Optical Methods in Quantum Information Processing With Quantum Dots

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Most physical realizations of quantum technologies could not be possible without optical methods and their applications in initialization, control and readout, among many others. Due to the relevant quantum system scales of order 10-100 nm, laser light of frequencies within the visible spectrum can be used. Here, quantum dot systems are the focus, as the phenomenology translates well to other optically controlled quantum systems. In semiconductor quantum dots, the nature of semiconductor band gap allows for engineering of finely tuned, distinct Raman transitions. Here, optical methods for initialization, control and readout will be presented, followed by discussion.

INTRODUCTION

There have been remarkable developments in optics and single photon control over the past 100+ years, allowing scientists to precisely characterize light by its discrete wavelength and devise spatially and temporally coherent light sources called lasers. Subsequently, optics-enabled quantum technologies have brought a multitude of hitherto unknown opportunities such as quantum sensing, communication and computation, all realized with quantum bits (qubits) and broadly categorized as quantum information processing (QIP). Generally speaking, a system is considered quantum if its properties are dominated by quantum effects such as Heisenberg uncertainty and superposition principles. Examples of such systems are single atoms, single vacancy centers or single photons, but even some many-particle systems such as group of electrons in a superconducting state, many-million atom states in Bose-Einstein condensate, or a collection of photons in a laser [1]. As a consequence of being quantum, those systems obey the Heisenberg uncertainty principle, which makes their measurements probabilistic in nature. On the contrary, the preparation, evolution and control of quantum systems is fully deterministic, with only noise errors caused by technical implementation imperfections.

Because of the relevant scales, optical methods have a variety of direct and indirect applications in QIP. One indirect example is photoluminescence spectroscopy, which lets one characterize the transition rates and functionality of nitrogen defects in diamond known as nitrogen-vacancy (NV) centers, widely researched for QIP applications [2]. Other examples are optical fibers, waveguides and linear optics circuits used for remote entanglement generation among many others, which show great promise for the future large-scale QIP realizations [3]. This papers explores direct applications of optical methods in preparation, control and measurement of electron spin 1/2 states in quantum dots (very small semiconductor particles). An electron in a quantum dot, confined by electrodes on a chip (Fig. 1) is an example of a reliable and widely-researched spin qubit. The qubit is typically prepared and read out into one of energy non-degenerate "spin-up" and "spin-down" eigenstates, but exists in the superposition of the two throughout the manipulation stage. These three processes - initialization, manipulation and readout - can all be accomplished using optical methods. In truth, many engineering and nanoscale manufacturing improvements must be made before useful, many-qubit quantum devices transpire, but the necessary ingredients for QIP with spin 1/2 qubits have been demonstrated in a variety of ways.

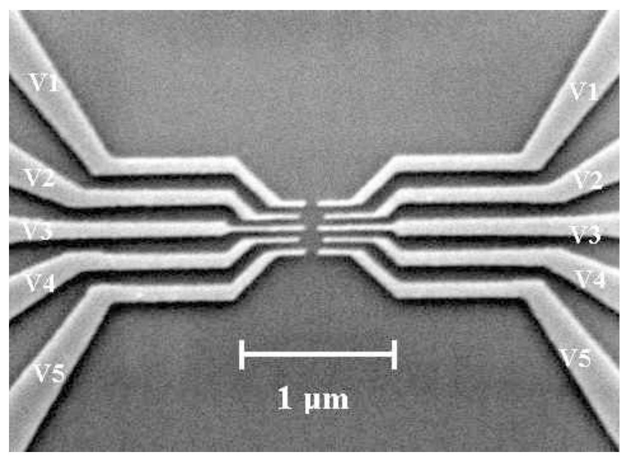

FIG. 1. Adopted from Jeong [4], electron scanning microscope image of the experimental setup. Two quantum dots on GaAs/AlGaAs heterostructure containing a 2D electron gas 80nm below the surface (electron reservoir). The dots are voltage-confined by 10 independently tunable surface gate electrodes.

