Optically manipulating spins in semiconductor quantum dots*

Wang Yao  
*Department of Physics, The University of Texas, Austin, Texas 78712-0264

Ren-Bao Liu  
Department of Physics, The Chinese University of Hong Kong, Shatin, N.T., Hong Kong, China

L. J. Sham  
Department of Physics, University of California San Diego, La Jolla, California 92093-0319

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Physics considered here is the active control of a quantum system and of its decoherence by its environment. The relevance is in the quantum nature of nanoscience and how coherent optics in semiconductor quantum dots can contribute to quantum control. This article reviews: (1) The more recent theory of control of a set of dot spins through cavity quantum electrodynamics and (2) the quantum basis for control of decoherence of the electron spin interacting with the nuclei in the quantum dot. © 2007 American Institute of Physics. [DOI: 10.1063/1.2723178]

I. INTRODUCTION

Semiconductor quantum dots, as they came into being, had been hailed as “giant atoms.” Hence, visions dance in our heads of the quantum control of dots with ease equal to that achieved in the quantum optics of atoms. We are here to examine in theory what could be done in the solid state version of control of quantum systems and what support experiments have shown so far. We wish this subject to reach a wide circle of semiconductor physicists and, therefore, we shall attempt to make the terminology and concepts of quantum operations in this review as self-contained as possible.

Optical control of electron spins in dots is the subject of this review. The coherent control of exciton and biexciton dynamics in a dot has been reviewed in a previous ICPS conference and elsewhere. The work paves the way to the optical control of a dot electron. A doped dot of one active electron presents us with a two-level quantum system with a very long lifetime (a spin flip time of several milliseconds), which gives rise to a sufficiently long coherence time for many operations of duration of the order of 10 ps. The fundamental physical properties of the doped dots have been studied with the purpose of single electron optoelectronics (see the recent review in Ref. 5) and optical control of the spins in the dots has been suggested for quantum computers.

The motivation for the study of the dot spins can be divided into two classes:

1. This is the Mount Everest of electron physics. Nanoscience brings us the ultimate quantum playground to make the electrons dance and to observe them. Control and measurement have to be carried out with blunt (macroscopic) instruments. The stake in basic physics is the uncharted area between quantum and classical physics.

2. Semiconductor optics should be able to contribute to quantum technology, such as information processing and computation. The difficulty of scaling from a toy of a few qubits to a computer is certainly nontrivial, starting with the transportation of qubits to distant sites. The current modus operandi is to experiment on a small system or on basic quantum telecommunication. The progress in the last decade across a broad range of research efforts has been very encouraging.

There are a number of experimental achievements important to optical control of dot spin: Single trion spectroscopy, polarizing electron spin weakly and to almost perfection, and coherent control of an ensemble of single spins. There have been theoretical proposals for control of the single dot spin and a pair of spins residing in neighboring dots. Experiments are proceeding with the optical control of single spins.

The central question here is whether the control and decoherence of a quantum unit, which involves the interaction with a macroscopic system, can be facilitated by an intermediary. For control of the spins with laser pulses, we review the research on the intermediaries of cavity and wave guide photons. The use of the cavity photons in a quantum network of dots connected by traveling photons, described below, is different from the pioneering use of the cavity as a bus for spins in a number of dots connected to the same cavity. For the electron spin decoherence, we review the recent quantum theories and make a case for control of the electron spin to restore the coherence lost due to the interaction with the nuclear spins in the dot as the mesoscopic shield from the larger environment.

II. ELECTRON SPIN IN A DOT AS TLS

TLS is the abbreviation for the “two-level system.” There is experimental evidence that the electron spin is a
robust isolated quantum TLS,\textsuperscript{5} even under optical excitation to trion.\textsuperscript{11} The effect of the spin interactions may be classified as decoherence, to be addressed in Sec. IV. The object of the quantum control of electron spins is to prepare a system of localized spins in a particular state and then to bring it to another designated state. The difficulty with trying to directly control a system, of always interacting spins, is that the physical resource required scales exponentially with the number of spins.\textsuperscript{21} A method to avoid such an impossible enormous expense of resource is known as the quantum circuit in which operations, each involving a small number of quantum units, are performed sequentially. It is proven that a desired quantum transformation can be designed with only operations which involve two spins\textsuperscript{22} or with the availability of a group of rotations of a TLS and an entangling operation with two TLS.\textsuperscript{23} Thus, a demonstration of control over one spin and two spins may be sufficient to ensure that the control of a large number of spins is feasible.

