Spin Blockade, Spin Relaxation and Spin Dephasing, in $^{12}$C and $^{13}$C Nanotubes

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Pauli Blockade in a Double Quantum Dot

A Forward bias: $\mu_\ell > \mu_r$

Reversed bias: $\mu_\ell < \mu_r$

Spin Blockade in a Double Dot
Control and Detection of Singlet-Triplet Mixing in a Random Nuclear Field

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Inhomogeneous Dephasing

the situation in GaAs

\[ \text{(0,2)s} \]

\( T_+ \)

\( S \)

\( T_0 \)

\( (1,1) \) is GS

Voltage-controlled tilt

\( (0,2) \) is GS

\[ \varepsilon \]

\[ E \]

\[ \tau_s \]

Experiment
- B=0 mT
- B=100 mT

Theory
- B=0 mT
- B=100 mT
Inhomogeneous dephasing due to hyperfine fields in GaAs is revealed in single-shot measurements.

\[ \tau_S (\text{ns}) \]

\[ \begin{pmatrix} 0 & 100 & 200 & 300 & 400 & 500 \\ -0.5 & -0.8 & -1.1 \end{pmatrix} \]

\[ \langle V_{rf} \rangle \quad V_T \]

\[ \langle V \rangle \]

\[ \Delta B_z \]

\[ B_{\text{nuc},l} \quad B_{\text{nuc},r} \]

\[ T_0 \]

\[ \text{S} \]

\[ \text{TRIPLET} \quad \text{SINGLET} \]

\[ \text{Sapphire Stripline} \quad \text{Directional Coupler} \]

\[ \text{LO} \quad \text{IF LPF} \quad \text{IF Amplifier} \quad \text{G} = 10 \text{dB} \]

\[ \text{Attenu.} \quad \text{215 - 240 MHz} \]

\[ \text{Scope} \quad \text{300K} \quad \text{4K} \]

\[ \text{SS coax} \quad \text{800mK} \]

\[ \text{Nb coax} \quad \text{100mK} \]

\[ \text{QPC - Bias} \]

\[ \text{100pF} \quad \text{100nH} \quad 820nH \]

\[ \text{Bias-Tee} \]

\[ \text{100pF} \quad \text{5kΩ} \]

\[ \text{Cp} \]

\[ \text{Monday, June 22, 2009} \]
Carbon nanotubes

Quantization around circumference gives 0 or 1.4 eV·nm/r gap

Roll up $rk_{\perp} = \text{integer}$

Perturbations induce small ~10s meV gaps
Controlling Hyperfine Coupling using Nanotube Quantum Dots

- CVD growth with 99% $^{12}$CH$_4$ or 99% $^{13}$CH$_4$
Controlling Hyperfine Coupling using Nanotube Quantum Dots

- Pd contacts
Controlling Hyperfine Coupling using Nanotube Quantum Dots

- Pd contacts
- NO$_2$ + Al$_2$O$_3$ ALD
Controlling Hyperfine Coupling using Nanotube Quantum Dots

- Pd contacts
- Al₂O₃ + NO₂ ALD
- Al top gates

![Image showing nanotube structures with different gaps and g values](image_url)
Devices

• CVD growth with $^{12}$CH$_4$ or $^{13}$CH$_4$

• Fe catalyst

• Pd contacts

• Al$_2$O$_3$ + NO$_2$ ALD

• Al top gates

Related work


$^{13}$CH$_4$: Liu and Fan, JACS (2001)

Devices

- CVD growth with $^{12}\text{CH}_4$ or $^{13}\text{CH}_4$
- Fe catalyst
- Pd contacts
- Al$_2$O$_3$ + NO$_2$ ALD
- Al top gates

Related work

DQDs:
- Biercuk et al., Nano Lett. (2005)
- Graeber et al., PRB (2006)

Single dot charge sensing:
- Biercuk et al., PRB (2006)

$^{13}\text{CH}_4$:
- Liu and Fan, JACS (2001)

NO$_2$:
- Williams et al., Science (2007)
Devices

- CVD growth with $^{12}$CH$_4$ or $^{13}$CH$_4$
- Fe catalyst
- Pd contacts
- Al$_2$O$_3$ + NO$_2$ ALD
- Al top gates

Related work

DQDs:  
Graeber et al. PRB (2006)  

Single dot charge sensing:  
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$^{13}$CH$_4$: Liu and Fan, JACS (2001)
Williams et al., Science (2007)
Tunable double dot

13C spin blockade

Figure –: Spin blockade in a 13C nanotube double dot. a Current $I_{dd}$ near zero detuning as a function of magnetic field for positive bias (non-blockade) red tracer and negative bias (blockade) for two values of $V_M$. 

