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Electronic structure of artificial atoms and molecules: spin-orbit coupling effects

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Abstract

This thesis describes effects of the coupling of spin and spatial degrees of freedom for electrons confined in quantum dots. The discussion is carried mainly in the context of spin control by orbital degrees of freedom. The dissertation consists of eleven chapters: introduction (1) and summary of published articles and manuscript that appeared as a result of PhD research (2). The papers are included in subsequent chapters (3-10) followed by the manuscript (11). Chapters (3-5) study single and multiple few-electron lateral quantum dots in the presence of Rashba and Dresselhaus spin-orbit coupling in terms of mixing of electron orbitals, anisotropy of spin exchange between neighboring quantum dots and impact of the dot orientation with respect to crystal host lattice on effective Landè factor. In next two chapters (6-7) we describe proprieties of quantum dots shaped as quantum rings in the context of breaking the circular symmetry of the charge density by spin-orbit coupling and possible application of such structures to perform controlled spin rotations. We refer to recent experimental results explaining the anisotropy of spin-orbit coupling observed in a single self-organized quantum dot (8) and the role of tunnel coupling on the observed spectrum of the electric dipole spin resonance in gated nanowires (9). Description of anisotropic spin polarization and lifting of the Pauli blockade due to spin relaxation and electric dipole spin resonance in nanowire quantum dots is given in the last two chapters (10-11).

Streszczenie

Poniższa rozprawa opisuje efekty sprzężenia pomiędzy spinowym i przestrzennym stopniem swobody elektronów uwięzionych w kropkach kwantowych. Dyskusja prowadzona jest głównie w kontekście kontroli spinu poprzez orbitalny stopień swobody. Praca zawiera jedenaście rozdziałów: wstęp (1) oraz podsumowanie artykułów i manuskryptu, które powstały jako efekt badań przeprowadzonych w trakcie doktoratu (2). Publikacje i manuskrypt tworzą kolejne rozdziały (3-11) rozprawy. W rozdziałach (3-5) badamy własności pojedynczych i wielokrotnych kilkuelektronowych kropek kwantowych w obecności oddziaływania Rashby oraz Dresselhausa w kontekście mieszania orbitali elektronowych, anizotropii wymiany spinu między sąsiadującymi kropkami oraz wpływu orientacji kropki w odniesieniu do materiału podłoża na efektywny czynnik Landègo. W kolejnych dwóch rozdziałach (6-7) badamy własności kropek kwantowych w postaci pierścieni kwantowych w kontekście łamania symetrii gęstości ładunku przez sprzężenie spin-orbita i możliwości wykorzystania tych struktur do przeprowadzania kontrolowanych obrotów spinu. Odnosimy się do nowych wyników eksperymentalnych wyjaśniając anizotropię oddziaływania spin-orbita obserwowaną w pojedynczej samozorganizowanej kropce kwantowej (8) oraz wpływ sprzężenia tunelowego na zmierzone widmo elektrycznych rezonansów spinowych w bramkowanych drutach półprzewodnikowych (9). Opis anizotropowej polaryzacji spinowej oraz znoszenia blokady Pauliego poprzez relaksację spinową i rezonanse spinowe w kropkach kwantowych zdefiniowanych w nanodrutach jest przedstawiony w dwóch ostatnich rozdziałach (10-11).

