Coherent control and probing of artificial atoms and molecules

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Supported by
US DOE & NSF NIRT
CONACYT, Mexico
Current Topics

• Quantum dots – confinement vs interactions – artificial atoms
  – Coulomb blockade & assorted IV characteristics
  – Optical effects – excitons and effects of external fields

• Photons, spins, energy transfer & entanglement in/between quantum dots

• Nanomagnetics and nanospintronics: Kondo effect
Quantum Dots: $L \sim \lambda$

good things come in small packages

Confinement: $\text{KE} \sim k^2 \sim L^{-2}$

Interactions: $\text{PE} \sim q^2/L$

For $L \sim 100\text{nm}$, $\text{PE} \sim 1\text{meV}$

while $\text{KE} \sim 0.5\text{meV}$ (in GaAs)

For $L \sim 5\text{nm}$, $\text{PE} \sim 20\text{meV}$; $\text{KE} \sim 200\text{meV}$

Total energy

$E = \text{KE} + \text{PE}$
Quantum dot fabrication

Self-assembly

- Stranski – Krastanow islands
- MBE
- in-plane densities
  \( \sim 10^{10} - 10^{11} \text{ cm}^{-2} \)
- size variations < 10%
- sharp photoluminescence features, frequency \( \propto \) size

\[\text{Figure 1} \text{ Scanning electron micrographs illustrating the experimental technique used for studying single self-assembled quantum dots. a, Scanning electron micrograph of a GaAs semiconductor layer on which In}_{0.65}\text{Ga}_{0.35}\text{As self-assembled quantum dots with a density of about } 10^{10} \text{ cm}^{-2} \text{ have been grown by molecular beam epitaxy. To permit their microscopic observation these dots—unlike those used for spectroscopy—have not}\]

Quantum dot fabrication

Colloidal dots

- Chemical synthesis
- CdSe, CdS, InP, etc.
- Size ~ 5nm diameter
- Uses for biotags

Quantum Dot Corp.
Quantum dot fabrication

Lithographic dots

Ensslin et al - ETHZ

Kouwenhoven et al - Delft
QD w/tunable size and e\textsuperscript{-} number
~ artificial \textbf{atoms}

**Electronic transport** -
- Low bias: ground state – \textbf{Coulomb blockade}
- High bias: excited states – selection rules(!)
- Rings, \textit{phases} & resonances

**Incident/outgoing photons** -
- Visible/optical: \textbf{exciton}
- Far infrared: internal \textbf{multi-electron} excitations
- \textbf{Raman}: excitons/confined phonons

**Capacitance** -
- Ground state vs B-field
- Combination w/optics & in-plane transport
FIG. 5. Current as a function of gate voltage $V_G$ and source-drain voltage $V_{DS}$, for different values of interdot tunneling: (a) $t = 0.01$, (b) $t = 0.1$, and (c) $t = 0.2$. Symmetric DDS case.

QD molecules

Blick et al. Science 2002

in series

future? multidots for quantum computing

in parallel

Ramirez et al., PRB 1999
Manipulating few electrons in a 2D SET array

Andreas Weichselbaum & SU
Motivation

- Charge qubit in a solid state device? (in lieu of spin)
- Array of coherently interacting islands
- Electrostatic control via gate voltages
- Constant number of excess electrons on array
- Q: given a starting configuration $\psi_0$, how to transform to a final state $\psi_f$ that is clearly distinguishable spatially?
Hamiltonian (Hubbard type)

\[ H = - \sum_{<i,j>,\sigma} t_\sigma c_{i\sigma}^+ c_{j\sigma} + \frac{1}{2} \sum_{i,j} \hat{n}_i V_{ij} \hat{n}_j + \mathcal{H}_{\text{ext}} (\text{gate voltages}) \]

\[ = - \sum_{<i,j>,\sigma} t_\sigma c_{i\sigma}^+ c_{j\sigma} + \frac{e^2}{2} \hat{n}_j C^{-1}_{(11)} \hat{n}_j + e \hat{n} C^{-1}_{(11)} \left( C_{12} \mathbf{V}_g \right) \]

electrostatic Gibbs free energy of the system

with a capacitance matrix

\[ C_{\text{total}} = \begin{pmatrix} C_{\text{sites}} & C_{\text{sites-leads}} \\ C_{\text{leads-sites}} & C_{\text{leads}} \end{pmatrix} \equiv \begin{pmatrix} C_{(11)} & C_{12} \\ C_{21} & C_{22} \end{pmatrix} \]

and elements

\[ C_{ii} = C_{\text{stray}} + \sum_{j=\text{sites,leads}} C_{i\to j} \equiv C_{\Sigma,i} \]

\[ C_{ij} = -C_{i\to j} \]
Geometries

one electron potential:

→ “quantum well”

→ “quantum barrier”
Eigenenergies (2e)

Structure #1; quantum well

Structure #2; quantum barrier

Energy splitting: ground state (singlet) to ...

