Optical manipulation of electron spins bound to donors in GaAs

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Searching for a quantum information processor

- **classical v. quantum information**
  - classical bit: 0 or 1
  - quantum bit: $\psi = a|0\rangle + b|1\rangle$, $|a|^2 + |b|^2 = 1$

- **why quantum information?**
  - certain problems can be solved faster with quantum algorithms (factoring large numbers)
  - quantum simulation
  - quantum communication (secure quantum key distribution for cryptography)
Motivation: spins and light for QIP

- Long lived stationary qubit
  \[ |\Psi\rangle = a|0\rangle + b|1\rangle \]
  SPINS

- Fast single qubit operations
  SPIN-LIGHT interaction

- Fast transmission of quantum information
  SPIN ↔ LIGHT transfer

Two level systems with strong optical transitions relax quickly
\( \Lambda \) systems for QIP

**Three-level system**

- Long lived stationary qubit
  \[ |\Psi\rangle = a|0\rangle + b|1\rangle \]

- Fast qubit operations
  SPIN-LIGHT interaction

- Fast transmission of information
  SPIN ↔ LIGHT transfer

\[ |0\rangle \quad |3\rangle \quad |1\rangle \]
QIP in semiconductors

Long-term potential: advanced device integration

Disclaimer: An optics-based quantum processor will probably not look like this Sun Microsystems chip.
Outline: GaAs electron donor experiments

1. Donor electrons (D\textsuperscript{0}) and excitons (D\textsuperscript{0}X)

2. Dephasing and relaxation in D\textsuperscript{0}-D\textsuperscript{0}X systems

3. Optical manipulation of the donor electron spin state.
Impurities can bind spins and act as atoms.

A^0, A^0X system is analogous
Atomic nature of impurities is exhibited by carbon $A^0X$ in GaAs

Samples from Y. Hirayama, NTT
Acceptor energy levels fit hydrogenic model

$$E(A^0 X) - E_n(A^0) = E_0 - E_{ion} \left(1 - \frac{1}{n^2}\right)$$

model fit:

However, hole relaxation times are too short for QIP
Homogeneity of $D^0X$ observed in magneto-photoluminescence spectrum

- $D^0X$
- $D^+X$
- $D^0X$ (TES)
- $A^0X$
- $2p^+$, $2p^0$, $2s$
- $2p^-$
- $2s$, $2p$
- $1s$
Using a magnetic field to obtain a $D^0X \Lambda$-system

Neutral donor bound exciton

Radiative excitation and recombination

Neutral donor
Basic requirements for QIP $\Lambda$-system

- Strong radiative coupling to excited state
- Optically thick (for ensemble experiments)
- Homogeneous (optical transitions)
- Long coherence time between two lower states
**D^0_X-D^0 \Lambda-system parameters**

Relaxation rates should be compared to 100 GHz-THz Rabi frequencies (qubit rotation times)

- Strong radiative coupling to excited state
- Optically thick
- Homogeneous
- Long coherence time between two lower states

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radiative relaxation rate</td>
<td>&gt; 1 GHz (10-100 MHz in atomic systems)</td>
</tr>
<tr>
<td>Density</td>
<td>$10^{13}$-$10^{15}$ cm$^{-3}$ (10$^{10}$-$10^{11}$ in atomic systems)</td>
</tr>
<tr>
<td>Inhomogeneous broadening</td>
<td>2-10 GHz (compare to THz in quantum dot systems)</td>
</tr>
<tr>
<td>Electron coherence time</td>
<td>$T_2^* = 1$-$10$ ns, $T_2 = \mu$s $-$ ms ?</td>
</tr>
</tbody>
</table>
Outline: GaAs electron donor experiments

1. Donor electrons (D₀) and excitons (D₀X) for QIP

2. Dephasing and relaxation in D₀-D₀X systems

3. Optical manipulation of the donor electron spin state.
Sample is high-purity MBE GaAs

B-field
1.5-2 K

excite
collect

GaAs 10 μm
n~5×10^{13} cm^{-3}

Al_{0.3}Ga_{0.7}As , 4 μm

GaAs SI substrate

*Sample from C. Stanley and M.C. Holland, University of Glasgow*
$T_2^*$ can be measured using Raman transitions

If $\delta \gg \gamma, \Omega_R, \gamma \gg \alpha,$

$$\Delta \nu \approx \alpha + \frac{\Omega^2 \gamma}{4 \delta^2}$$

$\Delta \nu$ gives $\alpha$ or $T_2$ for large detuning/low pump intensity.
Raman sidebands are clearly observed in PL-spectra.

