Ultrafast Spectroscopy of Quantum Dots (QDs)

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Outline:

- 1. Types of QDs and Techniques of Ultrafast Spectroscopy
Outline:

2. Application Aspects: Dynamics of Amplification in QD-Lasers

Monitoring of high-frequency optical operation in semiconductor nanostructures by ULTRAFAST SPECTROSCOPY

predicted advantages of QD-lasers:

- low threshold current density
- high characteristic temperature
- high differential gain
- large spectral tunability, from NIR to UV

D. Bimberg and coworkers, TU Berlin
3. Fundamental aspects: Semiconductor QDs as artificial atoms

Monitoring of the „discrete-level“ - structure of semiconductor nanostructures by ULTRAFAST SPECTROSCOPY

L. Banyai, S. W. Koch, *Semiconductor Quantum Dots*
Part 1: Types of QDs and Techniques...

Quantum Dots: Nanocrystals and epitaxially grown Islands

Precipitation of spherical nanocrystals in colloidal solution or glass, polymer etc. matrix

Lattice-mismatch induced island growth
CdSe QDs emitting in the visible (nanocrystals)

Wavelength (nm)

Photon Energy (eV)

CdSe QDs
T = 4.2 K
R = 38 nm

Optical Density

PL Intensity (arb. units)

CdSe QDs
R = 1.5 nm
T = 300 K

CdSe QDs
R = 2 nm
T = 300 K

CdSe in glass

5 nm
InGaAs self-assembled islands emitting in the NIR

Calculated confined eh-pair energies for InAs assuming pyramidal shape

D. Gerthsen et al., Karlsruhe

Grundmann, Bimberg et al., TU Berlin
Part 1: *Types of QDs and Techniques...*

**Femtosecond Heterodyne Technique**

![Diagram of femtosecond heterodyne technique](image)

- \(\omega_2\) probe
- \(2\omega_2 - \omega_1\) four-wave mixing (FWM)
- \(\omega_1\) pump
- Signal
- Waveguide
- Ti:Sa + OPO, 80 fs ... 2 ps
Femtosecond Ultrafast Spectroscopy

Usually:

![Diagram showing Ultrafast laser, Pump, Probe, Delay, Sample, Pump-probe reflection signal, FWM-signal, Pump-probe transmission signal, and Luminescence correlation signal.]

J. Shah, Ultrafast Spectroscopy
Femtosecond Heterodyne FWM- and PP-Spectroscopy

Usually:

Here:
AOM- Acousto-Optical Modulator

\[ I_{\text{det}} \propto \varepsilon_{\text{ref}} \varepsilon_{\text{signal}} \]

\( \varepsilon = \) electric field

150fs 76MHz laser

AOM1 79MHz

AOM2 80MHz

HF-Lock-in delay

probe beam

sample

pump beam

FWM 2MHz 3MHz 4MHz
Part 2: *Applied aspects: QD-laser...*

**Gain Dynamics in Quantum Dots**

InAs/InGaAs QDs
3 x stack,
20nm GaAs barrier
Gain Dynamics of InGaAs QDs

Ground State Emission (GS):
1070nm @ 25K, 1170nm @ 300K

Sample from TU Berlin, Prof. Bimberg

- ridge waveguide
  5x500μm,
  3 stacked QD layers

- areal dot density
  ~2x10^{10}cm^{-2}

- optical density ~ 1.5
  (α~30cm^{-1})

- Carrier injection electrically (0...20 mA)
Gain Dynamics of InGaAs QDs

Pump-induced gain change in a heterodyne pump-probe experiment at maximum gain (20 mA) and without electrical injection (0 mA)

Gain recovery in < 100 fs at 300 K!

**Gain Dynamics of CdSe QDs**

Ground State Emission (1): 605 nm @ 6K

Gain Dynamics of CdSe QDs

Gain recovery time spectrally varying, <1...100ps
Gain Dynamics of CdSe QDs

Gain spectrum inhomogeneously broadened:
Spectral hole burning in gain spectrum with two fs-pump and one fs-probe beam

Spectral hole width of a single gain process
~20 meV

Intrinsic limit of gain recovery below 100 fs!

