NANOESTRUCTURAS
V Escuela Nacional de Física de la Materia Condensada

Parte III

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Photons, excitons, spins, energy transfer & entaglement ….. all in QDs

- Gammon/Steel – optical control of quantum dot state
- Imamoglu – single photon source from quantum dots
- Klimov – Förster coupling between quantum dots
- Zrenner – coherent control of quantum dot photodiode
Naturally Formed GaAs QDs

- 42 Å GaAs layer
- 250 Å Ga$_{0.3}$Al$_{0.7}$As layer
- 500 Å GaAs cap layer
- 500 Å Ga$_{0.3}$Al$_{0.7}$As layer

STM image showing the island formation and elongation.


Samples are provided by D.S. Katzer, D. Park, and E. S. Snow, NRL
Photoluminescence of a Single Exciton

Weak field limit:

\[ H_{int} = g_{ex}^* \mu_B B_0 S_z + \alpha B_0^2 \]

\[ g_{ex}^* = g_e^* + g_{hh}^* \]

\[ g_{hh}^* = 6\kappa + \frac{27q}{2} \]

Excitons

Excited excitons (s- and p-shell)
Biexcitons, $X^-$, $X^{--}$, etc.

Photoluminescence in SINGLE QD

**Fig. 1.** Power dependent PL spectra from a single isolated quantum dot at zero magnetic field. Contributions from the s-shell and p-shell can be clearly distinguished. In the spectral region of the s-shell, the single exciton (1X) and biexciton lines (2X) are labelled.
Experimental Techniques

• **Linear Spectroscopy**: Photoluminescence
  Indirect probe of exciton resonances
  Requires spectral diffusion of excited carriers

• **Coherent Nonlinear Spectroscopy**:
  CW differential transmission
  Resonant excitation
  Probe coherent interaction in the system

\[ E_{NL}(\varepsilon) \sim \chi^{(3)}(\varepsilon) \, E_{pump} \, E^*_{pump} \, E_{probe} \]

\[ I_{signal} = \text{Re} \left( E_{NL} E^*_{probe} \right) \]

D Steel et al U Michigan
Non-degenerate Nonlinear Spectroscopy: Advantages

• Probe single excitonic state decay dynamics
  Measure both $T_1$ and $T_2$ *

• Probe coupling between different excitonic states
  Probe inter-dot energy transfer and dot-dot coupling
  Study excited states of excitons
  Probe multi-exciton correlation effects ;
  Study the coherent interaction between exciton doublet .

* Nicolas H. Bonadeo et al., PRL, 81, 2759 (1998)
Non-degenerate experiments can excite both $\sigma^+$ and $\sigma^-$ excitons by varying the frequency and the polarization of the excitation beams (pump and probe beam).
Exciting Two Electrons

Two contributions:

1. Incoherent:
   Ground state depletion

2. Coherent:
   Zeeman coherence between $\sigma^-$ and $\sigma^+$ state

Resonant and coherent excitation of two electrons

3-level diagram in 2-electron basis

\[
\begin{align*}
\sigma^- & : |-\frac{3}{2}\rangle + |\frac{1}{2}\rangle \\
\sigma^+ & : |-\frac{1}{2}\rangle + |\frac{3}{2}\rangle \\
\text{Pump} & : \sigma^- \\
\text{Probe} & : \sigma^+ \\
\text{Ground state} & : |\frac{3}{2}\rangle + |\frac{1}{2}\rangle
\end{align*}
\]
Optically Entangling Two Systems: the Importance of Coupling

- Without coupling → Product state of the two subsystems.
- A strong coupling allows one system to see the excitation of the other.

Coulomb interaction between charged particles: trapped ions

Magnetic dipole interaction: NMR systems

Exciton-exciton Coulomb coupling: excitons

- Mutual coherence between E and E′ is essential.
Coulomb Correlation*:
Coulomb interaction between electron-hole pairs within single QD

Two electrons are involved within a single QD.
Coulomb interaction between the two excitons is important.

But

Two-exciton state cancels the signal due to the symmetry in the level diagram.

