Spin-valley dynamics of electrically driven ambipolar carbon-nanotube quantum dots

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Abstract
An ambipolar $n$–$p$ double quantum dot defined by potential variation along a semiconducting carbon-nanotube is considered. We focus on the (1e,1h) charge configuration with a single excess electron of the conduction band confined in the $n$-type dot and a single missing electron in the valence band state of the $p$-type dot for which lifting of the Pauli blockade of the current was observed in the electric-dipole spin resonance (Laird et al 2013 Nat. Nanotechnol. 8 565). The dynamics of the system driven by periodic electric field is studied with the Floquet theory and the time-dependent configuration interaction method with the single-electron spin-valley-orbitals determined for atomistic tight-binding Hamiltonian. We find that the transitions lifting the Pauli blockade are strongly influenced by coupling to a vacuum state with an empty $n$ dot and a fully filled $p$ dot. The coupling shifts the transition energies and strongly modifies the effective $g$ factors for axial magnetic field. The coupling is modulated by the bias between the dots but it appears effective for surprisingly large energy splitting between the (1e,1h) ground state and the vacuum (0e, 0h) state. Multiphoton transitions and high harmonic generation effects are also discussed.

Keywords: carbon nanotubes, quantum dots, spin–orbit coupling, Pauli blockade

(Some figures may appear in colour only in the online journal)

1. Introduction
Manipulation of the spin degree of freedom for electrons confined in quantum dots (QDs) has been under extensive studies in the context of construction of spintronic single-electron devices [1] for over a decade. For QDs a successful implementation of the electron spin-resonance was performed with a microwave generator integrated into the device [2]. The magnetic field produced by ac currents [2] was soon replaced by the effective magnetic field due to the spin–orbit coupling for a driven electron motion [3–5], in the electric-dipole spin resonance [6] (EDSR). EDSR can also be induced by fluctuations of the Overhauser field [7] or inhomogeneous field [8] translated into an effective ac magnetic field by the driven electron motion [9]. The detection of the spin flip [2–5, 7] exploits the Pauli blockade [10] of the current that flows across a double QD.

The lifting of the Pauli blockade induced by ac electric field has been observed in double QDs defined within a semiconducting carbon nanotube (CNT) [11, 12]. The EDSR was observed for double QDs in an ambipolar work point: with one quantum dot storing an extra electron of the conduction band and the other quantum dot a hole (a single-unoccupied state) in the valence band. This charge configuration is denoted as (1e,1h) in the following. The energy spectrum of the (1e,1h) system was determined with the atomistic tight binding approach in [13]. The driven electron dynamics was discussed in [14] in a continuum approach strictly in the...
subspace corresponding to the (1e,1h) charge configuration. The dynamics of the (1e,1e) system in unipolar quantum dots has also been considered [15].

In CNTs the electron dynamics in resonant experiments involves both the spin and the valley [16, 17] degrees of freedom. In this work we solve the problem of the spin-valley transitions between the Pauli blocked and nonblocked states using a time-dependent configuration interaction approach and the Floquet theory [18, 19] for the ambipolar dots. We start from the CNT geometry that produces the best qualitative fit of the energy spectra to the observed transitions in external magnetic field. The spectral transitions overestimate by a factor of ~2 the experimental g factors for the axial magnetic field. By the study of the dynamics, we find that the states of the (1e,1h) charge configuration are strongly coupled by the ac potential with the ‘vacuum state’ (0e, 0h)—with an empty n-dot and a fully filled p-dot. The coupling, beyond the subspace considered in [14], produces a strong shift of the transition lines off the spectral energy differences. That, in turn, strongly modifies the effective g factors for the driven spin-valley transitions in axial magnetic field, removing the overshoot of the effective Landé factors for the axial field.

The present approach provides exact results for the coherent few-electron dynamics including the higher order effects, energy shifts and multiphoton transitions [4, 11, 20, 21]. The spin-valley dynamics enters the nonlinear regime already at relatively weak ac voltages. Motivated by this fact we look for the high harmonic generation (HHG) effects that are encountered in systems driven by strong laser fields (for recent studies of HHG in solids see [24–27]). Higher harmonics of the driven dipole moment are found but only in resonant conditions.

2. Theory

The description of the applied methodology is organized in the following way: in section 2.1 we discuss the model of the n – p quantum dot induced within a carbon nanotube. section 2.2 presents the time-dependent configuration interaction method that we use to describe the dynamics of the system. Finally, in section 3 we discuss the Floquet approach that is specifically suited for the treatment of the time-dependent problems with periodic time modulation of the Hamiltonian.

2.1. Model

We consider a carbon nanotube of length $L = 53.11\,\text{nm}$, diameter $2r = 1.33\,\text{nm}$ and the chiral vector $c_0 = (17, 0)$ for which the CNT is semiconducting and can confine electrons in quantum dots defined electrostatically. The nanotube is considered bent [28] above the electrostatic gates as in figure 1. We have found [13] that the bends of the CNT appearing at the area where the confinement potential is defined result in energy spectra which qualitatively agree with the experimental spectrum of Pauli-blockade-lifting transitions [12].

We model a double n – p quantum dot induced within the nanotube by external voltages. The shape of the electrostatic confinement potential is described by

$$W_{\text{QP}}(z) = -\frac{V_n}{1 + \left(\frac{z - z_0}{d}\right)^4} + \frac{V_p}{1 + \left(\frac{z - z_0}{d}\right)^4}, \quad (1)$$

where $V_n$ and $V_p$ are potentials on the n and p dots, respectively, $z_0$ is a shift of the QD centre from $z = 0$ and $d$ is a half width of the QD. In the calculations we use $V_n = V_p = 0.21\,\text{eV}$, $z_0 = 8\,\text{nm}$ and $2d = 16\,\text{nm}$. The experimental data [12] contain signatures of the intervalley mixing. In the present model the intervalley scattering is introduced by an effective potential peak of 1 $\text{eV}$ at a single atom at $z = -12.28\,\text{nm}$. The potential peak forms a short-range scattering centre which mixes the K and K' orbital states and, in result, enables the inter-valley transitions due to EDSR. In an experimental sample the short-range scattering centres are induced by defects of the crystal structure such as vacancies or impurities.