Samenvatting

Dit proefschrift beschrijft gevolgen van de koppeling van de spin en ruimtelijke vrijheidsgraden voor elektronen opgesloten in kwantumstippen. Het centrale thema betreft de spin controle door orbitale vrijheidsgraden. Het proefschrift bestaat uit elf hoofdstukken: inleiding (1) en samenvatting van gepubliceerde artikelen en manuscript, die verschenen als resultaat van dit promotieonderzoek (2). De papers worden in de volgende hoofdstukken (3-10) gevolgd door het manuscript (11). Hoofdstukken (3-5) bestuderen enkelvoudige en meervoudige-elektron laterale kwantumstippen in de aanwezigheid van Rashba en Dresselhaus spin-baan koppeling in termen van de menging van elektronorbitalen, anisotropie van spin-uitwisseling tussen naburige kwantumstippen en de impact van de stip oriëntatie met betrekking tot de kristal gastmatrix op de effectieve Landè factor. In de twee volgende hoofdstukken (6-7) beschrijven we de eigenschappen van kwantumstippen, die de vorm hebben van kwantumringen, in de context van het breken van de cirkelvormige symmetrie van de ladingsdichtheid door spin-baankoppeling en mogelijke toepassing van dergelijke structuren om gecontroleerde spin rotaties uit te voeren. We verwijzen naar recente experimentele resultaten en verklaren de anisotropie van spin-baan koppeling waargenomen in een zelf-georganiseerde kwantumstip (8) en de rol van tunnel koppeling op het waargenomen spectrum van de elektrische dipool spin resonantie in 'gated' nanodraden (9). Een beschrijving van de anisotrope spinpolarisatie en de opheffing van de Pauli blokkade ten gevolg van spin relaxatie en elektrische dipool spin resonantie in nanodraad kwantumstippen wordt gegeven in de laatste twee hoofdstukken (10-11).

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About the dissertation

The present dissertation is composed of a collection of articles that describes the effects of spin-orbit interaction on the electronic structure of artificial atoms and molecules, i.e. single and systems of quantum dots. The dissertation consist of eight published papers:

A.1 M. P. Nowak and B. Szafran, *Coupling of bonding and antibonding electron orbitals in double quantum dots by spin-orbit interaction*,
Physical Review B 81, 235311 (2010),

A.2 M. P. Nowak and B. Szafran, *Time-dependent configuration-interaction simulations of spin swap in spin-orbit-coupled double quantum dots*,
Physical Review B 82, 165316 (2010),

A.3 M. P. Nowak and B. Szafran, *Singlet-triplet avoided crossings and effective g-factor versus spatial orientation of spin-orbit-coupled quantum dots*,
Physical Review B 83, 035315 (2011),

A.4 M. P. Nowak and B. Szafran, *Spin-orbit coupling effects in two-dimensional circular quantum rings: Elliptical deformation of confined electron density*,
Physical Review B 80, 195319 (2009),

A.5 M. P. Nowak, B. Szafran, and F. M. Peeters, *Fano resonances and electron spin transport through a two-dimensional spin-orbit-coupled quantum ring*,
Physical Review B 84, 235319 (2011),

A.6 M. P. Nowak, B. Szafran, F. M. Peeters, B. Partoens, and W. J. Pasek, *Tuning of the spin-orbit interaction in a quantum dot by an in-plane magnetic field*,
Physical Review B 83, 245324 (2011),

A.7 M. P. Nowak, B. Szafran, and F. M. Peeters, *Resonant harmonic generation and collective spin rotations in electrically driven quantum dots*,
Physical Review B 86, 125428 (2012),

A.8 M. P. Nowak and B. Szafran, *Spin polarization anisotropy in a narrow spin-orbit-coupled nanowire quantum dot*,
Physical Rev B 87, 205436 (2013),

and manuscript that is under review at the stage of preparation of the thesis:

M.1 M. P. Nowak and B. Szafran *Spontaneous and resonant lifting of the spin blockade in nanowire quantum dots*,
arXiv:1303.0211 (2013).

The collection of articles is preceded by the introduction and summary of the works.

1 Motivation and context of the thesis

During the last two decades significant progress has been made in confining and isolating single and few charge carriers in three dimensional potential cavities within a semiconducting medium that are called quantum dots. This opened unprecedented opportunity to study quantum phenomena at the nanoscale and to take a step towards quantum information processing. Electrons trapped in quantum dots possess a discrete energy spectrum [1] and such systems are often referred to as artificial atoms. Coupled quantum dots – where the electron wavefunction forms extended orbitals – are called artificial molecules.