Scattering states

Bound state of multi-excitons

σ+ state

σ− state

Pump
σ−

Probe
σ+

|−1/2⟩|+1/2⟩

|−3/2⟩|+1/2⟩

|−3/2⟩|+3/2⟩

|−1/2⟩|+3/2⟩

|−3/2⟩

|+1/2⟩

|+3/2⟩

Turn on the Coulomb Correlation

Pump $\sigma^-$

Probe $\sigma^+\ gamma$

Ground-state Depletion

Zeeman Coherence

Total Signal

No Signal !!
Evidence for Zeeman Coherence in Single QDs

Creation of two-electron entanglement
**Ultimate Goal**

- **Combine:**
  - Optical control of individual QDs
  - Long spin lifetimes
  - QD nanostructure engineering

- **To produce:**
  - Qubit register of QD spins

Coherence of a single QD can be controlled – Gammon et al have demonstrated this for excitons [Stievator et al., PRL (2001)]  
Next: QNOT and other quantum gates

Spins have long coherence times: dephasing times $T_{2}^{*} = 5\text{ns-300ns}$  
[Dzhioev et al., PRL (2002)]  
Next: explore in single QDs; optical read-out and initialization schemes
Coupling and Entangling of Quantum States in Quantum Dot Molecules

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Science 2001
Imamoglu – single photon source from QDs

Michler et al., Science 290, 2282 (2000)

- It is difficult to isolate a single photon, or fix the number of photons in a pulse
- Fluctuations in photon number mask the quantum features of light
- A stable train of laser pulses have Poissonian (photon number) statistics

→ It is desirable to have *single photon* sources:

![Single photon turnstile]

Complete regulation of photon generation
Single Photon Sources

Quantum Cryptography: secure key distribution by single photon pulses
Quantum Computation: single photons + linear optical elements

Available sources:
- Highly attenuated laser pulse ⇒ Poisson fluctuations
- Parametric down conversion ⇒ Random generation of single photons

Possible solution: Deterministic (triggered) single photon emission:
Single Photon Turnstile Device

Experiments:
Coulomb blockade of electron/hole tunneling in a mesoscopic pn-junction:

Single Molecule at room temperature:

Single InAs Quantum Dot in a microcavity:
Signature of a triggered single-photon source

- Intensity (photon) correlation function:
  \[ g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2} \]

- Single quantum emitter (i.e. an atom) driven by a cw laser field exhibits photon antibunching.

- Triggered single photon source: absence of a peak at \( \tau = 0 \) indicates that none of the pulses contain more than 1 photon.
Photon antibunching

- Intensity correlation \( g^{(2)}(\tau) \) of light generated by a single two-level (anharmonic) emitter.

- Assume that at \( \tau=0 \) a photon is detected:
  - We know that the system is necessarily in the ground state \(|g>\n  - Emission of another photon at \( \tau=0+\varepsilon \) is impossible.
  \Rightarrow Photon antibunching: \( g^{(2)}(0) = 0 \). (nonclassical light)

- \( g^{(2)}(\tau) \) recovers its steady-state value in a timescale given by the spontaneous emission time.

- If three are two or more 2-level emitters, detection of a photon at \( \tau=0 \) can not ensure that the system is in the ground state \( g^{(2)}(0) >0.5\).
Single InAs Quantum Dots

InAs/GaAs Single QD

P (W/cm²)

s-Shell

p-Shell

T=4K

Energy (eV)

Intensity (a.u.)

P (W/cm²)

650

400

210

105

55

17

2X

1X

x2 in intensity

x5 in intensity

Two main wavelengths emitted from each shell
(For s-Shell)

• 2X recombination while there is already an e-h pair in the s-shell (biexciton)
• 1X recombination while there is no other e-h pair in the s-shell (exciton)

Due to carrier-carrier interaction
Typically $h\nu_{1X} = h\nu_{2X} + 2-3\text{meV}$

Unique wavelengths for 1X and 2X transitions
Exciton linewidth measured by a scanning Fabry-Perot

Under non-resonant pulsed excitation

Free Spectral Range: 62 µeV

linewidth: 5.6 µeV
Photon antibunching from a Single Quantum Dot

105 W/cm²
\[ g^2(0) = 0.1 \]
\[ \tau = 750 \text{ ps} \]

55 W/cm²
\[ g^2(0) = 0.0 \]
\[ \tau = 1.4 \text{ ns} \]

15 W/cm²
\[ g^2(0) = 0.0 \]
\[ \tau = 3.6 \text{ ns} \]

\[ \rightarrow \text{proof of atom-like behavior} \]
A single quantum dot excited with a short-pulse laser can provide single-photon pulses on demand

BUT

How about embedding quantum dots in a microcavity to increase collection efficiency and fast emission?
Microdisk Cavities