- +1 mV bias $B_{||} = 200$ mT
- -1 mV bias $B_{||} = 200$ mT

Charge sensing signal $g_s$ for $-z$. The transfer of charge from the left dot to the right is delayed until the excited state is reached at high detuning. 

Schematic of spin blockaded transport. Any spin may occupy the left dot, but only a spin singlet is allowed in the right dot, suppressing negative bias current once an electron enters the left dot and forms a triplet state.

\[ ^{13}\text{C} \text{ spin blockade} \]

\[ \begin{align*}
V_R (\text{mV}) & \quad \text{Current (pA)} \quad V_R (\text{mV}) \\
0 & \quad 0 & \quad 0 \\
10 & \quad 1 & \quad 1 \\
+1 \text{ mV bias} & \quad B_\parallel = 200 \text{ mT} & \quad -1 \text{ mV bias} \\
0 & \quad 0 & \quad 0 \\
10 & \quad 1 & \quad 1 \\
\end{align*} \]

\[ V_{MF} = 256 \text{ mV} \quad V_{MF} = 222 \text{ mV} \]

Magnetic field dependence of spin relaxation

\[ g\mu_B B_{\text{nuc}} = \frac{A}{\sqrt{N}} \]
\[ B_{\text{nuc}} = 6.1 \text{ mT} \]
\[ \rightarrow A \sim 100 \text{ } \mu\text{eV} \]

See Also
Expt:
Koppens, Folk, et al. (GaAs) Science 305, 1346 (2005).
Theory:
Jouravlev and Nazarov PRL 96, 176804 (2006)

See Also
Theory:

Hysteresis in $^{13}$C leakage current

![Graph showing hysteresis in leakage current](image)


sets independent lower bound on $A \sim 50 \mu$eV
Charge sensing

\[ V_L \quad V_R \]

\[ g_s \]

\[ V_S \]

double dot charges
‘gate’ sensor dot
Charge sensing

VL  VR

double dot charges
‘gate’ sensor dot

gs

VS

(0,0) (1,0) (2,0)
The double dot studied here is based on a single-wall carbon nanotube grown by chemical vapor deposition with a laser nanolithography functionalized feedstock. The device is coated with a bilayer of atomic layer deposition aluminum to gate oxide using the honeycomb pattern. On time-scales in fair agreement with the hyperfine coupling strength in carbon nanotubes, which allows phonon mediated relaxation and suppresses hyperfine mediated relaxation.

We find that dephasing occurs at zero magnetic field and goes through a minimum in a parallel field of 1.4 T. We associate these results with the spin-orbit modified electronic spectrum of carbon nanotubes, which allows phonon coupling dominates decoherence. Although recent protocols for coherent manipulation and detection of individual and pairs of Pd contacts, we argue that the absolute number of electrons in each dot is longest at zero magnetic field and displays a minimum in a parallel field of 1.4 T. We interpret these results within the context of the recently observed spin blockade regime for a carbon nanotube double dot to measure relaxation and dephasing times. The relaxation time is found to be longest to relaxation but lead to rapid dephasing. The double dot is defined by top-gates L, R, and M and a single dot between contacts 3 and 4 as a function of \( L \), \( R \), and \( N \).

Recent advances in nanoelectronics and spins of heavy isotopes free of nuclear spins have enabled the development of qubits which primarily comprise isotopes free of nuclear spins to mitigate decoherence due to nuclei and an attractive alternative is to base spin qubits on group IV elements. Developing these systems requires a strong interdot capacitive coupling of 10 meV. Capacitive coupling of charge to the double dot is inferred from the conductance of the charge sensor. Fast pulses are applied to \( L \) and \( R \). The conductance of the charge sensor is unambiguously identified from the conductance \( g_s \) measured between contacts 3 and 4 as a function of \( V_R \) and \( V_L \). The conductance \( g_s \) is measured with a lock-in amplifier and standard lock-in measurements are carried out in a dilution refrigerator electron temperature.

