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Chapter 3

On the ‘Three-Orders Time-Limit’ for Phase Decoherence in Quantum Dots

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Abstract

The recent huge increase of the interest in the implementation of quantum information processing (QIP) focused an attention on the semiconductor quantum dots (QDs) as nano-systems with controllable quantum states both in charge and spin degrees of freedom. This creates an opportunity for discussing and modeling of QD-based qubits and quantum-logic gates for prospective quantum computer. Despite of many efforts in this respect the progress is, however, disappointing and the barrier of so-called diVincenzo criteria required to be fulfilled for the QIP technology is not broken and even not approached in any realistic proposal of using QDs for qubits and quantum gates. The reason of this situation is linked with unavoidable restrictions imposed on the coherence and the coherent control of quantum states in QDs, which precludes scalable implementation of qubits and quantum gates based on QDs. The decoherence of charge and of spin degrees of freedom in QDs and QD-systems locates inconveniently straight in the middle of the required six-orders of magnitude diVincenzo window for time scale of the control in comparison to the decoherence rate. This leads to the so-called ‘three-orders time-limit’ for QIP implementations in QDs. Some advantages of spin degrees of freedom in QDs can be noticed, however. Even though the time scale of the decoherence of spin in of particular practical importance magnetic QDs still is located in the center of six-orders diVincenzo window, the amplitude of the phase decoherence of spin might be frozen on a small level in proximity of the zero temperature. Short critical overview of the dephasing restrictions imposed on the QD technology is the aim of this chapter.

Keywords: quantum dots, quantum information processing (QIP), dephasing

1. Introduction

The notion of a quantum dot [1–3] comprises various nanometre-size semiconductor structures, manufactured by means of different technologies and resulting in spacial
limitations on the carrier dynamics (electrons and holes), as well as excitations of electron-hole pairs (excitons). The Coulomb energy scales with the QD size $d$ as $1/d$ (and is of the order of meV for QDs), while kinetic energy scales as $1/d^2$, which leads to the shell properties of dots, distinct in comparison to atoms (more complicated Hund-type rules for QDs), since both energies remain within mutual proportions of $d$, which favours Coulomb energy for dots in contrast to atoms [1] at the scale of meV orders. The nanometre-scale limitations on quantum dynamics result in kinetic energy quantization,

$$\Delta E \geq \frac{(\Delta p)^2}{2m^*} \approx \frac{\hbar^2}{2m^*d^2} \approx \begin{cases} 10 \text{ meV}, & d \sim 10 \text{ nm}, \\ 1 \text{ eV}, & d \sim 0.1 \text{ nm}. \end{cases}$$

In the case of QDs, quantization energy locates thus within a range accessible for control by means of external fields (electric and magnetic), in contrast to atoms (for the latter, quantum state control by means of external fields requires such values that are beyond the reach of present technologies). This advantage of QDs—which are relatively easy to create due to a variety of existing technologies in addition to their parameters’ flexibility and the possibility of immersing them in various media or even creating or modifying them by means of external fields—makes them very promising objects of new nanotechnologies and spintronic practical projects.

Various semiconductor materials may be used to create QDs. Note that insulator or metallic nanoparticles are also manufactured (however, collective electron liquids in metallic nanoparticles manifest distinct physical properties in comparison with semiconductor QDs, which explains why metallic nanostructures are not called QDs). For opto-electronic use, semiconductor dots seem best-suited due to their localization within other nanostructures (e.g., in quantum wells), with well-established technologies for control over such systems. Semiconductor QDs may be manufactured by means of etching technologies after a high-resolution photolithographic process (with the use of an ion or electron beam) has been applied (ordinary optic lithography with a resolution of up to 200-300 nm is not sufficient). Other technologies used here include among others the Stransky-Krastanov dot self-assembling method consisting in the application of epitaxy layers by MBE or MOCVD [MBE, Molecular Beam Epitaxy; MOCVD, Metal Organic Chemical Vapour Deposition]. Various lattice constants in successive epitaxy layers result in the spontaneous creation of nanocrystals on the ultra-thin so-called ‘wetting layer’. Electrical focusing in a quantum well [1, 4, 5] consists of yet another promising technique which, despite being at an early stage (due to a lack of sufficiently precise electrodes), offers the highest dot parameter flexibility and allows for dots to switch on/off within the working time periods of devices based on them [1, 4–6].

The possibility of control over the quantum states of carriers in QDs and their coherent (deterministic, controllable) time evolution are vital for nanotechnological and spintronic applications (especially where this concerns so-called ‘single-electron’ or ‘single-photon’ devices) as well as for the quantum processing of information. The absence of decoherence or its significant reduction up to the lowest possible level, at least within the time periods of control realization, is essential for all these applications. However, decoherence is unavoidable due to irreducible dot-environment interactions (there is no means of a dot’s total isolation). In the case of nanostructures, QDs offer a new class of physical phenomena
within the decoherence and relaxation range, entirely distinct from analogous processes in bulk materials or in atomic physics. This is due to the characteristic nanoscale-confinement energy, reaching values close to the typical energy parameters of crystal collective excitations (the energy characteristics of band acoustics and optical phonons). This convergence of energy scales results in resonance effects, which is different from what is observed in atomic physics where the scale of the atom-confinement energy is three orders of magnitude higher than the energy of crystal collective excitation, resulting in the weak influence of phonons on atomic states (included as a very small perturbation only). Specific decoherence effects in QDs result from a strong (resonance) coupling effect between the carriers trapped in them and the sea of various types of phonons (as well as with other collective excitations, or with local degrees of freedom, e.g., related to admixtures). This is why the frequently-used notion of an ‘artificial atom’ in reference to QDs is, to some extent, misleading.

The same reasons are responsible for the fact that QD modeling which does not account for environment-induced collective degrees of freedom may give rise to false conclusions since significant hybridization-induced (decoherence) changes of energy levels can reach up to 10%. This reduces the modeling fidelity if the environmental effects are neglected. Therefore, the current physics of nanostructures should embrace the recognition of the complex decoherence and relaxation effects observed in QDs for trapped carriers’ spin and charge, which are essentially different from what is observed in bulk materials and atoms.

2. Limitations on the quantum processing of information

Unavoidable decoherence—uncontrolled quantum information leakage into the surrounding environment due to the system’s interaction with the environment—perturbs the ideal quantum procedures which ensure the running of quantum schemes [7–12]. If, however, decoherence is kept below a certain threshold, quantum error corrections can be made by applying so-called ‘quantum error correction schemes’ [13], which enables the realization of any quantum procedures of a quantum computer or any other deterministic quantum project.

In classical information processing, quantum error correction consists in multiplying classical information and verifying by comparison the multiplied (redundant) classical registers with arbitrary frequency, errors which appear from time to time are identified and corrected immediately. In the quantum case, the multiplication of quantum information is impossible (No-Cloning theorem) and quantum error correction is based on a different scheme:

- Seeking more decoherence-resistant areas of the Hilbert space (multi-qubit states which, in a pair of qubits, record symmetrically both ‘true’ and ‘false’ are decoherence-resistant, e.g., singlet-type qubit states; information (or quantum states) symmetrization requires, however, the multiplication of quantum registers, which makes decoherence rise exponentially).
- Attempting the replacement of an information carrier for a more decoherence-resistant one (e.g., temporarily, a state can be teleported onto a more resistant carrier).

In order to satisfy quantum error correction requirements, DiVincenzo formulated a set of conditions [7, 14–16] which allow for the possibility of the implementation of quantum
error correction (the typical decoherence time must be at least of six orders longer than
the typical times of quantum procedures). None of the currently suggested solutions
for quantum computers have satisfied these time restrictions. This situation may follow
from the fact that the same interactions which allow for qubit control (logical operations)
are also responsible for decoherence. The stronger (energetically) the interaction is, the
faster the logical operations can be carried out. However, the same interaction couples
the system with the surrounding environment and produces strong decoherence effects.
In nanotechnological and optical projects involving quantum computers (multi-qubit), the
difference in the time-rate of quantum operations in relation to decoherence still do not
exceed three orders of magnitude.

However, it is expected that further intensive research in this area should result in:

- Finding another method of quantum error correction (despite great efforts, there is still
  no relevant solution).
- Finding a combined solution with qubit conversion (between a fast, controlled carrier
  and a decoherence-resistant one—unfortunately, qubit conversion is also inconveniently
  long-lasting).
- Finding global, topological and thus decoherence-resistant carriers of quantum
  information in them.
  - Braid groups (and non-Abelian anyons)—herein, the durations of logical operations
    are expected to be of 30 orders of magnitude greater than those of decoherence
    processes [17] (however, this is still unclear and it is doubtful if it is experimentally
    viable).
- It is hoped that superconductive states may satisfy the DiVincenzo conditions as they
  have non-local properties to a significant extent.

| DiVincenzo criteria, which render a scalable quantum computer project feasible |
|---|---|
| 1 | a properly defined qubit—two quantum states separated from the remaining states of the system (by relatively wide energy gaps or by forbidden transitions), so that quantum information entered therein does not leak out |
| 2 | the possibility of information recording on the defined qubit—i.e., the possibility of generating any superposition of two qubit states by means of external macroscopically controlled fields (e.g., by Rabi’s oscillations within a realistic range of fields) |
| 3 | designing and implementing a universal two-qubit operation, which may enable the performance of any logical quantum operation (a CNOT gate, or any other one, may work as such a universal gate provided there exists an effective means of switching qubit interactions on and off, i.e., the entanglement of two qubits can be controlled |
| 4 | ensuring that the time-rate of the performance of logical operations must be at least six order faster than the decoherence time-rate |
| 5 | ensuring that output information can be read |
| 6 | ensuring that the whole system can be reset |

| DiVincenzo conditions that enable quantum cryptography |
|---|---|
| 1 | defining a free information carrier—a mobile qubit (e.g., a photon) |
| 2 | enabling the maintaining of the quantum properties of mobile qubits at a constant level over long distances |
| 3 | enabling the identification of a qubit state (measurement) |
In the case of quantum cryptography, equipment requirements [18] are more easily met in respect to decoherence and this is why this quantum technology (public key distribution via a quantum channel) has been used in practice in optical systems [fibre-optical ones over distances of 100 km (up to 1000 km), and outdoors over distances of 2 km].

3. Quantum dots—the prospective technology for quantum gates

The idea of employing quantum evolution for information processing corresponds with the feasibility of the deterministic control over a quantum system in order to execute a previously designed quantum algorithm. However, such a deterministic evolution (also called ‘unitary’ or ‘coherent’) requires a totally isolated quantum system. Unfortunately, no quantum system can be totally isolated from the environment. Any quantum system is susceptible to the environment’s influence. In consequence, unitary or coherent evolution is perturbed, and quantum information undergoes uncontrollable and irreducible leakage into the environment. Therefore, the feasibility of the construction of a scalable quantum computer is seriously hindered due to decoherence phenomena. The better recognition of decoherence processes in quantum systems may, however, enable the development of new technologies transcending these limitations and facilitating the attainability of quantum gates.

Quantum state decoherence progresses along two channels: relaxation, i.e., quantum state annihilation; and dephasing, i.e., phase relations change within a quantum state description. Relaxation (or ‘amplitude decoherence’) is related to the decrease in time of the diagonal elements of the quantum state density matrix, whereas dephasing (or ‘phase decoherence’) corresponds to the reduction of the off-diagonal elements of the density matrix. Both types of decoherence are caused by interaction with the environment and they become more significant the stronger the interaction is.

Solid-state technology (which is promising for new practical realizations of the quantum processing of information using nanometre-scale semiconductor QDs) is burdened mostly with phase decoherence processes. Both the charge (i.e., orbital) and the spin degrees of freedom of quantum states in QDs undergo dephasing due to their environment (however, it should be emphasized that the spin degrees of freedom seem to be more decoherence-resistant than orbital degrees of freedom, since they are less susceptible to direct crystal phonon-induced interaction; however, spin requires much longer periods of time-control than orbital degrees of freedom due to weaker interaction with spin).

