Optoelectronic manipulation of single spins in semiconductors

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ABSTRACT

In systems with sizable spin-orbit interaction intense optical illumination or an electric field can generate an effective “pseudomagnetic field” which replaces a true applied magnetic field for the efficient and rapid manipulation of spins. The theoretical characteristics of optically-induced spin precession in self-assembled quantum dots will be described, along with the potential for manipulating spins bound to donors and acceptors with electric fields.

Keywords: spin manipulation, spintronics, optoelectronics, Stark shift, quantum dot, dopant

1. INTRODUCTION

The stability of coherent spin states in solids, and the proposal of methods to encode and manipulate quantum information using spin states has motivated extensive exploration into scalable methods of manipulating single spins in solids. Magnetic fields oscillating with the appropriate frequencies to manipulate these spins have wavelengths orders of magnitude larger than desirable spin-spin separations for scalable devices in solids, so new methods are required to apply these oscillating fields to individual spins, and not to nearby neighbors. Both optical illumination and electric fields offer potential routes to such local addressing, but neither couple directly to the spin. Fortunately in solids the spin-orbit interaction provides a method of coupling optical illumination or applied electric fields to the spin degrees of freedom of a material. Recent progress on manipulating spins optically in quantum dots as well as dopant spins with electric fields will be described.

An intense pulse of light, detuned from an optical transition, will shift the electronic energies of the transition. If the light is circularly polarized, so that the oscillator strength of the electronic transitions depends on the spin of the electron and hole, then the shift of the electronic energies will depend on the spin. This spin-dependent shift behaves as an effective magnetic field to precess a spin in the electronic states of the quantum dot. As the spin-dependent shift only occurs when the optical field is on, this effective magnetic field can be focused to a spot size corresponding to the light wavelength, and can be temporally constrained to the duration of the optical pulse. The response of a spin confined to a self-assembled quantum dot grown by molecular beam epitaxy has been calculated, and the dependence of the pseudomagnetic fields on the dot size and composition has been found. Error rates for such dots have also been calculated, and the expected error rate is smaller than $10^{-6}$, which is sufficient for quantum computation error correction algorithms to succeed.

The use of electric fields to perform spin manipulation of electrons confined to donors or of a Mn-hole acceptor complex will also be described. All possible spin operations can be performed using electric fields alone on a nonzero integer spin, whereas manipulation of spin-1/2 electronic ground states requires at least a static applied magnetic field. Control of an individual spin-1/2 state with local electric fields can be achieved by changing the magnitude of the Landé $g$ tensor to bring spins into resonance with an extended AC magnetic field, moving spins in a fringe-field or a hyperfine gradient, modulating zero-field spin splittings, or with $g$-tensor modulation resonance ($g$-TMR). $g$-TMR uses the electric-field dependence of the Landé $g$ tensor anisotropy to manipulate the spin, and so does not require microwave magnetic fields or nanoscale magnetic materials or nuclear polarization gradients. Although $g$-TMR works by changing the orbital character of the wave function with an electric field, and thereby indirectly influencing the spin through the
spin-orbit interaction, it does not require zero-field spin splittings (so g-TMR could be performed in a silicon or diamond host).

Shallow donors have cubically-symmetric $g$ tensors in the absence of an electric field, so any $g$-tensor anisotropy is introduced through the applied electric field. Self-assembled quantum dots grown by molecular beam epitaxy, by contrast, have highly asymmetric $g$ tensors in the absence of an electric field. The built-in anisotropy also makes them very sensitive to applied electric fields.\textsuperscript{20} As the $g$ tensors of electronic spins bound to donors are isotropic in the absence of an electric field, the $g$ tensors depend \textit{nonlinearly} on applied electric and magnetic fields, and substantial $g$ tensor anisotropy and rapid spin manipulation can be achieved for a hydrogenic donor state. The most rapid Rabi oscillations are found at subharmonics of the Larmor frequency rather than the fundamental, which permits rapid spin manipulation using AC electric fields with frequencies far below the Larmor frequency.