The ability to control single carries opens a possibility for quantum computation that utilizes a new class of algorithms [2, 3] that take advantage of quantum parallelism. Spin of the electron confined in a quantum dot is considered as a good candidate [4] for a bit of quantum information – qubit – due to its weak coupling to the environment which increases the coherence time. For realization of quantum algorithms single and two-qubit gates are required. While the latter can be realized relatively simple – by controlling the exchange coupling between the spins [5] – the control of single spin raises more difficulties. First demonstration of single electron spin rotation employed Rabi resonances induced by oscillating magnetic field [6]. This method however cannot be used for the creation of scalable devices as the electrode used for the creation of a local AC magnetic field is of considerable size. For practical implementation of quantum gates electrical control of electron spin would be preferable. This is made possible by spin-orbit coupling. Spin-orbit interaction is an effect of relativistic origin where in the electrons reference frame the external electric field results in an effective magnetic field that depends on the electron momentum and affects the electrons spin. In semiconductor nanostructures

there are two possible sources of the spin-orbit interaction. First of them is Rashba [7] spin-orbit coupling which arises from the external electric fields in the structure. The second is Dresselhaus [8] spin-orbit interaction which results from the inversion asymmetry of the crystal structure in zincblende semiconductors. The strength of the spin-orbit interaction depends on the geometry of the device, and can be controlled by the strength of the external electric [9] or magnetic fields [10]. In recent years a great attention has been paid to spin-orbit coupling in quantum dots which resulted in measurements of i.e. spin precession in an effective magnetic field [11], avoided crossings in the energy spectra [12, 13], their dependence on the orientation of the external magnetic field [14] and first demonstration of coherent electrical spin control [15]. On the other hand spin-orbit interaction leads to coupling of the spin with the environment through orbital degrees of freedom which results in spin decoherence and relaxation [16, 17] and lifts the spin polarization in external magnetic field limiting the ability to store information in the spin degree of freedom.

The present thesis is devoted to spin-orbit interaction effects in quantum dots and its application to the control of the electron spin in artificial atoms and molecules. The present work describes effects of spin-orbit interaction in lateral structures and explains recent experimental results on self-organized quantum dots and gated nanowires.

2 Summary of the articles

2.1 Article A.1, *Coupling of bonding and antibonding electron orbitals in double quantum dots by spin-orbit interaction*

The first experimental demonstration of the control of a single [6] and a pair [5] of spins in a solid state exploited planar quantum dots. These structures are created by tailoring of the confinement potential of the two dimensional electron gas in a doped heterostructure. In article [A.1] we describe effects of spin-orbit interaction in lateral few-electron coupled quantum dots.

To study the two-dimensional quantum dots we developed a computational scheme in which the single electron spin-orbitals are obtained in a basis consisting of Gaussian functions distributed on a square mesh. This approach is applicable to systems without any symmetry and takes into account Rashba and Dresselhaus

spin-orbit interactions. We applied the configuration interaction scheme to obtain the solution for two- and three-electron in a numerically exact manner.

We describe experimentally measurable [18] consequences of spin-orbit interaction on the optical absorption spectra in a microwave regime. We find that spin-orbit coupling results in a mixed character of the states in the single-electron regime that posses bonding and antibonding components. For odd-number of electrons this results in opening avoided crossings between the states of opposite parity and spin visible in the absorption spectra. For the two-electron system the optical transitions are possible only in the presence of spin-orbit coupling that lifts the spin-polarization of the states and opens singlet-triplet anticrossing in the lowest part of the energy spectra.

2.2 Article A.2, *Time-dependent configuration-interaction simulations of spin swap in spin-orbit-coupled double quantum dots*

Laterally coupled quantum dots have been used [5] for for realization of two-qubit quantum gates that realize *SWAP* and *SQUARE ROOT OF SWAP* operations that are based on spin exchange [19] between the two electrons confined in adjacent coupled dots. Without spin-orbit coupling the process is described by isotropic Heisenberg Hamiltonian where the spin exchange is controlled by the strength of the tunnel coupling [20]. Spin-orbit interaction is known to introduce anisotropic corrections [21] to the spin exchange that results in a dependence of the process on the initial direction of the spin polarization and limits the fidelity of the *SWAP* operation. The previous works discussed ways to minimize the anisotropy [22, 23] or to utilize it for the construction of quantum gates [24, 25]. Recent work [26] showed that in zero magnetic field the anisotropy vanishes but only for a specific choice of the spin basis. In regard of the ongoing discussion in the work [A.2] we perform a numerical experiment to study the spin swap process as it takes place in time under the presence of spin-orbit interaction.

