^{\dagger}_{\mu,\nu,yz}, \hat{c}^{\dagger}_{\mu,\nu,yz}, \hat{c}^{\dagger}_{\mu,\nu,yz})^T$ is the vector of electron anihilation operators corresponding to states with the spin  $\sigma =\uparrow,\downarrow$  on the orbital  $d_{xy}, d_{xz}, d_{yz}$  at the position  $(\mu, \nu)$ . In Eq. (1),  $\hat{H}^0$ accounts for the splitting of the 3*d* orbitals degeneracy and the in-plane external potential  $V(\mathbf{r})$  defining the quantum dot confinement

$$\hat{H}^{0} = \begin{pmatrix}
4t_{l} - \Delta_{E} & 0 & 0 \\
0 & 2t_{l} + 2t_{h} & 0 \\
0 & 0 & 2t_{l} + 2t_{h}
\end{pmatrix} \otimes \hat{\sigma}_{0} \\
+ \begin{pmatrix}
V_{\mu,\nu} & 0 & 0 \\
0 & V_{\mu,\nu} & 0 \\
0 & 0 & V_{\mu,\nu}
\end{pmatrix} \otimes \hat{\sigma}_{0}.$$
(2)

The atomic spin-orbit coupling is defined by  $\hat{H}_{SO}$  and is given by

$$\hat{H}_{SO} = \frac{\Delta_{SO}}{3} \begin{pmatrix} 0 & i\sigma_x & -i\sigma_y \\ -i\sigma_x & 0 & i\sigma_z \\ i\sigma_y & -i\sigma_z & 0 \end{pmatrix} , \qquad (3)$$

where the matrix corresponds to  $\mathbf{L} \cdot \mathbf{S}$  with the orbital angular momentum represented in the  $t_{2g}$ -orbitals basis. Finally, the coupling of the external magnetic field to the spin and atomic orbital momentum of electrons is taken into account by the Hamiltonian

$$\hat{H}_B = \mu_B(\mathbf{L} \otimes \sigma_0 + g \mathbf{1}_{3 \times 3} \otimes \mathbf{S}) \cdot \mathbf{B}/\hbar, \tag{4}$$

where  $\mu_B$  is the Bohr magneton, g is the Landé factor,  $\mathbf{S} = \hbar \boldsymbol{\sigma}/2$  with  $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$  and  $\mathbf{L} = (L_x, L_y, L_z)$  with

$$L_x = \begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, L_y = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}, L_z = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & -i & 0 \\ (5) \end{pmatrix}$$

The hopping elements in Eq. (1) consist of the kinetic term (spin-conserving intersite hopping) and the Rashba SOC (mixing between the  $d_{xy}$  and the two other orbitals) and take the form

$$\hat{H}^{x} = \begin{pmatrix} -t_{l} & 0 & 0\\ 0 & -t_{l} & 0\\ 0 & 0 & -t_{h} \end{pmatrix} \otimes \hat{\sigma}_{0} + \frac{\Delta_{RSO}}{2} \begin{pmatrix} 0 & 0 & -1\\ 0 & 0 & 0\\ 1 & 0 & 0 \end{pmatrix} \otimes \hat{\sigma}_{0} ,$$

$$\tag{6}$$

and

$$\hat{H}^{y} = \begin{pmatrix} -t_{l} & 0 & 0\\ 0 & -t_{h} & 0\\ 0 & 0 & -t_{l} \end{pmatrix} \otimes \hat{\sigma}_{0} + \frac{\Delta_{RSO}}{2} \begin{pmatrix} 0 & -1 & 0\\ 1 & 0 & 0\\ 0 & 0 & 0 \end{pmatrix} \otimes \hat{\sigma}_{0}.$$
(7)

Finally, the hybridization between  $d_{xz}$ ,  $d_{yz}$  orbitals are taken into account by

$$\hat{H}_{mix} = \frac{t_d}{2} \begin{pmatrix} 0 & 0 & 0\\ 0 & 0 & 1\\ 0 & 1 & 0 \end{pmatrix} \otimes \hat{\sigma}_0.$$
(8)

In our simulations, we take on the tight-binding parameters  $t_l = 875$  meV,  $t_h = 40$  meV,  $t_d = 40$  meV,  $\Delta_E = 47$  meV, after Ref. [14, 18], the Landé factor g = 3 [22, 36] and the SO coupling parameters  $\Delta_{SO} = 10$  meV,  $\Delta_{RSO} = 20$  meV [19, 20].

