Theory of Coherent Optical Control of
Exciton Spin Dynamics in a Semiconductor
Dot

Pochung Chen, C. Piermarocchi, and L.J. Sham

Department of Physics, University of California San Diego,
La Jolla, CA 92093-0319, USA

Abstract

We use the spin-polarized excitons in a single quantum dot to design optical controls for basic operations in quantum computing. We examine the ultrafast nonlinear optical processes required and use the coherent nonlinear optical responses to deduce if such processes are physically reasonable. The importance and construction of an entangled state of polarized exciton states in a single quantum dot is explained. We put our proposal in perspective with respect to a number of theoretical suggestions of utilizing the semiconductor quantum dots.

Key words: exciton, spin, nonlinear spectroscopy, quantum computing.

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Corresponding author: L.J. Sham, Department of Physics, University of California San Diego, 9500 Gilman Drive, La Jolla, CA 92093-0319, USA. Email: lsham@ucsd.edu, Phone: 858 534-3269, Fax: 858 534-2232.
1 Introduction

Recently there has been remarkable experimental progress in optical control of exciton spins in semiconductor heterostructure [1]. In particular, optical experiments on a single quantum dot have been demonstrated [2–4]. It is thus an opportune time to consider in theory what optical processes on the exciton spins in one or a few quantum dots can lead to potential device applications. To focus the discussion, we choose to use quantum computing [5,6] as a paradigm. This approach has the benefit of not only raising relevant basic issues of optical control of spins for quantum computing but it may also be relevant to more semiclassical devices. There are possible extensions of these fundamental quantum processes to more semiclassical results for an ensemble of dots. Examples include the extension to quantum dots of spin oscillation effects [7] or spin-dependent conditional processes [8] for an ensemble of excitons in a quantum well which depend on exciton-exciton interaction [9].

In this paper, we start with the simplest example, a model of two interacting excitons of opposite spins in a single quantum dot and investigate what is an appropriate quantum bit of information (qubit) and how, in principle, to address the bit, to form entangle states and to make a logic gate of two qubits, all with ultrafast coherent optical control. These are some of the basic optoelectronic issues involved in making a device for quantum computing. We shall also show how a coherent nonlinear spectrum may help in reaching these goals. We have chosen the simplest possible system to increase the likelihood of an experimental demonstration.

We need to put our work in perspective in relation with the large number of suggestions for the implementation of quantum computing in the semiconductor media. The earliest suggestions use either two electrons in two connected dots [10] or two electron spins in two dots with exchange coupling [11], controlled respectively by infrared light [12] or by magnetic field. Another class makes use of the nuclear spins or electron spins of the implanted impurities to make a transistor [13,14]. Yet another suggests the interaction of dots via the electromagnetic field in a cavity.
There is one on optical control of excitons transferring between neighboring dots [17]. The group of proposals closest to ours uses optical control of interacting excitons in quantum dots [18,19]. There are interesting differences among the three which will be discussed below. They also have in common with the approach of optically driven evolution of a multilevel atom [20], although the difficulties with atoms as briefly discussed in Ref. [10] are absent in the dots.

2 Basic requirements of quantum computing

First, we need two quantum states to represent the 0 and 1 of a qubit [5]. Examination of what operations on $n$ pairs of states are needed to carry out a specific quantum computation, such as the solution of the Deutsch problem [21], factorization [6] or a quantum search [22], then gives us a target to design the nonlinear optical operations on the quantum dots. In general, the operations fall into three groups of steps (see Figure 1). The first group prepares the system of $n$ qubits into an entangled (often maximally entangled) state. This can be carried out with separate operations on each qubit. The reason for the entanglement is that the next group of operations is effectively a transformation into another state. This group of operation is named the Oracle. For those of us familiar with Fortran, CALL SUBROUTINE may be a more transparent analogy. The advantage of the quantum computing over the classical counterpart lies in this transformation being sort of a parallel processor for different possibilities. If your input state to the Oracle involves a small portion of the qubits, you are leaving out a lot of possibilities and thus, are not likely to get your money’s worth in the answer by the Oracle. Basic to this second group of operations is the conditional dynamics. It has been shown that combinations of the control-not (C-NOT) gate between two qubits and single bit operations are sufficient to carry out any quantum algorithm [6]. We shall explain the C-NOT gate in terms of the quantum dot. The third group of operations disentangles the output state, which can then be measured.

Nuclear magnetic resonance experiments have been performed to implement the
solution of the Deutsch problem in its simplest form [23,24]. It is instructive to compare the procedure in the quantum dot with the NMR experiments.