This Letter reports measurements of spin relaxation and dephasing in a two-electron quantum dot. Spin-qubits are described by quantized longitudinal modes and carbon nanotube double dots include observation of singlet-triplet and carbon nanotubes. Recent advances in nanoelectronics and spin blockade mitigate decoherence due to nuclei. Double quantum dots include observation of singlet-triplet states required to form qubits and spin blockade. Developing these systems requires strong interdot capacitive coupling of 10 meV.
Levels in a single dot

\[ \Delta_{SO} = 0 \]
\[ \Delta_{KK'} = 0 \]
\[ \Theta = 0 \]
Levels in a single dot, including spin-orbit coupling

\[ \Delta_{SO} = 0.17 \text{ meV} \]

\[ \Delta_{KK'} = 0 \]

\[ \Theta = 0 \]
Levels in a single dot, including spin-orbit coupling

\[ \Delta_{SO} = 0.17 \text{ meV} \]
\[ \Delta_{KK'} = 0 \]
\[ \Theta = 0 \]
Levels in a single dot, including spin-orbit coupling and valley mixing

\[ \Delta_{SO} = 0.17 \text{ meV} \]
\[ \Delta_{KK'} = 0.02 \text{ meV} \]
\[ \Theta = 0 \]
Levels in a single dot, including spin-orbit coupling, valley mixing, and misaligned field.

\[ \Delta_{SO} = 0.17 \text{ meV} \]
\[ \Delta_{KK'} = 0.02 \text{ meV} \]
\[ \Theta = 5^\circ \]
Levels in a single dot, including spin-orbit coupling, valley mixing, and misaligned field

See Also
Kuemmeth, Ilani et al.
Nature 452, 448 (2008)
Levels in a single dot

single-particle levels

\[
\begin{align*}
\Delta_{SO} & \quad \Delta_{KK'} \\
B_{\text{orb}} & \quad B_{\text{spin}} \\
E & \quad \Delta_{\theta}
\end{align*}
\]

addition spectrum

\[
\begin{align*}
\mu_{\text{orb}} & \quad -100 \\
V_L (\text{mV}) & \quad -140 \\
B (\text{T}) & \quad B (\text{T}) \\
(0,0) & \quad (1,0) \\
(1,0) & \quad (2,0) \\
(2,0) & \quad (3,0) \\
(3,0) & \quad (4,0)
\end{align*}
\]

slopes differ by \(2\mu_B\)

(1,1) and (0,2) nanotube double dot states

10 blocked and 6 unblocked two-electron states

\[
\begin{align*}
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes |K\rangle|K\rangle \otimes T_+ \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes |K'\rangle|K'\rangle \otimes T_- \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes (|K'\rangle\downarrow|K\uparrow) + (|K\uparrow|K'\downarrow) \\
&(|R\rangle|L\rangle + |L\rangle|R\rangle) \otimes (|K'\rangle\downarrow|K\uparrow) - (|K\uparrow|K'\downarrow) \\
\end{align*}
\]

\[
\begin{align*}
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes |K\rangle|K\rangle \otimes T_0 \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes |K'\rangle|K'\rangle \otimes T_0 \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes (|K'\rangle|K\rangle + |K\rangle|K'\rangle) \otimes T_- \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes (|K'\rangle|K\rangle - |K\rangle|K'\rangle) \otimes T_+ \\
&(|R\rangle|L\rangle + |L\rangle|R\rangle) \otimes |K'\rangle|K'\rangle \otimes S \\
&(|R\rangle|L\rangle + |L\rangle|R\rangle) \otimes |K\rangle|K\rangle \otimes S \\
&(|R\rangle|L\rangle + |L\rangle|R\rangle) \otimes (|K'\rangle|K\rangle - |K\rangle|K'\rangle) \otimes T_+ \\
&(|R\rangle|L\rangle + |L\rangle|R\rangle) \otimes (|K'\rangle|K\rangle - |K\rangle|K'\rangle) \otimes T_- \\
\end{align*}
\]

\[
\begin{align*}
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes |K\rangle|K\rangle \otimes T_- \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes |K'\rangle|K'\rangle \otimes T_+ \\
&(|R\rangle|L\rangle - |L\rangle|R\rangle) \otimes (|K'\rangle\uparrow|K\downarrow) + (|K\downarrow|K'\uparrow) \\
&(|R\rangle|L\rangle + |L\rangle|R\rangle) \otimes (|K'\rangle\uparrow|K\downarrow) - (|K\downarrow|K'\uparrow) \\
\end{align*}
\]