The external magnetic field induces the Aharonov-Bohm phase shifts for the electrons moving along the interface, that we account for using the Peierls approach, i.e. multiplying the elements of the Hamiltonian corresponding to the hopping between ions placed at points  $\mathbf{r}_a = (x_{\mu_a}, y_{\nu_a})$  and  $\mathbf{r}_b = (x_{\mu_b}, y_{\nu_b})$  by a factor  $e^{i\frac{e}{\hbar}\int_{\mathbf{r}_a}^{\mathbf{r}_b}\vec{A}\cdot\vec{d}l}$ , where  $\vec{A}$  is the vector potential taken in the symmetric form  $\vec{A} = (-y/2, -x/2, 0)B$  for  $\vec{B} = (0, 0, B)$ .

The in-plane external potential that models the double quantum dot system is assumed as a superposition of two Gaussian functions

$$V(x,y) = -V_0 \exp\left(-\frac{1}{2}\left((x+\frac{s_x}{2})^2+y^2\right)/R^2\right) - V_1 \exp\left(-\frac{1}{2}\left((x-\frac{s_x}{2})^2+y^2\right)/R^2\right), \quad (9)$$

where  $s_x$  is the distance between the minima of the separate functions (centers of the left and right QDs),  $V_0$  and  $V_1$  define the depth of each potential minimum and R is the size parameter of the separate Gaussians.

### B. The two-electron problem

The two-electron problem is defined by the Hamiltonian

$$\hat{H}_2 = \hat{H}(1) + \hat{H}(2) + \frac{e^2}{4\pi\epsilon_0\epsilon r_{12}},$$
(10)

with the dielectric constant  $\epsilon = 100$  [37]. The problem is solved with the exact diagonalization method with the basis spanned by the atomic orbitals

$$\Psi_q(x, y, \sigma) = \sum_j a_j^q d_j(x, y, \sigma)$$
$$= \sum_{r_j, o_j, s_j} a_j^q d_{r_j, o_j}(x, y) S_{s_j}(\sigma), \quad (11)$$



FIG. 1. The charge density for the two-electron singlet (a,c) and two-electron triplet (b,d) for B = 0. Results for a symmetric system with  $V_0 = V_1 = 50$  meV and the spacing parameter  $s_x = 29.64$  nm (a,b) and  $s_x = 31.2$  nm (c,d).

where the summation runs over the position of ions  $r_j$ , orbitals  $o_j$  on the ion and the z-component of the spin indexed by  $s_j$  while S is the spin-up or spin-down eigenstate. In the sums j is equivalent to the triple of indeces  $(r_j, o_j, s_j)$  and  $d_{r_j, o_j}$  is one of the 3d orbitals localized on the ion position  $r_j$ . Evaluation of the Coulomb integrals has been performed in real space using the two-center approximation [38] and integration over the d orbitals after Ref. [34]. In the calculations, we use up to 50 lowestenergy single-electron states that produce 1225 Slater determinants as a basis for the two-electron problem.

### C. Time stepping

We study the spin flips induced as singlet-triplet transitions between the eigenstates of the two-electron Hamiltonian with an external periodic perturbation of the potential  $V_{AC}(x,t) = -eFx \sin(\nu t)$ . The time evolution is determined by integration of the time-dependent Hamiltonian  $\hat{H}_2(t) = \hat{H}_2 + V_{AC}(x_1,t) + V_{AC}(x_2,t)$ . The solution is obtained in the basis of time-independent Hamiltonian eigenstates,  $\Psi(t) = \sum_m c_m(t) \exp(-iE_m t/\hbar) |m\rangle$ with  $\hat{H}_2|m\rangle = E_m |m\rangle$ . Substituting this form of the wave function to the Schrödinger equation and projecting the resulting equation on the eigenstates  $\langle n|$  one obtains a system of equations for  $c_n(t)$ ,