According to DiVincenzo's seven criteria for scalable quantum computation with spin states, well defined qubits must be reliably initialized to a particular state $|0\rangle$ (usually the lower energy spin-up), stay coherent for much longer than the manipulation (by gate operations) time and be selectively read out on demand with high fidelity [5]. Here, quantum dot spin systems will be the focus, as the phenomenology translates to many other optically active solid-state spin systems, such as energy-state-based ion qubit or nuclear spin in NV center. Optical methods for initialization of spin state will be discussed first. Then, schemes for optical manipulation and observation on spin dynamics, followed by optical readout techniques will be presented. Lastly, the discussion section offers insights into the limitations and future of optical QIP.

OPTICAL SPIN INITIALIZATION

The fundamentals of optical spin initialization were first studied in bulk semiconductors in the 1960s and 70s [6]. The conceptual understanding rests on the interband transitions in semiconductors, where spin-polarized (spin aligned in a particular direction) electrons and holes (absence of electron) are selectively excited into the conduction and valence bands, respectively. A schematic of the band structure of the cheap and commonly used for QIP applications gallium arsenide (GaAs) semiconductor, adopted from Gywat [7] is presented in Fig. 2.

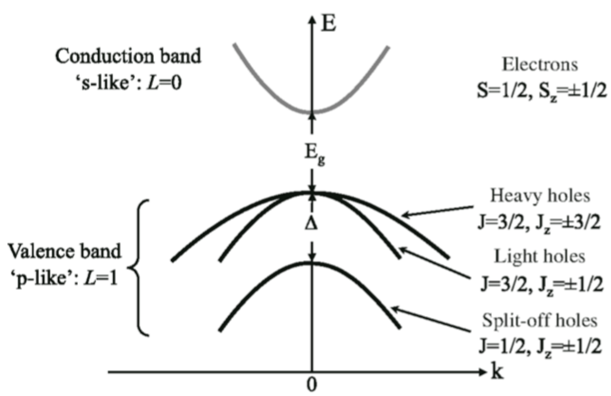

FIG. 2. Adopted from Gywat [7]. Band structure of GaAs. E_g is the band gap and Δ is the spin-orbit energy.

With reference to Fig. 2, possible optical transitions are from the heavy and the light hole bands (which are due to effective mass) and the split-off band (Δ is due to spin-orbit interaction), but the energy of light is usually chosen to not excite carriers from the split-off band. Hence, only the hole transitions play a role in electron excitation into the conduction band, which limits the number of possible transitions. In practice, circularly polarized light is used, because absorption of circularly polarized photon causes an angular momentum change of $\pm\hbar$ elsewhere and electron-hole pair is then only be created by specific transitions. The available transitions are represented in Fig. 3., adopted from Gywat [7].

Because heavy hole transition is three times more likely than light hole transition, circularly polarized light that excites both yields an ensemble with an average

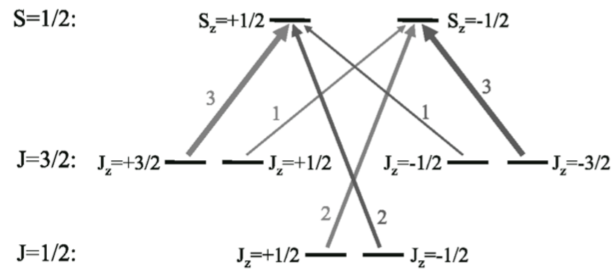

FIG. 3. Adopted from Gywat [7]. Transitions involving circularly polarized light from light and heavy hole bands into the conduction band. The numbers indicate the relative strengths of each transition.

spin polarization of 50% (total angular momentum J indeterminacy). However, introduction of strain into the lattice can lift the heavy-light hole degeneracy and by tuning the energy of laser light to only heavy or only light hole transition 100% spin polarization is, in theory, obtained [7]. In an undoped semiconductor the spin polarized electron and hole recombine within less than a nanosecond and emit a photon with polarization corresponding to the spin polarization. However, in doped semiconductors the recombining can be arranged with another optically polarized dopant electron. In that case, the spin polarized-electrons are left in the conduction band and can further controlled and used for QIP. This method is termed optical (here, laser) pumping and in practice, involves exciting electrons in a quantum dot, with an electron reservoir available to recombine with the holes [4].