We apply an external magnetic field of magnitude B within the $xz$ plane and an angle $\alpha$ with respect to the $z$ axis: $\mathbf{B} = (B_n, 0, B_l) = (B \sin \alpha, 0, B \cos \alpha)$. In part of the calculations (section 4.4) an additional bias electric field $F_z$ is introduced. The resulting electrostatic potential is described by $W_{\text{E}}(z) = eF_z z$ for $z \in [-2d, 2d]$, $W_{\text{D}}(z) = W_{\text{D}}(-2d)$ for $z < -2d$ and $W_{\text{D}}(z) = W_{\text{D}}(2d)$ for $z > 2d$.

2.2. Time-dependent configuration interaction approach

In order to study the spin-valley dynamics of the confined carriers we (i) calculate the single-electron energy spectrum, (ii) solve the Schrödinger equation for the few-electron states with the Slater determinant basis built from the single-electron eigenstates, (iii) solve the time-dependent Schrödinger equation in the basis of the few electron eigenstates under the influence of the ac electric field. The results presented below provide an exact solution of the dynamics of the system spanned by the last four electrons in the states near the Fermi level.

In order to determine the single-electron states we use the atomistic tight-binding approach with the $p_z$ orbitals. We solve the eigenproblem of Hamiltonian

$$H = \sum_{\{i,j,\sigma,\sigma'\}} \left( c_{ij}^\dagger c_{ij}^\sigma \right) \varepsilon_{ij}^{\sigma\sigma'} + \text{h.c.}$$

$$+ \sum_{i,\sigma,\sigma'} c_{i}^\dagger \left( W_{\text{QP}} + W_{\text{D}}^\dagger \right) \delta_{\sigma\sigma'} + \frac{q_{\text{e}} B}{2} \frac{H_{\text{F}}}{} \right) c_{i}^{\sigma\sigma'} . \quad \text{(2)}$$

The first sum in equation (2) accounts for the hopping between the nearest neighbor atoms, $c_{ij}^\dagger(c_{ij})$ is the particle creation (annihilation) operator at ion $i$ with spin $\sigma$, and $\varepsilon_{ij}^{\sigma\sigma'}$ is the spin-dependent hopping parameter. The second sum accounts for the external electric and magnetic fields, with $\delta_{\sigma\sigma'}$ standing for the Kronecker delta and $W_{\text{D}}^\dagger W_{\text{D}}$ for the QD/bias potential matrix elements at $i$th ion, $q_{\text{e}} = 2$ stands for the Landé factor, $\mu_{\text{B}}$ the Bohr magneton and $\mathbf{B}$ for the vector of Pauli matrices.
In carbon nanotube QDs [17, 29–31] the SO coupling is provided by mixing the π and σ bonds [17, 32–36]. The spin–orbit interaction due to the curvature of the graphene plane introduces the spin dependence in the hopping parameters \( t_{ij}^{\sigma\sigma'} \). We apply the form of the parameters that accounts for both the folding of the graphene plane into the tube [35] and the curvature of the tube as a whole [37]. The explicit form of the hopping parameters is given in the appendix A.

With the single-electron problem solved, we calculate the few electron eigenstates using the configuration interaction (CI) method. We are interested in (1e, 1h) charge configuration, in which in the \( n \)-type dot we have a single-electron (1e) in the conduction band and three electrons per four accessible states (or a single unoccupied state 1h) at the top level of the valence band in the \( p \)-type dot (see the bottom part of figure 2(a)). For CI calculations we build the basis consisting of the highest energy level of the valence band (4 single-electron states) and the 5 lowest-energy levels of the conduction band (20 states). We assume that all the lower valence band levels are fully occupied.

The Hamiltonian for the interacting electron system reads

\[
H_{\text{int}} = \sum_u \epsilon_u g_u^\dagger g_u + \frac{1}{2} \sum_{ab;cd} V_{abcd} g_a^\dagger g_b^\dagger g_c g_d
\]  

(3)

where \( \epsilon_u \) is the energy of the \( u \)th eigenstate of Hamiltonian \( H_{1e} \) while \( g_u^\dagger \) and \( g_u \) are the creation and annihilation operators of the electron in the \( u \)th state. The second term of equation (3) accounts for the electron–electron interaction (explicit form of the interaction matrix elements \( V_{abcd} \) is given in the appendix B).

The atomistic approach used here accounts for all inter-valley effects [38] that accompany the short range component of the Coulomb potential. Moreover, the atomistic tight binding is not limited by the low-energy continuum approximation, and covers an ample variation of the external potential necessary for formation of an ambipolar quantum dot within the tube.

The four-electron energy spectrum is given in figure 3 as a function of the bias field \( F_b \). The slope of the lines is
determined by the electric dipole moment of the system i.e. the electron distribution between the dots. The non-degenerate energy level that grows fastest with the bias field $F_0$ is the vacuum state (0e, 0h). At $F_0 = 0$ the ground-state corresponds to the (1e,1h) charge configuration. The nonzero bias field is considered in section 4.4, elsewhere we take $F_0 = 0$.

The present model assumes that the length of the both the $n$- and $p$-type quantum dot is similar. Then, for both the dots to be incompletely filled in order to form the (1e,1h) ground-state, the confinement potentials in both the dots should be of a similar width/depth. Note, that the confinement potential that is included in the Hamiltonian has an effective character, since it includes not only the electrostatic fields induced by the gates but also the electrostatic screening by the valence band electrons besides the topmost energy level that enters explicitly the configuration interaction basis and the electron dynamics.

We simulate the valley and spin transitions driven by external ac field by solving the time-dependent Schrödinger equation with Hamiltonian

$$H'(t) = H_{dc} + \sum_{j=1}^{4} e F_0 z_j \sin(\omega t),$$

(4)

where $F_0$ is the ac electric field amplitude and $\omega$ is its frequency. Using the eigenstates $\Psi_n$ of Hamiltonian $H_{dc}$ we construct basis in which the time-dependent Schrödinger equation is solved

$$\Psi_{n,\tau + \Delta \tau} = \sum_n c_n(t) \Psi_n(t) e^{-i E_n \Delta \tau / \hbar}.$$  

(5)

In this basis the Schrödinger equation $i\hbar \frac{\partial \Psi}{\partial \tau} = H' \Psi$ takes the form

$$i \hbar \frac{d \Psi}{d \tau} = \sum_n c_n(t) e F_0 \sin(\omega t) \Psi_n(t) \Psi_n(t) e^{-i E_n \omega \Delta \tau / \hbar}. $$

(6)

We discretize the time in equation (6) and calculate the coefficients $c_k(t)$ using the Crank–Nicolson algorithm.