$$i\hbar c'_n(t) = -eF\sum_m c_m(t) \exp\left[i\left(E_n - E_m\right)t/\hbar\right] \\ \times \sin(\nu t) \langle n|x|m\rangle,$$
(12)

that we solved using the Crank-Nicolson time stepping with the two-electron ground-state as the initial condition, i.e.  $c_n(t=0) = \delta_{n,1}$ .



FIG. 2. The low-energy spectrum for the electron pair in a symmetric quantum dot system  $V_0 = V_1 = 50$  meV for the spacing parameter  $s_x = 29.64$  nm (a,b) and  $s_x = 31.2$  nm (c,d). The color of the line indicates the z-component of the total spin. The singlet-triplet avoided crossing is enlarged in (b) and (d).

### III. RESULTS

#### A. The two-electron eigenstates

In Fig. 1 we plotted the charge density of the lowestenergy spin singlet (a,c) and the lowest-energy spin triplet (b,d) for a symmetric quantum dots with spacing between the Gaussian minima of  $s_x = 29.64$  nm (a,b) and  $s_x = 31.2$  nm (c,d). The results for a stronger coupling [Fig. 1(a,b)] and in particular the form of the densities in the barrier region indicate that the singlet (triplet) charge density has a bonding (anti-bonding) character. For weaker interdot coupling [Fig. 1(c,d)] the charge densities in both spin states are more similar and the bonding (anti-bonding) character of the states is less pronounced.

The two-electron energy spectra for the considered DQDs are displayed in Fig. 2(a,b) for  $s_x = 29.64$  nm and in Fig. 2(c,d) for  $s_x = 31.2$  nm. At B = 0 the ground-state is spin-singlet for any interdot distance  $s_x$ . The singlet-triplet energy splitting (the exchange energy) at B = 0 varies strongly with  $s_x$ , e.g. the interdot tunnel coupling, and for  $s_x = 29.64$  nm it is equal to 185.5  $\mu$ eV, decreasing to only 33.1  $\mu$ eV for  $s_x = 31.2$  nm. The applied external magnetic field promotes the spin-polarized triplet for the ground-state. The singlet-triplet ground-

state transformation occurs via an avoided crossing [Fig. 2(b,d)] that is opened by the spin-orbit coupling. The position of the avoided crossing at the magnetic field scale is determined by the exchange energy at B = 0. For  $s_x = 29.64$  nm ( $s_x = 31.2$  nm) the center of the avoided crossing occurs at 1.05 T (0.19 T) with the width (e.g. minimal energy spacing between the anticrossing energy levels) equal to 12.3  $\mu$ eV for the stronger coupling and only 2.68  $\mu$ eV for the weaker coupling.

To account for an intrinsic or intentional asymmetry in the confinement potential of the double quantum dot system we introduce unequal depths of the Gaussian quantum wells forming the artificial molecule. In Fig. 3(a) we plot the exchange energy at B = 0 as a function of the right quantum dot potential calculated for the left potential set at  $V_0 = 50$  meV and  $s_x = 29.64$ nm (black line) as well as  $s_x = 31.2$  nm (red line). The exchange energy is minimal for the symmetric system. For an asymmetric double dot in the spin-singlet state both the electrons tend to occupy the deeper quantum dot [Fig. 3(b,c)] which lowers the singlet energy with respect to the spin triplet for which the double occupancy of the lowest orbital is forbidden by the Pauli exclusion principle. In contrast, the reaction of the charge localization to the asymmetry in the triplet state is weak [Fig. 3(b,c)]. Note, moreover, that a small asymmetry  $V_1 \neq V_0$  does not introduce a significant charge redistribution to the charge in the spin-singlet state which is a result of the electronelectron interaction. Only for  $V_1 \leq 49$ nm or  $V_1 \geq 51$  nm the electrons in the singlet start to occupy the deeper dot. For the weaker interdot coupling the transition to the deeper dot is more abrupt [cf. Fig. 3(b) and (c)].