Part 2: *Applied aspects: QD-laser...*

**Quantum dots as active media in optical microcavities**

CdSe QDs linked to microspheres

Picture: M.V. Artemyev, I. Nabiev
"Dot - in - a - Dot" - Structure

CdSe nanodot

Glass microsphere

Cavity Modes of a CdSe-doped Microsphere

WGM

\[ \text{TM, } \ell=36, n=1 \]

\[ \text{TM, } \ell=36, n=2 \]

\[ R_{PD} = 2.5 \, \mu\text{m} \]

\[ R_{QD} = 2.5 \, \text{nm} \]

\[ 1.7814 \, \text{eV} \]

\[ 1.80605 \, \text{eV} \]

\[ 0.00009 \, \text{eV} \]

\[ 0.00008 \, \text{eV} \]

\[ Q=19793 \]

\[ Q=22573 \]

Optical Pumping of a CdSe-doped Microsphere

R_{PD} \sim 15 \mu m

cw-Ar laser, 488 nm
Excitation spot size 40 \mu m^2
T = 300 K
520 \text{ nm} < \lambda_{em} < 640 \text{ nm}

10 mW
CdSe nanocrystals (not on microsphere)

14 mW

See also:
Artemyev, Woggon et al.
Nano Letters 1, 309 (2001)
Part 3: *Fundamental aspects: Artificial atoms...*

**Rabi Oscillations in Quantum Dots**

![Bloch-sphere: population oscillation](image)
Rabi-Oscillations in Atoms

**Simple model: two coupled oscillators**

\[
\begin{align*}
|g> & |e> \\
3> & 2> \\
|1> & |0>
\end{align*}
\]

**Photon field**

**Atom states**

\[
\begin{align*}
|e> & |g> \\
\text{\vdots} & \\
|3> & |2> \\
|1> & |0>
\end{align*}
\]

**Rabi frequency**

\[
\omega_R = \frac{\vec{\mu} \cdot \vec{E}_0}{\hbar}
\]

**Two-level system in resonance with photon field**

\[
\begin{align*}
E_b & \quad \vec{E}_0 = \hbar \omega_0 = E_b - E_a \\
E_a & \quad E_0 : \text{electromagn. field vector} \\
\hbar \omega_0 & : \text{transition energy} \\
\vec{\mu} & : \text{transition dipole moment}
\end{align*}
\]
Here pulsed excitation!

Pulse area: time-integrated Rabi frequency (~ input field intensity)

Occupation probability of the ground (excited) state

Population oscillation
blue = -1
red = +1

Initial conditions:
for \( t \ll -t_0 \) in ground state
No dephasing!

\[
\theta = \int_{-\infty}^{+\infty} \frac{\mu \cdot \vec{E}_0}{\hbar} \, dt
\]
Rabi Oscillations in Rb-Atoms

Effect of Dephasing $T_2$ on Rabi oscillations

The effect of a damping $\gamma = 1/T_2$ of polarization:

$|b|^2$ vs $\omega_R t/2\pi$

Population flopping over many periods is possible in systems with long dephasing times and large transition dipole moments: $\gamma / \omega_R << 1$. 
Dephasing time $T_2$ of InGaAs quantum dots

From 300K to 100K the FWM decay is dominated by a short dephasing time $< 1\text{ps}$

Below $T=10 \text{K}$ a slow dephasing time $> 500 \text{ ps}$ is observed (suppression of LO-phonon scattering!)

Is the observed dephasing time $T_2$ large enough to observe population flopping, i.e. Rabioscillation in QDs ?

Rabi Oscillations in InGaAs Quantum Dots

**Experiment**

Use of spectrally shaped ps-pulses

⇒ a sharpened distribution of the spectral intensity improves the visibility of the oscillations.

**Rabi oscillation:**
two oscillation maxima can be clearly distinguished

Borri et al., Phys. Rev. B (Rapid Comm.), in press
Distribution in Transition Dipole Moments $\mu$

\[
\frac{\Delta T}{T} \propto \int_{-\infty}^{+\infty} d\omega |E_{\text{probe}}(\omega)|^2 \int_{0}^{+\infty} \Psi_{TE_0}^2(r) dr \int_{-\infty}^{+\infty} P(\mu) d\mu \int_{-\infty}^{+\infty} \alpha(\omega, \omega_\xi, \mu, \theta(E_{\text{pump}}, \mu)) f(\omega_\xi) d\omega_\xi
\]

\[
P(\mu) \mu^2 = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{(\mu-\mu_0)^2}{2\sigma^2}}
\]

in average
\[\mu = 35 \text{ D} \]
\[\sigma = 20\%
\]

Borri et al.,
Phys. Rev. B (Rapid Comm.),
in press
Part 3: *Fundamental aspects: Artificial atoms...*