### 2.3. Floquet approach

The direct solution of the time-dependent Schrödinger method is supported with the Floquet theory. We use the Floquet Hamiltonian method [18, 19] to describe the dynamics of the system. For Hamiltonian $H'(t)$ that is periodic in time with the period $T = 2\pi / \omega$, the Floquet theorem asserts the existence of the solution of the Schrödinger equation

$$i \hbar \frac{\partial \Phi}{\partial \tau} = H' \Phi$$

(7)

of the form

$$\Phi(t) = \phi(t) e^{-i \epsilon \tau / \hbar},$$

(8)

where $\phi(t)$ is periodic in time with the same period $T$ as the Hamiltonian $H'(t)$. The $\Phi$ functions are called Floquet modes and upon the Fourier series expansion can be expressed by

$$\Phi_0(t) = e^{-i \omega \tau / \hbar} \sum_{n=-\infty}^{\infty} \sum_b \phi_{nb} e^{-i n \omega \tau / \hbar} b, $$

(9)

where $a$ enumerates the Floquet mode, $\epsilon_a$ is the quasienergy, $b$ are eigenfunctions of the Hamiltonian $H_{dc}$ and $\phi_{nb}$ are coefficients expanding the Floquet mode in the basis of $b$ eigenstates. Since functions $\Phi_0$ satisfy the equation ($H' - i \hbar \frac{\partial}{\partial \tau}) \Phi_0 = 0$, after expanding it in the basis of $b$ eigenstates we obtain

$$\sum_n \sum_b (\epsilon_a + ma) \phi_{nb} \phi_{nb} = 0, $$

(10)

where

$$H^{(m)} = \frac{1}{T} \int_0^T H'(t) e^{i m \omega t} dt. $$

(11)

For the periodic perturbation $e F_0 \cos(\omega t)$ the matrix elements $\langle a | H^{(m-n)} | b \rangle$ read

$$\langle a | H^{(m-n)} | b \rangle = E_{a} \delta_{ab} \delta_{mn} - \frac{i}{2} \delta_{m-n} \langle a | e F_0 | b \rangle \delta_{ab}. $$

(12)

Finally, one writes the time-independent Floquet Hamiltonian $H_F$ with the matrix elements defined by

$$\langle am | H_F | bn \rangle = \langle a | H^{(m-n)} | b \rangle + n \hbar \omega \delta_{ab} \delta_{mn} $$

(13)

in a block form we can express the matrix of the Hamiltonian $H_F$ as

$$H_F = \begin{pmatrix}
A + 2 \hbar \omega I & B & 0 & 0 & 0 & \cdots \\
B & A + \hbar \omega I & B & 0 & 0 & \cdots \\
0 & B & A & 0 & \cdots & \cdots \\
0 & 0 & B & A - \hbar \omega I & B & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \ddots \\
0 & 0 & 0 & B & A - 2 \hbar \omega I & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix}
$$

with the diagonal matrix $A$ containing on the diagonal the energies of the eigenstates $|a\rangle$ and matrix $B$ built of the elements $-\frac{i}{2} \langle a | e F_0 | b \rangle$. We solve the eigenproblem of the Hamiltonian $H_F$ and obtain a set of eigenergies $\epsilon_i$ and eigenstates $|e_i\rangle$.

We are interested in the transition probability from the initial state $|a\rangle$ at time $t_0$ to the final state $|b\rangle$ at time $t$. We use matrix form of the time-evolution operator $U(t, t_0)$

$$U_{ab}(t, t_0) = \langle b | U(t, t_0) | a \rangle = \sum_n \langle bn | e^{-i H_F \Delta t / \hbar} | a0 \rangle e^{i n a t} $$

(14)

where $\Delta t = t - t_0$. The transition probability reads

$$P_{a\rightarrow b}(t, t_0) = |U_{ab}(t, t_0)|^2 = \sum_n \sum_m \langle bn | e^{-i H_F \Delta t / \hbar} | a0 \rangle e^{i m a t} \langle am | e^{-i H_F \Delta t / \hbar} | bn \rangle. $$

(15)

We first average over $t_0$ [19]
The third electron of \( F \) or \( pK \) or \( \uparrow \rightarrow \downarrow \) and \( pK \) is still much stronger (figure 4(b)) than for \([T]\). Both these \( pK \) and then over \( \bar{a} \) pulse duration \( \Delta t \), finally obtaining

\[
P_{a \rightarrow b}(\Delta t) = \sum_{n} |\langle bn|e^{-i\hbar \Delta t/\hbar}|a0\rangle|^2.
\]

and over \( \bar{a} \) pulse duration \( \Delta t \), finally obtaining

\[
P_{a \rightarrow b} = \sum_{n} \sum_{\bar{b}} |\langle bn|\epsilon_{\bar{b}}\rangle|^{2} |\langle \epsilon_{\bar{b}}|a0\rangle|^2.
\]

In order to obtain convergent results for the probabilities \( P_{\bar{a} \rightarrow b} \) a sufficient number of the harmonics \( \pm \hbar a \) must be included into the \( H_{\bar{F}} \) matrix. For the calculations presented in the present paper 800 harmonics have been used.