Below we present a decoherence analysis, in particular the phase decoherence of the charge (orbital) degrees of freedom and degrees of freedom of spin of excitations localized in QDs, dealing with the issues associated with limitations on the feasibility of QIP.

In the case of semiconductor QDs, decoherence is unavoidable due to strong dot-environment interaction (there are no means for the perfect isolation of a dot). In the case of nanostructures—QDs included—there appears to be a new class of physical phenomena related to decoherence and relaxation, distinct from analogous processes in bulk materials and atomic physics. This is due to characteristic meV-scale energy resulting due to nanoscale confinement, reaching values close to the typical energy parameters of band phonons in the surrounding medium. This coincidence of energy scales results in resonance effects, which is different from what is observed in atomic physics. For atoms, the incommensurability of the atom-confinement energy and phonon energy is of three orders of magnitude, resulting...
in a weak phonon impact on atomic states. Specific decoherence effects in QDs result from strong and resonant coupling between the carriers trapped in dots and the sea of various types of collective excitations in the surrounding medium, which highly modifies the QD states. Hybridization-induced changes of energy levels reach up to 10%. Therefore, the decoherence and relaxation effects observed in QDs and for trapped carrier spin or charge (which are essentially different from what is observed in bulk materials and atoms), seem to be of central importance for any possible QD applications, including QIP.

4. Phase decoherence of orbital degrees of freedom in nanostructures

Orbital degrees of freedom pertain to charge-type excitations, such as electrons and holes and charge-balanced electron-hole pairs—excitons. As charge carriers, these excitons interact with the electric field of the electromagnetic wave and so they can be controlled by means of quantum optics methods. Charge-type excitations can be localized in nanometre-scale artificial structures manufactured within various semiconductor heterostructures, namely in QDs. Excitons attract special interest as they can be precisely controlled by an electromagnetic wave within the visible (or near infrared) light range corresponding to the typical energy gap separating electron states from hole states in semiconductors (a typical material is GaAs and QDs will be, e.g., self-assembled structures of GaAs/InAs type). By accommodating the energy of (incident light) photons with the energy of the exciton, an exciton state in the Rabi oscillation regime can be created in which the superposition state of the charge qubit spanned on the states $|1\rangle$ (no exciton in a QD) and $|2\rangle$ (one exciton in a QD) can be selected. The techniques of ultra-high-frequency laser impulses (measured in femtoseconds) and the resulting application of a high intensity beam allowing for high-frequency Rabi oscillations [19] has attracted a lot of interest in QIP research. This interest has been centred on the fact that the lifetime of the excitons in the dots is measured in nanoseconds (this may suggest a difference of six orders of magnitude between the control time and the amplitude decoherence time, which is required by DiVincenzo’s criteria [7, 13–16, 20]). Nevertheless, in QDs the interaction between the excitons (electrons and holes) and phonons of the surrounding crystal is unavoidable and must be accounted for in all considerations, thus diametrically changing this ostensibly convenient situation. Phonons are quanta of the crystal oscillations; acoustical phonons refer to the oscillations of the density type (all the atoms in the unit cell oscillate in the same direction) and optical phonons are related with polarization oscillations (the opposite-sign ions in the unit cell oscillate in the opposite directions; polarization oscillations in such ionic crystals can be excited by means of light, and thus they are called ‘optical’ phonons). Both types of phonons can interact with charge-type degrees of freedom in QDs. Phonons can be of transversal or longitudinal polarization, but these are the longitudinal modes (LO and LA for optical and acoustic phonons, respectively) that contribute most substantially to the interaction with the electrons/excitons [21].

In polar materials (e.g., GaAs, a weakly polar semiconductor), LO phonon interaction prevails. The interaction of charges with LO phonons is characterized by means of the dimensionless Fröhlich constant [21, 22]. The higher the constant value, the stronger that the interaction is between the charges and the LO phonons, and for the semiconductor GaAs three-dimensional (bulk) the constant averages out at around 0.06. For QD GaAs/InAs,
experiments (infrared absorption in a magnetic field and the broadening of the satellite luminescence peak connected to LO phonons, expressed quantitatively via the so-called ‘Huang-Rhys factor’ [23]) show a double value of the constant, which suggests a substantial increase of the interaction with LO phonons. This phenomenon has been explained [24] with regard to a certain ambiguity [21] in the definition of LO phonon-electron interactions in crystals. The interaction between an LO phonon and an electron leads to the polarization of the crystal lattice by the moving electron. This polarization (i.e., an appropriate packet of optical phonons) is dynamic and leads to a reverse interaction with the polarization-inducing electron. It can be thought of as being composed of two components: an inertial one, which lags behind the moving electron; and a non-inertial one, which accompanies the moving electron. The latter component should be contained in the total crystal field which defines the electron itself (the electron in the crystal is not a free particle and includes, by its definition, the periodic crystal field—thus it can be characterized by the effective mass and quasi-momentum instead of momentum). The necessity of extracting only the inertial part of the polarization from the total interaction of the electron and the LO phonons leads to the above-mentioned ambiguity in the definition of the electron-LO phonon interaction. When the electron is trapped in a QD, it moves with a quasi-classical velocity [25] which exceeds the velocity of a free band electron. Thus, it better escapes from the dynamic polarization, which results in an increase in the inertial part of polarization and the interaction between the electron and LO phonons in QDs. The more localized the electron in a smaller QD, the bigger the quasi-classical velocity of the electron and the bigger the increase in the interaction with LO phonons. The quantitative analysis of the problem agrees well with the experimental data. It should be emphasized, however, that the marked increase in the value of the Fröhlich constant in QDs parallels the increase in the decoherence of electron/exciton states in dots due to the increase in the interaction between the small system of the QD and the sea of LO phonons in the crystal.

The energy scale corresponding to the nanometre localization of electrons (excitons) in QDs ranges from a few to several tens of meV. The same energy scale also characterizes the phonons in crystals, in which the energy of LA phonons ranges from 10 to 20 meV at the edge of the Brillouin zone and the energy of LO phonons at the centre of the Brillouin zone (a gap in the LO phonon spectrum at point \( \Gamma \) [21, 22]) reaches 30 meV. Thus, in the case of QDs we deal with a strong coupling regime for an interaction of QD charge degrees of freedom with phonons (of all types). The same energy scale of both types of excitations—local in QDs and collective in the surrounding crystal—results in the strong mutual hybridization of these excitations or in the dressing of electrons (holes) or excitons with phonons and in the creation of composite particles (quasi-particles)—polarons [21, 22, 26–29].

The creation of polarons in QDs is a strongly decoherent process (much more than it is in bulk materials). The dynamics of this process can be investigated by employing the Green function technique [28]. By means of this technique, the correlation function of the exciton (electron) in the QD can be expressed, which defines the overlap (the scalar product) of the state of the carrier gradually dressed by phonons with the initial state of the bare exciton (or electron) in the dot. Thus, it is possible to quantitatively characterize the leakage of quantum information (fidelity loss) due to the entanglement (in a quantum sense) of the QD’s charge with the deformation and polarization degrees of freedom of the whole crystal, which are entirely beyond our control.
The inertia of the crystal lattice is so disadvantageous that it makes it impossible to maintain
the coherence of orbital degrees of freedom dynamics (the unitary quantum evolution of the
excitations) within the time periods required by the DiVincenzo conditions. The typical times
of dressing charge-type excitations with phonons are located within the time-range of single
picoseconds, which is the middle of the six-orders time window between the amplitude
decoherence time for excitons in QDs and the time-scale of the quickest techniques for
their excitation. On both sides of this window, there appear windows of three-orders of
magnitude, which precludes the implementation of the quantum error-correction scheme
due to the non-fulfilment of the DiVincenzo conditions.

These strongly unfavourable estimations indicate that it may be impossible to scale a
quantum computer in a QD technology with only by light control unless more effective
quantum error-correction schemes would be proposed [7, 13, 14].

It should be emphasized that LA phonons are of greater importance to the process of dressing
the excitons with phonons (polaron decoherence effects), despite the fact that their interaction
with excitons is energetically much weaker (at least by one or two orders of magnitude)
than in the case of LO phonons. Strong dephasing due to LA phonons corresponds with a
wide linear dispersion of acoustic phonons, which in turn leads to a more immediate and
significant induced change in the wave functions of charge-type excitations in QDs than in
other phonon modes.

LA phonons-induced decoherence (phase decoherence or dephasing, corresponding to the
reduction of the off-diagonal elements of the density matrix [8–12]) is—and as can be shown
by means of a microscopic analysis—a relatively fast process and its time-scale is of the order
of the ratio of the dot diameter and the sound velocity (it is of the order of picoseconds).
Acoustic phonons are especially inconvenient as they are present in any crystal (as well as in
any amorphous material), and this is why the above-presented mechanism of decoherence
is unavoidable by its nature [strong dephasing also exists at a temperature of 0 K due to
phonon emission; at higher temperatures, the dephasing effects are enhanced due to phonon
absorption effects, which become more important with the increase in temperature].

Strong decoherence restrictions on the quantum evolution of the charge degrees of freedom in
QDs encouraged the researchers to concentrate their attention on the spin degrees of freedom
in nanostructures (spin does not interact directly with phonons) instead of pursuing the idea
of constructing an quantum computer based on QDs that is only controlled by light [7, 14, 28].

4.1. Phonon-induced dephasing of excitons localized in quantum dots

An exciton created in a QD by means of an non-adiabatic process (in the sub-picosecond
order) [4, 5, 30] is a bare particle (an electron-hole pair) which is gradually dressed with
phonons until it becomes a polaron. The time within which the polaron is created depends
upon the lattice inertia. It is relatively long and its accurate evaluation is an important task.
The process of the hybridization of a QD-localized exciton with the collective excitations
of the crystal lattice surrounding the QD is, in fact, a time-dependent evolution of a
non-stationary state, which at the initial time (the moment of the excitation’s creation) is
identical with the state of the bare exciton. The bare exciton is not the stationary state
of the whole system, the QD exciton and the sea of phonons in the surrounding crystal
interacting with it (a polaron represents a stationary state of such a complex system). The
non-stationary initial state (the bare exciton) [the electric field of the e-m wave interacts with the charge and, in consequence, excites a bare electron from the valence band into the QD; the resulting hole is also captured by the QD—a bare QD exciton is thus created] undergoes further non-stationary evolution. In the non-stationary state, the energy is not determined; however, the mean energy is shared over time between the subsystems, the QD and the phonon sea. The mean energy of a bare QD exciton is higher in comparison with the polaron energy (whose energy is lower and therefore the polaron is created by means of interaction with phonons due to energy minimization). The excess energy of the lattice deformation (for acoustical phonons) together with the polarization energy (for optical phonons) is carried outside the QD by the LA and LO phonons, respectively (by their wave packets). A QD polaron is created—a hybridized state of an exciton dressed with an LA and LO phonon cloud [actually, the name of the polaron refers to electrons dressed with LO phonons [22]—a process dominating in strongly polar materials; here, the name refers generally to an electron or exciton dressed with all types of phonons]. The time-scale of QD polaron creation is of the same order as the time that a phonon-wave packet needs to leave the QD area. It should be emphasized that this process is not to be interpreted in terms of Fermi’s golden rule [in such an approach, quantum phase transitions resulting from a time-dependent perturbation refer to transitions between stationary states, which is not the case here] [25]. The process of polaron creation is a non-stationary state evolution, in which the elementary processes of phonon absorption or emission contribute in the virtual sense (without energy conservation).

Note that the polaron energy is shifted with respect to the bare QD exciton energy by a few meV [28], while the LO phonons energy has a much greater gap, $\bar{\hbar} \Omega \approx 36.4$meV (in GaAs).

The kinetics of polaron creation correspond with the coherent evolution of an entangled state of two interacting systems, namely a QD exciton and the sea of phonons (of various types), and this state in non-separable [28].