We adopt a calculation scheme of [A.1] to obtain eigenstates of coupled quantum dots and used them to prepare the initial state for time evolution such that the two electrons are in opposite spin configuration and are localized in adjacent dots. We present that for neglected Coulomb interaction the spin exchange involves

tunneling of spin-opposite electrons between the dots which is accompanied by the precession of their spins in the spin-orbit effective magnetic field. This results in an appearance of additional spin components that depend on the initial spin polarization direction even without the magnetic field. The Coulomb interaction blocks the electron motion but the tunneling of the spin densities accompanied by spin precession is still observed. We find that the Coulomb interaction results in the generation of spin components in the direction of the spin-orbit field which are maximal at the half-time of the spin *SWAP* – altering the *SQUARE ROOT OF SWAP* operation. Moreover we present a way to restore the isotropy of the exchange process by proper choice of the double dot orientation with respect to crystal lattice due to cancelation of the effective Rashba and Dresselhaus fields.

2.3 Article A.3, *Singlet-triplet avoided crossings and effective g-factor versus spatial orientation of spin-orbit-coupled quantum dots*

The orientation of the lateral quantum dots with respect to the crystal axes is defined at the stage of fabrication of the device [27]. In the work [A.3] we demonstrate that in the presence of both Rashba and Dresselhaus coupling the energy splitting between the spin opposite states depends on the orientation of the dot. We explain that in the presence of spin-orbit coupling the strength of the Zeeman interaction that polarizes the spins depends on the extension of the charge density in the (001) plane provided that the length of the dot is comparable with the spin-orbit length (that is inversely proportional to the coupling strength). This results in changes of the effective g-factor calculated from the spin splittings. If the strength ratio of the Rashba and Dresselhaus coupling is detuned from 1 the dependence becomes weaker. The change of spin polarization results in changes of the width of the avoided crossing between singlet and triplet which is observed in the lowest part of the two-electron energy spectrum.

2.4 Article A.4, *Spin-orbit coupling effects in two-dimensional circular quantum rings: Elliptical deformation of confined electron density*

Special case of quantum dots are the circular structures with removed center that form quantum rings. Closed quantum rings have been studied in the context of persistent currents [28] and magnetization [29, 30, 31] in an external magnetic field. Previous studies on spin-orbit coupled quantum rings focused on the one-dimensional approximation of the structure [32]. Recent study [33] showed that spin-orbit interaction leads to breaking of the rotational symmetry of the charge density in the quantum ring. We employ configuration interaction calculations on a Gaussian mesh to describe a realistic quantum ring with finite width channels. We present that the one-dimensional approximation results in several artificial results and find that in the presence of both Rashba and Dresselhaus spin-orbit coupling rotational symmetry of the charge density is broken but *only* in the presence of external magnetic field or in the presence of both couplings with unequal strengths. We find that even for radically reduced channel width the exact results do not tend to the ones obtained in the one-dimensional approximation [33].

Moreover we show that the deformation of the charge density is increased by the electron-electron repulsion in the two electron regime. The breaking of the rotational symmetry of the charge density by the spin-orbit coupling corresponds to changes in experimentally accessible quantities such as the chemical potential of the ring and the magnetization that resembles the dependencies known for rings with defects [34].

2.5 Article A.5, *Fano resonances and electron spin transport through a two-dimensional spin-orbit-coupled quantum ring*

Open quantum rings allow for the realization of two-path interferometers where the phenomena related to phase change in the vector potential of the external magnetic field (Aharonov-Bohm effect [35]) or in the spin-orbit field (Aharonov-Casher effect [36]) results in modification of the conductance. Moreover the spin-orbit-coupled rings are considered for performing controlled rotations of transported electron spin [37]. The work [A.5] studies electron transport through the quantum ring in the

presence of Rashba spin-orbit coupling that results from the electric field present solely in the ring area. This approach allows for well defined spins in the leads and is realized by the addition of an electrode above the ring [38].