1\textsuperscript{st} excited singlet: 2.86 meV ~ 33.2 mK

lowest triplet: 1.57 meV ~ 18.2 mK

$t_\sigma = 10 \, \mu eV \sim 0.12 \, K \sim 2.4 \, GHz$

$C_{ij} = C_{ig} = 45 \, aF \ (U \sim 1 \, meV; \ V_{12} \sim 0.2 \, meV)$
Eigenspectra (2e)

Gate sweep for spectrum change
Array dynamics

**Singlet (ground) state**

- Singlet ground state is singly degenerate (*anti-crossing*)
- Very small applied voltages can make use of this anti-crossing to get *quantum beating* within effective 2-level subspace
- However, rotation to the second state cannot be made absolutely perfect …
Single qubit gates

NOT gate $X$ "flips" qubits: $X|0\rangle = |1\rangle$ and $X|1\rangle = |0\rangle$

Need to implement "real" $H$ and two-bit gates such as a CNOT
A. Weichselbaum & SU

![3D diagram of $V_{pot}$ (meV) with voltages]

![Diagram of singlet and triplet states]

$b)$ $\Delta V_{pot}$ ($\mu$eV)
Klimov – Förster coupling between QDs

Non-Radiative Energy Transfer Mechanism

\[ D^* + A \rightarrow D + A^* \]

Coulomb-driven interaction

Dipole-dipole interaction (Förster 1946)
Higher multipoles interaction (Förster – Dexter)

Exchange-driven interaction (Dexter)
A acceptor: larger dot
D donor: smaller dot

NQDs have better characteristics than biological light harvesting compounds, eg LH2

<table>
<thead>
<tr>
<th>c)</th>
<th>resonant NQDs</th>
<th>LH2 [Ref.12]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dipole $\mu_A$ ($\mu_D$)</td>
<td>25 (4.4) Debye</td>
<td>8.2 (8.2) D</td>
</tr>
<tr>
<td>Distance $R_{DA}$</td>
<td>54 Å</td>
<td>18 Å</td>
</tr>
<tr>
<td>Coupling $J$</td>
<td>2.4 cm(^{-1})</td>
<td>26 cm(^{-1})</td>
</tr>
<tr>
<td>Overlap integral $\Theta$</td>
<td>0.004 cm</td>
<td>0.0004 cm</td>
</tr>
<tr>
<td>Estimated rate $\Gamma_{et}$</td>
<td>(38 ps(^{-1}))</td>
<td>(3 ps(^{-1}))</td>
</tr>
</tbody>
</table>

trioctylphosphine oxide (TOPO) $\sim$11 Å
(a) PL decays from a dense film of monodisperse R=12.4A/9A CdSe/ZnS NQDs at the energies specified in the inset. Inset: cw PL spectra from film (solid) and original solution (dashed).

(b) Dynamic redshift of the peak emission. Inset: PL spectra at the specified times.

Crooker et al PRL 2002
(a) Schematic of NQD energy-gradient bilayer for light harvesting—13 Å dots on 20.5 Å dots.

(b) “Instantaneous” PL spectra at 500 ps intervals (from 0 to 5 ns), showing rapid collapse of emission from 13 Å dots.
Coherent Spin Transfer Between Molecularly Bridged Quantum Dots

Min Ouyang and David D. Awschalom*

SCIENCE VOL 301 22 AUGUST 2003

Femtosecond time-resolved Faraday rotation spectroscopy reveals the instantaneous transfer of spin coherence through conjugated molecular bridges spanning quantum dots of different size over a broad range of temperature. The room-temperature spin-transfer efficiency is ∼ 20%, showing that conjugated molecules can be used not only as interconnections for the hierarchical assembly of functional networks but also as efficient spin channels. The results suggest that this class of structures may be useful as two-spin quantum devices operating at ambient temperatures and may offer promising opportunities for future versatile molecule-based spintronic technologies.