No roughness on the sidewall up to 1nm!
Q > 18000 for 4.5 μm diameter microdisk
Q = 9000 for 2 μm diameter microdisk


Fundamental whispering gallery modes cover a ring with width ~ λ/2n on the microdisk
A single quantum dot in a microdisk

Larger width of the peaks due to larger lifetime of the quantum dot

P = 20 W/cm²
T = 4 K

Q = 6500

Pump power well above saturation level
Tuning the quantum dot into resonance with a Cavity Mode

Cavity coupling can provide better collection

- Small peak appears at $\tau=0$:
- **Purcell effect**: reduction of emission time
Klimov – Förster coupling between QDs

Non-Radiative Energy Transfer Mechanism

D* + A

Coulomb-driven interaction

D + A*

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

Exchange-driven interaction (Dexter)
NQDs have better characteristics than biological light harvesting compounds, eg LH2

trioctylphosphine oxide (TOPO) ~11Å

A acceptor: larger dot

D donor: smaller dot
(a) PL decays from a dense film of monodisperse R=12.4Å/9Å 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 A dots on 20.5 A dots.

(b) “Instantaneous” PL spectra at 500 ps intervals (from 0 to 5 ns), showing rapid collapse of emission from 13 A dots.
Our Goal

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

\[ H = \sum_{i,j} N (T_e c_i^\dagger c_j + T_h d_i^\dagger d_j) + \sum_i N c_i^\dagger c_i d_i^\dagger d_i^\dagger + \sum_{i,j=NN} U_{NN} c_i^\dagger c_j^\dagger d_i d_j + \sum_{i,j} V_s c_i^\dagger d_i^\dagger d_j c_j \]
Calculations for two dots

Probability of each basis state as a function of time

Period of small oscillation \( \sim 1.3 \sim \frac{1}{2T_e/V_c} \)

Period of large oscillation \( \sim 15 \sim \frac{1}{2V_f/V_c} \)

Probability of each basis as a function of time without Förster

Probability of each basis as a function of time without tunneling
Time evolution of the oscillator strength of an exciton initially localized at dot 12 in a 24 dot chain

The “movie” of the 24 dots

exciton probability at each dot
Förster rate for polymer

\[
\bar{t}_{n \rightarrow n+1} = \frac{h}{8|U|} = \frac{2\pi \hbar/V_c}{8|U|/V_c} \approx 3.1 \frac{2\pi \hbar}{V_c}
\]

From graph/calculation

\[
\bar{t} \approx 2.2 \frac{2\pi \hbar}{V_c}
\]

efficient interdot transfer rate
Zrenner – coherent control of quantum dot photodiodes

Zrenner et al. Nature 2002

Possibles measurements

★ Photoluminescence (PL) spectrum (for \( \tau_{\text{rad}} < \tau_{\text{tunnel}} \))
★ Photocurrent (PC) spectrum (for \( \tau_{\text{rad}} > \tau_{\text{tunnel}} \))
Rabi Oscillation in a two level system

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

\[ \Omega(t) = \frac{\vec{\mu} \cdot \vec{E}(t)}{\hbar} \]

For \( \omega = \omega_0 \) and using RWA

\[ P_{0 \to X} = \sin^2 \left( \frac{\Theta}{2} \right) \]

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

Exciton Population

Pulsed Area (\( \Theta \))
Time scales for the device

\[ \tau_{\text{rad}} \approx 1 \text{ ns} \quad \tau_{\text{tunnel}} \approx 3 \text{ ps} \rightarrow \infty \]

Tuned by Vb

**Photoluminescence** \( \tau_{\text{rad}} < \tau_{\text{tunnel}} \)

NeHe laser

**Photocurrent** \( \tau_{\text{rad}} > \tau_{\text{tunnel}} \)

Ti:sapphire laser

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

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

Artificial ion (charged exciton)

$\tau_{\text{rad}} < \tau_{\text{tunnel}}$

How does the Zrenner device work?

\[ \tau_{\text{rad}} > \tau_{\text{tunnel}} \]

How does the Zrenner device work?

- Pulse $\tau_{\text{pulse}} \simeq 1 \text{ ps}$
- Frequency $f_{\text{pulse}} \simeq 82 \text{ MHz}$

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

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

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

\[ \hbar \omega \]

\[ |10\rangle \rightarrow |1x\rangle \rightarrow |10\rangle \]
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?

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

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


Modeling in terms of density matrix with tunneling

★ How much was population inverted?
★ Can this be controlled by gates? Detuning?