\[
\begin{align*}
&\approx t^2/4d \\
\end{align*}
\]

\[
\begin{align*}
&\Delta_{SO} = 0.17 \text{ meV} \\
&\Delta_{KK'} = 0 \\
&\Theta = 0 \\
\end{align*}
\]

\[
\begin{align*}
tunneling &= t = 0.03 \text{ meV} \\
\text{spin-orbit} &= \text{SO} = 0.17 \text{ meV} \\
\text{m_orb} &= 0.33 \text{ meV/T} \\
\text{m_spin} &= 0.058 \text{ meV/T} \\
\text{no KK' scattering} \\
\text{no exchange} \\
\theta &= 0 \\
B &= 0
\end{align*}
\]
Pauli blockade in carbon nanotube double dot despite spin-orbit coupling

lowest two-particle states have different spin & valley symmetries

\[
\begin{align*}
&\langle R | L \rangle - \langle L | R \rangle \otimes | K' \rangle | K' \rangle \otimes T_+ \\
&\langle R | L \rangle - \langle L | R \rangle \otimes (| K' \uparrow \rangle | K \downarrow \rangle + | K \downarrow \rangle | K' \uparrow \rangle) \\
&\langle R | L \rangle - \langle L | R \rangle \otimes | K \rangle | K \rangle \otimes T_- \\
&\langle R | L \rangle + \langle L | R \rangle \otimes (| K' \uparrow \rangle | K \downarrow \rangle - | K \downarrow \rangle | K' \uparrow \rangle)
\end{align*}
\]
The lowest electronic states in carbon nanotube quantum dots are characterized by their energy levels and the ability to tunnel between different states. The figure illustrates the T1 sequence, which is a pulse protocol used to manipulate and detect the electronic states of the quantum dots.

In the T1 sequence, the system is pulsed from an initial state to a final state, and the relaxation of the system is monitored. The relaxation time, $\tau$, is defined as the time it takes for the system to relax to the ground state from an excited state.

The figure shows the variation of the sensor conductance, $g_s$, as a function of the applied magnetic field, $B$, and the voltage, $V_R$. The conductance is measured in units of $(e^2/h)$, where $e$ is the charge of an electron and $h$ is Planck's constant.

The figure contains several panels, each illustrating different aspects of the T1 sequence. The panels show the evolution of the conductance with respect to the applied magnetic field and voltage, highlighting the transition between different electron states. The pulse triangle, which is marked by the solid white lines, indicates the sequence of pulses applied to the system.

The data points are marked with blue circles, and the transitions between states are indicated by white arrows. The labels $E$, $M$, and $R$ represent the different states of the quantum dot system. The figure also includes annotations for the relaxation times, $\tau_M$, associated with each pulse cycle.

By analyzing the conductance as a function of time and magnetic field, it is possible to extract information about the electronic states of the quantum dots and their relaxation properties. The T1 sequence is thus a powerful tool for studying the quantum dynamics of nanoscale devices.
B-dependence of relaxation rate

Charge sensing

Transport

Leakage Current (fA) vs $T_M$ (µs)

$g_s(e^2/h)$ vs $B$ (mT)

$\tau_M = 0.5 \mu s$

Leakage Current (fA) vs $B$ (mT)

$\Delta_{SO}$, $\Delta_{kk'}$, $\Delta_{\theta}$

$B_{orb}$, $B_{spin}$
B-dependence of relaxation rate

\[ T_1 \propto \sqrt{\text{splitting}} \]

Bulaev et al. PRB 77, 235301 (2008)
Inhomogeneous Dephasing

\[ T_2^* = \frac{\hbar}{g\mu_B} \delta B_{\text{nuc}}^{||} = 3.2 \text{ ns} \]

\[ \delta B_{\text{nuc}}^{||} = 1.8 \text{ mT} \approx B_{\text{nuc}}/2 \]
Summary

nanotubes can form gate-controlled dots with controlled hyperfine coupling

few-electron regime accessible using charge sensing readout

Both many-electron Pauli Blockade and two-electron T2* measurements indicate large hyperfine coupling in nanotubes