We developed a computational scheme that allows for the solution of the electron transport problem through a quantum ring with two-dimensional channels by the solution of time-independent Schrödinger equation with boundary conditions that assume the electron to enter from one of the leads. We find that in contrary to the one-dimensional studies [37] that in the presence of spin-orbit interaction Fano resonances appear in the conductance of the ring. The resonances occurs for Fermi energy that matches the energy of resonance states localized in the ring – whose wavefunctions vanish in the leads – and appear due to the breaking of the symmetry of the localized states by the spin-orbit coupling. We find that in the resonances the spin polarization of transported electron is altered which is due to the coupling to the spin of the resonance states. We demonstrate that the observed Fano resonances are the narrowest when the spin-orbit coupling strength is tuned to the maxima of the Aharonov-Casher oscillations. When the strength is detuned from the maxima the resonances become broader and the spin polarization at the output of the ring strongly depends on the electron Fermi energy which was not the case in the previous studies [37].

2.6 Article A.6, *Tuning of the spin-orbit interaction in a quantum dot by an in-plane magnetic field*

In 2010 the first transport spectroscopy measurement of a *single* self-organized quantum dot was reported [14]. Self-organized quantum dots are formed in the Stranski-Krastanov growth and are known for an almost a decade. The previous studies however were limited to the optical spectroscopy that were performed on ensembles of this objects. The study of Ref. [14] revealed broad anticrossings in the few-electron energy spectra of a single quantum dot opened by spin-orbit interaction that is exceptionally strong in these structures due to their limited height (that increases the strength of Dresselhaus coupling) and potential profile in the dot [39] that results in strong built-in electric fields (that increase the strength of Rashba coupling). The measured anticrossings [14] changed their width as the orientation of the external magnetic field was varied. The experimental work concluded that they are due to

Rashba coupling but did not provide any further explanation. Article [A.6] is the first theoretical description of a spin-orbit coupled three-dimensional quantum dot in a single and two-electron regime and provides a theoretical explanation of the experimental findings of Ref. [14].

For studies performed in work [A.6] a new computational scheme was developed that allowed for the inclusion of the full Rashba and Dresselhaus Hamiltonians into a three dimensional calculation in an efficient way. Two-electron eigenstates are obtained using a configuration interaction scheme where the electron-electron interaction matrix elements are calculated through the solution of the Poisson equation in the quantum dot.

In one-electron energy spectra we find avoided crossings opened separately by Rashba and Dresselhaus couplings whose width changes with the orientation of the in-plane magnetic field. We explain that the width of the anticrossing depends on the mixing ration between the states of opposite spin and parity and that due to the different form of the Rashba and Dresselhaus Hamiltonians this mixing vanishes for different orientation of the magnetic field. Under the presence of both couplings the magnetic field orientation for which the anticrossing is the narrowest depends on the relative strength of Rashba and Dresselhaus couplings. We compared results of our calculation in the two electron regime with the experimental data and explained that the results indicate the presence of Dresselhaus coupling in the structure. Furthermore we derive analytical formula which allows for the calculation of the relative strength of the spin-orbit couplings from the experimentally accessible data.

2.7 Article A.7, *Resonant harmonic generation and collective spin rotations in electrically driven quantum dots*

For the realization of a complete set of quantum gates the implementation of single qubit quantum gates that require single spin rotations is necessary. Electrical spin rotations are performed by electric dipole spin resonance which relies on transitions between spin states induced by oscillating electric field mediated by spin-orbit coupling when the oscillation frequency matches the Larmor frequency in weak external magnetic field [6]. A lot attention was paid [40, 41, 42, 43, 44, 45] to InAs and InSb nanowire quantum dots that allow for efficient electrical control of electron spin due the strong spin-orbit interaction in this systems [12, 13]. The work [40]

measured maps of the current in function of the magnetic field and electric field frequency that revealed central resonance line due to transition from spin-parallel triplet states to degenerate spin-antiparallel state. Recent experiment [42] presented detailed spectroscopy of the electric dipole spin resonance spectrum revealing the additional resonance line at the fractional frequency and the splitting of the central line. We developed a computational scheme that allowed for simulation of electric dipole spin resonance in a two-electron nanowire artificial molecule for the explanation of this observation and to study the impact of the electron-electron interaction on the spin rotations.