3 Antiparallel-spin Excitons as Qubits

Consider first a simple model of a quantum dot which is a pill box with the shortest dimension along the growth axis (the $z$ axis). It houses the ground state $|0\rangle$, the spin-polarized heavy-hole exciton state $|+\rangle$ which can be excited by an electromagnetic wave of circular polarization $\sigma^+$, the heavy-hole exciton state $|-\rangle$ associated with the wave of circular polarization $\sigma^-$, and the antiparallel spin biexciton state $|+\rangle$, as shown in the inset of Figure 2. Both single exciton states have energy $E_X$ and the biexciton energy is $2E_X - \Delta$, where $\Delta$ is the biexciton binding energy.

We use the absence and presence of each single exciton as a qubit. Thus $|00\rangle$ which denotes the 0 state for both qubits corresponds to the ground state $|0\rangle$ and $|10\rangle$, $|01\rangle$ and $|11\rangle$ correspond to the physical states $|+\rangle$, $|-\rangle$ and $|+\rangle$.

A single qubit operation such as flipping the first bit from 0 to 1 requires a sum of two optical excitations $|0\rangle \rightarrow |+\rangle$, $|-\rangle \rightarrow |+\rangle$. This corresponds to the matrix transformation

$$X_1 = \begin{bmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 \end{bmatrix}.$$ (1)

To accomplish this, we might use two sharp resonance excitations with frequencies corresponding to $E_X$ and $E_X - \Delta$. Conceptually, this requires two Rabi flops [25] or, to borrow the spin language [26], a $\pi-$pulse of each frequency.

A C-NOT operation changes, say, the second qubit (the target) depending on the
state of the first qubit (the control). If the first is 0, the second is unchanged and if the first is 1, the second bit is flipped. The matrix transformation is

\[
X_1 = \begin{bmatrix}
1 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & 0 & 1
\end{bmatrix}.
\] (2)

This is accomplished by a single \(\pi\)–pulse, resulting in \(|+\rangle \leftrightarrow |+\rangle\).

The ability to address resonantly only the transition between the exciton and biexciton states without affecting the transition between the ground state and the single exciton state is rooted in the strong interaction between two excitons. In the limit of no interaction between two excitons, a resonant excitation or rotation of one pair of states would give rise to an equal amount on the other pair \([27]\). There would then be no possibility of conditional exciton spin dynamics.

The proposal of resonance addressing of two levels for a C-NOT operation in a two-qubit system with sufficiently strong interaction was first put forward by Barenco et al. \([10]\) for intrasubband transitions for electrons in the conduction band of two coupled dots. Such operations in the optical range is, in the current state of the art, simpler than in the infrared range needed for the intrasubband transitions. The working principle for two strongly interacting qubits is different from that for two weakly interacting ones as is used in NMR \([23,24]\). In the latter, the target spin is allowed to precess under the spin-spin interaction due to the control spin. The time it takes is much longer than the Rabi oscillation time scale for addressing each spin individually. The NMR system can afford such long times since they are still shorter than the dephasing time of the spins in the millisecond range. By contrast, the optical dephasing time is about 40 ps \([3]\). This can only be accommodated by a judicious design of a sequence of femtosecond pulses.

The fundamental experimental requirement for being able to carry out one and two
qubit operations is to demonstrate Rabi flops in a quantum dot. Rabi oscillation has long been established for the spin [26] and the atomic system [25] but has not been established experimentally in a quantum dot. In the quantum well, Rabi oscillations are claimed to have been seen [28]. However, since it is possible to excite a large number of extended excitons, a likely scenario is the oscillation of the coherent exciton density or the average polarization, as indicated by the theoretical analysis of the experiment in the same paper. It is driven by the electric field, unlike the collective density oscillation of the exciton gas. [29]. Thus, it might be a coherent mixture of the Rabi rotations between pairs of multiexcitonic states with roughly equal transition frequencies. If so, this finding in a quantum well could be a harbinger of the Rabi oscillations in a quantum dot when the excitonic states become discrete.