3. Spectra in external magnetic field

In figures 4(a)–(c) we plot the calculated lowest energy levels of the (1e,1h) charge configuration as functions of \( \bar{B}=B_x \) and \( \alpha \), respectively. We denote the considered four-electron energy levels as N1, N2, B1 and B2, with the dominant Slater determinants listed in table 1. In all the four lowest-energy (1e,1h) states of figure 4, in the dominant configurations, two of the electrons of the \( p \) dot occupy the two-lowest energy states of the valence band \( pK \uparrow \) and \( pK' \downarrow \). The third electron of the \( p \) dot occupies one of the two-highest energy levels of the valence band \( pK' \downarrow \) or \( pK \uparrow \). Finally, the single electron in the \( n \) type dot occupies one of the two-lowest energy states of the conduction band \( nK \downarrow \) or \( nK' \uparrow \). The low-energy spectrum at \( B = 0 \) (figures 4(a) and (b)) consists of a ground-state singlet and an excited-state triplet. In the states B1 and B2 the last two electrons are spin-valley polarized, i.e. in B1 state the last electron in both \( n \) and \( p \) dot occupies the \( K' \uparrow \) energy level, and for B2 the occupied spin-valley level is \( K' \downarrow \). Both these states are blocked, i.e. the electron of the \( n \) dot is forbidden to pass to the \( p \) dot by the Pauli exclusion principle, since the state with the same spin and valley is occupied in the \( p \) dot. This is not the case for the other two states, denoted by N1 and N2. These states are referred to as ‘nonblocked’ in the following. The nonblocked states enter into an avoided crossing that is opened at \( B = 0 \) by the exchange integral \([13, 39]\), hence their non-linear dependence on \( B \) near 0T.

The orbital magnetic dipole moment due to the electron circulation around the tube \([40]\) is oriented parallel or antiparallel to the axis of the tube \([32]\). The circumferential spin–orbit interaction fixes the projection of the electron spin on the orbital magnetic moment \([17]\). Thus, the spins of the lowest-energy states are nearly polarized along the axis of the tube and a weak magnetic field applied in the \( x \) direction does not affect the energy levels of electrons confined within a straight CNT. The entire dependence of the energy levels on \( B = (B_0,0,0) \) that is visible in figure 4(a) results from the bend of the CNT. For the considered radii of the bend \( (R_o, R_p) \) in figure 2) the reaction of the energy levels on the magnetic field \( B = (0,0,B_z) \) is still much stronger (figure 4(b)) than for \( B = (B_0,0,0) \). The reaction anisotropy to the magnetic field vector leads to the distinct dependence of the spectrum on the angle \( \alpha \) (figure 4(c)).

4. Spin-valley transitions

4.1. Weak ac field

The lifting of the Pauli blockade in states B1 and B2 is achieved via ac electric field driven transitions to the nonblocked states N1 and N2 \([11–14]\)—see the arrows in figure 4.

Figure 5 shows the time-resolved occupation probabilities obtained for the \( B_2 \rightarrow N1 \) transition at the resonant driving frequency. For a weak ac field amplitude of 1kV cm\(^{-1}\) the transition is much faster for the magnetic field oriented perpendicularly to the axis of the CNT \( B = (B_0,0,0) \) (figure 5(a)) than for (0,0,\( B \)) (figure 5(b)). The transition \( B_2 \rightarrow N1 \) involves both the valley and the spin inversion with the transition of the occupied valence band state \( pK' \downarrow \rightarrow pK \downarrow \). For a straight CNT the spins are polarized in the \( z \) or \( -z \) directions. \( B \) oriented along the \( x \) direction contributes in mixing the \( \sigma_\zeta \) eigenstates and to opening the channel for the spin inversion, hence the shorter transition times.
functions of the driving ac frequency for a number of magnetic field values (figures 6(a) and (b)) and orientation angles (figure 6(c)) for a weak ac field $F_0 = 1 \text{ kV cm}^{-1}$. The dashed lines indicate the nominal transition energies which agree very well with the position of calculated probability peaks. The width of the peaks for $B_z$ is larger than for $B_x$ which is consistent with the shorter transition times (figure 5). For the case of figure 6—weak oscillations amplitude—the $g$ factors of the transition lines can be exactly estimated from the energy spectra.

Let us briefly comment on the relation of the energy differences between the blocked and nonblocked energy levels and the experimental EDSR transitions (figure 2 of [12]). (i) The dependence over the orientation angle of the magnetic field in the $(x,z)$ plane of both the experimental (figure 2(c) of [12]) and the present (figure 6(c)) results exhibits a pair of lines near zero transition energy/frequency separated by a gap from another pair of levels. The lines change in phase with the maxima near 0 and 180° and a minimum near 90°. (ii) In the upper pair of lines as a function of $B_x$ (figure 2(a) of [12]) one of the lines increases and the other decreases with the magnetic field. The effective $g$ factor extracted from the slope for the increasing line $g = \frac{\Delta E}{\mu_B \Delta B}$ of the experimental paper is 1.8 while for the data of figure 6 the value is 1.67 (we take the derivative near 0.05 T). For the decreasing line we find $-0.68$. Although no $g$ factor estimate has been given for that line in the experiment, from figure 4(b) in [14] we can assert that the slope of the decreasing line is less steep than for the increasing one, thus $|g| < 1.8$. For the lower lines we find $g$ factors of 1.49 and 0.86, while the experimental data provides 1.9 for one of the lines. For nonzero $B$ only a single lower line is resolved in the experiment. (iii) The largest deviation for the $g$ factors is found for $B_x$ orientation of the field: we find 8.67 and 4.42 for the $g$ factors in the upper branch, while the experimental paper produces 4.5 and 3, respectively. A possible explanation for the deviation of the actual transition spectrum from the energy spectra as due to the dynamics of the transitions is provided below.

4.2. Strongly driven system

We analyze the effect of the ac driving field amplitude on the dynamics. In figure 7 we plot the transition spectra for amplitudes $F_0 = 1, 2, ..., 5 \text{ kV cm}^{-1}$ at two different magnetic field orientations—$B_z = 0.05 \text{ T}$ and $B_x = 0.05 \text{ T}$. In both cases the increase of $F_0$ yields a few interesting effects: (i) broadening of the resonant peaks, (ii) shifts of the resonant transition energies (especially large for the upper branch lines), (iii) emergence of the $B_2 \rightarrow N_2$ transition at $B_2 \rightarrow N_1$ resonant peaks, and (iv) appearance of fractional resonances at the fractions of the resonant frequencies.
The transition $B_2 \rightarrow B_1$ is strongly shifted off the energy levels, although the $B_2 \rightarrow N_2$ transition is only times smaller than the one for $B_2 \rightarrow N_1$, and the dipole matrix elements for the transitions to $N_1$ and $N_2$ are similar (see table 2).