Linewidths are measured by a combination of a grating and Fabry-Perot etalon (resolution < 20 MHz)
No dependence of $\Delta \nu_{\text{Raman}}$ on B-field

$T_2^* = 2 - 3$ ns

→ No magnetic field dependence is observed within the measurement uncertainty.
$T_2^*$ is believed to be limited by the nuclear hyperfine interaction.

$$E_{HF} = m_s m_n \frac{2 \mu_0}{3} g_0 \mu_B \gamma_n \hbar |\psi_e(0)|^2$$
A simple model can give insight into the hyperfine dependence on nuclear polarization

Spin $\frac{1}{2}$, single species lattice, constant electron density over some finite volume $V$.

$$E_{HF} = m_s m_n \frac{2 \mu_0}{3} g_0 \mu_B \gamma_n \hbar |\psi_e(x_n)|^2$$

$$E_{HF}^{tot} \propto \rho_e \sum_i m_n^i$$

$$\Delta E \propto \frac{1}{V_e} \sqrt{N} \sqrt{p(1-p)} = \Delta E \propto \frac{1}{\sqrt{N}} \sqrt{p(1-p)}$$

1. The larger the electron spatial wavefunction, the smaller the hyperfine broadening!
2. Need extremely high nuclear polarization for a significant increase in $T_2^*$!
A simple calculation gives dephasing time consistent with Raman measurements.

For maximum HF field broadening for spin $3/2$ particles:

\[ \Delta B_{nuc} = b_0 \sqrt{I_0(I_0 + 1)/N_{nuc}} \]

- $b_0 = 3.5 \text{ T}$
- $I_0 = 3/2$
- $a_B = 100 \text{ Å}$
- $N_{nuc} = 2 \times 10^5$

- $\Delta B_{nuc} = 15 \text{ mT}$
- $\Delta \nu = 85 \text{ MHz}$
- $T_2^* = 2 \text{ ns}$

Magnitude and B-field dependence are consistent with Raman measurement (2-3 ns).
Overcoming nuclear dephasing

- **Nuclear polarization** ❌
- **Different semiconductors**
  - Isotope purified II-VI spin-0 matrix
  - CdSe investigated ❌
- **Spin Echo**
  - Need refocusing pulses fast compared to 1 ns dephasing - motivation for fast pulse experiments
  - Can be used to reach fundamental homogeneous $T_2$ decoherence time.
  - $T_2$ time is fundamentally limited by $T_1$ processes.
The $T_1$ time of the system can be determined by a pump/probe experiment.

- **Thermal equilibrium**
- **Optical pumping**
- **Pumped state, laser off**
- **$T_1$ relaxation**
- **Probe level 2 population**

Detect!
A diagram of the optical pumping/$T_1$ experiment
Optical pumping can obtain > 0.95 polarization at 9.9 T

$\rho_{11} = 0.95$
max observed (0.98)
Pump-probe data shows nice recovery of population

\[ T_1 = 770 \mu s \]
Millisecond $T_1$ times are observed.