As an example of an integer spin system we consider Mn in GaAs. The core spin $5/2$ of the Mn combines with a bound heavy hole state ($j = 3/2$) to form a $J = 1$ ground state complex. Due to the inversion asymmetry of the GaAs host lattice, the spin states can be split linearly with an applied electric field. The strength of this splitting is substantial, corresponding to a 1 Tesla field for a 40 kV/cm applied electric field.

2. ULTRAFAST OPTICAL SPIN MANIPULATION IN QUANTUM DOTS

The AC Stark effect with unpolarized light is a nonlinear effect whereby light with photon energy tuned near to the absorption transition between two states induces a level repulsion between the two states.\textsuperscript{6, 7} In perturbation theory\textsuperscript{8} this level repulsion depends on $I f/\delta$, where $I$ is the light intensity, $f$ is the oscillator strength of the transition, and $\delta$ is the detuning of the photon energy from the transition energy (see Fig. 1). When circularly polarized light illuminates a transition from the first valence state pair to the first conduction state pair in a crystal with spin-orbit interaction the oscillator strengths of the transitions differ considerably; this is the source of the effective spin splitting of the conduction state pair. We will examine Stark splittings in CdSe and InAs colloidal quantum dots (CQD), as well as in InAs/GaAs and InAs/InP self assembled quantum dots (SAQD). This set, while not exhaustive, spans a broad range of parameters\textsuperscript{21} by including unstrained spherical and strained lens-like dots with widely varying bandgaps.

![Figure 1. (Left) Schematic of optical process leading to optical spin manipulation. In the absence of the optical field the spin states are doubly degenerate. Optical illumination below the band gap (detuned) causes an effective spin-dependent shift of the electronic states, which can be detected in polarization-dependent absorption.\textsuperscript{22} A spin oriented perpendicular to the effective spin quantization axis precesses in this effective “pseudomagnetic” field. (Right) Band edges for an InAs quantum dot embedded in a GaAs host. The band edges are the consequence of material composition and strain.\textsuperscript{23}]

We calculated optical Stark shifts non-perturbatively using a restricted basis of quantum dot wave functions calculated with eight-band $k \cdot p$ theory in the envelope approximation using a method described previously.\textsuperscript{23}
Because of the larger bandgap and lack of strain, the CdSe calculations were performed with a zincblende single-band model for the conduction band, and a four-band model for the valence band.

Calculation of the energy shifts under illumination was done by constructing a restricted set of Fock states for the combined electron-photon system. Eight valence states and the lowest conduction doublet were used for the quantum dot states. The states in the basis for the electron-photon system were \(|1,0; 1,1,1,1,1,1,1,1; N_γ\rangle, |0,1,1,1,1,1,1,1; N_γ\rangle, |1,1,1,1,\ldots 0,\ldots,(N-1)_γ\rangle\) for a total of 10 states. For the first two Fock states all valence states and one conduction state are occupied, and there are \(N_γ\) photons in the photon field. For the other eight Fock states, both conduction states are occupied, only one valence state is unoccupied, and there is one less photon than in the first two states. Inclusion of additional states did not alter the results. Coherent states for the light field can be naturally constructed from the mixed electron-photon states obtained above.

Figure 2. Energy of conduction and valence states for a CdSe spherical quantum dot (left) and an InAs self-assembled molecular-beam-epitaxy quantum dot (right) as a function of detuning. The incident power density is \(10^9\) W/cm².

In Fig. 2 are shown the energies of the conduction states for two representative dot systems: an InAs/GaAs SAQD and a CdSe CQD. In both cases, the splittings increase as the detuning approaches zero, and change sign when the detuning changes sign. The first notable feature is the difference in spin selectivity, which can be traced to the differing strain and shape. For CdSe both spin states are shifted, but by different amounts, while for InAs/GaAs only one spin state is shifted. Because the CdSe dot has approximate spherical symmetry and is unstrained, the highest valence states are closely spaced (<1 meV), with comparable amounts of heavy hole (HH) and light hole (LH) character. As a result, both the HH and LH transitions contribute to the Stark shift, and both spin directions for the conduction state are shifted. The difference between the shifts reflects the difference in oscillator strengths for HH and LH transitions, whose ratio of 2:1 is approximately the ratio found in bulk band-to-band transitions, 3:1. This 2:1 ratio, and the dependence of the splitting on detuning, agrees with experimental measurements. In contrast, for InAs/GaAs dots the highest valence state is almost entirely HH, and separated from the next valence state by several tens of meV (depending on size). Hence, the Stark shift is dominated by the (doubly degenerate) highest valence state which gives a Stark shift for only one spin direction of the conduction electron.