To better understand the spin-selective transitions one needs to recall the Pauli exclusion principle. In a quantum dot containing a spin-up electron, transitions to the spin-down state may occur, but transitions to spin-up state are forbidden. Hence, by arranging two distinct trion states (localized excitation of three charged quasiparticles - in ref. to Fig. 4, negative trion is two electrons and one hole) spin-selective transition to only one of them will be observed for a given helicity of light. This conceptual understanding can be extended to drive coherent oscillations between the spin states, as outlined in the 'Optical Control' section. An picture representing transitions for the single electron state to the negatively charged trion is presented in Fig. 4 [7].

In practice, magnetic field large enough to suppress the nuclear induced spin flips needs to be present to lift the energy degeneracy of spin up and down states. By additionally laser cooling the excited electrons, the desired spin state was shown to be initialized with fidelity of 99.8%, as inferred using a differential absorption technique with Pauli spin blockade [8].

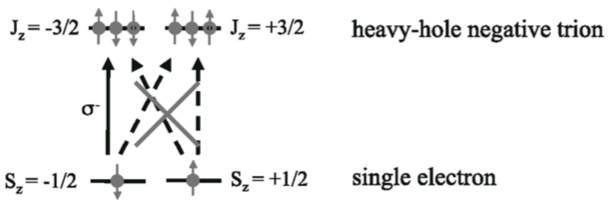

FIG. 4. Adopted from Gywat [7]. Spin state transition involving left circularly polarized light (σ^-). The dashed arrow transitions are not allowed for the depicted system. Only the solid arrow transition is allowed and accomplished with σ^- polarized light due to Pauli exclusion principle or $\pm\hbar$ change in total angular momentum.

In short, optical spin state initialization in quantum dots can be made straight-forward with exploitation of spin dependent optical selection rules commonly present in semiconductors. Those pumped electrons have been engineered to have spin coherence times (T_2^*) in hundreds of nanoseconds [9], long enough for many picosecond (ps) light pulses to be applied for manipulation. From a QIP standpoint, the initialization into a spin eigenstate must be followed by spin rotations (around the Bloch sphere), which is the focus on the next section.

OPTICAL CONTROL

Control of solid-state spin has been best accomplished with ultrafast (ps) optical pulses[10–13]. Though, electrical control has also been demonstrated [14, 15], such gate operation times are in the nanosecond range, which limits the number of possible operations within the coherence time. The light pulse manipulation technique can be described in terms of the optical (or ac) Stark effect [16] or avoided crossing between excitons and photons [17], but their phenomenologies follow that of optical pulses very closely and will not be presented in detail. Optical Stark effect can been used to control orbital coherence in a quantum dot [18] or observe ensemble spin manipulation in a quantum well [19]. Instead of Raman scattering, it relies on the magneto-optical Kerr effect to probe the spin [20]. Generally, with addition to manipulation, the spin state can be monitored and temporally resolved using various techniques discussed in 'Observing spin dynamics' subsection.

Coherent control of spin can be achieved by driving the qubit with timed ultrafast laser pulses at resonant frequency ω (where energy difference between two spin states is $\Delta E = \hbar\omega$). In that scenario, the axis of qubit rotation is determined by the arrival time of the pulse with respect to its oscillation period. This necessitates a large magnetic field (B_{ext} up to several Tesla) to increase spin precession frequency upwards of 10GHz. Because of the pulse time scales, an arbitrary gate operation is then completed within a single Larmor precession period. This spin rotation scheme relies on stimulated Raman transition (inelastic scattering of monochromatic light) through two independent Λ systems (named for their shape on a diagram like Fig. 4), whereby the laser photon falls on the electron, interacts with its dipole moment to change the spin state and gets emitted at lower energy. Fig. 5 shows the experimental manipulation setup used by Press [21].