Note, that the dipole matrix elements between $B$ and $N$ states are non-zero only due to the presence of the intervalley and spin–orbit coupling, which introduce small admixtures of the opposite spin and valley in the single-electron states. The diagonal elements of table 2 are proportional to the electric dipole moments of the states. For the quadruple of $(1e,1h)$ states the dipole moments are similar, and for $(0e,0h)$ it is larger, which agrees with the slope of the energy levels in figure 3.

A deeper insight into origin of the resonant frequency shifts and appearance of the $N_2$ energy level can be drawn from an analysis of the convergence of the basis. Figure 9 presents the average occupation probability for $F_0 = 5 \text{ kV cm}^{-1}$ and $B_z = 0.05 \text{T}$ as obtained from the Floquet theory and a growing number of basis elements. We can see that the transition spectrum gets blue-shifted from the two-level Rabi transition (figure 9(a)) with the inclusion of the entire quadruple $(B_1, B_2, N_1, N_2)$ of the lowest-energy $(1e,1h)$ levels (figure 9(b)). Moreover a two-photon resonance for $N_1 \rightarrow N_2$ transition is observed at half the frequency of the direct resonance. Inclusion of the vacuum state $(0e,0h)$ (figure 9(c)) as the fifth element to the basis provides even stronger blueshift and produces a spectrum nearly identical with the convergent one (figure 9(d)). The transitions are blue-shifted as in the Bloch–Siegert shift [18, 41], however the effects of figure 7 do not follow the dependence on $F_0$ for the Bloch–Siegert transitions since they involve more than two energy levels.

We can see that the electron tunneling from $(1e,1h)$ quadruple to the nondegenerate $(0e,0h)$ state has a pronounced effect on the transition spectrum. Analyzing the right hand side of equation (6) we found (see appendix C) that for the transitions of figure 8 the electron from the initial state $B_2$ is transferred mostly to the vacuum state $(0e,0h)$—for which the transition matrix element (table 2) is the largest. Note, that this transition is off resonance, since the ac frequency in figure 8 is tuned to $B_2 \rightarrow N_1$ transition ($\sim 0.85 \text{ meV}$) and the $(0e,0h)$ state is about $20 \text{ meV}$ higher in the energy. Also, both $N_1$ and $N_2$ states get occupied by transitions from the vacuum state $(0e,0h)$. The vacuum state is more strongly coupled to $(1e,1h)$ states than are among each other (table 2). Thus the vacuum state serves as an intermediate state for transitions to $N_1$ and $N_2$. The transition $(0e,0h) \rightarrow N_1/N_2$ is immediate which prevents accumulation of the electron in the vacuum state $(0e,0h)$. The $(0e,0h)$ occupation probability in the conditions of figure 8(b) is $2\%$ at most (see figure C1 in appendix C). The $N_2$ state has also been observed in figure 5(b) for the smaller amplitude of $F_0 = 1 \text{ kV cm}^{-1}$. For this amplitude the vacuum state $(0e,0h)$ does participate in the transition with adequately lower maximal occupation probability of about $0.05\%$.

4.3. The effective $g$ factors for transitions lifting the spin-valley blockade

We investigate what is the influence of the ac field amplification on the $g$ factors obtained in the calculations. The dependence of the transition spectra on the magnetic field for a strong ac field of $F_0 = 5 \text{ kV cm}^{-1}$ is given in figure 10. Let us focus on the direct $B_1/2 \rightarrow N_1$ and $N_2 \rightarrow N_1$ transitions—the ones of the high energy branch of the plots.

For $B_z$ orientation of the field the three maxima are blue-shifted with respect to the energy difference, but the shift does not strongly depend on the magnetic field. Hence, the effective $g$ factors for the $B_z$ orientation are similar to the ones obtained from the energy spectra—see the upper half of table 3. Only a slight reduction of the absolute value of the $g$ factors is observed in the transition lines for $F_0 \geq 3 \text{ kV cm}^{-1}$.

For the magnetic field oriented along the $z$ axis the $g$ factors deviate more significantly from the ones obtained from the energy spectra. In the energy spectra for $B_z$, field the $N_2$ level shifts promptly up the energy scale off $B_2$ and $B_1$ energy levels, and the transition $N_2 \rightarrow N_1$ energy separates from $B \rightarrow N_1$ energy—on contrary to what is observed for $B_x$ field. When $N_2 \rightarrow N_1$ shifts off the $B_1/2 \rightarrow N_1$ transition energies, the blueshift of their energies is reduced, hence the reduction for the $g$ factors (lower part of table 3). The $g$ factors—as taken from the spectra were by a factor of $1.5–2$ larger than in the experiments. The $g$ factors as calculated from the transition spectra are significantly decreased for larger $F_0$. Moreover, the values for both $B \rightarrow N_1$ transitions produce similar $g$ factors at larger $F_0$, while for the $g$ factors extracted from the energy spectra one factor was nearly 2 times larger then the other.

The first row of table 3 presents the $g$ factors as obtained from the energy spectra. In the time dependent dynamics these values are reached for small AC field amplitude. The systematic dependence of the energy spectra for the CNT geometry was given in the previous paper [13]. The $g$ factors for the $z$ orientation of the magnetic field—the axis of a straight tube—were found relatively weakly affected [13] by deflection of the CNT. However, the details of the bend for the spectra are crucial for the $g$ factors for the magnetic field oriented in the $x$ direction [13]. For a straight CNT the discussed $g$ factors for $B_z$ are simply zero [13].

![Figure 8](image-url)
In order to check the nonlinear transitions for a modified CNT geometry we increased both $R_n$ and $R_p$ radii by 5% and the evaluated $g$ factors are given in the third column of table 3. The $g$ factors for $B_z$ are still only weakly affected. The factors for $B_x$ are more substantially changed (increased). Nevertheless, the same decreasing trend of the Landé factors as functions of the ac field amplitude is observed for modified radii.