The exciton-phonon system is represented by the following Hamiltonian:

$$H = \sum \varepsilon_n a_n^+ a_n + \sum \hbar \omega_s(q) c_{q,s}^+ c_{q,s}$$

$$+ \frac{1}{\sqrt{N}} \sum_{q,m,n_1,n_2,s} F_s(q,n_1,n_2,q) a_{n_1}^+ a_{n_2} \left( c_{q,s} + c_{q,s}^+ \right),$$

where the LO interaction ($s = o$) and the LA interaction ($s = a$) are represented by the following functions:

$$F_o(n_1,n_2,q) = -\frac{e}{q} \sqrt{\frac{2\pi \hbar \Omega}{\varepsilon \varepsilon'}} \int \Phi_{n_1}(R_e,R_h)$$

$$\times \left( e^{i q \cdot R_e} - e^{i q \cdot R_h} \right) \Phi_{n_2}(R_e,R_h) d^3R_e d^3R_h,$$

and:

$$F_a(n_1,n_2,q) = -\frac{\hbar q}{2M \varepsilon_o} \int \Phi_{n_1}(R_e,R_h)$$

$$\times \left( c_{e} e^{i q \cdot R_e} - c_{h} e^{i q \cdot R_h} \right) \Phi_{n_2}(R_e,R_h) d^3R_e d^3R_h.$$
Here, $c_{q,s}^{(+)}$ denote annihilation (creation) boson operators for LO ($s = o$) and LA ($s = a$) phonons with quasi-momentum $q$; the phonon frequency $\omega_o(q) \equiv \Omega_q \simeq \Omega - \beta q^2$ ($\Omega$ represents an energy gap for LO phonons at $\Gamma$ point) and $\omega_a(q) = C_a q$, $C_a$ is the sound velocity (for LA phonons); $M$ represents the mass of ions in the unit cell; $\sigma_{e,h}$ is the deformation potential constant of an electron and hole, respectively; $v$ is the volume of the unit cell; $N$ is the number of cell in the crystal; $\tilde{\epsilon} = (1/\epsilon_\infty - 1/\epsilon_0)^{-1}$ is the effective dielectric constant; $R_e, R_h$ represent the coordinates of an electron and hole; $\Phi_n(R_e, R_h)$ denotes the QD exciton (electron) wave function and $a_n^{(+)}$ is the exciton or electron annihilation (creation) operator in $n$-th state (of boson type for excitons and of fermion type for electrons). The interaction between a charge and longitudinal phonon modes is considerably stronger than with transversal modes, which is why only the first ones are further considered [21, 22].

The interaction between an exciton and phonons from both branches (LO and LA) has the simplest linear form with respect to the phonon operators (the third element in the Hamiltonian). It can be represented by means of graphs, as in Fig. 1.

Vertices of this type (as in Fig. 1) result in the mass operators of Green functions, both for the exciton (electron) and the phonon (note that without the linear term with respect to the interaction due to the specific form of the vertices); for the exciton, this is illustrated by the graphs in Fig. 2.

The graphs in Fig. 2 correspond to the complete expressions for the mass operator (thick lines represent the full Green functions; the effective vertex of the exciton-phonon interaction is also marked (the shaded vertex)). It is an accurate form of the mass operator. Within the first approximation, the effective vertex can be replaced with a bare one (it is an approximation with controlled accuracy—the terms of a higher order than the quadratic
with respect to the interaction are omitted [subsequent bare vertices enter the effective vertex]. Given that the charge-phonon interaction is weak, this approximation leads to a small error. The bare vertices of this interaction (the corresponding functions appearing in the Hamiltonian) attain the form (which results from the mechanism of interaction between the charge and LO phonons—by means of polarization, and with LA phonons—by means of deformation) [21, 22] given by the (3, 4) formulae, with the integrals representing the overlap integral of the localized exciton states (initial and final) with the phonon plane wave. These integrals represent the bottleneck effect, typical for QDs [32, 33], resulting from the absence of the translational invariance of a QD system and leading to the non-conservation of the momentum (quasi-momentum). The overlap integral with the plane wave favours the momentum \( q \approx \frac{\hbar}{d} \), where \( d \) denotes a QD size. If the exciton is not localized—i.e., is represented by a plane wave—this integral would yield the law of momentum conservation, corresponding to the translational invariance of the system in that case. In the case of a QD-localized exciton, this integral does not become Dirac’s delta but rather defines those quasi-momenta \( q \) of phonons which were involved in the interaction. At the same time, the law of conservation of energy holds true for each vertex (i.e., for the interaction process), which results from the unperturbed uniformity of time in the case of QD-localized states. Due to the above functions, the fact of the selection of the fixed values of quasi-momentum for QD-localized exciton (electron) states is called ‘the bottle neck effect’. The presence of the above-mentioned integrals results in the elimination of all the phonon modes except for those found within the range of \( q \approx \frac{\hbar}{d} \) (\( d \) a QD size, typically of the order of 10 nm); thus, the range of the significant quasi-momentum of a phonon interacting with a QD charge is of the order of 1-10% of the Brillouin zone close to its centre (as illustrated in Fig. 3).

The model (variational) ground state exciton wave function in a parabolic QD assumes the following form (including Coulomb interaction e-h):

\[
\Phi_0(r_e, r_h) = \frac{1}{(\pi)^{3/2}L_e L_h L_z} \exp \left[ -\frac{r_{e,\perp}^2}{2L_e} - \frac{r_{h,\perp}^2}{2L_h} - \frac{z_e^2 + z_h^2}{L_z} \right],
\]

(5)

where \( r_{e,h,\perp} \) denotes the positions of the exciton components (e and h) in the xy plane of the QD. The numerically estimated parameters for a QD characterized by the values in Tab. 1 are \( L_e = 6.6 \text{ nm}, L_h = 5.1 \text{ nm} \) and \( L_z = l_z \) (which agrees sufficiently well with a more accurate numerical calculation and diagonalization [28]). The noticeable difference between the electron and the hole’s effective lateral dimensions results from the fact that the e-h Coulomb interaction energy is comparable to the inter-level energy of the heavier holes, while the energy of the lighter electrons is quantized with greater inter-level gaps.

The above form of the ground state QD exciton wave function yields the following phonon coupling functions:

\[
|F_0(0,0,k)|^2 \simeq \frac{\pi \hbar^2 \Omega^2}{16 \alpha \epsilon_e} \left( L_e^2 - L_h^2 \right)^2 e^{-\alpha k^2} = \frac{\chi_0}{\chi_e} e^{-\alpha k^2},
\]

\[
|F_0(0,0,k)|^2 \simeq \frac{\pi \hbar^2 \Omega^2}{16 \alpha \epsilon_h} \left( \epsilon_e - \epsilon_h \right)^2 e^{-\alpha k^2} = \frac{\chi_0}{\chi_e} e^{-\alpha k^2},
\]

(6)
where $k_m = (6\pi^2/v)^{1/3}$ denotes the Debye wave vector ($\simeq 1.1 \cdot 10^{10} \text{m}^{-1}$), $a = l^2/2$, $l$ is the QD size averaged over all directions (this is the averaged ground-state dimension of an exciton), and the same for e and h ($l$ is significantly smaller than the lateral dimensions $L_e(L_h)$ but greater than the vertical one $L_z$). The exponential factor $e^{-ak^2}$ corresponds to the above-mentioned bottleneck QD effect. These functions are illustrated by the lines in Fig. 3. Both these functions (often called the ‘form-factors’ of the exciton-phonon interaction) assume non-zero values in the vicinity of point $\Gamma$ and reach the maximum for quasi-momentum $p \simeq \hbar / d$ ($d \sim l$). What matters here is the fact that this behaviour closely corresponds with the bottleneck effect, which replaces the momentum-conservation condition for the system without translational invariance [32, 33].

The bottleneck effect (which seems to limit the importance of phonons in nanostructures) led to an under-evaluation of the phonons’ role in QDs and of their input in the total interaction in nanostructures [32]. This mistaken view often resulted in the underestimation of phonon-induced phenomena in many physical processes in QDs. Despite the fact that the coupling constants (and form-factors including the bottleneck effect) are rather low-valued, the resonant coincidence (proximity) of the energy levels of the quantum states for carriers localized in the QDs and bulk phonon energy characteristics results in a strong increase in the non-perturbative effects of the mutual hybridization of both subsystems (excitons/electrons and phonons), leading to significant polaron-type effects, both for LA and LO phonons. It is a vital process in the case of QDs, for which such hybridization effects result in the change of quantum states by as much as 10% and lead to significant effects regarding time-dependent processes of amplitude decoherence (relaxation, i.e., a decrease in the diagonal elements of the density matrix [27, 34, 35]), and phase decoherence (dephasing, i.e., a decrease in the off-diagonal elements of the density matrix) [8–12]. From the list of parameters for the GaAs/InAs system (Tab. 1), one can notice that the interaction between the exciton and the LO phonons is significantly bigger (by one order of magnitude) than with LA phonons (cf. the values of the parameters $g$ in formulae (6)).

In order to give the description of fidelity loss [8, 13], we will discuss the exciton correlation function $\langle a^\dagger_n(t)a^\dagger_n(0) \rangle$. For $n_1 = n_2$, this corresponds to the overlap of the exciton state at time $t = 0$ with this state at the initial moment $t = 0$ (for $n_1 = n_2 = 0$—for the ground state of the exciton changing into a polaron after being gradually dressed with LA and LO phonon clouds). The modulus of this correlation function thus gives a measure of
fidelity of the time-dependent (non-stationary) exciton state. The averaging (...) denotes the temperature-dependent averaging over the phonon states, assuming the exciton vacuum state [36]—that is, without a change of state of a bare exciton—which corresponds to the fact that the great canonical averaging sector without exciton, vacuum, it energetically distant from the exciton state [36]—that is, without a change of state of a bare exciton—which corresponds to the fact that the great canonical averaging sector without exciton, vacuum, it energetically distant from the next exciton sectors. The energy separation is here of order of 1 eV.

The Fourier transform of the correlation function is called the ‘spectral density’ [37, 38]:

$$I_{n_1,n_2}(\omega) = \int_{-\infty}^{\infty} \langle a_{n_1}(t)a_{n_2}^+(0) \rangle e^{i\omega t} dt, \quad (7)$$

The spectral density function can be expressed by the imaginary part of the retarded Green function:

$$\text{Im} G_r(n_1,n_2,\omega) = -i(n_1,n_2,\omega)/(2\hbar), \quad (8)$$

where:

$$G_r(n_1,n_2,t) = -\frac{i}{\hbar} \langle [a_{n_1}(t),a_{n_2}^+(0)] \rangle = \frac{1}{\hbar} \int_{-\infty}^{\infty} G_r(n_1,n_2,\omega)e^{-i\omega t}d\omega, \quad (9)$$

is the commutation-retarded Green function which describes the linear dielectric response to the electromagnetic wave coupled to an exciton [in the case of the instant creation of an exciton, the time-dependent electromagnetic signal is assumed to be Dirac’s delta \(\delta(t)\)].

In our case, the Green function \(G_r(n_1,n_2,t)\) and the correlation function can be obtained by including the interaction between the exciton and the LA and LO phonon sea via the standard temperature-dependent Matsubara-Green function techniques [37, 38]. The advantage of the Matsubara-Green function approach over the others consists of the derivation of the Dyson equation with an appropriate mass operator, which accounts for the interaction of the exciton with the sea of phonons. Both these methods lead to the Dyson equation with an appropriate mass operator, which accounts for the interaction of the exciton with the see of phonons.

