UNIVERSITÄT DORTMUND

Ultrafast Spectroscopy of Quantum Dots (QDs)

Ulrike Woggon

FB Physik, Universität Dortmund

With thanks to:
M.V. Artemyev, P. Borri, W. Langbein, B. Möller, S. Schneider

Fruitful cooperations:

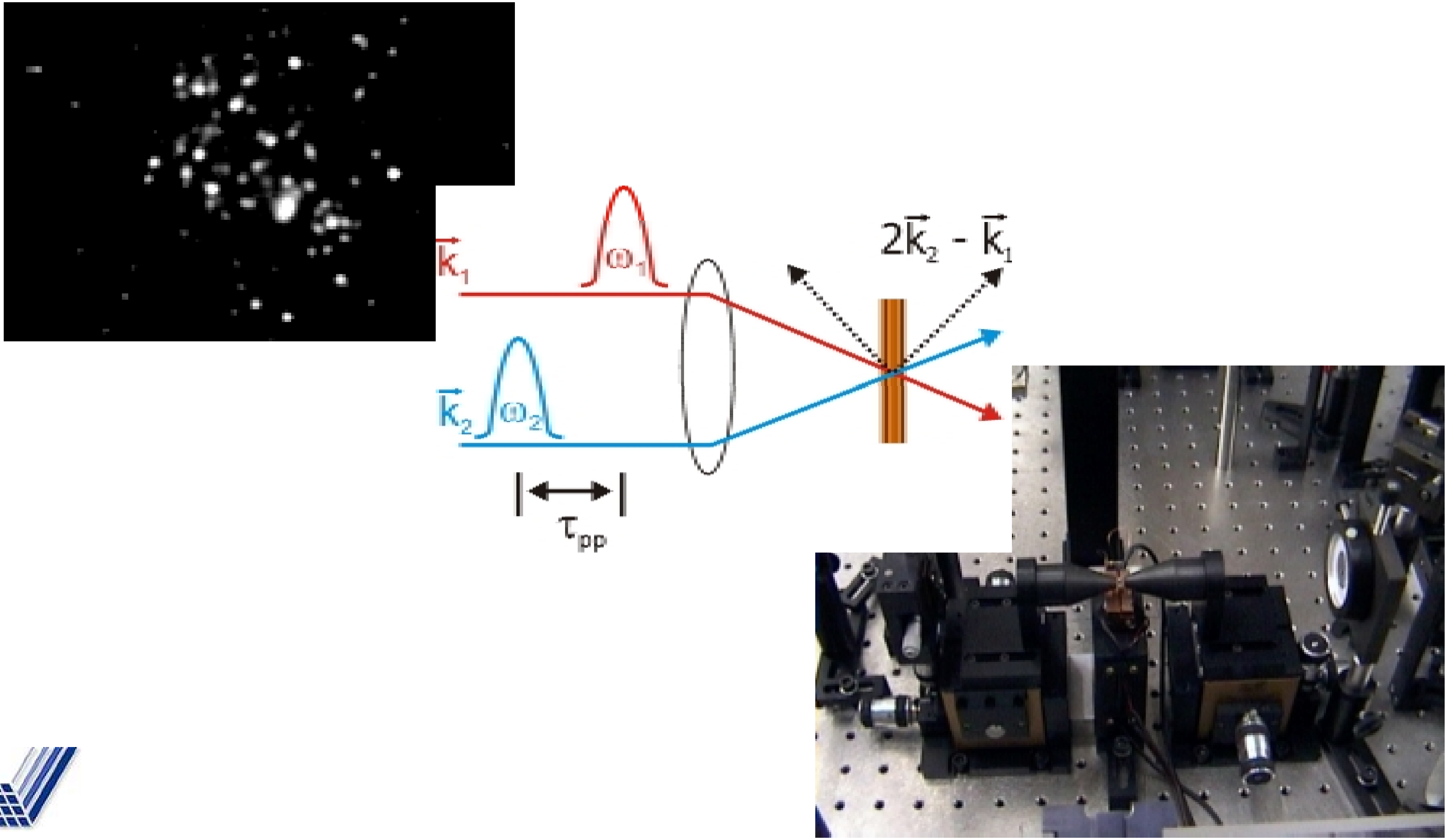
calculations: R. Wannemacher, Leipzig,

samples: D. Bimberg and coworkers, Berlin
D. Hommel and coworkers, Bremen
A. Forchel and coworkers, Würzburg