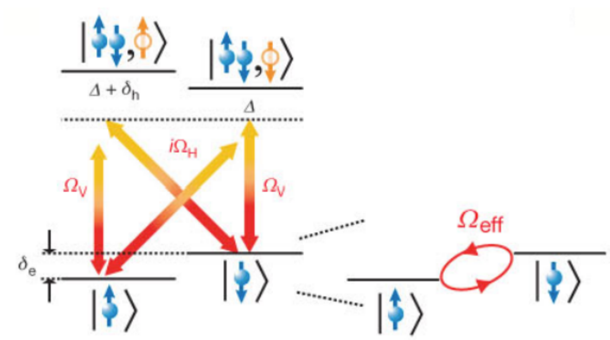

FIG. 5. Adopted from Press [21]. Picosecond pulse to an electron in a quantum dot. The electron spin state are split by B_{ext}= 7T aligned upward in the diagram, providing Larmor frequency of 26.3 GHz.

The 2008 experiment with InGaAs quantum dots by Press [21] was the first to demonstrate these mechanisms experimentally, and it is worth a closer analysis. Here, the lowest interband transitions are two trion states - spin singlet electron pair and unpaired heavy hole - split by frequency δ_h. Each trion state forms an independent Λ system with two metastable states (occupied most of the time) $|\uparrow\rangle$ and $|\downarrow\rangle$, with vertical and diagonal transitions coupled to vertically or horizontally polarized light (Ω_V,Ω_H), $\frac{\pi}{2}$ out of phase with each other. The transitions have Rabi frequencies $\Omega_V = \mu E_V/\hbar$, $\Omega_H = \mu E_H/\hbar$, where μ is the dipole strength and E_V, E_H are complex field amplitudes of the rotation pulse in polarization basis. Shining circularly polarized laser light ensures that the probability amplitudes from the two Λ systems add constructively and large detuning Δ ensures no undesired population of the excited states. Therefore, a single pulse will coherently drive the transition between $|\uparrow\rangle$ and $|\downarrow\rangle$ by a stimulated Raman transition. In that way, controlled Rabi oscillations are accomplished with effective Rabi frequency of $\Omega_{eff} \approx \Omega_H\Omega_V/\Delta$, and can be confirmed by scattered photon count rate (Fig. 6a) [21].

The above figures of Rabi oscillations show the possibility of rotating a qubit about a single axis by an arbitrary angle. However, full Bloch sphere control requires rotation about a second axis. Conveniently, Larmor precession of the spin along z axis accomplishes that task. Hence, through appropriate timing of a

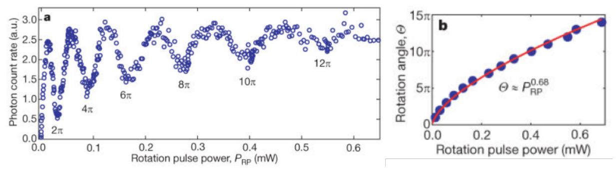

FIG. 6. Adopted from Press [21]. **a**. Rabi oscillations between spin states evident by oscillating photon count rate. A photodetector counts single photons emitted by Raman transition. Here, the photon count rate is proportional to the instantaneous population in $|\downarrow\rangle$. **b**. The rotation angle as a function of pulse power. A power law function is a good fit to the dependence.

pulse the spin is simultaneously rotated about two axes, and the rotation can be investigated using Ramsey interferometry (Fig. 7) [21].