III. CONTROL WITH CAVITY QED

A key issue in implementing quantum control of a large system of quantum units is the transfer of unit state information between two distinct units which must be able to survive without a significant loss of coherence.\textsuperscript{9} An alternate approach is to build a quantum network of small clusters of a few quantum units connected by quantum channels carrying photons.\textsuperscript{24} This could be the blueprint for quantum telecommunication and distributed quantum computing.\textsuperscript{25,26} The idea is that: (1) Each cluster, called a node, is isolated from the others and can be controlled quantum mechanically; (2) the information of the quantum state can be coded in photons transported along a channel to the next node; (3) the information carried by the photon wavepacket is used to build a state of sending and receiving nodes. The entanglement of states in two nodes is of fundamental interest to quantum applications. Strongly correlated electron states can be built from the entanglement.\textsuperscript{27}

The first proposed node\textsuperscript{24} utilizes the cavity quantum electrodynamics (QED) of atoms. The sending and receiving of the photons would be carried out using the symmetry between the two nodes. A theory of the Raman process assisted by the cavity photon\textsuperscript{28} maps the motional state of a trapped atom to a photon state and vice versa. The single atom state is replaced by a collective state of atoms for its adiabatic transfer with a single photon state which lessens the strong coupling requirement between the atoms and the cavity.\textsuperscript{29} An adiabatic mapping scheme for the single trapped atom was also given.\textsuperscript{30}

We describe below a method of control with a shaped optical pulse which can convert a spin state in a dot to a wave packet of a linear combination of zero and one photon states propagating along a wave guide and vice versa.\textsuperscript{31,32} This quantum theory of evolution in the system of a coupled dot, cavity, and wave guide, shown in Fig. 1(a), does not depend on the symmetry between the sender and the receiver nor the adiabatic approximation.

### A. Basic optical control process

The goal is to convert an electron spin state to a photon wavepacket which propagates down the waveguide shown in Fig. 1(a) and to perform the reverse of converting a photon state carried by the wave packet into an electron spin state. To be specific, the electron spin state $\beta_+|+\rangle + \beta_-|\rangle$ is a linear combination of the eigenstates $|\rangle$ along the $x$ axis. The photon state $\beta_+|\rangle + \beta_-|\rangle$ is the same linear combination of zero-photon state $|\rangle$ and one photon state wavepacket $|\rangle$, given by

$$|\rangle = \int d\omega \alpha(t,\omega)|\rangle$$

composed of one-photon frequency states $|\rangle$ with the propagating description understood and the coefficient $\alpha(t,\omega)$ at time $t$ normalized.

Light excites the spin states $|\rangle$ to the lowest trion state $|T\rangle$, which are composed of a singlet pair of electrons and a heavy hole state $|h_{3/2}\rangle \pm |h_{3/2}\rangle$ (normalization omitted), respectively. The selection rules connect the $X$ polarized light to the vertical transitions in Fig. 1(b), namely between $|+\rangle$ and $|T\rangle$ or $|\rangle$ and $|T\rangle$ and connect the $Y$ polarized light to the slanted transitions in Fig. 1(b), namely between $|+\rangle$ and $|T\rangle$ or $|\rangle$ and $|T\rangle$. An ac Stark pulse is used to bring the trion state $|T\rangle$ into resonance with the cavity mode plus the spin-down state, $|\rangle\rangle$. Then a shaped laser pulse with $Y$ polarization is used to move the $|+\rangle$ state to $|T\rangle$, which evolves into $|\rangle\rangle$ by the coupling of the $X$ cavity mode photon $|\rangle\rangle$ to the exciton component of the trion state $|T\rangle$. The coupling of the cavity to the waveguide changes the cavity photon into a propagating wavepacket, leaving the spin-down part of the electron state $|\rangle$ unchanged. The sequence of transformations, with the additional specification of the zero cavity photon state $|\rangle$, is, in the order of the spin, cavity, and waveguide

\begin{equation}
(\beta_+|+\rangle + \beta_-|\rangle)|\rangle\rangle \rightarrow (\beta_+|T\rangle + \beta_-|\rangle)|\rangle\rangle \rightarrow |\rangle\rangle(\beta_+|\rangle + \beta_-|\rangle)\rangle\rangle \rightarrow |\rangle\rangle(\beta_+|\rangle + \beta_-|\rangle)\rangle\rangle.
\end{equation}
qubit (0,1) in the wave guide via the cavity mode.