We now examine the effect of dot size on the spin splitting. Shown in Fig. 3 are the splittings for detunings from 30 meV - 70 meV for the four systems under consideration. Since we are ultimately interested in manipulating spins through the effective magnetic field, it is useful to consider the precession angle associated with a light pulse of duration \(\delta t\), given by \(\theta_s = \Delta E \delta t / h\) where \(\Delta E\) is the Stark spin splitting between up and down conduction states. In the results that follow, we give Stark splittings in meV as well as the corresponding \(\theta_s\) for a 200 fs pulse with power density \(10^9\) W/cm², which we refer to as a reference pulse. The ~2 meV spin splitting seen in CdSe CQD’s agrees with that measured experimentally in undoped dots.

The trend in Fig. 3 is surprising; for a given material system larger dots have larger splittings, in spite of the fact that oscillator strengths decrease with increasing dot size. The reason is that the interaction term in the Hamiltonian is proportional to \(1/\omega\). For a fixed detuning, the increase in \(1/\omega\) for larger dots dominates the decrease in the dipole matrix element.
Eg (eV)

Laser field strength

\[ \theta_s = \Delta E \delta / \hbar \]

The angle \( \theta_s = \Delta E \delta / \hbar \) is for an incident 200 fs pulse with \( 10^9 \) W/cm².

The importance of the bandgap suggests selecting the dot material to minimize the gap. One possibility is to use a small bandgap material, such as InAs, but to avoid the bandgap-increasing effect of strain by using an InAs CQD. Confinement increases the bandgap over that of bulk InAs, but for typical CQD sizes \( E_g < 1 \) eV, which is substantially less than \( E_g \approx 2 \) eV for CdSe CQDs.

Fig. 3 confirms the expectation that InAs CQDs have larger splittings than CdSe CQDs over most of the range of sizes, but the increase in splitting is at most a factor of 1.5. While InAs CQDs have larger splittings than CdSe, they are still a factor of 5 smaller than for InAs/GaAs. In spite of a smaller bandgap, the lack of strain in InAs CQDs decreases the HH/LH splitting so much that the Stark splitting is substantially decreased as well.

An alternative to the complete elimination of strain is to choose a substrate material that has a lattice constant closer to InAs. InAs/InP is an excellent candidate, with a lattice mismatch of 4% (as opposed to 7% for InAs/GaAs), and a edge-to-edge bandgap of approximately 0.5 eV. The highest valence state is still predominantly HH, and the first excited valence state ranges from 10 meV - 30 meV below, depending on size. In addition, InAs/InP has a bandgap at the technologically important 1.55 µm. Fig. 3 shows that the splittings in InAs/InP are substantially larger than those for InAs/GaAs. Some of the improvement is simply due to the smaller bandgap and the factor of \( 1/\omega \) in the interaction as discussed earlier. However, the discontinuity in the curves indicates the improvement due to the increase in strain and the resulting increased HH/LH splitting.

At our standard illumination of \( 10^9 \) W/cm² with a 200 fs pulse width, a \( \pi \)-rotation is possible at even large detunings, \( \delta = -70 \) meV.

Bit errors may arise from a variety of sources, such as nonlinear effects and free-carrier excitation. Here we wish to focus on bit errors due to a persistent problem with quantum dot systems, nonzero linewidths. For measurements on an ensemble of dots, inhomogeneities in the dot size will yield a different rotation angle for different dots. More relevant for a quantum computer, in which individual dots will be selected, the finite line width of the states will cause some uncertainty in \( \theta_s \). To address this question, we have calculated numerical estimates of

\[
\frac{1}{\Delta E} \frac{\partial \Delta E(E_g, \hbar \omega)}{\partial E_g} = \frac{1}{\theta_s} \frac{\partial \theta_s(E_g, \hbar \omega)}{\partial E_g},
\]