For a symmetric potential the single-electron Hamiltonian eigenstates are also eigenstates parity operator  $\Pi = \text{diag}[P, -P, -P, P, -P, P]$ , where P is the scalar parity operator  $P\psi(r) = \psi(-r)$ . The lowest-energy singlet and the lowest spin-polarized triplet that participate in the avoided crossing have the same – negative –  $\Pi$  parity – see Fig. 4. The same symmetry of the lowest singlet and triplet states has a direct consequence for the spin transitions driven by AC electric field, namely the dipole transition matrix elements between the two lowest-energy levels vanish so the first-order (single-photon) transitions is forbidden and only the second-order processes (twophoton) can occur. Similar behavior has been observed for two electrons in single quantum dot and discussed in details in our previous paper [34].

### B. EDSR in weakly asymmetric system

Let us first study the case of weakly asymmetric system with  $V_1 = 51$  meV. For small asymmetry the electron charge is still more or less evenly distributed over the dots, but the asymmetry lifts the parity selection rule that forbids the single-photon, first-order transitions.



FIG. 3. (a) The exchange energy, i.e. the energy difference between the spin-triplet and singlet states for B = 0 and  $s_x = 29.64$  nm (black line) as well as  $s_x = 31.2$  nm (red line). The charge localization in the left (red line) and right dot (blue line) in the singlet (solid line) and the triplet state (dotted line) for  $s_x = 29.64$ nm and  $s_x = 31.2$  nm are displayed in (b) and (c), respectively.



FIG. 4. Same as Fig. 2(a) with the color of the lines indicates the  $\Pi$  parity – the negative one is plotted in blue, and the positive one in red.

Fig. 5(a) shows the spectrum for  $s_x = 29.64$  nm near the singlet-triplet avoided crossing that is shifted to higher magnetic field by the asymmetry due to the increased exchange energy [Fig. 3(a)] (to B = 1.22 T from 1.05 T in the  $V_0 = V_1$  case). The width of the avoided crossing is also enlarged: from 12.3  $\mu$ eV (for  $V_1 = 50$ meV) to 13.8  $\mu$ eV (for  $V_1 = 51$  meV). The essential quantity for the first-order spin transitions is the dipole matrix element  $\langle \psi_n | x | \psi_m \rangle$  between the two-lowest energy levels which is plotted by the black curve in Fig. 5(b). For an ideally symmetric system  $V_1 = V_0$  a similar singlet-triplet avoided crossing of energy levels is obtained with the interchange of the average values of the spin between the two lowest-energy levels, but the transition dipole matrix element is zero. For the system with lifted parity symmetry, the matrix element reaches its maximum in the center of the avoided crossing which allows the first order transition possible.

We studied the transitions from the ground-state to the first-excited state near the single-triplet avoided crossing induced by the external AC sine electric field, with the initial state set to the ground-state. The maximal occu-



FIG. 5. (a) Same as Fig. 2(b) only for the right quantum dot made deeper by 1 meV, e.g.  $V_1 = 51$  meV. (b) The average z-component of the total spin – with respect to the right vertical axis – in the ground-state (in blue) and the first excited state (in red). The black line shows the dipole matrix element between the two lowest-energy states (referred to the left vertical axis).

pancy of the first-excited state for the simulation lasting 5 ns is given in Fig. 6 for B = 0.434 T (a), B = 0.94 T (b), B = 1.5 T (c), and B = 2 T (d) with the amplitude of the AC field increasing in steps of 5  $\mu$ V/nm. For the magnetic fields in Fig. 6(a,b) and Fig. 6(c,d) the ground-state is singlet and triplet, respectively. At the amplitude of 5  $\mu$ eV the largest peak corresponds to the singlet-triplet transition that occurs at the driving energy