4.4. Detuning and the transition energy shifts

The reduction of $g$ factors for the axial magnetic field discussed above has been obtained as a result of the energy shifts of the transition lines that occur for a relatively large amplitude of 5 kV cm$^{-1}$ (potential drop of 0.5 mV along 100 nm), while in the experiment [12] the amplitude of 0.5 kV cm$^{-1}$ was applied. However, we would like to emphasize that the widths of the quantum dots in the present calculations are a few times smaller than in the actual experimental setup, since the atomistic description sets the limits to the size of the system that can be effectively simulated. Therefore, in order to obtain the same effect for the energy levels a stronger ac electric field has to be applied in the modeled system. The factor which is crucial in the energy shifts discussed above is the participation of the vacuum (0e,0h) state in the transitions, and it varies not only with the amplitude but also with the bias $F_b$ between the dots. The latter shifts the position of the (0e,0h) state on the energy scale with respect to the (1e,1h) ground-state.

![Figure 9](image9.png)  
**Figure 9.** The transition spectra for $B_z = 0.05$ T and $F_0 = 5$ kV cm$^{-1}$ for varied basis of few-electron stationary Hamiltonian eigenstates: (a) 2-element basis, (b) 4-element (1e,1h) basis, (c) 5-element basis (4 states of (1e,1h) and a single (0e,0h) state), (d)–(e) 21-element basis. Spectra (a)–(d) are obtained within the Floquet approach and (e) through the solution of time-dependent Schrödinger equation.

![Figure 10](image10.png)  
**Figure 10.** Maximal occupation probability obtained for a solution of the Schrödinger equation for 200 ns as a function of the driving field frequency and the magnitude of (a) $B_x$ and (b) $B_z$ magnetic field. The ac driving field amplitude was set to 5 kV cm$^{-1}$.

| N1     | B1      | B2      | N2      | (0e,0h) |
|--------|---------|---------|---------|---------|
| 15.84  | 9.53·10^-5 | 3.23·10^-4 | 2.12·10^-2 | 1.03    |
| 9.53·10^-5 | 15.79  | 1.62·10^{-6} | 8.22·10^{-5} | 2.94·10^{-3} |
| 3.23·10^-4 | 15.79  | 5.21·10^{-4} | 15.80  | 0.433   |
| 2.12·10^-2 | 8.22·10^{-5} | 7.31·10^{-3} | 0.433  | 33.12   |

| N1 (0e,0h) | B1 (0e,0h) | B2 (0e,0h) | N2 (0e,0h) | (0e,0h) (0e,0h) |
|------------|------------|------------|------------|-----------------|
| 1.03       | 2.94·10^-3 | 7.31·10^{-3} | 0.433      | 33.12           |

| Table 2. Dipole matrix elements $\langle \psi_i | z | \psi_f \rangle$ between the energy levels B1, B2, N1 and N2 of (1e, 1h) charge configuration and the ‘vacuum state’ (0e, 0h) for $B_z = 0.05$ T. The results are given in nanometers. |
The position of these two states is controlled in the EDSR experiment by detuning voltage applied between the dots [4, 12]. The role of the detuning for the energy shifts can be estimated from Figure 11(a), where we plotted the result for $F_0 = 5\, \text{kV cm}^{-1}$ and $B_z = 0.05\, \text{T}$ (same as figure 9(e)) but for the bias $F_0 = 1\, \text{kV cm}^{-1}$. The (0e,0h) state is now about 40 meV above the (1e,1h) ground-state. The blue-shift of the transition peaks with respect to the energy splitting is reduced from large—as in figure 8—to barely visible—as in figure 11(a). For a weak ac field the transition lines at $F_b = 0$ coincide with the energy differences (see figure 7(b) for $F_0 = 1\, \text{kV cm}^{-1}$) and a stronger amplitude is required to produce the blue shifts of the transition lines. However, for $F_0 = -3\, \text{kV cm}^{-1}$—for which the vacuum energy level (0e,0h) is closer to the (1e,1h) ground-state (figure 3), the blue shifts appear—see figure 11(b) already at $F_0 = 1\, \text{kV cm}^{-1}$.

Concluding, the transition energy shifts that stand behind the variation of the $g$ factors appear also for small amplitudes provided that the coupling with the (0e, 0h) state is activated in the detuning bias.

In the simulations the vacuum (0e, 0h) state gets never very strongly occupied, and serves only as a transition channel between the (1e, 1h) energy levels of the lowest-energy quadruple. The coupling between the vacuum (0e, 0h) state and (1e, 1h) states has a tunnelling character. The effects of the tunneling for a quantum wire—quantum dot in the EDSR phenomenon was studied in [42]. The authors [42] found that the spin flipping probability gets below 1 for stronger ac field and that the transition times are lower than expected for the Rabi oscillations. The first effect was encountered in figure 8 as, according to the present study, a result of participation of a third state in the dynamics. We also reproduce the second feature for larger ac fields. The B2 → N1 transition time for the

Table 3. Effective Landé factors $g = \frac{1}{\mu_B} \frac{df}{d\Omega}$ for B → N1 transitions for the magnetic field oriented in $x$ (upper part) and $z$ direction (lower part of the table) as calculated from energy and transition spectra and the experimental results of [12]. In the calculated results the derivative is taken at 0.05 T. The second column contains the results for the CNT parameters applied in the entire paper and the third column the values for the bent radii increased by 5%. Due to a finite width of the transition peaks—at the energy scale—we estimate the precision in the evaluation of the $g$ factors to ±0.1.

| $g$ factors for $B_x$ | B2 → N1 | B1 → N1 | B2 → N1 (Rn, Rp increased by 5%) | B1 → N1 |
|----------------------|---------|---------|---------------------------------|---------|
| $E$ spectrum         | 1.7     | -0.7    | 2                               | -0.9    |
| Transitions for $F_0 = 3\, \text{kV cm}^{-1}$ | 1.6     | -0.6    | 1.9                             | -0.9    |
| Transitions for $F_0 = 5\, \text{kV cm}^{-1}$ | 1.3     | -0.5    | 1.7                             | -0.8    |
| Experiment [12]      | 1.8     | —       | 1.8                             | —       |

| $g$ factors for $B_z$ | B2 → N1 | B1 → N1 | B2 → N1 (Rn, Rp increased by 5%) | B1 → N1 |
|----------------------|---------|---------|---------------------------------|---------|
| $E$ spectrum         | 8.7     | 4.4     | 8.6                             | 4.5     |
| Transitions for $F_0 = 3\, \text{kV cm}^{-1}$ | 7.4     | 4.2     | 7.4                             | 4.2     |
| Transitions for $F_0 = 5\, \text{kV cm}^{-1}$ | 3.5     | 3.7     | 3.9                             | 3.7     |
| Experiment [12]      | 4.5     | 3       | 4.5                             | 3       |