as shown in Fig. 4. For the materials and sizes considered this quantity varies from \(-0.008 \) meV\(^{-1}\) to \(-0.014 \) meV\(^{-1}\), depending on detuning. The results of Fig. 4 may be used to estimate the uncertainty in the rotation angle, \( \Delta \theta_s \). For example, an ensemble measurement of InAs/InP dots with an inhomogeneous linewidth of 50 meV would give \( \Delta \theta_s \approx 0.5 \) for a \( \pi \)-pulse, which should be sufficient to permit observation of a spin echo. For a single dot with a linewidth of 0.1 meV, \( \Delta \theta_s \approx 0.001 \), corresponding to a bit error rate of \( 10^{-6} \). The spin decoherence time for ensembles of undoped quantum dots can exceed several nanoseconds; spin coherence times for single doped dots are expected to be significantly longer, corresponding to a bit error rate of less than \( 10^{-6} \). This is...
sufficiently small for error correction algorithms to apply\(^{10}\) (desirable error thresholds have been estimated\(^{10}\) at \(10^{-5} - 10^{-6}\)).

![Figure 4](image-url)

Figure 4. (Left) Schematic source of error from dot size (and transition energy) distribution. (Right) Sensitivity of \(\theta_s\) to variations in the bandgap as a function of bandgap and detuning, estimated using a finite difference approximation. Note that larger detunings than in Fig. 3 were used so there would be values of \(\theta_s\) at fixed values of \(\hbar \omega\) for every bandgap value. \(\theta_s\) is computed assuming a reference pulse.

Errors in \(\theta_s\) may also arise from shot noise in the laser pulse. The electric field has some uncertainty due to variations in the number of photons in the pulse. Assuming the laser is focused to 1 \(\mu m^2\) with a photon energy of 1 eV, a reference pulse contains approximately \(10^7\) photons. The uncertainty in the incident intensity is \(\Delta I \approx I/\sqrt{N}\), giving \(\Delta \theta_s \approx 2\pi/\sqrt{N} \approx 10^{-3}\). The corresponding bit error rate is approximately \(10^{-6}\), which is still acceptably small for error correction.\(^{10}\) This estimate gives a lower limit on the error rate since it neglects additional sources of laser noise that would increase the error rate.

We conclude that use of the spin-AC Stark effect is a viable approach to single qubit manipulation in quantum dots. The magnitude of the Stark splitting in SAQDs is 5-10 times larger than in CQDs due to the strain-induced HH/LH splitting. The Stark splitting also increases with decreasing bandgap, though the effect is smaller than that of strain. Because strain increases both the HH/LH splitting and the bandgap, finding the optimal system involves a seeking a system with sufficient strain to induce HH/LH splitting, but not so much as to make the bandgap too large. We propose as a candidate system InA/InP dots, which are strained, but have a relatively small bandgap of 0.7-0.9 eV. This energy range includes 1.55 \(\mu m\), which may be important for interconnection within a quantum computer, or for quantum communication applications. We find that for such dots, \(\pi\)-pulses may be obtained for experimentally realistic pulses (\(10^9\) W/cm\(^2\), 200 fs) and detunings (-70 meV). For typical inhomogeneous broadening (50 meV) the variation in rotation angle is \(\Delta \theta_s \approx 0.5\). For a single dot with an intrinsic linewidth of 0.1 meV we estimate the bit error rate to be on the order of \(10^{-6}\), and we estimate the bit error rate due to laser shot noise is also on the order of \(10^{-6}\). These error rates are sufficiently low for quantum error correction.

### 3. MANIPULATION OF A SI DONOR IN A GAAS HOST

The geometry of g-TMR for a single electron spin bound to a donor is shown in Fig. 5(a), with the electron wave function indicated by the lighter shade, and the silicon nucleus indicated by the darker dot at the center. A static magnetic field along with a gated time varying electric field is applied to the crystal containing the Si\(_{Ga}\) donor. We considered all orientations of the field and found that the most rapid Rabi oscillations occur when the magnetic field is applied at an angle \(\theta = 45^\circ\) to the electric field, which is the configuration shown in Fig. 5(a).