FIG. 6. The results of simulation of the time evolution for the weakly asymmetric ( $V_1 = 51 \text{ meV}$ ) double quantum dot system with spacing of  $s_x = 29.64 \text{ nm}$  in an AC external electric field  $eFx \sin(\nu t)$ . The subsequent lines in the upper part of the plots correspond to the AC field amplitude increased by 5  $\mu$ V/nm offset by +1 each along the vertical axis. The simulation lasts 5 ns and starts from the ground-state. The lines show the maximal occupancy of the first excited state. The panels correspond to B = 0.434 T (a), B = 0.94 T (b), B = 1.5 T (c) and B = 2 T (d).

near the singlet-triplet energy difference. For B = 0.94 T and 1.5 T, where the dipole matrix element is large, at the lowest-amplitude, we also resolve the peaks of higherorder transitions at half and 1/3rd of the frequency for the single-photon transition processes. As the AC field amplitude is increased the frequency of the spin-flip transition [Fig. 6(a,b)] for the magnetic field below the singletriplet avoided crossing is blueshifted at higher AC field amplitudes. On the other hand the frequency for the spin transition for the magnetic field above the single-triplet avoided crossing gets redshifted for high AC amplitudes [Fig. 6(c,d)]. In both cases, in an intense AC field, we find an effective upbeat of the triplet energy with respect to the singlet. In Fig. 6(c) we find that the regular transition spectrum gets deteriorated at higher frequency with the disappearance of the single-photon peak. For B = 0.94 T at higher amplitude the AC field drives the system across the singlet-triplet avoided crossing so that the process for the spin-flip acquires the Landau-Zener-Stueckelberg-Majorana (LZSM) character [39].

Fig. 7 shows the two lowest-energy states for the double dot system with a time-independent electric field of



FIG. 7. The lowest singlet and triplet energy levels for B = 0.94 T (a), 1.5 T (b) and 2 T (c) for the weakly asymmetric system and a constant electric field +eFx. In the considered field range a singlet-triplet avoided crossing opens for B = 1.5 T and 2 T that changes. In the AC simulation the avoided crossing changes the spin-flip probability at low frequency from the multiphoton transitions to the LZSM interference – see Fig. 6(c,d) and Fig. 8 at higher amplitude.



FIG. 8. Same as Fig. 6(d) only as a function of the inverse of the driving energy.



FIG. 9. The results for weakly asymmetric double quantum dot with  $V_1 = 51$  meV and  $s_x = 29.64$  nm. (a) shows the position of the spin-flip singlet-triplet resonance in the AC electric field as a function of its amplitude for selected values of the external magnetic field. (b) The spin-flip time as a function of the amplitude in the first-order transition. The line for 1.5 T is interrupted since the first order peak can not longer be resolved at large F. (c) The maximal share of the states other than the two lowest-energy ones in the time evolution of the wave function for the system driven by the external AC field.

eFx added to the potential. The avoided crossing for B = 1.5 T [Fig. 7(b)] is reached at  $|F| \simeq 20 \mu V$ . Note, that in AC field with the amplitude of  $F \simeq 20 \ \mu V/nm$ the maximal occupancy of the singlet-state is no longer zero at low  $h\nu$  limit [cf. Fig. 6(c)]. At higher amplitude the low-frequency limit in Fig. 6(c) corresponds to an adiabatic sweep of the potential across the singlet-triplet avoided crossing with 100% spin flip probability for a slow sweep. For nonzero AC frequency the LZSM interference between the contributions from both the states appear with a diabatic and an adiabatic components [39] generating the interference comb visible in Fig. 6(c) at higher amplitude and low frequency. For B = 2 T we reach a similar point at  $F \simeq 30 \ \mu V/nm$  [cf. Fig. 6(d) and Fig. 7(c)]. The change of the character of the triplet-singlet transition is better illustrated by the transfer probability plotted as a inverse of the driving energy presented in Fig. 8 for B = 2 T. As the amplitude increases from  $5 \,\mu V/nm$  to  $25 \,\mu V/nm$  higher-order peaks corresponding to multiphoton transitions appear but the corresponding peaks fall below 100% at large  $1/h\nu$ . For 30  $\mu$ V/nm and above, the transition probability peaks acquire the height of 100% that does not fall below at large  $1/h\nu$ , and appear periodically on the  $1/h\nu$  scale, which are characteristics of the LZSM interference [39].