In the case of weak exciton-phonon coupling (which is the nature of the case currently being discussed), the mass operator attains the form of [39] as illustrated by the graphs in Fig. 2. For the QD exciton (similarly as for the bulk semiconductor [40]), with an accuracy up to \(g_s\) [note that \(F_s(n_1,n_2,k) \sim g_s\), where \(g_s\) is an exciton-phonon constant], both real and imaginary parts of the mass operator \(M\) are obtained in the following form (for the effective vertex, the components of a higher-order are omitted, i.e., multi-phonon processes are not included):
Quantum Dots - Theory and Applications

where:

\[ N_k \] population of the phonon mode

Taking \( \omega_s \) as \( \text{GaAs} \) polar material). Note that equations (10)–(11) contain the full Green function (in accordance with the graph in Fig. 2). For exciton-LO polaron interactions prevails in a polar material (GaAs is a weakly polar material). For exciton-LO phonon interactions, the Fröhlich constant is of importance [21, 22]:

\[ \gamma_n(\omega) = \frac{1}{N} \sum_{k,s,n_1} |F_k(n,n_1,k)|^2 \left[ \frac{1}{(1+N_k)\gamma_n(\omega+n_1,k)} + \frac{N_k}{\gamma_n(\omega+n_1,k)} \right] \]

\[ \gamma_n(\omega) = \frac{1}{N} \sum_{k,s,n_1} |F_k(n,n_1,k)|^2 \left[ \frac{1}{(1+N_k)\gamma_n(\omega+n_1,k)} + \frac{N_k}{\gamma_n(\omega+n_1,k)} \right] \]

(11)

where: \( N_{k,s} \) is the Bose-Einstein distribution function defining the temperature-dependent population of the phonon mode \( k,s \), \( M_{n,n}(\omega) = \Delta_n(\omega) - i\gamma_n(\omega) \), \( G_t(n,n,\omega) = [\hbar \omega - E_n - M_{n,n}(\omega) + i\epsilon]^{-1} \) (for \( T = 0 \), \( N_{k,s} = 0 \)). The above system of equations enables the time-dependent analysis of the dressing of the exciton with a cloud of phonons.

For GaAs in the surrounding medium, the material parameters are taken from Ref. [41], while a InAs/GaAs QD is modelled in terms of parabolic approximation [1] with a curvature \( \hbar \omega_0^e = 20 \text{ meV}, \hbar \omega_0^h = 3.5 \text{ meV}, l_e = \sqrt{\frac{\hbar}{m_e\omega_0^e}} = l_h = \sqrt{\frac{\hbar}{m_e\omega_0^h}} = 7.5 \text{ nm} \), which results in the identical size of the ground state of the electron and the hole (when the Coulomb interaction is not accounted for); the QD vertical dimension (the QD is significantly flattened) is \( l_e^{(h)} \simeq 2 \text{ nm} \) (with a suitably adjusted parabolic curvature \( \omega_e^{(h)} \)) [numerical estimations including particularities of the QD shape show that they only weakly affect the QD structure and the polaron characteristics [28].

For exciton-LO phonon interactions, the Fröhlich constant is of importance [21, 22]:

\[ \alpha_e = \frac{e^2}{\bar{e}} \sqrt{\frac{m_e^*}{2\hbar^3 \Omega}}. \]

(12)

Its value grows in nanostructures [24, 26], which significantly influences QD polaron-related processes.

In equation (10), the first component provides the main contribution—polaron red-shift resulting from exciton-LO polaron interactions prevails in a polar material (GaAs is a weakly polar material). Note that equations (10)–(11) contain the full Green function (in accordance with the graph in Fig. 2). Taking \( \gamma_n(\omega) = 0 \) on the right-hand side of equation (10), the first-order approximation for the energy shift is:

\[ \Delta_n(\omega) = \frac{1}{N} \sum_{k,n_1} |F_k(n,n_1,k)|^2 \left[ \frac{1}{\hbar \omega - E_n - \Delta_n(\omega - \Omega) - \hbar \Omega} + \frac{N_k}{\hbar \omega - E_n - \Delta_n(\omega - \Omega) + \hbar \Omega} \right] \]

\[ \Delta_n(\omega) = \frac{1}{N} \sum_{k,n_1} |F_k(n,n_1,k)|^2 \left[ \frac{1}{\hbar \omega - E_n - \Delta_n(\omega - \Omega) - \hbar \Omega} + \frac{N_k}{\hbar \omega - E_n - \Delta_n(\omega - \Omega) + \hbar \Omega} \right] \]

\[ \Delta_n(\omega) = \frac{1}{N} \sum_{k,n_1} |F_k(n,n_1,k)|^2 \left[ \frac{1}{\hbar \omega - E_n - \Delta_n(\omega - C_3 k) - \hbar C_3 k} + \frac{N_k}{\hbar \omega - E_n - \Delta_n(\omega - C_3 k) + \hbar C_3 k} \right]. \]

(13)
As has already been pointed out, the first term of the equation provides the dominating contribution while the second—of a significantly smaller order of magnitude [due to the smaller value of the LA phonon coupling constant]—can safely be neglected here. However, this term—to a greater extent than the first one—contributes substantially to the derivative \( d\Delta/d\omega\mid_{\omega=E+\Delta} \). The derivative of the first term \( \sim F^2/(h\Omega)^2 \) is small due to the gap in dispersion of the LO phonons, but this derivative is important for estimating the residuum of the Green function at its pole—in equation (16). Moreover, in the first term of this equation, the weak dispersion of the LO phonons is neglected due to its insignificant contribution to the energy shift \( \Delta \) (which has been verified via numerical methods) [28]. The numerical solution of equation (13) for \( n=0 \) yields the polaron energy shift \( \Delta_0 \approx -5 \text{ meV} \) (for the structure parameters listed in Tab. 1).

For the description of the kinetics of polaron creation, i.e., of the process of the dephasing of an non-adiabatically excited exciton (this is experimentally observed at the picosecond time-scale [30]), the imaginary part of the mass operator and the out-of-pole form of the imaginary part of the Green function is of especially importance—it provides complete information about the spectral intensity (not limited to the poles defining the energy and lifetime of quasi-particles, here polarons). The Fourier transform of the spectral intensity yields the unknown correlation function. The imaginary part of the mass operator is given by Eq. (11). Taking \( \gamma = 0 \) on its right-hand side:

\[
\gamma_\alpha(\omega) = \frac{\omega}{\pi} \sum_{n,k} \left[ |F_a(n,n_1,k)|^2 \right. \\
\left. \left[ (1 + N_{k,\alpha}) \delta(\hbar\omega - E_{n_1} - \Delta_{n_1} - \hbar\Omega_{k}) \right. \\
\left. + N_{k,\alpha} \delta(\hbar\omega - E_{n_1} - \Delta_{n_1} + \hbar\Omega_{k}) \right] + |F_a(n_1,n,k)|^2 \left[ (1 + N_{k,\alpha}) \delta(\hbar\omega - E_{n_1} - \Delta_{n_1} + \hbar\Omega_{k}) \right. \\
\left. \left. + N_{k,\alpha} \delta(\hbar\omega - E_{n_1} - \Delta_{n_1} - \hbar\Omega_{k}) \right] \right]
\]  

(14)

The first term in equation (14) defines the polarization energy transfer to the LO phonon sea, while the second one defines the deformation energy transfer (smaller) to the LA phonon sea during the process of the gradual exciton-dressing with both types of phonon modes. The term \( \gamma \) can be estimated for the ground state of the exciton \( (n=0) \) [higher excited levels are neglected]; integrating over \( k \) yields:

\[
\gamma_0(\omega) \simeq A x^3 e^{-\frac{x^2}{\Delta_{n_1}^2}} \left[ \Theta(x)(1 + N(x)) - \Theta(-x)N(-x) \right] \\
+ B \left[ \Theta(h\Omega - x)(h\Omega - x)^{3/2} e^{-\frac{a(h\Omega - x)}{\delta}} \Theta(-0.6h\Omega + x)(1 + N(x)) \right. \\
\left. + \Theta(h\Omega + x)(h\Omega + x)^{3/2} e^{-\frac{a(h\Omega + x)}{\delta}} \Theta(-0.6h\Omega - x)N(-x) \right],
\]  

(15)

where \( x = \hbar\omega - E_0, E_0 = E_0 - \Delta_0 \) is the energy of an excited polaron, \( N(x) = (e^{\frac{x^2}{\Delta_{n_1}^2}} - 1)^{-1} \);
Figure 4. The spectral intensity versus energy ($x = \hbar \omega - \tilde{E}_0$) for a QD of a dimension averaged over all directions $l = 6$ nm (upper); the temperature evolution of the side-band due to the LA phonons (middle) and the satellite peak due to the LO phonons (bottom). Only the side-band (due to the LA phonons) increases in value as the temperature rises (within the mentioned range of temperature). LO absorption processes are negligible within this temperature range (the left-hand satellite peak corresponds to LO phonon absorption and is smaller by several orders of magnitude than the right-hand one corresponding to emission of LO phonons; the emission contribution becomes more significant for $T > 80$ K); the satellite peak that is LO phonon-induced increases significantly with the increase in the value of the QD’s Fröhlich constant [expressed by the Huang-Rhys factor][23]—Fröhlich constant in bulk (a), in a QD (b).

\[
A = \frac{(\sigma_e - \sigma_h)^2}{4\pi\rho\hbar^2 c^2}, \quad B = \frac{e^2\hbar \Omega (L_e^2 - L_h^2)^2}{36e^2(h\beta)^{5/2}}
\]

(LO phonon dispersion as defined in [31] $\Omega_k = \Omega - \beta k^2$ and at the Brillouin zone edge, i.e., for $k = k_m$ attains the value $\Omega_{k_m} = 0.6\Omega$). The first term of equation (15) corresponds to the LA phonon channel of energy dissipation, while the second one corresponds to the LO phonon dissipation channel. The numerical parameters of this equation for the structure under investigation (Tab. 1) are listed in Tab. 2.

As $\gamma_0$ equals 0 at $x = 0$ [which results from equation (15)], this point is understood as the the well-defined Green’s function pole (it corresponds to a stable quasi-particle—the polaron, this is the exciton dressed in phonon clouds [a generalization of an ordinary electron polaron]...
dressed only in an LO phonon cloud [22, 34]). The time-dependent evolution of the phonon dressing is given by the correlation function, corresponding to the Green function in the form of:

\[ G_r(0,0,\omega) = \frac{1}{\hbar \omega - E_0 - \Delta(\omega) + i\gamma(\omega) + i\epsilon} \]

where:

\[ a = 1 - \left. \frac{d\Delta(\omega)}{d\omega} \right|_{\omega=E_0} \]

\[ = 1 + \frac{1}{N} \sum_{k,s} \left| \frac{E_{s}(0,0,k)}{\hbar \omega_{s}(k)} \right|^2 \left[ 1 + 2N_s(k) \right], \]

\[ \gamma'(x) = \frac{\gamma(x)}{a}(x = \hbar \omega - \tilde{E}_0, \tilde{E}_0 = \bar{E}_0/a), \epsilon = 0+. \]

The imaginary part of this retarded Green’s function (16) attains the form:

\[ \text{Im} G_r(0,0,\omega) = -a^{-1} \pi \delta(x) - \frac{a^{-1}\gamma'(x)/x^2}{1 + (\gamma'(x)/x)^2}. \]

The inverse Fourier transform of the spectral intensity (the imaginary part of the retarded Green’s function) gives the correlation function in the time domain:

\[ I(t) = -2\hbar \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \text{Im} G_r(0,0,\omega)e^{-i\omega t}, \]

(the indices \( n_1 = n_2 = 0 \) of function \( I \) are suppressed). The first term in (18) yields:

\[ I^{(1)}(t) = a^{-1}e^{-\frac{\tilde{E}_0}{\hbar}t}. \]

Notice that in the second term of equation (18), for temperatures \( T < 100 \text{ K} \), the second term in the denominator can be safely neglected within the perturbative treatment. This allows us to exchange the order of integration with respect to \( \omega \) and \( k \)—the inverse Fourier transform can be calculated first and, simultaneously, Dirac’s delta can be employed in equation (14). Such integration over frequencies (energy) yields a convenient representation of the correlation function:

\[ I^{(2)}(t) = \frac{1}{N} \sum_{k,s} \left| \frac{E_s(0,0,k)}{\hbar \omega_s(k)} \right|^2 \left\{ [1 + N_s(k)]e^{-i(E_0/\hbar + \omega_s(k))t} + N_s(k)e^{-i(E_0/\hbar - \omega_s(k))t} \right\}. \]
Figure 5. The evolution of the side band due to acoustic phonons (LA) versus the QD’s dimension and temperature for $T \geq 0$ K (the same temperatures for each QD dimension $l = 3, 6, 12$ nm).