LONG $T_1$ times observed (1000× longer than previously reported)

$T_2^* = \text{ns} < T_2 < T_1 = \text{ms}$

KMC Fu, W Yeo, SM Clark, C Santori, C Stanley, MC Holland, Y Yamamoto, PRB 74, 121304 (2006)
Summary of D⁰ relaxation

- Raman experiment indicates nanosecond electron spin dephasing times that are not dependent on magnetic field.
- Magnitude and field dependence of Raman linewidths are consistent with nuclear hyperfine theory.
- Dephasing due to static, inhomogeneous nuclear environment theoretically can be overcome with spin-echo techniques.
- $T_2$ is then fundamentally limited by measured millisecond $T_1$ times.
- Needed: (for both QIP and to reach longer coherence times) **SPIN CONTROL**
Outline: GaAs electron donor experiments

1. Donor electrons ($D^0$) and excitons ($D^0X$) for QIP

2. Dephasing and relaxation in $D^0$-$D^0X$ systems

3. Optical manipulation of the donor electron spin state.
The ‘dark state’: a simple view of EIT and coherent population trapping (CPT)

In RWA, interaction frame

\[
H = \begin{bmatrix}
0 & 0 & \Omega_p^* \\
0 & 0 & \Omega_c^* \\
\Omega_p & \Omega_c & 0
\end{bmatrix}
\]

Eigenstates:

\[
\frac{1}{\sqrt{\Omega_p^2 + \Omega_c^2}} \left( \Omega_c |1\rangle - \Omega_p |2\rangle \right) \quad \text{dark}
\]

\[
\frac{1}{\sqrt{2} \sqrt{\Omega_p^2 + \Omega_c^2}} \left( \Omega_p^* |1\rangle + \Omega_c^* |2\rangle + \sqrt{\Omega_p^2 + \Omega_c^2} |3\rangle \right) \quad \text{bright}
\]

\[
\frac{1}{\sqrt{2} \sqrt{\Omega_p^2 + \Omega_c^2}} \left( \Omega_p^* |1\rangle + \Omega_c^* |2\rangle - \sqrt{\Omega_p^2 + \Omega_c^2} |3\rangle \right) \quad \text{bright}
\]
EIT-based QIP

- Slow light and light storage
- Quantum optical delay lines
- Quantum optical memory (needed for long distance quantum communication)
EIT-based QIP

- Slow light and light storage
- Non-linear optics
- Large, conditional phase shifts (4-level)
- Single photon detection (4-level)
EIT-based QIP

- Slow light and light storage
- Non-linear optics
- Study of $|1\rangle \leftrightarrow |2\rangle$ coherence and control

- Measurement of 1-2 quantum coherence
- Adiabatic passage between 1-2
Experimental Procedure: Photoluminescence excitation (PLE) spectroscopy to detect $|3\rangle$, $D^0X$ population
TES lines are enhanced under resonant excitation

- Diagram showing energy levels and transitions:
  - 1s
  - A*
  - A
  - A1
  - 2p-
  - 2s
  - 2p0

- Intensity graph with wavelength (nm) ranging from 817.5 to 821.5
  - Aboveband
  - Resonant, A

- Legends for intensity and wavelength are provided.
A modest dip on two-photon resonance is observed

\[ \Omega_p, A^* \quad \Omega_c, A \]

\[-1/2 \quad +1/2 \]

Probes and coupling determine \( T_2^*, \Omega_c \)

We use the density matrix description in which the $3 \times 3$ density matrix $\rho$ evolves according to

$$\frac{\partial}{\partial t} \rho = -\frac{i}{\hbar} [\hat{H}, \rho] + \mathcal{L}(\rho)$$

- $\Omega_p$: probe Rabi frequency
- $\Omega_c$: coupling Rabi frequency
- $\Delta$: probe detuning from $|1\rangle \leftrightarrow |3\rangle$
- $\delta$: is two photon detuning

Data can be fit to a three-level density matrix model.
Model incorporates all relevant relaxation terms in $\mathcal{L}(\rho)$

- Relaxation term $\mathcal{L}$ includes:
  - $\Gamma_{31}, \Gamma_{32}$: radiative relaxation rates
  - $\Gamma_{21}, \Gamma_{21}$: lower state population relaxation rates
  - $\gamma_3$, additional $|3\rangle$ broadening
  - $\gamma_{21}$, lower state decoherence rate
Model fits $\Omega_C$ dependence if $T_{12}$ increases with $\Omega_C$.