Figure 9(a) shows the position of the first order transition on the energy scale  $E_r = h\nu_r$  as a function of the AC amplitude. Note that, the magnetic fields considered in Figs. 9(a) as well as 6 were chosen to produce equal singlet-triplet energy splitting before and after the avoided crossings, hence the coalescent lines at the limit of zero amplitude. The frequency shifts before and after the singlet-triplet avoided crossing have opposite signs but similar magnitudes. The line for 1.5 T is interrupted above 35  $\mu$ V/nm, since the resonance line can be hardly recognized in the spectrum [cf. Fig. 6(c)].

A strong AC amplitude induces appearance of other states than the lowest-energy couple in the time evolution. The maximal share of the higher energy states as a function of the amplitude is given in Fig. 9(c) as the leakage of the wave function outside the lowest-energy singlet and triplet. The dependence was determined at the first-order resonant transition and as we can see the leakage on the amplitude is similar for all the magnetic fields considered. At the amplitude of 20  $\mu$ V/nm the maximal share of the higher energy states is about 2.5%.

The dependence of the spin-flip time on the amplitude for the first-order transition is plotted in Fig. 9(b). The minimal flip time of about 245 ps for B = 1.5 T is obtained at the amplitude of 25  $\mu$ V/nm. For even stronger amplitudes the flip time no longer decreases which is related to the potential sweeps across the singlet-triplet avoided crossing and the leakage of the wave function to higher energy states for which the evolution no longer have a two-state Rabi resonance character [40].

# C. Optimization of the dipole matrix moment

In the Rabi oscillations regime the rate of the firstorder spin-flip transition is proportional to the value of the dipole matrix element. The latter is strictly zero for an ideally symmetric double quantum dots. In the precedent section we described the results obtained in a system with a weak asymmetry but the value of the matrix element can be optimized with the system parameters.

The dipole matrix element for  $s_x = 29.64$  nm is displayed in Fig. 5(b). In a system with larger interdot barrier ( $s_x = 31.2$  nm), but still with the small asymmetry  $V_1 = 51$  meV as in Fig. 5(b), the singlet-triplet avoided crossing occurs at lower magnetic field of B = 0.23 T [cf. Fig. 10(a)] than for the stronger interdot coupling due to a reduction of the exchange energy [Fig. 3(a)]. The width of the singlet-triplet avoided crossing is 3.2  $\mu$ eV for  $s_x = 31.2$  nm. The maximal value of the dipole matrix element in the center of the avoided crossing is about 0.323 nm, e.g. about 3 times lower than for  $s_x = 29.64$  nm [Fig. 5(b)].

The position of the singlet-triplet avoided crossing and the maximal value of the dipole matrix element strongly depends on the asymmetry of the confinement potential. In Fig. 10(b) and (c) we plotted the average spins and the dipole matrix element near the singlet-triplet avoidedcrossing for  $s_x = 31.2$  nm with  $V_1 = 54$  meV (b) and  $V_1 =$ 60 meV (c). For  $V_1 = 54$  meV [Fig. 10(b)] the maximal value of the dipole matrix element is 40 times larger than for the small asymmetry  $V_1 = 51$  meV [Fig. 10(a)] and about 4 times larger than for the large asymmetry  $V_1 =$ 



FIG. 10. Same as Fig. 5(b) but for  $s_x = 31.2$  nm and  $V_1 = 51$  meV (a),  $V_1 = 54$  meV (b) and 60 meV (c).

60 meV [Fig.10(c)].