Notice that, when comparing with Eq. (17), for $t = 0$ this leads to $I^{(1)}(t = 0) = a^{-1}$ and $I^{(2)}(t = 0) = 1 - a^{-1}$, which in effect results in the appropriate normalization of the correlation function.

The spectral intensity and its reverse Fourier transform (its modulus) are plotted in Figs. 4—9 for various temperatures and QD dimensions. The fact that the numerically calculated correlation function agrees well with the experimentally obtained data [30]—cf. Fig. 7 (upper), for a small QD and sub-picosecond excitations, may confirm the validity of the theory developed here. The LA channel (although negligible in terms of energy compared to the LO channel in GaAs) is the fastest and most effective in the dephasing process. The LO channel is slower and accompanied by fast oscillations (beats of $\approx 100$ fs corresponding to the existence of the LO gap) [the LO channel of dephasing can be significantly intensified due to the anharmonic decay of the LO phonons, e.g., for GaAs/InAs up to 10 ps] [42]. Dephasing produced by LO phonons is significantly weaker than that for LA phonons (contrary to the energy shift). The inclusion of the LO channel results in the weak modification of strong LA dephasing—cf. Figs 8 and 9. Figure 9 (right) presents the type of the scaling of the dephasing time versus a QD dimension—linear for the LA channel and quadratic for the LO channel. This behaviour agrees well with the simple relationship: dephasing time $\approx \frac{l}{v_g}$, $v_g$—phonon group velocity, $l$—QD dimension. For LA phonons $v_g = C_a$, which yields a linear function
Figure 6. Evolution of the satellite peak (the right-hand one—corresponding to LO phonon emission)—versus the QD’s dimensions; in practical terms, it is not dependent on temperature for $T < 80$ K.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>electron effective mass in GaAs</td>
<td>$m_e^* = 0.067 m_e$</td>
</tr>
<tr>
<td>hole effective mass (heavy) in GaAs</td>
<td>$m_h^* = 0.38 m_e$</td>
</tr>
<tr>
<td>dielectric constant in GaAs (static)</td>
<td>$\varepsilon_0 = 12.9$</td>
</tr>
<tr>
<td>dielectric constant in GaAs (dynamic)</td>
<td>$\varepsilon_\infty = 10.9$</td>
</tr>
<tr>
<td>electron deformation potential in GaAs</td>
<td>$\varphi_e = 6.7$ eV</td>
</tr>
<tr>
<td>hole deformation potential in GaAs</td>
<td>$\varphi_h = -2.7$ eV</td>
</tr>
<tr>
<td>LO phonon energy at $\Gamma$ point in GaAs</td>
<td>$\hbar \Omega = 36.4$ meV</td>
</tr>
<tr>
<td>density of GaAs</td>
<td>$\rho = 5.36$ g/cm$^3$</td>
</tr>
<tr>
<td>sound velocity (LA) in GaAs</td>
<td>$C_s = 4.8 \times 10^5$ cm/s</td>
</tr>
<tr>
<td>electron confinement energy in a GaAs/InAs dot</td>
<td>$\hbar \omega_{el} = 20$ meV</td>
</tr>
<tr>
<td>hole confinement energy in a GaAs/InAs dot</td>
<td>$\hbar \omega_{hh} = 3.5$ meV</td>
</tr>
<tr>
<td>lateral dimension of a QD (electron)</td>
<td>$l_e = 7.5$ nm</td>
</tr>
<tr>
<td>lateral dimension of a QD (hole)</td>
<td>$l_h = 7.5$ nm</td>
</tr>
<tr>
<td>dot height (electron and hole)</td>
<td>$l_z = 2$ nm</td>
</tr>
<tr>
<td>Debye wave vector in GaAs</td>
<td>$k_D = (\frac{2 \pi}{\lambda_D})^{1/3} = 1.1 \times 10^{10}$ m$^{-1}$</td>
</tr>
<tr>
<td>Fröhlich constant in a GaAs-bulk (electron)</td>
<td>$\alpha_e = 0.07$</td>
</tr>
<tr>
<td>Fröhlich constant in a GaAs/InAs dot (electron)</td>
<td>$\alpha_e' = 0.15$</td>
</tr>
</tbody>
</table>

Table 1. Quantum dot and material parameters for GaAs/InAs

of $l$, and for LA phonons $\nu_L = 2\beta k \sim 2\beta/l$, which results in a quadratic dependence on the QD dimension $\sim l^2/(2\beta)$. Conclusive proof of this result follows below.

5. The universal rule for the estimation of the dephasing time of localized excitons in nanostructures

In order to estimate the dephasing time of a QD’s (or other nanostructures) localized excitation (e.g., an exciton) due to hybridization with collective excitations in the surrounding
Table 2. Parameters of exciton-phonon (LA and LO) interaction for a GaAs/InAs dot

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>exciton-LA phonon coupling constant</td>
<td>$A = \frac{\hbar}{2\pi}\left</td>
</tr>
<tr>
<td>exciton-LO phonon coupling constant</td>
<td>$B = \frac{\hbar}{2\pi}\left</td>
</tr>
<tr>
<td>lateral exciton dimension (electron)</td>
<td>$L_e \approx 6.6\text{[nm]}$</td>
</tr>
<tr>
<td>mass operator exponent (LA)</td>
<td>$\alpha_{LA} \approx (\text{1 mm})^21.8$ meV$^{-2}$</td>
</tr>
<tr>
<td>mass operator exponent (LO)</td>
<td>$\alpha_{LO} \approx (\text{1 mm})^2149$ meV$^{-1}$</td>
</tr>
<tr>
<td>lateral exciton dimension (hole)</td>
<td>$L_h \approx 5.1\text{[nm]}$</td>
</tr>
</tbody>
</table>

Figure 7. Modulus of the correlation function $|\langle a(t) a^+ (0) \rangle|$ (the fidelity measure of the ground excitonic state) versus time for rising temperatures. The three plots correspond to small, medium and large QDs, and contain curves related to the same set of temperatures as given in the upper plot. For the small QD, the experimentally observed fidelity loss for a non-adiabatically excited exciton (for 0.2 ps pulse) [30] is reproduced well in the upper figure.
which is expressed (with multi-phonon effects neglected) by the following formula [37, 38]:

$$\gamma \sim \int dk |F(k)|^2 \delta(\omega - E - \omega(k)),$$

(21)

where the interaction vertices assume their general form (the interaction between the QD localized excitation and the non-localized crystal excitations expressed by means of plane waves):

$$F(k) \sim \langle \Psi_0 | e^{ikr} | \Psi_0 \rangle,$$

(22)

where $|\Psi_0(r)\rangle$ is a wave function of a QD localized exciton corresponding to its ground state with energy $E$ (for simplicity of description, a single particle-localized excitation is considered here, e.g., an electron [one-dimension picture, $\hbar = 1$]). Thus, the correlation function:

$$I(t) \sim e^{-iEt} \int dr |\Psi_0(r)|^2 \int dk F^*(k) e^{i(kr - \omega(k)t)},$$

(23)

appears to attain the form of a time-dependent overlap of a probability density of a QD localized particle:

$$|\Psi_0(r)|^2,$$

(24)

with a collective excitation wave packet (phonons) escaping from the QD-space region:

$$\int dk F^*(k) e^{i(kr - \omega(k)t)}$$

(25)

(where $k$ is the centre, $k \sim 1/l$, $l$ is the QD diameter due to the above-mentioned QD bottleneck effect entered here via $F(k)$). The wave packet carries off the excess (deformation or polarization) energy of the particle being dressed to the QD’s surrounding region in the crystal with the group velocity $v_g = \frac{\partial \omega(k)}{\partial k}$ (for $k \sim 1/l$). Thus, the dephasing time corresponds to the time for the decrease in value of the modulus $I(t)$, which here is of the order of $\tau \simeq \frac{l}{v_g}$, where $l$ is the QD dimensions averaged over all directions [the QD exciton state dimension] (as illustrated in Fig. 10).

In this representation, the dephasing time is of the order of the proportion of a QD dimension $l$ to the phonon group velocity, i.e., the velocity of the phonon packet carrying off the excess energy from the QD to the surrounding medium (this is the evolution of a non-stationary QD state of a non-adiabatically excited bare exciton). For LA phonons, the group velocity remains constant and equals the sound velocity $v_s = C_s$, which results in a linear dependence of the dephasing time with respect to the QD’s dimension,

$$\tau \simeq \frac{l}{C_s},$$
Figure 8. The typical shape of the modulus of the correlation function of an exciton interacting simultaneously with LO and LA phonons. The oscillations, corresponding to the gap in the LO phonon spectrum [the frequency is \( \sim 1/\Omega \sim 100\text{fs} \)], are significantly stronger for a QD due to the increase in the value of the effective Fröhlich constant (upper).

Figure 9. Left: the modulus of the correlation function for, respectively, LA (upper) and LO (lower) phonons only. Right: the exciton dressing time vs the averaged QD dimensions \( l \) for the LA channel (upper), the linear dependence, and for the LO channel (lower), which is a quadratic dependence on \( l \).

For optical phonons \( v_g = 2\beta k \sim 2\beta/l \) leading to a quadratic dependence of the dephasing time on \( l \),

\[
\tau \simeq \frac{l}{v_g} = l \left( \frac{\partial \epsilon}{\partial p} \right)^{-1} = \frac{\hbar l}{2\beta k} \simeq \frac{\hbar l^2}{2\beta},
\]

as \( k \simeq 1/l \) (due to the bottleneck effect on the centre of the wave packet in the momentum space). In the case of LO phonons, the dephasing time-scales quadratically with the dot’s dimensions, and thus for the state-of-art structures it attains values much larger than the dephasing time for the LA channel (with the linear scaling) (cf. Fig. 9).
Figure 10. The correlation function’s decrease in value corresponds to the decrease in the overlap of the local distribution of a QD particle (exciton) with the phonon (LA) wave packet carrying off the excess energy; the correlation function is the overlap of a QD particle (exciton) probability density, $|\Psi(r)|^2$ (upper left), with the wave packet escaping from the QD-space region $A(r,t) = \int \int e^{-ikr'}|\Psi(r')|^2dr'dk e^{i(r\pm d \cdot \epsilon(k) t)k \cdot dr} dk$ (illustrated here as $A(r)$ for a sequence of times $\tau$—time-scale of dephasing).

6. Decoherence of the degrees of freedom of spin in quantum dots

Spin do not interact directly with phonons—the spin of the QD excitations interacts weakly with the lattice oscillations due to their links to orbital (charge) degrees of freedom via:

- spin-orbit coupling [43],
- specific Hund-like rules for multi-electron QDs [1]—the filling of the subsequent shell in the multi-electron QD depends upon the total electron (hole) spin of a given shell [the generalization of singlet and triplet states] which, in effect, link spin and orbital degrees of freedom.