Increase in $T_{12}$ is consistent with sample heating and simple phonon-bath model. $T = 1.5, 1.5, 2.0, 3.1, 4.6, 6.0$ K ($P_C = 0.5-8$ mW)
CPT Summary

- Coherent population trapping of electrons spins in a semiconductor is observed for the first time.
- Nanosecond dephasing rate limits the CPT effect (consistent with Raman experiment).
- Although coherence is observed, fast pulse techniques are necessary to achieve larger coherences.
Outline: GaAs electron donor experiments

1. Donor electrons ($D^0$) and excitons ($D^0X$) for QIP

2. Dephasing and relaxation in $D^0$-$D^0X$ systems

3. Optical manipulation of the donor electron spin state.
Needed: fast control of the electron spins

- Options
  - Microwave electron spin resonance
    - slow, ns pulse times
    - non-local, $\lambda = \text{millimeter to centimeter}$
Needed: fast control of the electron spins

- Options
  - MW electron spin resonance
  - Pulsed two-color Raman transition
    - faster- speed is limited by $|1\rangle$-$|2\rangle$ splitting (in our system $\sim 100$ ps)
    - local, limited to laser spot size
    - rotation angle is controlled by relative phase between the two fields- need good phase control!

```
|3\rangle

|1\rangle  |2\rangle
```
Needed: fast control of the electron spins

Options
- MW electron spin resonance
- Pulsed two-color Raman transition
- Single ultra-fast pulse
Previously demonstrated semiconductor single-pulse ESR techniques

- Time-resolved Faraday rotation, differential transmission
  - Fast, passive

- Resonant stimulated Raman two-photon process (SR2P) in quantum dots.
  - Fast, active, fidelity of pulses is fundamentally limited

- Optical-Stark effect
  - Fast, active, control over the rotation axis is limited
Spin control with off-resonant ultra-fast pulses

- Spin state is manipulated through a two-photon Raman process with a single, far-detuned pulse
  - Local, limited to laser spot size
  - Fast (ps-100 fs)
  - Active (any rotation angle possible theoretically)
  - Control over the rotation axis is achieved with pulse arrival timing
Ultra-fast rotations can be achieved with a single broadband pulse

Three level system:

$$H = \hbar \begin{pmatrix} 0 & 0 & \Omega_1/2f(t) \\ 0 & \delta & \Omega_2/2f(t) \\ \Omega_1^*f(t) & \Omega_2^*f(t) & \Delta \end{pmatrix}$$

Effective two level system:

$$H = \hbar|\Omega|f^2(t) + \hbar \begin{pmatrix} 0 & \Omega f^2(t) \\ \Omega^*f^2(t) & \omega_L \end{pmatrix}$$

$$\Omega = \frac{\Omega_1 \Omega_2^*}{4\Delta}$$
Pulse timing determines the rotation axis

2 level approximation

\[
H = \hbar \begin{pmatrix}
0 & |\Omega|e^{i\phi}f^2(t) \\
|\Omega|e^{-i\phi}f^2(t) & \omega_L
\end{pmatrix}
\]

rotating frame

\[
H = \hbar \begin{pmatrix}
0 & |\Omega|e^{i(\phi - \omega_L t)}f^2(t) \\
|\Omega|e^{-i(\phi - \omega_L t)}f^2(t) & 0
\end{pmatrix}
\]

Bloch picture

\[
\frac{\partial \vec{s}}{\partial t} = \vec{\Pi} \times \vec{s}
\]

\[
\Pi = \begin{pmatrix}
|\Omega| \cos(\phi - \omega_L t) \\
|\Omega| \sin(\phi - \omega_L t) \\
0
\end{pmatrix}
\]
3-level simulations with relaxation show $\pi$ and $\pi/2$ pulses are possible.

\[ \Delta = 1 \text{ THz} \]

\[ \tau_{\text{pulse}} = 2 \text{ ps} \]

\[ \omega_L = 2\pi \times 39 \text{ GHz} \]

\[ B = 7 \text{ T} \]
Experimental demonstration of ultrafast optical spin control

Experiment

1. Optically pump spins
2. Apply pulse
3. Measure population in state $|2\rangle$

Population after pulse

Population

$\Omega_\gamma$, THz

$\pi/2$

$\pi$
Pulse experiment

Optical pumping
Fast pulse population transfer and saturation
1 THz detuning, 7T

- Population transfer is observed
- Saturation indicates dephasing process at large pulse areas
- Is transfer coherent at smaller areas?