Fig. 1. Multilayer CdSe QD artificial solids. (A) Schematic bilayer configuration with two QDs of different sizes denoted as AB. (B) Typical optical absorption spectra taken at 4.5 K of sample ABAABA (black), isolated 7.0-nm CdSe QDs in PVB matrix (red), and isolated 3.4-nm CdSe QDs in PVB matrix (green). Blue and red arrows highlight the positions of absorption peaks corresponding to the first exciton states of 3.4- and 7.0-nm QDs, respectively. (Inset) Typical AFM image (170 nm by 170 nm) of CdSe QD monolayer structure. High-resolution transmission electron microscopy (TEM) images (scale bar, 5 nm) and schematic energy diagrams of individual 7.0- and 3.4-nm CdSe QDs are also shown.
Our Goal

Study the excitation energy transfer in quantum-dot arrays using an appropriate model Hamiltonian

\[ H = \sum_{i,j} \left( T_e c_i^\dagger c_j + T_h d_i^\dagger d_j \right) + \sum_i N U c_i^\dagger c_i d_i^\dagger d_i + \sum_{i,j=NN} U_{NN} c_i^\dagger c_i d_j^\dagger d_j + \sum_{i \neq j} N V_s c_i^\dagger d_j^\dagger d_j c_j \]
The “movie” of the 24 dots

exciton probability at each dot

V_c t/2π
This term is defined as

\[ V_F = \frac{\mu_D \mu_A}{\varepsilon_0 R^3} \kappa \]

The dipole moments entering \( V_F \) are modeled by a hard well confinement potential, proven successful in the description of these quantum dots with rescaling the gap.

Estimation of Förster coupling as a function of dot size
The acceptor is shown to act as an ideal “eavesdropping” point that can effectively monitor the coherent (Rabi) oscillation between the two donor dots without collapsing them.

*Time Evolution of a Trimer Cluster*

The system oscillates with a characteristic time of the splitting between two levels with detuning, what is the characteristic damping time?
Energy Diagram for Trimer

\[ \delta \]

- Donor
- Donor
- Acceptor

\( V^b_F \)
\( V^d_F \)
\( V^{db}_F \)

\( \Gamma_D \sim 1\,ps \)

\( \Gamma_A \)
$\delta$  
\[ \Gamma_D \]  
\[ V_{F}^{db} \]  
\[ \Gamma_A \]  
\[ \delta \]  
\[ \Gamma_A \]  
\[ V_{F}^{db} \]  
\[ \Gamma_{eff} \]  
\[ \Gamma_{A} \gg V_{F}^{db} \]  
\[ \Gamma_{eff} = \frac{\left(V_{F}^{db}\right)^2 \Gamma_A}{\left(\frac{\Gamma_A}{2}\right)^2 + \delta^2} \]  
Valid for Dimer
Probability of finding lower state exciton at the acceptor dot

$\delta = 0$

Excitation started at D1

a)

Excitation started at D2

$\delta = 0$

b)

Excitation started at D1

$\delta = 0.1$ mev

c)

Excitation started at D2

$\delta = 0.1$ mev
d)
Conclusion

- The dynamics of excitonic states in dimer and trimer arrangements of colloidal quantum dots using the density matrix approach.

- The dots are assumed to be in close proximity, thanks to molecular linkers or spacers that allow dipole Förster coupling but yet prevent direct carrier hopping.

- Our results for the dimer clusters show that the effective damping of a donor dot depends on the Förster coupling (itself a function of dot separation and sizes), and on the width of the bright exciton state of the acceptor dot.

- The Rabi oscillations of the tuned donors appear as plateaus in the acceptor level, and therefore in the photoluminescence response.
Zrenner – coherent control of quantum dot photodiodes

Possibles measurements

- Photoluminescence (PL) spectrum (for $\tau_{\text{rad}} < \tau_{\text{tunnel}}$)
- Photocurrent (PC) spectrum (for $\tau_{\text{rad}} > \tau_{\text{tunnel}}$)

Diagram:
- Laser focus
- Shadow mask
- Top contact: 5nm Ti (semitransparent)
- QDs InGaAs
- Back contact

Device layers:
- i-GaAs-Buffer
- n-GaAs-Substrate
Rabi Oscillation in a two level system

For $\omega = \omega_0$ and using RWA

$$H = \frac{\hbar \omega_0}{2} \sigma_z + \frac{\hbar \Omega(t)}{2} \cos(\omega t) \sigma_x$$

$$\Omega(t) = \frac{\bar{\mu} \cdot \bar{\varepsilon}(t)}{\hbar}$$

$$P_{0 \rightarrow X} = \sin^2 \left( \frac{\Theta}{2} \right)$$

$$\Theta = \int_{-\infty}^{\tau} \Omega(t) dt$$

Exciton Population

Pulsed Area ($\Theta$)
How does the Zrenner device work?

\[ \tau_{\text{pulse}} \approx 1 \text{ ps} \]

\[ f_{\text{pulse}} \approx 82 \text{ MHz} \]

\[ I = e \rho_{XX} f_{\text{pulse}} \]

\[ \rho_{XX} = \sin^2 \left( \frac{\Theta}{2} \right) \]
Rabi Oscillation in the photocurrent

\[ I = e \rho_{XX} f_{\text{pulse}} \]

\[ \hbar \omega \]

\[ |x\rangle \]

\[ |0\rangle \]
What we are doing...