The solid lines in Fig. 11(a) show the absolute value of the dipole element at the center of the singlet-triplet avoided crossing as a function of the right-dot potential  $V_1$ . The asymmetry of the confinement potential pushes the avoided crossing to higher values of the magnetic field (dashed lines referred to the right axis). The dependence of the matrix element on the asymmetry is nonmonotonic which is related to the charge distribution in the singlet and triplet states. Fig. 11(b) shows the charge in the left and right quantum dot calculated for B = 10 T- near the value of the magnetic field for which the maximum of the dipole matrix element is achieved in Fig. 11(a) for  $V_1 = 54$  meV. The gap in the lines of Fig. 11(b) corresponds to the region of the singlet-triplet avoided crossing as a function of  $V_1$  that we skipped for the clarity of the plot. At the left (right) hand side of the avoided crossing the ground-state for B = 10 T is the spin triplet (singlet).

Comparing Fig. 11(a) and Fig. 11(b) we can conclude



FIG. 11. (a) The solid lines show the dipole matrix element for the transition between the two lowest-energy states at the center of the singlet-triplet avoided crossing. The dashed lines show the position of the avoided crossing (right vertical scale). The red and black lines correspond to the  $s_x$  parameter equal to 31.2 nm and 29.64 nm, respectively. (b) The charge localized in the right and left quantum dots for  $s_x = 31.2$  nm in the singlet (red lines) and triplet (blue lines) for B = 10 T. The gap in the lines was left for the avoided crossing of the singlet and triplet energy levels for the clarity of the plot. The ground-state is singlet (triplet) at the right(left)-hand side of the figure.

that the maximal value of the dipole matrix element is achieved for  $V_1$  which on the one hand localizes the singlet entirely in the deeper dot but for which the triplet charge distribution is still evenly distributed in both dots. In the region of the avoided crossing the two-lowest energy states have hybridized wave functions that due to the contribution of the triplet are delocalized over both dots which produces a large value of the dipole matrix element. For large  $V_1$ , on the other hand, both singlet and triplet states are confined within a single deeper quantum dot with potential perturbed by the potential of the left Gaussian. Due to this perturbation the dipole matrix element is non-zero. Since the system is localized in a single dot the matrix element does not achieve values as large as for the delocalized triplet state. Comparing Fig. 3(b) and Fig. 11(a) we also notice that at higher B the triplet state is more easily localized in the deeper dot: for  $V_1 = 60$  meV the charge in the right dot is 1.2eat B=0, but already 1.7e at B = 10 T.

For the studies of the transition times we chose  $V_1 =$ 



FIG. 12. Singlet-triplet transition time for the double-dot system with  $s_x = 31.2$  nm and  $V_1 = 54$  meV driven by the AC field of  $eFx \sin(\nu t)$  for three values of the magnetic field corresponding to different values of the dipole moment, see Fig. 10(b).

54 meV – near the maximum of the dipole matrix element [Fig. 11(a)] and three values of the magnetic field after the singlet-triplet avoided crossing, where the ground-state is the spin triplet - 7.9 T, 8 T and 9 T - see Fig. 10(b). The singlet-triplet transition times as functions of the AC amplitude is given in Fig. 12. The transition times reach minima for lower values of the AC field amplitude than in Fig. 9(b) and start to grow for the F values that exceeds the one for the LZSM transition regime. The LZSM regime is open for the amplitudes of  $F = 1.95 \ \mu V$ , 2.7  $\mu V$  and 9.5  $\mu V/nm$  for B = 7.89 T, 8 T and 9 T, respectively. Note, that for each value of the magnetic field the minimum spin flip time of about 330 ps is achieved for some value of the AC field amplitude. Since the amplitudes of the AC field near the minimal spin flip time are smaller than in Fig. 9(b), so are the wave function leakage values near the minimum which are only 0.5%, 0.02%, 0.0043% for  $F = 17.5 \ \mu V/nm$ , 5  $\mu V/nm$ , and 2.43  $\mu V/nm$ , corresponding to B = 7.89 T, 8 T and 9 T, respectively.