Weak spin coupling with phonons suggests that the spin of a QD electron constitutes a well-isolated quantum system (an insignificant spin-orbit interaction results in an extremely slow spin decoherence, the same is due to weak interaction with nuclear spin) that is suitable for a qubit’s definition. One can expect that for spin qubits in QDs, DiVincenzo’s conditions would be satisfied [7, 14, 15]. Due to the minor influence of the surrounding medium, the QD’s spin coherence is maintained until the time period of order of $\mu$s [16]. However, a difficulty arises when Rabi oscillations are implemented (for single qubit operations). Because of the low value of the gyromagnetic factor in semiconductors, qubit spin control (a qubit spanning across two spin orientations in an external constant magnetic field) via Rabi oscillations is extremely slow and the DiVincenzo conditions are again not satisfied (the Pauli term, $g\mu_B B_z$, leads to very slight Zeeman splitting of only 0.03 meV/T, in GaAs).
For two-qubit operations of spin qubits, no such disadvantage exists—there is an effective procedure for switching spin qubit interactions on and off [15, 16] resulting in qubit entanglement control at the time-scale of picoseconds. The idea of spin interaction control follows from the phenomenon of exchange interaction between two spins, being induced by strong Coulomb interaction [44]. The exchange energy for it is the singlet-triplet energy gap for the spin pair [44], and consequently it is of (several) meV in magnitude, resulting in a picoseconds time-scale for the control of the entanglement of qubits. The scheme of this control relies upon the singlet and triplet states of an electron pair (each electron captured in an individual QD but located closely enough to maintain their quantum indistinguishability [their localized wave functions must overlap]) and their relation with the orbital structure of the corresponding wave functions. Due to the fermionic nature of electrons:

- the singlet state \( \frac{1}{\sqrt{2}}(|↑⟩_1 ⊗ |↓⟩_2 − |↓⟩_1 ⊗ |↑⟩_2) \) corresponds to the symmetric orbital wave function,

- the triplet states \( \frac{1}{\sqrt{2}}(|↑⟩_1 ⊗ |↑⟩_2 + |↓⟩_1 ⊗ |↓⟩_2) \) correspond to the antisymmetric orbital wave function,

so that the complete spin-orbital wave function remains antisymmetric, as is required for fermions. In the absence of a magnetic field, the singlet state is the ground state [44], but as the field increases in value [i.e., the magnetic field breaks the symmetry of time-reversion], it becomes less energy-efficient and, finally, the triplet state is preferred (with parallel spin orientation) [a parallel spin orientation is also preferred due to the Pauli term, which is, however, of very low value, and its contribution to energy competition is negligible; the triplet spin state is preferred due to the minimization of the energy of Coulomb interaction by the antisymmetric orbital state in the presence of the magnetic field breaking the time-reversion symmetry [44]]. For the critical field (of the order of a few T for QDs), both the singlet and the triplet states have the same energy, which means that the exchange qubit interactions is switched off (the exchange interaction constant is expressed via the difference in the energy value for the singlet and the triplet) [44]. It can be switched on again by varying the value of the applied external magnetic field and shifting the system out of the degeneracy point. The exchange interaction equal to the energy gap between the triplet and singlet states is originated by the relatively strong Coulomb interaction and varies within the range of several meV or more, which allows for the rapid entanglement of qubits.

Let us also mention that the possibility of using the exchange interaction to implement and exert control over single-spin qubits has been investigated, which has resulted in:

- a singlet- and a triplet-defined qubit on a two-electron QD of He-type [45],
- a spin qubit defined by the spin states with \( S_z = \pm 1/2 \) for three electrons [16] but separated energetically by a strong exchange-like interaction—i.e., a qubit defined by a pair \( |1/2, −1/2⟩_s, |1/2, 1/2⟩_t \) or a pair \( |1/2, 1/2⟩_s, |1/2, −1/2⟩_t \) [the spin states of three electrons can be classified according to the spin addition rule: first, two spins of 1/2 are added, yielding a singlet \( |0, 0⟩ \) and a triplet: \( \{ |1, −1⟩ \} \) and then the third
spin of 1/2 is added yielding eight three-electron spin states:
\[
\begin{align*}
&|1/2, -1/2 >_s \\
&|3/2, -3/2 >_t \\
&|1/2, -1/2 >_t
\end{align*}
\]
\[
\begin{align*}
&|1/2, 1/2 >_s \\
&|3/2, 1/2 >_t \\
&|1/2, 1/2 >_t
\end{align*}
\].

In both cases, however, for the collective qubit’s definition, the number of particles must be increased, which results in the enhancement of local decoherence \( \sim e^{N} \) \((N—number of qubits)\). On the other hand, however, it might be conveniently directed towards the application of collective-global (and thus more robust against decoherence) subspaces of the Hilbert space for multi-particle systems for QIP [13].

The scheme for the entangled qubits’ control relies upon the symmetry-induced close connection between spin exchange interaction and the orbital (thus, strong) Coulomb interaction of electrons (leading to an energy gap between the singlet and triplet states of an order of magnitude in meV). Due to this strong Coulomb interaction, and following the spin exchange interaction, the time-rates of double-qubit gate unitary operations are of the order of picoseconds, which suggests the convenience of spin degrees of freedom for QIP in QDs. A model of a quantum gate based on the above-mentioned idea was proposed by DiVincenzo [15, 16, 20]. In his model, a pair of H-type QDs (a single qubit spanned across electron spin states \( |1/2, -1/2 >, |1/2, 1/2 > \)) was analysed for two-qubit operations. However, for the implementation of a quantum computer, single-qubit operations are necessary as well, which unfortunately are extremely slow for the defined spin qubit on single-electron spin states. The idea of accelerating single-qubit operations can be associated with the enhancement of the gyromagnetic factor in semiconductor environments with magnetic dopants (as in so-called ‘diluted magnetic semiconductors’) [46–50] in a magnetically ordered phase. In such materials with low concentrations of magnetic ions (of a few percent range), typically Mn\(^{2+}\) ones, phase transition to a magnetically-ordered phase takes place due to the mediating role of band holes [49]. Such a phase transition takes place even at temperatures exceeding 100 K (in Ga(Mn)As) [49, 50] and, additionally, can be controlled via the hole concentration in the semiconductor [47, 48, 51–53]. The magnetic ordering of the material produces an extremely strong Weiss-like magnetic field internally, which acts exclusively on the degrees of freedom of the spin (i.e., it does not act on orbital ones), thus leading to a significant increase in value of the Pauli term, i.e., in the enhancement of the effective gyromagnetic factor. This suggests that single-qubit operations could be accelerated up to the level required by the DiVincenzo criteria in QDs in DMSs (such structures have already become available).

However, introducing an additional spin subsystem (and such is the magnetic dopant part of the semiconductor) causes a new problem. Such a subsystem is a source of collective excitations—spin waves, which interact directly with the qubit spin. In this system, spin waves (magnons) behave like phonons and produce similar spin decoherence effects (as presented for phonons and the charges in the previous paragraphs).

A more detailed analysis of the problem (as will be presented below) shows that spin waves cause harmful decoherence within time-rates of the order of 500-1000 ps, which is a serious negative side-effect threatening the feasibility of the spin logic gate (again, in the centre of the six-order time window between the control time and the amplitude decoherence for spins [54]). It exists, however, the promising opportunity to diminish the amplitude of this spin dephasing, which is in contrast to the phonon-induced effect in the case of spin waves
and can be achieved at very low temperatures, as will be demonstrated in the following paragraphs. The essential elements of the analysis are as follows: (1) the averaging over the random distribution of magnetic admixtures in a DMS [55] yields a spin waves spectrum in a Holstein-Primakoff representation [56, 57] [the averaging restores the effective translational invariance in a randomly doped system [55] and allows for the application of momentum representation]; (2) the determining of the spin wave dispersion in a DMS with respect to the hole and the magnetic dopant concentration; (3) analyzing the structure of the interaction between the spin waves and the spin of a QD exciton. The quadratic dispersion of spin waves found as the result of this analysis turns out to be crucial for the timing of the phase decoherence of spin in QDs. The estimation of the time rate of this process is given as the QD dimension divided by the spin wave group velocity, according to the general rule presented in the paragraph 5, \( v_g \sim \frac{\hbar}{3} \), \( \tau \sim \frac{d^2}{17a} \); the time-rate scales as the square of the QD dimension (marked as \( d \)), similarly as in the case of LO phonons (and similarly as for LO phonons rapidly increases with the dot dimension growth) and for typical dots of dimension of about 10 nm reaches values of the order of 500 ps.

6.1. Dephasing induced by the dressing of QD exciton spin with magnons in a diluted magnetic semiconductor’s surroundings

An interesting question arises with regard to the QD spin in the magnetic surroundings when the Pauli term causing the Rabi oscillations for spin can be strengthened due to an increase in the effective gyromagnetic factor. Spin does not interact directly with phonons, and thus it is free from phonon-induced dephasing. Nevertheless, the dephasing role of phonons may be played by spin waves in magnetically-ordered media which, on the other hand, are convenient for accelerating single-qubit QD spin control to the level required by the DiVincenzo conditions. Spin waves (frequently called ‘magnons’) are collective spin-type excitations in the ferromagnetic or anti-ferromagnetic medium (or in any other magnetically-ordered spin system), and possess similar band properties to phonons in crystalline structures. The spin-exchange interaction between the magnons and the local QD spin (of an exciton trapped in a QD) is relatively strong and causes the dressing of the QD spin with the magnons in a similar fashion to the dressing of the QD charges with phonons. The opportunity for the experimental study of such a spin dressing phenomenon may be linked to the so-called ‘diluted magnetic semiconductors’ of the type III-V (e.g., Ga(Mn)As) or II-VI (e.g. Zn(Mn)Se). In these magnetically and weakly doped semiconductors, some relatively small part of the cations (usually a few %) is randomly substituted by transition metal ions (typically of Mn). The admixture spins interact with the spins of band holes and as a result the ferromagnetic ordering of the admixture spins is observed. The related Weiss field enhances the effective gyromagnetic factor in the Pauli term, describing the spin action of the external magnetic field conveniently for the acceleration of the control over the local QD spin.

6.2. Spin waves in the diluted magnetic semiconductor

To describe the dephasing of QD spin caused by magnons in DMSs quantitatively, the analytical expression of the spin wave spectrum in the DMS is needed. This can be found in the paper [64]. For the relevant theoretical description of the spin subsystem of the DMS, the model of dopant spin exchange mediated by band holes is utilized [58, 59], assuming
that the $p - d$ exchange between the band holes and the impure magnetic atoms may lead to the ferromagnetic alignment of the magnetic dopants (Mn). Note that the holes taking part in the spin exchange with the dopant Mn ions cause an indirect exchange beyond the weak, direct, short-range and anti-ferromagnetic exchange between the magnetic dopants [48]. The hole-induced indirect coupling, even for low concentrations of holes $x_p$ (lower than the magnetic dopant concentration $x$) occurs strong and leads to the ferromagnetic ordering of Mn spins even at relatively high temperatures, ~ $110$ K in Ga$_{0.947}$Mn$_{0.053}$As [58, 60].

Let is be emphasized that, in III(Mn)V DMSs, the Mn atoms of the magnetic dopants are simultaneously shallow acceptor centres, whereas in II(Mn)VI-type DMSs, the Mn dopants are not acceptors and the holes must be supplied by additional $p$ doping.

To briefly sketch the derivation of the magnon spectrum in DMSs, let us set out the Hamiltonian for the DMS system with magnetic dopants Mn$^{2+}$ (with spin $S = 5/2$) in the form:

$$\hat{H} = \hat{H}_s + \hat{H}_p,$$

where $\hat{H}_s$ describes the spin subsystem of the DMS:

$$\hat{H}_s = -2 \sum_{j=1}^{N_p} \sum_{n} A_p \left( R_j - R_n \right) \hat{s}_j \cdot \hat{S}_n,$$

where $\hat{s}_j, R_n$ and $\hat{S}_n, R_n$ are operators for the spin and position of the $j$-th hole and the $n$-th impurity atom (in the lattice point $R_n$; the interstitial positions of the dopants are not accounted for); $A_p \left( R_j - R_n \right)$ is the $p - d$ exchange integral; $A_p \left( R \right) < 0$ (antiferromagnetic) and $|A_p\left(R\right)| \sim 1eV$; $n$ summation goes over lattice points occupied by magnetic dopants; hole concentration $x_p = \frac{N_p}{N}$, magnetic dopant concentration $x = \frac{N_i}{N}$, $N_p$ is the number of band holes which contribute to exchange with the impurity atoms, $N_i$ is the number of magnetic impurities, $N$ is the number of elementary cells and $\hat{H}_p$ is the fermionic Hamiltonian of the holes.