Coherence and rotation axis control can be demonstrated with a double pulse experiment
Double pulse experiment

7T, 1.5K GaAs

AOM1 → power stabilizer → Ti:Sapph ring Laser

ACM2 → EOM → power stabilizer → Ti:Sapph pulsed Laser

Data Generator

camera

spectrometer

SPCM

Multichannel Scaler

Detect!

Optical pumping

A

A*
Small angle rotations around arbitrary axes using ultra-fast pulses

Pulse spacing $\tau = \tau_{\text{Larmor}}$

Two rotations about x-axis (rotating frame)
Small angle rotations around arbitrary axes using ultra-fast pulses

pulse spacing $\tau = \frac{\tau_{\text{Larmor}}}{2}$
2nd rotation about -x-axis

pulse 1 | pulse 2

population

$|1\rangle$, $|2\rangle$, $|3\rangle$
Oscillations will be observed with a frequency equal to the Larmor frequency.
Oscillations observed at 7 T and 5 T

- Ultrafast rotations about an arbitrary axis are demonstrated.
- Rotations maintain some degree of coherence
Pulse rotation angle and fidelity

Red curve: 2-level density matrix model

1) rotation + decoherence
2) wait (Larmor precession)
3) rotation + decoherence

→ adjust rotation angle and decoherence until model fits data
Pulse rotation angle and fidelity

![Graph showing 2-pulse amplitude versus delay, ps]

\[ \rho_{22} \]

<table>
<thead>
<tr>
<th></th>
<th>1 pulse</th>
<th>2 pulses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rotation angle</td>
<td>( \pi/3 )</td>
<td>( 2\pi/3 )</td>
</tr>
<tr>
<td>Fidelity ( = \langle \Psi_{\text{ideal}}</td>
<td>\rho</td>
<td>\Psi_{\text{ideal}} \rangle )</td>
</tr>
</tbody>
</table>
Fast decoherence must be present during pulse duration

- 2-pulse visibilities $< 0.65$
- 1-pulse experiment shows saturation
  $\Rightarrow$ Fast decoherence process! $\approx ps$ pulse duration
- Band-edge effects - not truly a 3-level system
  - Pulses are detuned by
    - 1 THz from lowest $D^0X$ transition
    - $\sim 1.3$ THz from free exciton transitions
    - $\sim 2$ THz from band edge
  - Spin exchange/interaction between virtual carriers and donor electrons/excitons?

Fast dephasing effects may be overcome with
- 100 fs pulses
- Emitters further from the band edge
Summary of spin control with far detuned ultra-fast Raman pulses

- spins can be probed AND manipulated with picosecond resolution
- theoretically arbitrary rotation angles
- experimentally significant rotation angles observed ($\sim \pi/3$)
- complete control of rotation axis with pulse timing
Summary: D⁰-D⁰X systems exhibit nice properties for optics-based QIP

1. Donor electrons (D⁰) and excitons (D⁰X)

1. Little inhomogeneous broadening of optical transitions.
2. Lambda system can be isolated with electron spin qubit
Summary: Coherence properties are promising

2. Dephasing and relaxation in D⁰-D⁰X systems

1. Nanosecond inhomogeneous dephasing of the electron spins
2. Dephasing due to ‘slow’ nuclear environment
3. Ultimately limited by ms T₁ times
Summary: Electron spin control is demonstrated with Raman transitions

1. Coherent population trapping is demonstrated.
2. Ultra-fast spin-rotations are demonstrated
   1. Single pulse areas of $>\frac{\pi}{3}$ are demonstrated
   2. Full control of the rotation axis is demonstrated
   3. Spin manipulation/probing time $\ll$ Larmor period

3. Optical manipulation of the donor electron spin state.
A neutral donor quantum computer?

Not quite yet....
Acknowledgments

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