Figure 11. Maximal occupation probability obtained for a solution of the Schrödinger equation for 200 ns for $B_z = 0.05\, \text{T}$ and (a) $F_0 = 5\, \text{kV cm}^{-1}$, $F_b = 10\, \text{kV cm}^{-1}$, (b) $F_0 = 1\, \text{kV cm}^{-1}$, $F_b = -3\, \text{kV cm}^{-1}$.
Figure 12. Fourier transform of the electron dipole moment for $F_0 = 1$ kV cm$^{-1}$ and $B_z = 0.05$ T at no bias field. Panel (a) shows the driving frequencies $\omega_c$ considered in this plot. In (b) and (c) B2 is the initial state. In (b) the driving frequency is $\omega_{ac} = 0.067$ meV—resonant for the B2–N1 transition. In (c) $\omega_{ac} = 0.05$ meV we are off resonance. The dashed line in (c) shows the position of the resonant frequency of (b). In (d)–(f) the initial state is N2. In (d) and (e) resonant ac frequencies are set for the direct ($\omega_{ac} = 0.0712$ meV) and two-photon ($\omega_{ac} = 0.0356$ meV) N2–N1 transition. In (f) an off resonant driving frequency is applied ($\omega_{ac} = 0.05$ meV). The dashed line in (f) shows the position of the N2–N1 direct resonance frequency.

4.5. Resonant transitions versus high harmonic generation

The results presented above contained a number of features characteristic to nonlinear optics. Besides the shifts of the direct transition lines also fractional resonances were observed, i.e. resonances at fractions of the direct transition frequency (see figures 7, 9 and 10). These transitions correspond to multiphoton absorption that is observed in atomic optics at intense laser fields. For the gated nanodevices the conditions for observation of the phenomena characteristic to nonlinear optics [4, 14, 43] appear at decent excitations of the order of 1 kV cm$^{-1}$ (or the potential drop of 1 mV over 100 nm). In gaseous phase [23] and in solids [24] strong laser fields ionize atoms and the ionized electrons are accelerated in the electric field of the laser. The oscillations of the dipole moment of the ionized electrons give rise to high harmonic generation [22] that is used in generation of ultrashort pulses [23].

We looked for high harmonics of the electron dipole moments that are driven by the ac field in our system [22]. We considered $F_0 = 1$ kV cm$^{-1}$, $B_z = 0.05$ T and no bias. Once the dynamics of the system is known we calculate the dipole moment $\mathbf{z}_d(t) \equiv e'(z_1 + z_2 + z_3 + z_4)$. Next, we Fourier analyze the dependence of the dipole moment on time. The results are presented in figure 12. We set the initial state to B2 in figures 12(b) and (c) and N2 in figures 12(d)–(f). The applied driving frequency of the electric field is taken resonant for the B2 → N1 transition figure 12(b) and resonant for the direct figure 12(d) and two-photon 12(e) N2 → N1 transitions as well as off resonances (figures 12(c) and (f)).
conditions we resolve up to 5th harmonic of the driving frequency (figures 12(b), (d) and (e)). Off resonances the highest harmonic of the spectrum is the 3rd one (figures 12(c) and (f)).

In off resonant conditions we notice a peak that correspond to the direct resonant transition B2–N1 (a tiny feature in figure 12(c)) and N2–N1 (a pronounced feature in figure 12(f)). For the latter plot also other lines are observed. The N2–N1 transition line is wide at the energy scale and couples strongly to other transitions.

Note, that in strong laser fields the high harmonic generation is a non-resonant phenomenon [22–24]. In the present conditions the yield of the high harmonics is still strongly related to the resonant transition spectrum. In resonant conditions the 2nd harmonics is by one (figure 12(e)), two (figure 12(d)) or three orders (figure 12(b)) of magnitude lower than the driving frequency. Off resonance the peak for the 2nd harmonic of the driving frequency is 4 (figure 12(f)) or 6 (figure 12(c)) orders of magnitude lower.

Obviously, high harmonics generated in our systems have only moderate order in comparison to HHG generated in atoms or molecules where the orders of 100 can be achieved [44]. Still, the new mechanism discussed by us, when optimized could in principle lead to generation of ‘truly high’ harmonics.

5. Summary and conclusions

We have analyzed spin-valley dynamics of the four last electrons in a n–p ambipolar quantum dot using a time-dependent configuration interaction method and the Floquet theory based on the single-electron states determined within the atomistic tight-binding approach. We studied the transitions lifting the Pauli blockade of the current within a quadruple of lowest-energy states of the (1e,1h) charge configuration. We discussed the results in the context of the accessible experimental data.

We demonstrated that the dynamics is significantly influenced by the coupling of the states of the (1e,1h) charge configuration with the nondegenerate vacuum (0e,0h) state. The vacuum state serves as a channel for transitions inside the (1e,1h) subspace and its participation in the transitions is determined by both the amplitude of the ac electric field and the bias electric field. The effect of the coupling are transitions energy shifts off the values expected from the eigenenergy spectra. The most significant consequence of the transition energy shifts is a strong modification of the g factors characterizing the dependence of the transitions on external axial magnetic field which removes the extensive overshoot of the effective Landé factors for the transition derived from the energy spectrum alone with respect to the experiment. We indicate that the transition energy shifts and thus the g factors can be manipulated by adjusting the strength of the ac field or the bias between the n and p dots. Emergence of other nonlinear effects has also been reported, such as fractional resonances or the high harmonic generation for resonant driving frequencies.