We emphasize that the process described above has been shown to be a unitary evolution. In particular, the formation of the photon wavepacket is deterministic, controlled by the laser pulse, unlike the probabilistic spontaneous emission. For any desired shape of the wavepacket, reasonably smooth, we can calculate the pulse shape of the laser which will produce it. The unitary transformation also guarantees the process to be reversible. The state of the photon wavepacket can be substituted with the spin state in a second dot connected to the wave guide via a cavity. With the known system parameters of the second node, the laser pulse shape can be found to engineer the reverse process.

B. Entanglement, initialization, and measurement

Unlike population transfer, the basic process described in the previous subsection can be extended. For example, a shorter laser pulse can transform the spin state |+⟩ to a linear combination of it with a trion state, |+⟩+|T⟩, the trivial normalization being understood. The basic process is then modified to

\[
|+\rangle \langle 0|_\text{vac} \rightarrow (|+\rangle + |T\rangle) \langle 0|_\text{vac} \\
\rightarrow (|+\rangle \langle 0| + |\rangle \langle -\rangle|_\text{C}) \langle 0|_\text{vac} \\
\rightarrow |0\rangle \langle +|_\text{vac} + |\rangle \langle -\rangle|_\text{α}. \tag{3}
\]

The result is a quantum marvel—an entangled state of spin and photon, leaving the cavity in its ground state. Since the process can be reversed, we can entangle one dot spin with a distant dot spin connected by the wave guide. In this way, quantum control of the whole spin system may be built.

The basic process is easily modified to put the spin in, say, the ground state, |−⟩. Suppose we start with an unpolarized state or in total ignorance of the spin state, represented by the density matrix 0.5(|+⟩⟨+| + |−⟩⟨−|). The cooling of the spin temperature from infinite to zero consists in four steps:

1. An X polarized ac Stark pulse is adiabatically switched on, bringing the states |T⟩ and |−⟩, C into resonance. The two states are split into two polariton states by the cavity-dot coupling.
2. A Y polarized pulse moves the spin-up state |+⟩ to a linear combination of the two polariton states.
3. The polariton state relaxes to the spin down state |−⟩ rapidly, emitting a photon into the waveguide.
4. The ac Stark pulse is adiabatically switched off.

This process dumps the spin entropy into the environment. The advantage of this method of initialization is its short time of the order 100 ps.

In the above process, a change of the pulse sequence can be used to measure the spin state. First a π-rotation pulse of polarization X moves the spin from |−⟩ to |T⟩ and then a longer Stark pulse of polarization Y brings the trion state into resonance with |−⟩C. The rapid emission of an X photon in the waveguide can be collected by a photon detector and the spin returns to |−⟩. If the spin before measurement is in the up state |+⟩, there will be no photon emitted and no change in state. This measurement process, which does not change the spin state, is known as a quantum non-demolition measurement. It can be recycled many times to accumulate photons in the detector for a definitive measurement.

The theory of the processes above assumes a precise knowledge of the dot-cavity and cavity-waveguide couplings and a perfect performance of the shaped pulses. An error analysis of these factors shows that a 10% error in any of these quantities gives no more than a 0.5% increase to the deviation from perfect fidelity. Also a feedback loop can be designed with the measured outcome of a process used to adjust the pulse shape required.

The experimental progress is striking. In addition to the quantum dots in cavity work reviewed in Ref. 20, there are now a fabrication of waveguides coupled to high quality microcavities and a demonstration of strong coupling between exciton in a dot and a cavity mode.

IV. QUANTUM CONTROL OF DECOHERENCE

The spin state deteriorates in the presence of outside influences in two ways. Relaxation of the spin polarization along the magnetic field to its equilibrium value is known as longitudinal relaxation, characterized by a time \(T_1\). The long \(T_1\) measured at a low temperature and high magnetic field is consistent with the theory of spin-orbit interaction assisted by electron-phonon scattering. The superposition between the spin-up and down components of a state along the field may also deteriorate in time, known as the transverse spin relaxation, characterized by a time \(T_2\). Contributions to finite \(T_1\) also limit \(T_2\). In measurement either of an ensemble of spins or of a single spin repeated in time, the inhomogeneous effect of the distribution of paths shortens the \(T_2\) time, denoted by \(T_{2}^\text{in}\). The remaining contribution to \(T_2\) is known as the pure decoherence mechanism.

Measured \(T_{2}^\text{in}\) in magnetic fields of several Teslas is of the order of 10 ns, much shorter than \(T_1\). It is, therefore, of a great deal of interest to quantum control to be familiar with the pure \(T_2\) and also to know if it can be prolonged.