4 Coherent Nonlinear Spectra

Figure 2 shows the coherent nonlinear spectra for two cw laser beams, with a strong pump beam with $\sigma-$ circular polarization and with a weak probe beam with $\sigma+$ polarization. The top curve is the linear absorption spectrum of the $\sigma+$ probe when the pump is turned off. The others are the corresponding spectra when the $\sigma-$ pump beam is focused at the exciton resonance and two frequencies below resonance as indicated in energy units on the right hand margin. The damping constant is taken to be 15 $\mu$eV, approximately that of the measured 20 $\mu$eV [3]. The pump beam intensity corresponds to a Rabi energy of 0.05 meV. It is taken to be much stronger than was used in the nonlinear measurement [3] in order to produce a discernable Rabi splitting as shown in the lower three curves where the resonance exciton line in the linear absorption spectrum is now split into two lines about twice the Rabi energy apart. The additional doublet 1 meV below the exciton line is the Rabi splitting of the exciton to biexciton transition, $|-\rangle \rightarrow |+\rangle$. Our use of the 1 meV biexciton binding energy is conservative compared with the 4 meV found for the dot used by Steel’s group [30].

The coherent cross-polarized pump and probe spectroscopy in a dot represents the
mesoscopic analogous of the Autler-Townes spectroscopy in atomic systems. Therefore, the presence of the four peaks in the nonlinear response can be interpreted in a dressed exciton picture, similar to the well known dressed-atom picture [25]. The strong $\sigma_-$ pump produces the Rabi oscillation between the two states $|0\rangle \otimes |N\sigma_-\rangle$ and $|-\rangle \otimes |(N-1)\sigma_-\rangle$, where $|N\sigma_-\rangle$ represent the state of the radiation field with $N$ photons with $\sigma_-$ polarization. Due to this strong coupling two dressed states result that can be written in the form:

$$|\text{lower}\rangle = \cos\theta |0\rangle \otimes |N\sigma_-\rangle - \sin\theta |-\rangle \otimes |(N-1)\sigma_-\rangle$$

$$|\text{upper}\rangle = \sin\theta |0\rangle \otimes |N\sigma_-\rangle + \cos\theta |-\rangle \otimes |(N-1)\sigma_-\rangle ,$$

with the angle $\theta$ defined as

$$\tan(2\theta) = -\frac{\sqrt{(\Omega)^2 + (\delta)^2}}{\delta} ,$$

$\delta = \omega_{\text{pump}} - E_X/\hbar$ being the detuning frequency. The energy of the lower and upper states are split by $2\hbar \sqrt{(\Omega)^2 + (\delta)^2}$, where $\hbar\Omega$ is the Rabi energy of the pump field.

The two peaks at the excitonic energy in one of the nonlinear spectra are due to transitions from the $|\text{lower}\rangle \rightarrow |+,N\sigma_-\rangle$ and $|\text{upper}\rangle \rightarrow |+,N\sigma_-\rangle$ by absorption of a $\sigma_+$ photon from the probe. The two peaks at the minus biexciton binding energy originate from transitions $|\text{lower}\rangle \rightarrow |+,-,(N-1)\sigma_-\rangle$ and $|\text{upper}\rangle \rightarrow |+,-,(N-1)\sigma_-\rangle$. The characteristic shape of the doublet in presence of detuning can also be interpreted within the dressed exciton picture. At resonance pumping, the Rabi oscillation gives a fifty-fifty probability of having the system in the lower or upper states, giving equal intensity to the two peaks of the Rabi splitting. On the other hand, when the pump has negative detuning, the full inversion of the $|0\rangle$ and $|-\rangle$ levels is not realized, and the system will more likely be in the lower state, which is the one with a bigger $|0\rangle$ component. This gives a bigger intensity for the peak corresponding to the $|\text{lower}\rangle \rightarrow |+,N\sigma_-\rangle$ transition (the higher energy peak in the doublet). In presence of a positive detuning the situation is reversed: the state $|\text{lower}\rangle$ is the one at higher energy and the lower energy peak in the Rabi doublet dominates. (This case is not shown in the figure.)
The coherent nonlinear spectra are useful in designing the laser pulses for the time
dependent operations to carry out the processes whose basic units are described in
the last section. Because of space limitation, we shall publish the time sequence work
in another paper [31]. The spectra give the frequencies for the resonance operations.
The Rabi splitting would give an indication of sufficient electric field strength for
Rabi oscillations. The pulse width and biexciton binding energy would provide the
limitation of the pulse width to be used in the qubit operations within the confines
of the dephasing time and of the error caused by affecting levels not intended to be
in the operation.