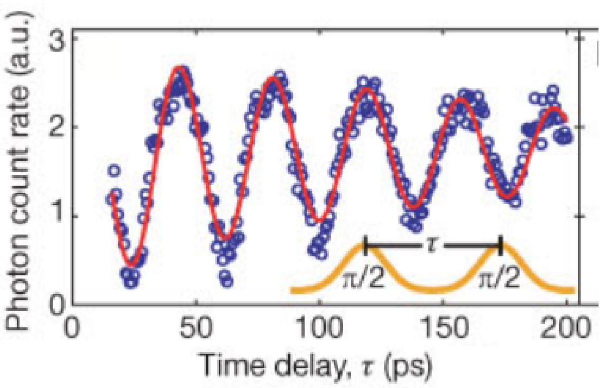

FIG. 7. Adopted from Press [21]. Demonstration of successful spin state manipulation with described methods. Ramsey interferometry for a $\pi/2$ pulse rotating the spin to perpendicular to the magnetic field, which then precesses for time τ before second $\pi/2$ pulse is applied.

Observing spin dynamics

Spin dynamics of a single quantum dot electronic spin can be resolved in real time using at least three approaches: the Hanle Effect, and Faraday and Kerr rotation spectroscopies. A good experimental example of the first can be found in Bracker [22]. In the experiment, electrons and hole are excited into the quantum well with circularly polarized laser. Then, the quantum dot is characterized by photoluminescence spectroscopy, which lets one identify three distinct lines corresponding to different excitation intensities to a neutral (X°), negatively charged (X^{-}) and positively charged excitons (X^{+}) (Fig. 8a). The degree of spin polarization can be inferred by monitoring the polarization of these three luminescence lines while sweeping an applied magnetic field perpendicular to spin polarization axis. An image from Bracker [22] of photoluminescence from a single quantum dot is presented in Fig. 8a, and Hanle effect measurements are shown in Fig. 8b.

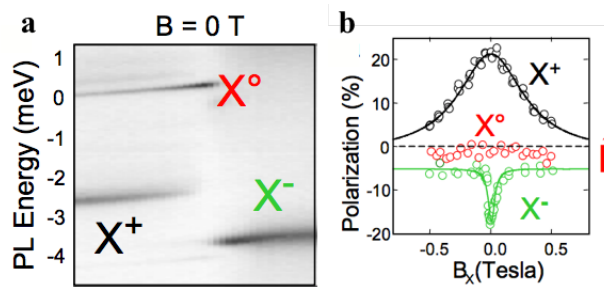

FIG. 8. Adopted from Bracker [22]. **a**. Photoluminescence of a quantum dot as a function of applied bias with the three excitation intensity lines. **b**. Hanle curves from the three transitions observed in the quantum dot.

An example application of the Time-Resolved Faraday Rotation Spectroscopy (TRFR) for spin dynamics measurement for ensemble of electronic spins in CdSe quantum dots is found in Gupta [19]. The measurement procedure occurs in a magneto-optical cryostat at temperatures of several Kelvin. The measurement relies on the polarization axis changes of linearly polarized light during transmission though matter with different refractive indices for right and left-handed circularly polarized light. This effect is known as Faraday rotation. An analogous effect for light reflecting off the surface is known as Kerr rotation and is a basis of Kerr Rotation Spectroscopy, discussed in the next paragraph [7]. In TRFR, the pump beam has circular polarization to excite spin polarized electrons and hole and the linearly polarized probe beam is used to measure Faraday rotation. After passing through the sample, the pump beam is blocked and the now rotated polarization probe beam is directed into the collection path. The polarization rotations are on order of 1mrad or less and can be used to infer the spin polarization of the sample [7]. An example of TRFR measurement result is presented in Fig. 9 [19].

In the TRFR example above, the measurements have been performed on large ensembles of spins. However, measurements on single quantum dot spins are also possible using both TRFR and Spin Kerr Rotation (SKR) spectroscopy. In that case the laser has to be focused tightly on the dot as the signal from a single dot is spectrally concentrated in a small region. Similarly to TRFR, the rotation angle in SKR is determined by the difference of the dynamic dielectric response functions for right and left-handed circularly polarized light [7].