Spin decoherence by spin-orbit interaction assisted by phonon scattering in quantum dots is suppressed at a temperature below a few Kelvins. The electron hyperfine interaction with nuclear spins in the dot is widely regarded as the dominant mechanism for decoherence. Until a few years ago, the theory of pure decoherence, due to nuclear spins, was dominated by spectral diffusion which requires a stochastic assumption. In recent years, the pure quantum theory has made progress. In the low field regime, the motion of the electron spin is complicated by longitudinal as well as transverse fluctuations and has been treated in the perturbation theory neglecting the nuclear spin-spin interaction. In the high field regime and with the inclusion of nuclear spin-spin interaction, the nonperturbative theory and numerical simulation of a few nuclear spins have been given.

Here we shall give a brief review of our quantum treatment of the electron spin decoherence dynamics with hyperfine coupling to a mesoscopic system of \(N\) mutually interacting nuclear spins in order to make the case that it is possible to control the electron spin decoherence.
coherence may be defined as follows. If the electron is initially in a quantum state $\beta_+|+\rangle + \beta_-|\rangle$, where $|\pm\rangle$ are the eigenstates in the high magnetic field, the system of the electron spin and all the nuclear spins in the dot may be regarded as in the product state

$$\langle \beta_+ | + \rangle \langle \beta_- | \rangle | J \rangle,$$

(4)

where $| J \rangle$ is the many-nuclear spin state. As a function of time $t$, the two nuclear terms associated with the two electron spin states evolve differently, yielding an entangled state of the form

$$\beta_+|J^*(t)\rangle + \beta_-|J^*(t)\rangle.$$

(5)

The electron spin coherence is measured by

$$L = \langle |J^*(t)\rangle |J^*(t)\rangle \rangle,$$

(6)

which is the off-diagonal coefficient of the reduced electron density matrix when the nuclear spin degrees of freedom are traced out. Decoherence occurs as the two nuclear spin paths diverge. On the time scale where the longitudinal relaxation is negligible, the Hamiltonian terms involving the electron spin flip may be transformed away leaving an effective Hamiltonian of the whole system to be in the form

$$\sum_{\pm} |\pm\rangle \tilde{H}\langle \pm|,$$

(7)

where $\tilde{H}$ are the nuclear spin Hamiltonian predicated on the electron spin state being $|\pm\rangle$. The coherence is simplified to

$$L = \langle |J^*(t)\rangle |J^*(t)\rangle \rangle.$$

(8)

For the nuclear spins in a mixed state, e.g., in a thermal ensemble, we can simply average over the initial nuclear states $J$. The form of the coherence in Eq. (8) shows that if the electron spin is rotated at time $\tau$, the nuclear spins would sense the electron spin up and down states to exchange roles so that $\tilde{H}_+$ is switched to $\tilde{H}_-$ and vice versa. Then at echo time $2\tau$, the coherence is

$$L = \langle |J^*(t)\rangle |J^*(t)\rangle \rangle.$$

(9)

Our theory consists in a direct attack of the many nuclear spin dynamics. We have found that the nuclear spin pair correlation is all the correlated motion needed under the condition that the number of nuclear spins $N$ is bounded above by the ratio of the hyperfine interaction to nuclear-nuclear dipole interaction, which is well satisfied for a dot of $N \approx 10^6$. A nonperturbative solution is then found.

Computation shows that the free induction decay time of the electron spin coherence is noticeably shorter than the decay time of the spin echo. This shows that the flip of the electron spin not only reverses the inhomogeneous effects but also partially the intrinsic dynamics. This is in agreement with the findings in NMR. More important is that quantum control of decoherence may be effected simply by a sequence of judiciously timed flips of the electron spin. The optical control is advantageous because of its speed relative to the decoherence time.

Amelioration of decoherence becomes important in any sustained quantum process. To put the method of restoring coherence by electron spin flip in perspective, we compare it with the other methods. The closest is dynamical decoupling, which uses a sequence of electron spin flips to erase all effects of the hyperfine coupling. Our use of the spin flips requires less frequent spin flips because the nuclear spin dynamics is slow whereas the reverse is true for fast bath dynamics. The methodology of designing sequences in dynamical decoupling works extremely well in our case also. The methods of decoherence-free subspace, which uses a number of physical TLS to represent one TLS, quantum error correction (for a review, see Ref. 8) and feedback control can be used in tandem with the spin-flip restoration of coherence.

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