5 An Entangled Exciton State

We have explained how an entangled exciton state is an important asset in quantum
computing. It is also an asset in quantum information in general, especially in
cryptography and teleportation [5]. In the scheme for general entanglement in the
last section, we require the Rabi oscillation. The collaboration of the Steel group
and the Gammon group has experimentally achieved the entanglement between the
two single exciton states $|+\rangle$ and $|−\rangle$ [32] without Rabi oscillations. In the pump and
probe spectroscopy described in the last section, the two laser beams of opposite
circular polarizations are phase-locked and kept at sufficiently low intensity to be
in the third order regime. A single exciton state produced by one of the beams can
be coupled to the single exciton state of opposite polarization by the second order
process of de-excitation by the same beam to the ground state and excitation by
the other beam to the second exciton state. It was found experimentally that the
dehphasing is due mostly to recombination with negligible contribution from pure
dehphasing mechanisms. This leads to a very stable entangled state within the 50 ps
time scale. This use of the spin coherence between two excitons (also called the
Zeeman coherence [33]) is an interesting contrast to the optical coherence used in
the Rabi oscillations. Both types of coherence exist in spin-polarized excitons [34].
The resultant state

\[ |\Psi\rangle = C_0|0\rangle + C_+|+\rangle + C_-|-\rangle \]  

(6)

still involves the ground state with the coefficients \(|C_\pm|\) of the order 0.3. It contains negligible amount of the biexciton state because of the resonance condition on the exciton energy. This is a strong interaction effect in the sense explained in Sec. 3. The linear combination of the two exciton states is an entanglement for one of two reasons below. In our two-qubit system, this is the combination \(|10\rangle\) and \(|01\rangle\). Alternatively, if the exciton is regarded as a pair of conduction electron and valence hole, the linear combination of two excitons of opposite polarization is an EPR state for the electron and hole, each capable of one of two spin states.

The existence of the entangled state is deduced from the measured coherent pump and probe spectra. A Tesla field is used to split the two spin exciton states. As the pump beam of one circular polarization is detuned below the exciton resonance of that polarization, the probe spectra of the exciton of opposite polarization acquired a change in line shape, indicating an interference effect between the third order transition involving the same exciton state and the third order process described above from the ground state to the exciton of the pump polarization and then to the exciton of opposite polarization via the ground state. The interaction between the two excitons which form the biexciton states prevents a similar third order process via the biexciton state to cancel the one via the ground state. The cancellation would be complete in the absence of interaction [32,27] and the pump would have no effect on the probe beam.

6 Discussions

We have given a theoretical prescription of the optical processes to build the basic operations for quantum computing in a two qubit system using two excitons of opposite spins in a single quantum dot. The advantage lies in the simplicity of the theory which could lead to a certain degree of simplicity for the experiment.
The proposal is very similar to that of Troiani et al. [18] and of Biolatti et al. [19] in the use of optical control in quantum dots. It differs from both in using excitons of opposite spins. At the two qubit level, our proposal is simpler since any interference between light of opposite polarization must come from the exciton-exciton interaction. Undesired interference effects between two color light beams of the same polarization via noninteracting sources may need to be ameliorated. Nonetheless, this is an important problem because we would encounter it, if we wish to increase the number of qubits in a single dot.

This brings us to the question of scaling. Clearly, while we could increase the number of qubits in a single quantum dot, the process would terminate rather quickly. We could consider two avenues to scale our two qubit systems to many dots, either by interdot interaction or by using the cavity as a data bus [16,15]. The exciton interaction can be strong enough for the biexciton to be seen directly in the interaction with the cavity mode [35]. The required interaction between dots needs to be as strong as the intradot interaction (a few meV’s). Thus a scheme of enhancing the interdot interaction, such as that of Biolatti et al. [19], might be followed.

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Figure Captions

Fig. 1. Circuit diagram for a generic quantum computation. The three qubit number controls via a function in the subroutine the change in the target qubit. The squares denote single qubit operations. The Oracle involves at least one C-NOT operation.

Fig. 2. Coherent nonlinear spectra. The vertical axis is the intensity of absorbed light in arbitrary units. The horizontal axis is $E_{probe}$, the frequency of the probe beam times $\hbar$ measured from the exciton energy. The damping constant is $\Gamma = 15 \mu$eV. The biexciton binding energy is 1 meV. The inset depicts the energy levels in a dot.
CALL SUBROUTINE

Entanglement
Control qubits

Disentanglement

Measurement
Read-out

ORACLE

Target qubit
\[ E_{\text{probe}} - E_x \text{(meV)} \]

\[ \Gamma_{-2} \]

\[ \Gamma_{-5} \]

\[ \Gamma \]

\[ -1.5 \]

\[ -1 \]

\[ -0.5 \]

\[ 0 \]

\[ 0.5 \]

\[ |0> \]

\[ |-> \]

\[ |+-> \]

\[ 2E_x \]

\[ \text{pump} \]

\[ \text{probe} \]

\[ \text{pump + probe} \]

\[ \text{intensity} \]