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Appendix A. Hopping parameters

In the present model the spin–orbit interaction due to the curvature of the graphene plane is accounted for in the spin dependence of the hopping parameters $t_{ij}^{σσ}$, $t_{ij}^{σν}$, $t_{ij}^{νσ}$, $t_{ij}^{νν}$, $t_{ij}^{νν} = i\delta \cos \phi_{ij} (x_{i} | H | z_{j}) - i\delta \cos \phi_{ij} (x_{i} | H | z_{j}) + i\delta \sin \phi_{ij} \sin \theta_{ij} (x_{i} | H | y_{j}) - i\delta \sin \phi_{ij} \sin \theta_{ij} (x_{i} | H | y_{j}), t_{ij}^{νσ} = (z_{i} | H | z_{j}) - i\delta \cos \phi_{ij} (x_{i} | H | z_{j}) + i\delta \cos \phi_{ij} (x_{i} | H | z_{j}) - i\delta \sin \phi_{ij} \sin \theta_{ij} (x_{i} | H | y_{j}) + i\delta \sin \phi_{ij} \sin \theta_{ij} (x_{i} | H | y_{j}), t_{ij}^{νν} = -i\delta \sin \phi_{ij} (x_{i} | H | y_{j}) + i\delta \sin \phi_{ij} (x_{i} | H | y_{j}) - \delta (\sin^{2} \frac{\phi_{ij}}{2} e^{i\theta_{ij}}) (z_{i} | H | y_{j}) + \delta (\sin^{2} \frac{\phi_{ij}}{2} e^{i\theta_{ij}} + \cos^{2} \frac{\phi_{ij}}{2} e^{-i\theta_{ij}}) (x_{i} | H | z_{j}).$
Appendix B. Electron–electron interaction

The electron–electron interaction is taken into account in the second term of equation (3) of the main text with the matrix elements

\[ V_{abcd} = \langle \psi_d(r_1, \sigma_1) \psi_b(r_2, \sigma_2) | H_c | \psi_i(r_1, \sigma_1) \psi_j(r_2, \sigma_2) \rangle = \sum_{i, j, k, l} \beta_{ik}^a \beta_{jl}^d \beta_{il}^b \beta_{kj}^c \times \langle p_i^\dagger (r_1) p_j^\dagger (r_2) | H_c | p_k^\dagger (r_1) p_l^\dagger (r_2) \rangle, \] (B.1)

where \( H_c \) is the Electron–electron interaction potential

\[ H_c = \frac{e^2}{4\pi \varepsilon \epsilon_0 r_2} \]

with \( r_{12} = |r_1 - r_2| \) and the dielectric constant \( \varepsilon = 9 \) as for \( \text{Al}_2\text{O}_3 \)—material which has been used as a substrate in the experimental setups [12]. The coefficients \( \beta_{ik}^a \) define contributions of \( p_i^\dagger \) orbitals of spin \( \sigma_i \) to the single-electron eigenstate \( i \). In the calculations we use the two-center approximation [46]. The on-site integral \( (i = j) \) we approximate by \( \langle p_i^\dagger p_i^\dagger | \frac{e^2}{4\pi \varepsilon \epsilon_0 r_2} | p_i^\dagger p_i^\dagger \rangle = 16.522 \text{ eV} \) [47] and for \( i \neq j \) we use the formula \( \langle p_i^\dagger p_j^\dagger | \frac{e^2}{4\pi \varepsilon \epsilon_0 r_2} | p_j^\dagger p_i^\dagger \rangle = \frac{e^2}{4\pi \varepsilon \epsilon_0} \delta_{ij} \) [39].

Appendix C. Role of \((0e,0h)\) state

This section contains additional analysis of the role of the vacuum state \((0e,0h)\) and the emergence of \( B_2 \rightarrow N_2 \) transition for ac driving frequency to \( B_2 \rightarrow N_1 \) resonance for \( B_z = 0.05 \text{ T} \) and \( F_0 = 5 \text{ kV cm}^{-1} \)—as in figure 8(b) of the paper. We limited the basis to the 5 states: \( N_1, B_1, B_2, N_2, (0e,0h) \) that govern the dynamics of the system (see the convergence test of figure 9 in the paper). In figure C1 we repeat a very beginning of the simulation presented in figure 8(b) only using the limited basis. We plot the vacuum state \((0e,0h)\) occupation—previously omitted. The occupation of the vacuum state appears in peaks which are followed by a step of the occupation of the \( N_2 \) state.

The role of the vacuum state \((0e,0h)\) in the dynamics of the system can be followed by an analysis of the solution of the Schrödinger equation in the configuration interaction method

\[
\ni\hbar \dot{c}_n(t) = \sum_n c_n(t) e F_0 \sin(\omega t) \langle \Psi_1 | \hat{\mathbf{r}}_n \Psi_1 \rangle e^{-\frac{\mu E_n - E_0}{\hbar}}. \tag{C.1}
\]

We integrate both sides over time with limits 0 and \( \tau \) and obtain

\[
c_n(\tau) = c_n(0) + \frac{1}{i\hbar} \int_0^\tau \sum_n c_n(t) e F_0 \sin(\omega t) \langle \Psi_1 | \hat{\mathbf{r}}_n \Psi_1 \rangle e^{-\frac{\mu E_n - E_0}{\hbar}} dt \equiv c_n(0) + \sum_n J_{n0}(\tau). \tag{C.2}
\]

The integrated matrix elements \( J_{n0}(\tau) \) contain information on the transitions between the basis states.

In table C1 we label the five states of the limited basis \((N_1,B_1,B_2,N_2,(0e,0h))\) by their energy order in the spectrum as 1st, 2nd, 3rd, 4th and 17th state in the basis, respectively. In the initial condition the system occupies state \( B_2 \) (i.e. the 3rd one). The third column of table C1 indicates that the driving—resonant with \( B_2 \rightarrow N_1 \) transition (or 3 → 1) induces most effectively the transition to the vacuum state \((0e,0h)\) (17th). The transition flux \( J_{17,4} \) is about 30 times more effective than \( J_{1,4} \). The last column of table C1 shows that transitions from the vacuum state go to both \( N_1 \) (1st) and \( N_2 \) (3rd) states. The direct transitions between the \( N_1 \) and \( N_2 \) states are by 1.5 or 2 more effective than \( N \rightarrow (0e,0h) \) transitions—see \( J_{4,1} \) and \( J_{4,17} \) and \( J_{17,4} \) and \( J_{17,1} \), etc.

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