Although many properties of shallow impurities (such as the energy spectrum) can be treated to an excellent approximation by two-band effective mass theory,\(^{27}\) \(g\)-tensor calculations require a multi-band treatment as
the coupling among multiple bands needs to be considered, and the spin-orbit interaction must be treated accurately. Moreover, the electric field breaks the spherical symmetry of the impurity site (Fig. 5(b)). These complexities are best handled numerically.

Our calculations of \( g \)-TMR for Si\(_{\text{Ga}}\) donors were carried out using 8-band \( \mathbf{k} \cdot \mathbf{p} \) theory in the envelope approximation using finite differences on a real space grid. Unlike quantum dots, however, there is no strain in this system. Material parameters correspond to \( T = 0 \). On a grid with sites located at points \( \mathbf{r} \), the hydrogenic impurity potential is given by

\[
V_c(\mathbf{r}) = \begin{cases} 
-C / 4\pi\varepsilon_0 |\mathbf{r} - \mathbf{r}_o|^2 & r = \mathbf{r}_o \\
-C / 4\pi\varepsilon_0 |\mathbf{r} - \mathbf{r}_o| & r \neq \mathbf{r}_o
\end{cases}
\]

where \( \varepsilon_0 \) is the low frequency dielectric constant of GaAs and \( C \) is the sum of the screened Coulomb potential and a central cell correction (CCC) averaged over the voxel centered on the donor located at \( \mathbf{r}_o \). The CCC arises due to the differing chemical nature of various impurities. \( C \) was adjusted until the binding energy of the 1s donor state matched experiment. A value of \( C = 0.84 \text{ eV} \) results in a binding energy of 5.8 meV and a Bohr radius of 9.5 nm for a 1 nm grid spacing and 130 sites. The Landé \( g \) tensor for the impurity ground state was then obtained from the calculated Zeeman splitting of the 1s level in a uniform magnetic field.

![Figure 5.](image_url) (a) Proposed geometry for \( g \)-tensor modulation, with a single silicon atom (dark dot) surrounded by the wave function of the electron shallowly bound to the silicon donor (light shade). (b) Closeup view of the impurity potential (Coulomb potential plus a central cell correction) shown along with the applied electric field’s biasing effect on the zone center band energies.

The \( g \) tensor components were calculated for various directions of \( \mathbf{B} \) with \( \mathbf{E} \) applied along [001], as shown in Fig. 6. Note that \( \partial g / \partial E \) decreases with increasing \( B \). The variation in \( \partial g / \partial E \) as a function of \( B \) is greater when \( \mathbf{E} \perp \mathbf{B} \). However at an intermediate \( B (\approx 2\text{ T}) \), \( \partial g / \partial E \) is identical in all directions. These results imply that an electric field induces a \( g \) tensor anisotropy oriented relative to \( \mathbf{E} \), which makes it possible to modulate the \( g \) tensor using an alternating electric field in addition to the static electric and magnetic fields.

We next solve for the donor atom’s spin dynamics by explicitly integrating the time-dependent Schrödinger equation. The nonlinear nature of \( g \) complicates a quantitative treatment within the rotating-wave approximation. The directional dependence of \( g \) (Fig. 6) can be used to obtain an analytical form of the \( g \) tensor by fitting each tensor component to a 4th order polynomial in \( E \), \( a_0(\mathbf{B}) + a_1(\mathbf{B})E^2 + a_2(\mathbf{B})E^4 \). A time-dependent \( g \) tensor can then be constructed for the time-varying electric field \( E(t) = E_{dc} + E_{ac}\sin(\omega t) \). The maximum amplitude of \( E(t) \) is always held constant at 2 kV/cm, so as not to exceed the breakdown field of the GaAs host. The spin dynamics of the donor atom can then be calculated using the effective time-dependent Hamiltonian,

\[
H(t) = \frac{\mu_B}{2} \sigma \cdot \tilde{g}(t) \cdot \mathbf{B}
\]
where $\mu_B$ is the Bohr magneton. As the Hamiltonian is explicitly time dependent, the state of a spinor, $S_j$ (where $j = \downarrow, \uparrow$) at time $t$ can be obtained by evolving $S_j(t = 0)$ forward in time in $n$ steps of $\Delta t = t/n \ll 1/2\omega$ as follows,

$$
|S_j(t)⟩ = \hat{T} \prod_{\nu=0}^{n} \exp \left( \frac{iH(t_\nu)\Delta t}{\hbar} \right) |S_j(0)⟩ \tag{4}
$$

where $\hat{T}$ is the time-ordering operator. For sufficiently small $\Delta t$ this is equivalent to

$$
|S_j(t)⟩ = \hat{T} \exp \left( \int_0^t \frac{iH(t')}{\hbar} dt' \right) |S_j(0)⟩, \tag{5}
$$