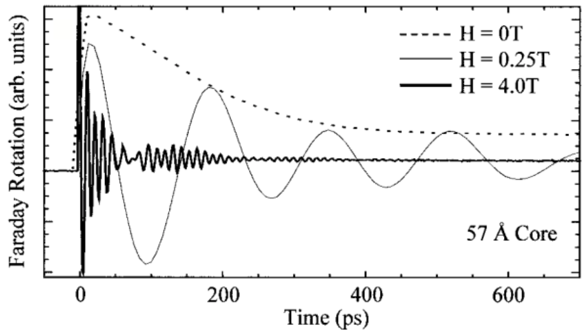

FIG. 9. Adopted from Gupta [19]. Field-dependent TRFR at $T = 6K$. As magnetic field is made stronger, TRFR becomes shorter due to inhomogeneous dephasing. At high magnetic field presence of multiple precession frequencies can be inferred.

OPTICAL SPIN READOUT

The same spin dependent optical selection rules used for initialization can also be exploited for readout of a quantum state by the polarization of light emitted upon recombination of an electron and a hole (Fig. 4). In the presence of spin-orbit interaction, the polarization of outgoing luminescence can often yield information about the dot spin state. The apparatus for a simple scheme measuring the sample spin polarization is depicted in Fig. 10 [7].

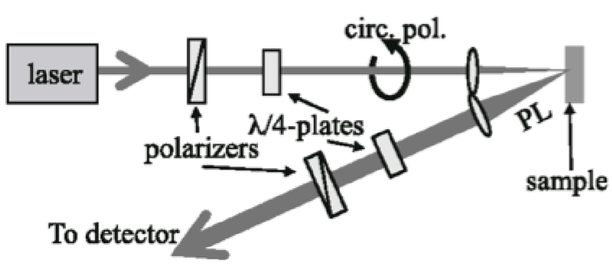

FIG. 10. Adopted from Gywat [7]. Apparatus for measuring polarized photoluminescence (PL) as described in the text.

Naturally, circularly polarized light is first focused onto the sample to pump spin polarized electrons and holes, as discussed in 'Optical Spin Initialization' section. The polarization is achieved by passing the laser light through a linear polarizer followed by a quarter wave plate, tilted 45° to the polarizer axis. This yields either a left or right circularly polarized beam, depending if the tilt is plus or minus 45°. Once it interacts with the sample, this excitation beam is blocked to prevent it from entering the detector.

The positioning of the detector is irrelevant under the assumption that luminescence is emitted isotropically. However, since the spins will be polarized along the pump beam axis, one can measure the polarization along the same axis by making the excitation and collection paths as parallel as possible. The luminescence, whose source is of course due to Raman transitions, is then collected and collimated by a lens focus on the excitation point and one circular polarization mode is made linearly polarized by the reverse of the process explained above. The intensity of linearly polarized light is then measured at the detector. To measure the relative intensities of both circularly polarized modes (corresponding to spin up or down states) the linear polarized can be rotated by 90° or alternatively, the quarter wave plate may be rotated 90° to switch the helicity of optical excitation.

DISCUSSION

One drawback of the discussed laser pumping for spin initialization method is that it necessarily has to be performed in a potentially undesirable magnetic field, as to suppress the hyperfine interaction and create energy non-degeneracy between the electron spin-up and spin-down states. This requirement has been overcome experimentally by looking at hole spin in the valence band as opposed to electron spin in the conduction band. Due to their p-like symmetry, holes have nodes at the lattice ions, resulting in very little coupling between the hole and nuclear spins. The pumping scheme described above works at zero magnetic field and yields 99% initialization fidelities [23]. The lack of nuclear spin interaction makes holes interesting for their potentially longer spin dephasing times.

There exists a multitude of important and successful initialization, control and measurement techniques that could simply not be mentioned in a few-page-long overview of the subject. For example, the single-shot quantum nondemolition readout schemes are of paramount importance for the future of QIP. Those methods do not only measure the spin state with great fidelity, but they are also crucial for future implementation of quantum error correction protocols in a quantum register [24]. Other interesting and highly sophisticated measurement mechanism rely on the use of ancillary qubits, real-time feedback and probabilistic analysis [25].

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