The calculations are based on a quasi-one-dimensional model of the nanowire with the exact inclusion of the Coulomb interaction. The simulation reveals that for non-zero interdot coupling the exchange interaction results in the splitting of the central resonance line which is accompanied by the collective rotation of the spins of both electrons. We also find that in the fourier transform of the driven electron momentum there are harmonics present of the driving frequency. When one of the harmonics matches the resonant frequency a transition appears which is the origin of the experimentally resolved resonances at the half frequency [42]. The fractional resonances appear only if the exchange coupling between electrons is nonzero, i.e., they are accompanied by the splitting of the central resonance line. Moreover we find that the fractional transitions appear also in a single-electron regime also without the spin-orbit coupling – the resonant generation of harmonic frequency is an intrinsic propriety of an electrically driven electron.

2.8 Article A.8, *Spin polarization anisotropy in a narrow spin-orbit-coupled nanowire quantum dot*

The electric dipole spin resonance experiments measure the main resonances at the frequency that corresponds to the energy separation due to Zeeman splitting in an external magnetic field. In experiments [41, 42] this allowed for measurement of the effective Landè factor for different in-plane orientation of the magnetic field. The measurements revealed strong anisotropy of the effective g-factor. In the work [A.8] we study effects of anisotropic spin polarization in nanowire quantum dots in the context of the experimental findings.

We solve the three dimensional Schrödinger equation for single and two-electrons

confined in a cylindrical quantum dot in the presence of Rashba coupling. We find that the degree of spin polarization of the states depends on the orientation of the external magnetic field. Surprisingly we find that the spin polarization becomes almost complete for decreasing radius of the wire. We present an analytical solution for the phenomena in the one-dimensional limit where we present analytical forms of the spin-orbitals that allows us to explain the changes of the effective g-factor. In the two-electron regime we perform configuration interaction calculation and relate our results to the experimental findings [41, 42]. We find that the form of the g-factor dependence on the magnetic field orientation matches the one obtained in the experiment. Moreover calculated width of the avoided crossing between the energy levels of the singlet and triplet states and its dependence on the magnetic field orientation agrees with the findings of Ref. [42].

2.9 Manuscript M.1, *Spontaneous and resonant lifting of the spin blockade in nanowire quantum dots*

Observation of the electric dipole spin resonance [40, 41, 42, 43, 44, 45] relies on the measurement of the current increase associated with the lifting of the Pauli blockade in the two electron coupled quantum dots. The single electron current is blocked when the two electrons initialize in spin parallel configuration forming a triplet state. Spin rotation drives the system into a $S = 0$ state which relaxes to the singlet ground state (with double occupancy of a single dot), which is followed by the escape of one of the electrons to the drain electrode. However, in the presence of spin-orbit coupling, phonon mediated spin-relaxation occurs [17, 46, 47] as a concurrent process to the electric dipole spin resonance, lifting of the blockade also from spin parallel states. So far the latter process has been considered in the context of the leakage current [48, 49]. In work [M.1] we present results of our simulation that describes the time dependent process of electric dipole spin resonance along with phonon mediated relaxation in the presence of spin-orbit coupling.

We find that the spin-nonconserving relaxation occurs provided the energy separation between initial and final states for the relaxation is small enough. This leads to spontaneous lifting of the blockade from one of the Zeeman split triplets. The only remaining blocked state is the spin triplet with spins polarized antiparallel to the magnetic field and in a weak magnetic field the resonances are observed solely

from this state. When the external magnetic field is increased the spin positive triplet becomes the ground state which results in the restoration of the blockade from this state. This results in the appearance of an additional resonance which is related to the spin rotation accompanied by charge reconfiguration. We identify the latter resonance in the recent experimental results [44].

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