The time dependent probability of making a spin-flip transition is $|⟨S_↑(0)|S_↓(t)⟩|^2$. Rabi oscillations are obtained when spin flip transitions are made resonantly (i.e. $|⟨S_↑(0)|S_↓(t)⟩|^2_{\text{max}} = 1$). Resonant spin flip transitions are usually made when $E(t)$ is driven at the Larmor frequency $\Omega_L$. However in case of the hydrogenic impurity system considered here, the donor electrons spin can be resonantly flipped at any sub-harmonic of the Larmor frequency: $\Omega_L/N$, where $N$ is an integer. This is illustrated in Fig. 7 top left, where the peak spin-flip transition probabilities are shown as a function of the driving $E$-field frequency $\omega$. Multiple resonance lines are apparent, located at $\Omega_L$ and its sub-harmonics. This unusual behavior arises from the highly nonlinear dependence of $g$ on the applied electric field (Fig. 6). For sub-harmonics higher than $N=2$, the Rabi frequencies $\Omega_R$ are lower than those at $N < 2$ and hence are not considered further for spin manipulation. The largest $\Omega_R$ can be achieved by driving $E$ at the second sub-harmonic ($N = 2$) of $\Omega_L$. Due to the smaller DC component of the electric field the Rabi oscillations are less rapid at $\Omega_L$, than at its second sub-harmonic. The resonance lines in Fig. 7 top left at $\omega = \Omega_L/N$, have a full width at half maximum of $\Delta \omega = 2\Omega_R/N$.

The Rabi frequencies are calculated next as a function of $E_{\text{dc}}$ and $\theta$ and are shown in Fig. 7 bottom left with the electric field driven at $\Omega_L$. For all $\theta$, and $\omega = \Omega_L$, $\Omega_R$ is largest when the AC and DC components of the electric field are equal. If the electric field is driven at $\Omega_L/2$, however, as shown in Fig. 7 bottom right, then $\Omega_R$ is largest if $E_{\text{dc}} = 0$. In both bottom panels of Fig. 7, the optimal angle of the magnetic field to the electric field is $\theta = 45^\circ$. Although the maximum $\Omega_R$ in the bottom panels of Fig. 7 are identical, driving $E$ at $\Omega_L/2$ offers two key advantages. When the peak value of $E$ is close to the breakdown of the host crystal, a pure AC field with an adjustable duty-cycle is much less likely to ionize the donor electron, as the carriers can recover during a thermal relaxation time. This allows for higher driving fields, which result in higher $\Omega_R$. It also may be experimentally more feasible to resonantly flip the spin at the lower frequency of the subharmonic $\Omega_L/2$ than the fundamental $\Omega_L$.
Figure 7. Spin dynamics of the donor atom as a function of various parameters. $E_{ac} + E_{dc} = 2 \text{kV/cm}$ and is [001] oriented. $\theta$ is the angle between $\mathbf{B}$ and $\mathbf{E}$. (Top left) Peak spin-flip transition amplitudes as a function of $E$’s driving frequency, for $E_{ac}/E_{dc} = 9$ and $\theta = 45^\circ$. Resonant transitions appear at sub-harmonics of the Larmor frequency $\Omega_L$. (Bottom) Rabi frequency $\Omega_R$ as a function of $E_{dc}$ and $\theta$ for: (Left) $E$ driven at $\Omega_L$, $\Omega_R$ is maximum at $\theta = 45^\circ$ and $E_{dc} = 1 \text{kV/cm}$. (Right) $E$ driven at $\Omega_L/2$, $\Omega_R$ is maximum at $\theta = 45^\circ$ and $E_{dc} = 0$. (Top right) $\Omega_R$ as a function of $B$ for optimal $\theta$ and $E_{dc}$ for bottom plots. Note that above $B = 2T$, $\Omega_R$ increases monotonically.