The important property of the considered system is the randomness of the distribution of the magnetic impurity atoms and the acceptors. The averaging over these random distributions conveniently results, however, in the effective restoring of the translational symmetry broken at any particular dopant distribution, though it is again present after averaging. This allows for momentum-representation for collective excitations, which can be utilized in the diagonalization of the spin part of the Hamiltonian. This procedure is originally presented in [64] and results in the explicit form for the low-temperature excitation spectrum of the DMS spin subsystem. This spectrum of magnons in DMSs consists of two branches: the gap-less branch $\varepsilon_1 \left( k \right)$ and the "optical-type" gapped branch $\varepsilon_2 \left( k \right)$. The value of the gap is given by the formula $\varepsilon_2 \left( 0 \right) = -\tilde{A}_p \left( 0 \right) \left( x_p + 2Sx \right)$ ('tilde' denotes the Fourier picture). The energy of the magnon branch $\varepsilon_1 \left( k \right)$ grows with increasing $k$, whereas the energy for the branch $\varepsilon_2 \left( k \right)$ diminishes with increasing $k$. Within the long-wave limit, i.e., for small $k$, the spin wave energy spectrum can be written as follows [64]:

On the 'Three-Orders Time-Limit' for Phase Decoherence in Quantum Dots

http://dx.doi.org/10.5772/60685
\begin{align*}
\begin{cases}
\epsilon_1(k) = D k^2, \\
\epsilon_2(k) = D_0 - D k^2,
\end{cases}
\end{align*}

(28)

with: \( D_0 = -\tilde{\hat{A}}_p(0) ( x_p + 2 S x ) \), \( D = -\tilde{\hat{A}}_p(0) \frac{2 S x x_p}{x_p + 2 S x} l_{ex}^2 \), where \( l_{ex} \) expresses the space-range of the exchange interaction.

### 6.3. Dressing of the QD’s local exciton spin by magnons from the surrounding DMS

In analogy to the creation of the polaron in a QD by the dressing of the QD exciton with band phonons, one can observe the formation of the excitonic-magneto-polaron (EMP), i.e., of the localized QD exciton spin dressed with magnons in the surrounding DMS (in the case of the magnetically ordered phase of the DMS at a sufficiently low temperature). Similar to the ordinary polaron, the dephasing of the spin caused by the magnons expresses the inertia of the collective spin wave system. For the non-adiabatically and rapidly excited bare exciton to the QD one deals with a non-stationary state of the whole system of QD with surrounding DMS. Similarly to the dephasing of the QD charge by phonons, in the case of step-by-step formation of the exciton magneto-polaron (EMP) in QD one deals with the outflow of an excess energy to the region outside the QD. This excess energy is linked now with the spin exchange interaction and the carriers for the energy outflow are magnons. The corresponding quantum evolution of the total system is the evolution of the non-stationary state (originally at \( t = 0 \) defined by the non-stationary initial state of the bare exciton rapidly excited in the QD). During this evolution, the mean energy is shared between the QD and the surrounding magnons in the DMS (the spin wave sea). The final EMP localized in QD and dressed with the cloud of magnons has a lower mean energy than the initial bare exciton. Together with the magnons escaping from the QD region (these ones remove the excess of the spin exchange energy outside the QD), the whole system remains in the non-stationary state, although the local EMP might be treated as the ‘stationary’ component of the eventual state of the system. In order to estimate the time-rate corresponding to the dressing of the local spin in the QD by magnons resulting in the formation of the EMP, the same rule as that for the QD charge with phonons can be utilized.

In analogy to phonon dressing, one arrives at the formula for the correlation function,

\[
I(t) \sim e^{-\frac{i E t}{\hbar}} \int dr \lvert \Psi_0(r) \rvert^2 \int dk F^*(k) e^{i(kr - \epsilon(k)t)},
\]

describing the time-dependent evolution of the overlap of the local QD particle distribution, \( \lvert \Psi_0(r) \rvert^2 \), with the wave-packet of the spin waves escaping the dot region step-by-step, \( \int dk F^*(k) e^{i(kr - \frac{\hbar \partial \epsilon(k)}{\partial k})} \) (where \( k \) corresponds to the centre of the wave-packet being of the order of \( \sim 1/l \), as caused by the QD bottleneck effect similar to that for phonons and which is visible from the form of \( F^*(k) \)). The group velocity of this wave-packet is equal to \( v_g = \frac{\partial \epsilon(k)}{\partial k} \) (at \( k \sim 1/l \)). Thus, one can conclude that the dephasing time defining the time-scale of the
dressing of the QD exciton spin with magnons and expressed by the function \( I(t) \) is given by that according the general rule, \( \tau \approx \frac{l^2}{2D} \).

The dressing of the QD exciton spin with magnons is similar to the dressing of the QD charge with LO phonons, since both LO phonons and magnons have a quadratic dispersion. Utilizing now the formulae for magnon energies as given by Eqs. (28), we arrive at the assessment for the time of formation of the EMP in form,

\[
\tau \approx \frac{l}{\sqrt{2g}} = l \left( \frac{\partial \varepsilon}{\partial k} \right)^{-1} = \frac{l}{2Dk} \approx \frac{l^2}{2D},
\]

\( k \approx \frac{1}{l} \) because of above-mentioned QD bottleneck. Note that the derived time for dressing with magnons scales as \( l^2 \). Therefore, it is relatively lengthy for state-of-the-art QD sizes (typically of the order of 10 nm for the diameter). The dephasing time of the exciton spin given by the above estimation depends upon the magnetic admixture and band-hole concentrations in the DMS because of the concentration-dependence of the factor \( D \). To illustrate this behaviour, several examples are presented in Fig. 11.

For the experimentally observed QD in the structure \( \text{Zn(Mn)Se/CdSe} \) with a dimension of \( \sim 10 \text{ nm} \) and with concentrations of \( x = 0.25, x_p = 0.025 \), one can estimate using the above formula the spin dephasing time as being of the order of 150 ps, which agrees with the timing of the formation of the EMP experimentally observed in \( \text{Zn}_{0.75}\text{Mn}_{0.25}\text{Se/CdSe} \) (note that, for this structure, the time for exciton annihilation is considerably longer, \( \sim 600 \text{ ps} \), and the complete formation of the EMP can thus be observed) [65–67].
6.4. Explicit form of the interaction of magnons with QD spin causing the spin pure dephasing

An important observation might be made that it is a considerable difference between the phonon- and magnon-assisted dephasing phenomena of charges and spin in QDs, respectively. To demonstrate this difference, it is necessary to analyse the exchange-spin interaction of QD excitons with magnetic admixture spin waves. This interaction can be expressed as follows:

\[
\hat{H}_{sd}(\mathbf{R}_e, \mathbf{R}_h) = -2\beta_0 \sum_n A_e(\mathbf{R}_e - \mathbf{R}_n) \hat{s}_e \cdot \hat{S}_n - 2\beta_0 \sum_n A_h(\mathbf{R}_h - \mathbf{R}_n) \hat{s}_h \cdot \hat{S}_n, \tag{29}
\]

where \(\hat{s}_e(h)\) is the spin operator of the electron (hole) of the exciton in the QD, \(\mathbf{R}_n\) is the position of the magnetic dopant (Mn\(^{2+}\)), \(\hat{S}_n\) is the spin operator of the dopant, and the \(n\) summation goes over the lattice sites occupied by magnetic dopants. The term \(A_{e(h)}(\mathbf{R}_{e(h)} - \mathbf{R}_n)\) describes the exchange-spin interaction between the electron (hole) spin of the QD exciton and the spin of the dopant, whereas an effective coefficient \(\beta_0\) accounts for the additional decrease of the exchange integrals due to the dot-structure proximity in real system (\(\beta_0\) is assumed to be 0.1 as estimated from the experimental data [65, 66]).

For the four-fold spin structure of the QD exciton, we use the notation \((j, n, s_z = \pm \frac{1}{2})\), where \(j = 1, 2\) correspond with the anti-parallel and parallel spin alignments of the e-h pair, respectively, and \(s_z\) is the spin projection of the electron in the e-h pair. Taking these representations for e-h pair spins \(\hat{s}_{e(h)}\) and the magnon representation (in the Holstein-Primakoff picture) for dopant spins \(\hat{S}_n\) [64], one can rewrite the Hamiltonian (29) in the manner presented below.

By \(a_{jns_z}^{(+)}\) we denote the bosonic annihilation (creation) operator of the exciton in the \((jns_z)\) state \((j = 1, 2\) indicates the opposite and the same spin mutual alignment of electron-hole pair creating an exciton, correspondingly; \(s_z\) denotes the spin alignment of the electron in electron-hole pair). One can now rewrite the Hamiltonian (29) in the basis of these excitonic states, namely, \(\sum_{\mu, \mu'} |\mu|H_{ad}|\mu'\rangle a_{\mu'}^\dagger a_{\mu}\), where \(\mu = (jns_z)\).

Taking the Holstein-Primakoff representation [68] for the spins of dopants and averaging over the dopant random distributions [64] and changing to the magnons—\(\hat{q}_i^{(+)}\), \(i = 1, 2\)
(diagonalization transformation is given explicitly in Ref. 64)—we arrive at the following form of the exchange Hamiltonian (29) [69]:

$$\hat{H}_{sd} = \hat{H}_{sd}^1 + \hat{H}_{sd}^2,$$

(30)

$$\hat{H}_{sd}^1 = -\left(\frac{\mu_0}{(2\pi)^7}\right)^2 2S\hat{q}_1\beta_0 \int d^3k_1 \int d^3k_2 \left[ v_{k_1}\hat{a}_1^+(k_2) + u_{k_1}\hat{a}_2^+(k_2) \right]$$

$$\times \left[ v_{k_2+k_1}\hat{a}_1(k_2+k_1) + u_{k_2+k_1}\hat{a}_2(k_2+k_1) \right]$$

$$\times \sum_{n,n',s_z=-1/2}^{1/2} s_z \left[ \left[ F_{nn'}^e(k_1) - F_{nn'}^h(k_1) \right] \hat{a}_{1n's_z}^+ \hat{a}_{1n's_z}^+ + F_{nn'}^h(k_1) \hat{a}_{2n's_z}^+ \hat{a}_{2n's_z}^+ + F_{nn'}^e(k_1) \hat{a}_{2n's_z}^+ \hat{a}_{1n's_z}^+ + h.c. \right],$$

(31)

$$\hat{H}_{sd}^2 = -\sqrt{2}\frac{S\hat{q}_1\beta_0}{(2\pi)^7} \int d^3k \sum_{n,n'} \left[ \left[ v_{k}\hat{a}_1^+(k) + u_{k}\hat{a}_2^+(k) \right]$$

$$\times \left[ F_{nn'}^e(k) \left( \hat{a}_{1n1/2}^+ \hat{a}_{1n1/2}^+ - \hat{a}_{1n1/2}^+ \hat{a}_{1n1/2}^+ \right)$$

$$+ \hat{a}_{2n1/2}^+ \hat{a}_{2n1/2}^+ \right]^+ + h.c. \right].$$

(32)

In the first part, $\hat{H}_{sd}^1$ we can identify the term describing the interaction without any change in the exciton spin (neither the electron nor the hole in the e-h pair). This term thus describes the interaction channel when the spin of the created (annihilated) magnon has to be cancelled by an annihilated (created) magnon (spin is conserved—it is different compared to phonons). It should be emphasized that, in the second part, $\hat{H}_{sd}^2$, the exciton spin-flip processes are included only with involvement of a single magnon. Therefore, the term, without any change of the state (the spin state here) and which leads to a pure dephasing of spin, contributes only to $\hat{H}_{sd}^1$. The Hamiltonian part $\hat{H}_{sd}^2$ does not cause any pure dephasing because this part of the interaction does not conserve the exciton state (spin). The term $\hat{H}_{sd}^2$ always causes an amplitude (diagonal) decoherence as a changing of the exciton spin state.

With the first term in $\hat{H}_{sd}^1$, and significantly for the pure dephasing of the QD exciton spin, one can associate the vertex which gives an imaginary part of the mass operator, cf. Fig. 12 (leading to the pure dephasing of the exciton ground state). Note that this is different from that for phonons, cf. Fig. 2.