**Quantum Beats in Quantum Dots**

Discrete Level-System

\[ |2\rangle \quad \Delta E \quad |1\rangle \quad \Delta E \quad |0\rangle \]

\[ \Delta E = 25 \text{ meV} \]
\[ \Delta t_{\text{beat}} = 168 \text{ fs} \]
Exciton-Biexciton Quantum Beats in QDs

Quantum Beats between two optical transitions:

$|G\rangle \rightarrow |x\rangle$ with $E_X$

$|x\rangle \rightarrow |xx\rangle$ with $E_{XX}$

$E_X - E_{XX} = E_{bin}$ (biexciton binding)
Exciton-Biexciton Quantum Beats in QDs

Determination of biexciton binding energy in CdSe/ZnSe QDs by femtosecond quantum beat spectroscopy

\[ \Delta E = 21 \text{ meV} \]

Summary

CdSe QDs in microspheres

InGaAs QDs in waveguides

Types of Quantum Dots and Techniques of Ultrafast Spectroscopy

Application Aspects: Dynamics of Amplification in Quantum Dot Lasers

Fundamental aspects: Semiconductor Quantum Dots as Artificial Atoms
For discussion
Rabi oscillations in differential transmission

Absorption coefficient of an inhomogeneously broadened ensemble:

\[ \alpha(\omega) = \int_{-\infty}^{+\infty} \sigma_0 \left( \frac{1/T_2}{(\omega - \omega_\xi)^2 + (1/T_2)^2} \right) \Delta N \int f(\omega_\xi) d\omega_\xi \]

\( \alpha \) is probed by a weak probe pulse after the pump: differential transmission intensity of the probe

dehasing time \( T_2 = 1.5 \text{ ps} \) 
\((T_2 = 0 \text{ ps})\),
pulse length \( t_0 = 5 \text{ ps} \)

Borri et al., Phys. Rev. B (Rapid Comm.), in press
Non-degenerate FWM with ns-pulses

asymmetric line shape at $T < 10 \, \text{K}$

Example: $t_0=1\text{ps}$

The biexciton population oscillates with a different period compared to the exciton population.

The dephasing reduces the amplitude of the oscillations.

Many oscillation periods are present, even when the averaging over the inhomogeneous broadening and the spatial mode profile are included.

Borri et al., Phys. Rev. B (Rapid Comm.), in press
Analysis of homogeneous line broadening - InGaAs

Fourier transform of the TI-FWM signal

- Energy (meV)
- $|E(\omega)|^2$ (a.u.)

sharp Zero-phonon line (long dephasing time) + broad Non-Lorentzian band (fast initial dephasing)

Extrapolation to $T = 0K$:

$\gamma_0 = 0.66 \mu eV$ (diff. Transm. $T_1=1$ ns)

Dephasing close to the radiative limit!

CdSe QDs attached to a glass $\mu$-sphere

Here: CdSe-shell on glass $\mu$-sphere

$R=3.1 \, \mu$m

Nano Lett. 1, 309 (2001)
CdSe Quantum Dots

T=6K
R~2.5 nm
I_0=5 mJ/cm^2

PL Intensity (arb. units)

Photon Energy (eV)

T=6K
R~2.5 nm
I_0=5 mJ/cm^2

PL Intensity (arb. units)

Excitons

1S\text{3/2} 1s_e

2S\text{3/2} 1s_e

Biexcitons

1S\text{3/2} 1s_e, 1S\text{3/2} 1s_e, 2S\text{3/2}

1S\text{1/2} 1s_e, 1S\text{3/2} 1s_e, 1S\text{3/2}

Spectra of PL and optical gain at high excitation

CdSe QDs
R ~ 3.0 nm
T = 10 K

Luminescence (arb. units)

Gain

\( \Delta \varepsilon \)

Wavelength (nm)

700 650 600 550

Probe (4) (3)

Pump (1) (2)

two-pair states

one-pair states

ground state

Energy (eV)

0 1.8 2.0 2.2 2.4

(i) 10 kW/cm²
(ii) 100 kW/cm²
(iii) 400 kW/cm²
(iv) 1400 kW/cm²

J. Lum. 70, 269 (1996).
Temperature-dependent dephasing time $T_2$

Temperature-dependent FWM
InGaAs SA-QDs

From 300K to 100K the FWM decay is dominated by a dephasing time below 1ps

Below 100K a slow component appears with an exponential decay time that increases with decreasing temperature.

At $T=7K$ the slow component dominates the dynamics with a dephasing time of 630 ps corresponding to only $2 \mu eV$ homogeneous broadening!

Spherical Microcavities - Photonic Dots