Fig. 7(top right) shows $\Omega_R$ as a function of $B$ for $\theta = 45^\circ$ and $E$ driven at $\Omega_L$ or $\Omega_L/2$. For magnetic fields greater than $2T$, $\Omega_R$ increases monotonically, whereas below $2T$ $\Omega_R$ exhibits a non-monotonic behavior. This feature can be explained by Taylor-expanding the time-dependent Hamiltonian to first order in the rotating wave approximation,

$$H(E) \approx \frac{\mu_B \sigma}{2} \cdot \left( \mathbf{\Omega}_0 + \frac{E_{ac}}{2} \frac{\partial \mathbf{\Omega}}{\partial E} \bigg|_{E=E_{dc}} \right) \cdot \mathbf{B}. \quad (6)$$

Here the Larmor frequency is given by the time independent static precession vector, $\mathbf{\Omega}_0 = \mu_B \mathbf{\Omega} \cdot \mathbf{B}/\hbar$ and the electron’s spin dynamics in the rotating frame is described by the time-dependent spin precession vector, $\mathbf{\Omega}_1(t) = \mu_B E_{ac} (\partial \mathbf{\Omega}_0 / \partial E) \cdot \mathbf{B}/2\hbar$. $\mathbf{\Omega}_1$ can be resolved into components that are parallel ($\mathbf{\Omega}_{||}$) and perpendicular ($\mathbf{\Omega}_\perp$) to $\mathbf{\Omega}_0$. In the rotating frame, $\mathbf{\Omega}_{||}$ is equivalent to $\mathbf{\Omega}_R$ (in the lab frame), as driving $E$ at $\mathbf{\Omega}_0$ leads to spin precession about $\mathbf{\Omega}_{||}$ or Rabi oscillations. As the tensor components $\partial g/\partial E$ decrease with increasing $B$ (see Fig. 6), the magnitudes of $B$ and $\partial g/\partial E$ have opposing effects on $\mathbf{\Omega}_{||}$ (and hence $\mathbf{\Omega}_\perp$). For $B < 1T$ the contribution from $\partial g/\partial E$ decreases over $B$ and hence the Rabi frequencies increase. For $1T < B < 2T$ the competing contributions of $B$ and $\partial g/\partial E$ make the $g$ tensor increasingly isotropic and the Rabi frequencies smaller. At $B \approx 2T$ the $g$ tensor becomes isotropic and the Rabi frequency vanishes. Above $B \approx 2T$, the effects of a much larger magnetic field dominate and the spin flip times decrease monotonically. Two key inferences, consistent with other work on $g$-TMR, can be drawn from this behavior. For spintronic applications the highest magnetic field possible is desirable in order to generate the largest possible Rabi frequencies. Secondly, the amount of $g$-tensor anisotropy induced is crucial to achieving shorter spin-flip times, not the degree of change in $g$ as a function of $E$.

4. MANIPULATION OF A MN ACCEPTOR IN A GAAS HOST

Calculations of the defect properties in GaAs are done using a tight-binding theory based on the deep level model of Vogl and Baranowski. In this model the interaction of the acceptor occurs via $T_2$-symmetric states that hybridize with the dangling bonds of the nearest-neighbor As. In order to calculate the defect wave functions we
use the Koster-Slater technique\textsuperscript{37} to calculate the Green’s function whose imaginary part gives the local density of states. This method has been proven to give the correct chemical trend of impurity levels in semiconductors.\textsuperscript{38} Starting with the tight-binding Hamiltonian $\hat{H}_0(k)$ of homogeneous GaAs, one first calculates the retarded Green’s function, $\hat{G}_0(k, \omega) = [\omega - \hat{H}_0(k) + i\delta]^{-1}$. To obtain a good description of the valence band structure, we use the $sp^3$ model including spin-orbit interaction.\textsuperscript{39} Then, by Fourier transforming $\hat{G}_0(k, \omega)$, we construct the homogeneous Green’s function in coordinate space, $\hat{G}_0(R_i, R_j, \omega)$, where $R_i$ and $R_j$ label the zincblende lattice sites. The final Green’s function is obtained by solving the Dyson’s equation,