# D. Spin flips for an ideally symmetric system

For completeness we present the results for an ideally symmetric system, for which the direct spin-flip transition is forbidden by the selection rule. Fig. 13 shows the probability of the spin-flip-singlet for  $s_x = 29.64$  nm and the external magnetic field set at 2 T, for a simulation lasting 5 ns and starting from the ground-state triplet as the initial state. The subsequent lines at the upper part of Fig. 13 correspond to a growing amplitude of the AC field. The most pronounced peak that is observed in Fig. 13 at weak AC field amplitude occurs at roughly half the singlet-triplet energy splitting that at the field of 2 T is equal to  $\Delta E_{ST} = 0.1663$  meV [cf.



FIG. 13. Same as Fig. 6 only for a symmetric system with  $V_0 = V_1 = 50$  meV at B = 2 T.

Fig. 2(b)]. The spin-flip occurs for AC frequency set at this peak by a two-photon second-order process. Also, no third-order (three-photon) peak at 1/3 of  $\Delta E_{ST}$  is observed. For higher amplitude the position of this peak gets red-shifted and the peaks corresponding to even denominators, e.g. 1/4 and 1/6 of  $\Delta E_{ST}$  are formed.

The study of the AC driven spin-flips in a symmetrix system with  $s_x = 29.64$  nm is summarized in Fig. 14 for two values of the external magnetic field B = 1.5 T and B = 2 T, both corresponding to the ground-state triplet. The position of the resonant second-order transition on the energy scale is redshifted with the amplitude [Fig. 14(a)]. In the studied range of the AC amplitudes – the spin-flip time is a decreasing function of the amplitude [Fig. 14(b)]. For 1.5 T the LZSM regime opens at 45  $\mu$ eV/nm, and for 2 T at still higher amplitudes which are outside the range studies in Fig. 14.

## IV. SUMMARY AND CONCLUSIONS

We have studied two electrons confined in a lateral double quantum dot defined within the two-dimensional electron gas on the (001)-oriented LAO/STO surface. Utilizing the three-band model of 3*d*-electrons localized at Ti ions on a square lattice, we have analyzed the energy spectrum of the system, paying particular attention to the singlet-triplet avoided crossing induced by the SO interaction. The calculated eigenstates have been employed as initial conditions for time-dependent calculations. In the magnetic field range close to the avoided crossing, the spin-flip singlet-triplet transition induced by an AC electric field has been analyzed in details in the context of the electric dipole spin resonance experiment.

We have demonstrated that for a fully symmetric system the first order singlet-triplet transition are forbidden



FIG. 14. (a) The position of the second-order (two-photon) singlet-triplet resonance in the AC field of  $eFx \sin(\nu t)$  for two values of the magnetic field. For both *B* values the ground-state of the system is the spin triplet. (b) The spin-flip transition time at the singlet-triplet resonance as a function of the driving field amplitude. Results for a symmetric double quantum dots with  $s_x = 29.64$  nm and  $V_0 = V_1 = 50$  meV.

due to the parity symmetry of the Hamiltonian which results in zeroing of the dipole matrix elements. To induce the spin-flip transition we introduce the asymmetry in the confinement potential in the form of unequal depths of the Gaussian quantum wells. Our simulations for a weakly asymmetric system show that the spin-flip singlet-triplet transition has the character of the Rabi oscillations for a low AC field amplitude, with minimal flip time of about 245 ps. Interestingly, when we increase the AC filed amplitude the frequency of the transition is blueshifted (redshifted) for the magnetic field below (above) the single-triplet avoided crossing. Moreover, we have noticed that for a magnetic field above the anticrossing the regular transition spectrum is highly deteriorated at a low frequency regime and the high AC field amplitude. This behavior has been explained as resulting from the Landau-Zener-Stueckelberg-Majorana transitions between the ground and first excited states which occur in the system when the electric field is large enough to drive the system across the avoided crossing.

The optimization of system geometrical parameters shows that the appropriate potential asymmetry between the dots highly reduces the leakage of transition to the higher states which can reach the value of about 0.05% with the spin flip time at the order of 330 ps. Finally our simulations for a fully symmetric system provide insight into the possibility of the spin-flip transition by the multiphoton processes. The singlet-triplet transition studied here will be relevant for future EDSR experiments aimed at reading the spin state of the dot and paving the way to a spin-orbit qubit at the oxides interface.

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