Estimating the tunneling time

Which one tunnels first, electron or hole? And, after one tunnels, how long before the other tunnels? What is the effect of Coulomb interaction?

\[
W = \frac{2\pi}{\hbar} \sum_\alpha \langle \psi_\alpha | V_{\text{Dot}} | \psi_0 \rangle \delta(E_\alpha - E_0)
\]

\[
\tau_{\text{tunnel}} = \frac{1}{W}
\]


Modeling in terms of density matrix with tunneling

- How much was population inverted?
- Can this be controlled by gates? Detuning?
We study: coupled DQD molecule

Shadow mask

$\hbar \omega$

Top contact (semitransparent)

Self-assembled QDs

Back contact

$V$

(a)

(b)
Model Hamiltonian

\[ H = \sum_j \varepsilon_j |j\rangle \langle j| + T_e (|1\rangle \langle 2| + |2\rangle \langle 1|) + \hbar \Omega \left( e^{-i\omega t} |0\rangle \langle 1| + e^{i\omega t} |1\rangle \langle 0| \right) \]

\[ \Omega(t) = \langle 0| \frac{\vec{\mu} \cdot \vec{E}(t)}{2\hbar} |1\rangle \]
Unitary transformation

\[ |\psi\rangle = \exp \left[ -\frac{i\omega t}{2} \left( |1\rangle\langle 1| - |0\rangle\langle 0| + |2\rangle\langle 2| \right) \right] |\psi'\rangle \]

\[ H' = \frac{1}{2} \begin{pmatrix} -\delta_1 & 2\Omega & 0 \\ 2\Omega & \delta_1 & 2T_e \\ 0 & 2T_e & \delta_2 \end{pmatrix} \]

\[ \delta_1 = \omega_{01} - \omega \]
\[ \delta_2 = 2\omega_{12} + \delta_1 \]

(c)
Results for $\delta_1 = \delta_2 = 0$, time evolution

$$\lambda_0 = 0,$$

$$\lambda_{\pm} = \pm \sqrt{\Omega^2 + T_e^2}$$

$$|\lambda_0\rangle = \cos \theta |0\rangle - \sin \theta |2\rangle,$$

$$|\lambda_{\pm}\rangle = \frac{1}{\sqrt{2}} (\sin \theta |0\rangle \pm |1\rangle + \cos \theta |2\rangle)$$

$$\cos \theta = \frac{T_e}{\sqrt{\Omega^2 + T_e^2}}$$

Ocupation Probability

$$P_0(t) = |\sin^2 \theta \cos(\Omega't) + \cos^2 \theta|^2,$$

$$P_1(t) = \sin^2 \theta \sin^2(\Omega't),$$

$$P_2(t) = \sin^2 \theta \cos^2 \theta |\cos(\Omega't) - 1|^2$$

$$\Omega' = \sqrt{\Omega^2 + T_e^2}$$
Varying the gate voltage

(a) $T_e = 0.01 \omega$
$\Omega = 0.05 \omega$
$\delta_1 = 0$

(b) $T_e = 0.01 \omega$
$\Omega = 0.05 \omega$
$\delta_1 = 0.1 \omega$
Average occupation of state $|2>$
Varying the pulse intensity

Enhance the tunneling between the dots

Supress the tunneling between the dots

\( \delta_1 = 0 \)
\( T_e = 0.01 \omega \)

\( \omega_{12} = 0 \)
\( \omega_{12} = 0.03 \omega \)
\( \omega_{12} = 0.06 \omega \)
Summary

★ Coupled QD molecule

★ Analytical solution

★ Controlling the tunneling by tuning some parameter as pulse intensity

J. M. Villas-Bôas, A. Govorov, and S. E. Ulloa, cond-mat/0403445
Support & Collaborations

- National Science Foundation NIRT & IMC
- Department of Energy
- Indiana 21st Century Fund

- Klaus Ensslin, ETH Zurich, Switzerland
- Enrique Anda, PUC-Rio, Brazil
- Pablo Tamborenea, U Buenos Aires, Argentina
- Ernesto Cota, CCMC-UNAM Ensenada, Mexico
- Gilmar Marques & Nelson Studart, UF Sao Carlos, Brazil

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