$$\hat{G}(\omega) = \hat{G}_0(\omega) + \left[ 1 - \hat{G}_0(\omega) \hat{V} \right]^{-1} \hat{G}_0(\omega),$$

where $\hat{G}(\omega)$ is the full matrix representation using all atomic orbitals at all lattice sites. The LDOS at each site $R_i$ is given by

$$A(R_i, \omega) = -\frac{1}{\pi} \text{Im} \left[ \text{tr}_\alpha \hat{G}(R_i, R_i, \omega) \right],$$

where the $\text{tr}_\alpha$ is taken with respect to the orbitals of the $\alpha$ atom, depending on which type of atom is actually located at the site $R_i$. Shown in Fig. 8 is a schematic of the core spin combined with hole spin and orbital angular momentum, as well as the acceptor state contour surface for spin-averaged and spin-polarized states.

Figure 8. (Left) Schematic of the Mn-hole complex in GaAs. The core spin 5/2 is oriented antiparallel to the hole spin, which is parallel to the orbital angular momentum of the hole state. The combination has angular momentum $J = 1$. (Middle) Spin-averaged contour surface for the Mn acceptor wave function. (Right) Spin-polarized contour surface for the Mn acceptor wave function.

The degeneracy of the $J = 1$ Mn ion can be substantially split by external electric fields, as shown in Fig. 9, and the eigenstates depend strongly on the electric field direction. This will be the source both of state splitting (analogous to the static magnetic field in traditional ESR) and state coupling (analogous to the oscillating perpendicular magnetic field in traditional ESR). We find the following electric-field-dependent Hamiltonian:

$$H_I(E) = \gamma [E_x (J_y J_z + J_z J_y) + \text{c.p.}],$$

where $E$ is an electric field, c.p. stands for cyclic permutation, and $\{x, y, z\}$ stand for the 3 major axes of the cubic crystal. Note that this Hamiltonian does not break time-reversal symmetry for the angular momentum operators $J$ always appear in pairs. We calculate, using the probability densities of the hole state found in our tight-binding calculations and first-order perturbation theory, $\gamma = 6.4 \times 10^{-30}$ Cm, corresponding to $\gamma E = 160 \mu$eV for $E = 40$ kV/cm. This exceptionally large splitting is equivalent to that generated by applying a 1 Tesla magnetic field using the measured $g$-factor,$^40$ 2.77. The linear dependence on electric field, critical to producing a large splitting, originates from the lack of inversion symmetry of the substituted ion in a tetrahedral host. The energy splittings from an electric field applied to bound states at inversion-symmetric sites in crystals, or
electrons bound in atoms or ions in vacuum, would depend quadratically on the electric field and would be correspondingly much smaller. The other essential element causing this large splitting is the large (∼10 Å) Bohr radius of the bound valence hole.35, 41 Recent progress in theory and scanning tunneling microscopy of Mn dopants in III-V semiconductors has confirmed the large spatial extent of the bound hole wavefunction.35, 41–43 Thus the response of the Mn wavefunction to electric fields is substantial compared to other ion levels associated with transition-metal (magnetic) dopants.

![Figure 9](image.png)

Figure 9. (Left) Proposed configuration for the electric resonances of a single Mn dopant in GaAs. A dc electric field $E_d$ is applied via the electrical gates and the STM tip. The resonance is driven by an additional small ac field. (Right) Schematic of the splitting of states with the applied electric field.

5. CONCLUSIONS

Three methods of spin manipulation of localized spins confined in semiconductors have been considered. Optical illumination can generate an effective magnetic field through the spin-orbit interaction that can be used to reorient electronic spins. Electric fields can be used to manipulate the electronic spin of a conduction electron bound to a donor in a static magnetic field, using $g$-tensor modulation resonance. Electric fields can also be used, in the absence of any magnetic field, to manipulate the integer spin of a Mn dopant in GaAs.

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REFERENCES