The imaginary part of the mass operator given by the graph in Fig. 12 can be written as follows:

$$\gamma_n(\omega, T)$$

$$= \pi \sum_{i,j=1}^{2} \sum_{k_1, k_2} \left| V(n, k_1, i; n, k_2, j) \right|^2 [n_i(k_1) + 1] n_j(k_2) \delta(\omega - E_n - \epsilon_i(k_1) + \epsilon_j(k_2)),$$

(33)
\[ n = 0 \text{ comprises here all the exciton ground state quantum numbers } (1, 0, \frac{1}{2}), \text{ and:} \]

\[ V(n, k_1, i; n, k_2, j)|_{n=0} = \beta_0 2 S x \left[ \begin{array}{c} v_{k_1}^* u_{k_1} \vspace{2mm} \\
\v_k^* u_k \end{array} \right] \left( F_{00}^0(k_1 - k_2) - F_{00}^0(k_1 - k_2) \right), \]

The matrix is addressed to the magnon branches: \( \{ij\} = \left[ \begin{array}{c} 11, 12 \\
21, 22 \end{array} \right] \), \( n_i(k) \) is Bose-Einstein distribution for the \( i \)-th magnon branch, \( u_k, v_k \) are coefficients of the DMS magnon diagonalization transformation [64], and

\[ F_{mn}^{(h)}(k) = A_{e(h)}(k) \int d^3 R_e \int d^3 R_h \Psi_n^*(R_e, R_h) e^{i k \cdot R_e} \Psi_n(R_e, R_h), \]

where \( \Phi_0 = \frac{1}{(2\pi)^3} \frac{1}{L_{\perp} L_x L_y} \exp \left[ -\frac{\delta_{1}^2}{2 L_{\perp}^2} - \frac{\delta_{1}^2}{2 L_x^2} \right] \), with the range of the electron and hole in the \( xy \) plane, \( L_{e(h)} \) denotes variational parameters introduced in order to account for e-h Coulomb attraction [64, 70], \( A_{e(h)}(R) = A_{e(h)}(R) e^{-2R/L_{e}} \) with the range of the exchange

\[ l_{ex} \approx a, (a^3 = v_0), \tilde{A}_{e(h)}(k) = \frac{\tilde{A}_{e(h)}(0)}{[1 + k^2 L_{e}^2/4]}, \tilde{A}_{h(h)}(0) = \frac{\tilde{A}_{h(h)}(0)}{4 \pi} A_{e(h)}. \]

The imaginary part of the mass operator can be rewritten in the form:

\[ \gamma_0(\omega, T) = \pi \sum_{k_1, k_2} |V(k_1 - k_2, 0)|^2 \]

\[ \times \left\{ n_1(k_1) + 1 \right\} n_1(k_2) v_{k_1}^2 v_{k_2}^2 \delta(\omega - E_0 - \varepsilon_1(k_1) + \varepsilon_1(k_2)) \]

\[ + [n_2(k_1) + 1] n_2(k_2) u_{k_1}^2 u_{k_2}^2 \delta(\omega - E_0 - \varepsilon_1(k_1) + \varepsilon_1(k_2)), \tag{34} \]

with \( \varepsilon_1(k) = Dk^2, \varepsilon_2(k) = D_0 - Dk^2 \), \( V(k_1 - k_2, 0) = \beta_0 2 S x (F_{00}^0(k_1 - k_2) - F_{00}^0(k_1 - k_2)) = f(k_1 - k_2) = A_{t} \frac{e^{-2\kappa k^2 L_{e}^2}}{[1 + k^2 L_{e}^2]}, A = \beta_0 2 S x (\tilde{A}_{h}(0) - \tilde{A}_{h}(0)), \kappa = l_{ex}^2 /4, \alpha = \kappa /2 (l^{-}\text{the dimension of the exciton in the ground state averaged over all directions} [70]). \]

Finally:

\[ \gamma_0(\omega, T) = \pi A^2 \sum_{k_1, k_2} |V(k_1 - k_2, 0)|^2 \]

\[ \times \left\{ n_1(k_1) + 1 \right\} n_1(k_2) v_{k_1}^2 v_{k_2}^2 \delta(\omega - E_0 - (Dk_1^2 - k_2^2)) \]

\[ + [n_2(k_1) + 1] n_2(k_2) v_{k_1}^2 u_{k_2}^2 \delta(\omega - E_0 - (Dk_1^2 + k_2^2)) \]

\[ + [n_2(k_1) + 1] n_2(k_2) u_{k_1}^2 u_{k_2}^2 \delta(\omega - E_0 - (D(k_1^2 - k_2^2))) \tag{35} \]}
Due to the different types of vertex interactions responsible for pure dephasing, namely exciton-phonon (triangle vertex) and exciton-magnon (quadratic vertex), they lead to significantly different phenomena. Spin conservation in the vertex (during interaction) requires the participation of two magnons (the exciton spin state remains unchanged), unlike phonons, in which case single-photon emission or absorption is feasible with the exciton state unchanged. For magnons, magnon emission must be accompanied by magnon absorption in order to balance the loss of spin in the vertex due to the spin wave emission—equation (33). Although factors corresponding to the emission (of type $1 + n_i$) assume non-zero values even at $T = 0$ (this is true for phonons as well), the absorption factor (of type $n_i$) the probability of magnon absorption is proportional to the number of magnons) falls to zero for $T \to 0$, and this is the reason why exciton spin dephasing by magnons becomes smaller and smaller ($\to 0$) as the temperature falls to $T = 0$ (in the case of phonons, dephasing remains non-zero even at $T = 0$ due to only phonon emission, even in a phonon vacuum for $T = 0$). Moreover, for magnons, $n_2 << n_1$ in the limit $T \to 0$ (due to a magnon gap) and therefore at low temperatures only terms with factors $n_1(1 + n_i)$ contribute.

At higher temperatures, when the number of thermodynamic magnons (liable to absorption) increases sufficiently, magnon-induced spin dephasing is as effective as phonon-induced charge dephasing. The difference between both effects presented herein emphasizes the fact that spin is more resistant to DMS-magnon-induced dephasing at low temperatures due to spin conservation constraints in comparison to phonon-induced charge dephasing (which is strong even at $T = 0$).

7. Magnon-induced versus phonon-induced QD exciton dephasing

Due to the different types of vertex interactions responsible for pure dephasing, namely exciton-phonon (triangle vertex) and exciton-magnon (quadratic vertex), they lead to significantly different phenomena. Spin conservation in the vertex (during interaction) requires the participation of two magnons (the exciton spin state remains unchanged), unlike phonons, in which case single-photon emission or absorption is feasible with the exciton state unchanged. For magnons, magnon emission must be accompanied by magnon absorption (in order to balance the loss of spin in the vertex due to the spin wave emission—equation (33)). Although factors corresponding to the emission (of type $1 + n_i$) assume non-zero values even at $T = 0$ (this is true for phonons as well), the absorption factor (of type $n_i$) the probability of magnon absorption is proportional to the number of magnons) falls to zero for $T \to 0$, and this is the reason why exciton spin dephasing by magnons becomes smaller and smaller ($\to 0$) as the temperature falls to $T = 0$ (in the case of phonons, dephasing remains non-zero even at $T = 0$ due to only phonon emission, even in a phonon vacuum for $T = 0$). Moreover, for magnons, $n_2 << n_1$ in the limit $T \to 0$ (due to a magnon gap) and therefore at low temperatures only terms with factors $n_1(1 + n_i)$ contribute.

At higher temperatures, when the number of thermodynamic magnons (liable to absorption) increases sufficiently, magnon-induced spin dephasing is as effective as phonon-induced charge dephasing. The difference between both effects presented herein emphasizes the fact that spin is more resistant to DMS-magnon-induced dephasing at low temperatures due to spin conservation constraints in comparison to phonon-induced charge dephasing (which is strong even at $T = 0$).

8. Conclusions

In conclusion, we can state that in the case of QDs we deal with a specific type of phonon-induced phenomenon, which is essentially distinct from the phonon-induced effects.
in bulk semiconductors. This difference is caused by the compatibility of the energy scale for carriers trapped in QDs with the band-phonon energy scale. Owing to this energy coincidence, the coupling of carriers in QDs with phonons always meets its strong regime limit. This coupling cannot be treated perturbatively, in general, and the resonance effects are of primary importance, resulting in strong polaron-type modifications of the QD electron and exciton spectra. The typical energy shift due to the formation of electron-polaron is of the order of 10%, while for exciton-polaron it is of the order of 5% with respect to the bare energy levels in QDs treated as separated from the environment. The confinement of carriers, as in the case of QDs, also causes the significant enhancement of the effective Fröhlich constant due to non-adiabatic effects, which additionally strengthens the electron-LO phonon interaction. The dressing of the electrons/holes/excitons in QDs with band phonons from the surrounding crystal induces the dephasing of charge (orbital degrees of freedom) in QDs (the off-diagonal decoherence). The typical time-scale of this dressing process in the case of the formation of an exciton-polaron in QDs turns out to be of the one picosecond time-scale (for a typical QD of 10 nm diameter). This dephasing is caused by the exciton dressing with LA phonons. Worth noting is the observation that the dephasing due to LO phonons is considerably smaller and slower—of the 100 ps scale. This phenomenon is caused by the relatively weak LO phonon dispersion near the Γ point (in the Brillouin zone), in contrast to the dispersion of LA phonons. Nevertheless, the outflow of the excess polarization energy to the space region outside the QD, as a typical process during LO polaron formation, is eventually accelerated by the anharmonic coupling (LO-T A is the most important anharmonic channel in GaAs), which results in a few ps time-scale. It is important to note that these effects of QD-charge dephasing by band phonons refer not only to QDs but also to all nanostructures in solids, because the carrier localization (the space-confinement of trapped carriers) plays the essential role here.

We have observed also that in magnetically-ordered media (like in DMSs), magnons (spin waves) play a similar destructive role to phonons. Spin waves cause the dephasing of the exciton spin in QDs, in a process of the formation of excitons-magnetos-polarons by the step-by-step dressing of the local spin of exciton in QD with the magnon cloud from the surrounding magnetically-ordered medium. By using the Green function technique, we have estimated the time for the dressing of the local exciton spin with band magnons in the case of DMSs which surround QDs by analogy to the dressing of QD charge (i.e., QD orbital degrees of freedom) with band phonons from the surrounding crystal. Nevertheless, the significant difference between these two phenomena is observed and elucidated, namely, in the case of spin dressing two magnons are needed (creation and annihilation) owing to the spin conservation in the interaction vertex, which results in the complete freezing (vanishing) of the spin pure dephasing at $T = 0$. This is in contrast to the phonon-induced pure dephasing, which maintains strong even at $T = 0$.

The dephasing scheme for the exciton charge and spin in QD structures is important for the feasibility assessment of QIP implementations in QDs. The picoseconds time-rate for QD charge dephasing probably precludes the feasibility of the implementation of error-correction schemes for all optically-controlled gates in QD technologies. The dressing of a localized spin with magnons in a DMS (i.e., the time corresponding to the formation of EMP in a QD) takes place at a longer time-scale in comparison to dressing charges with phonons, and the related time-scale is of the order of 150-200 picoseconds due to the relatively weak quadratic magnon dispersion, similar to as was the case for LO phonons. Nevertheless, the time-rate for QD spin dephasing induced by magnons in the surrounding DMS is also inconvenient for QIP
applications, similarly to the case of the dephasing of QD charges by phonons. The overall time-scale for QD spin kinetics (QD/DMS-embedded structures) is shifted by three-orders of magnitude to longer periods in comparison to QDs’ orbital degrees of freedom, though again with the same inconvenient dephasing time-rate falling right in the middle between the control-timing and the relaxation-timing (which does not allow the satisfaction of the DiVincenzo conditions). In this way, the ‘three-orders time-limit’ caused by the dephasing phenomena is repeated for spin in the QD/DMS.

The pure dephasing of spin in QD/DMS structures disappears, however, at $T = 0$, and is strongly suppressed in amplitude at low temperatures (in contrast to the charge dephasing), which supports expectations of some advantages of spin degrees of freedom in QDs for QIP applications.

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**References**


[38] A. L. Fetter, J. D. Walecka, Quantum Theory of Many Particle Systems, PWN, Warszawa, 1982