Experimentelle Physik IIb

Outline:

- 1. Types of QDs and Techniques of Ultrafast Spectroscopy

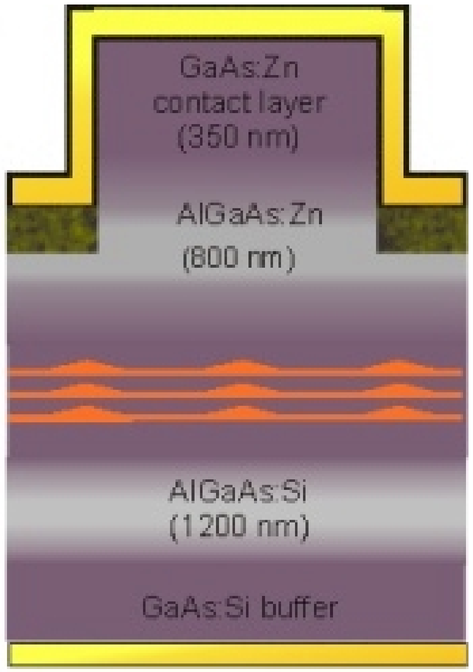


Outline:

- *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

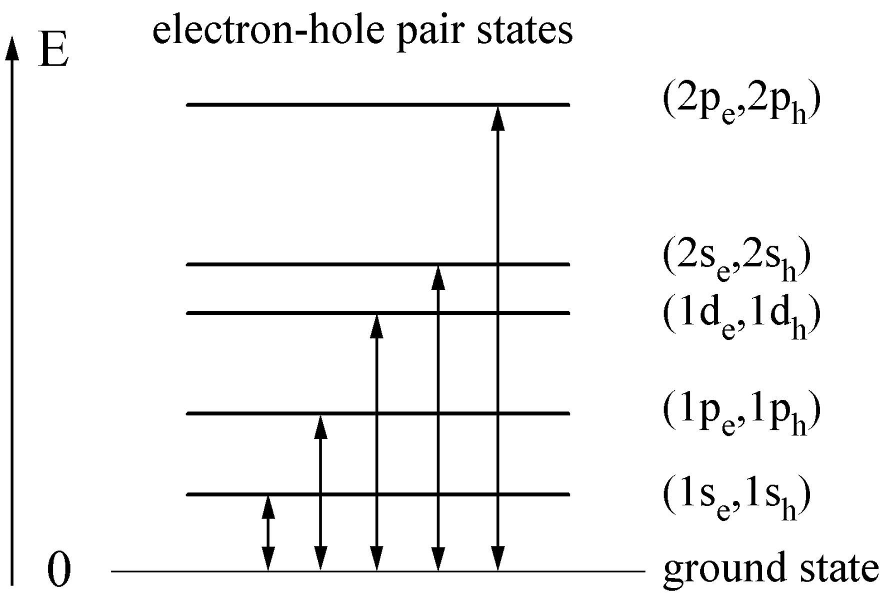
D. Bimberg and coworkers, TU Berlin

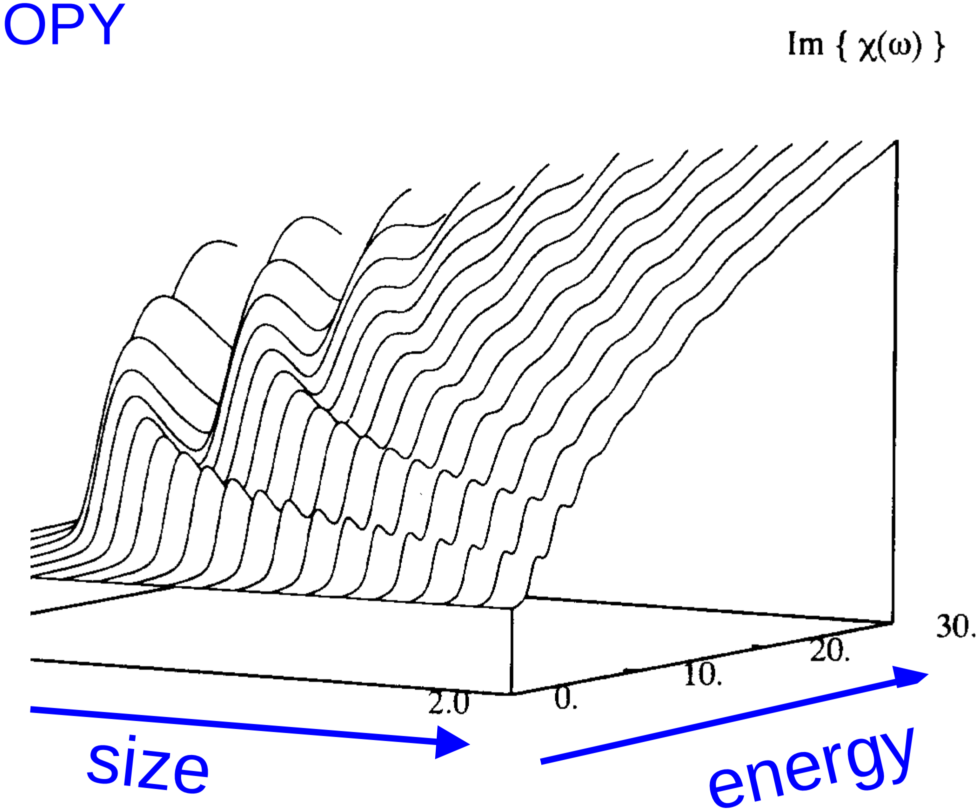
Outline:

- *3. Fundamental aspects: Semiconductor QDs as artificial atoms*

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

→ ULTRAFAST SPECTROSCOPY

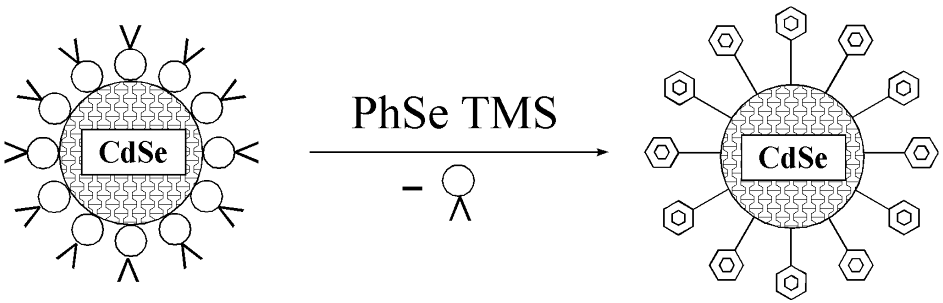




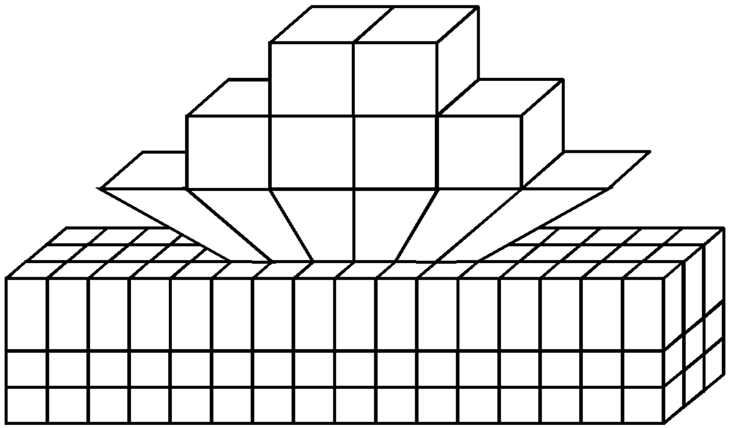
L.Banyai, S.W. Koch, *Semiconductor Quantum Dots*

Part 1: *Types of QDs and Techniques...*

Quantum Dots:
Nanocrystals and epitaxially grown Islands

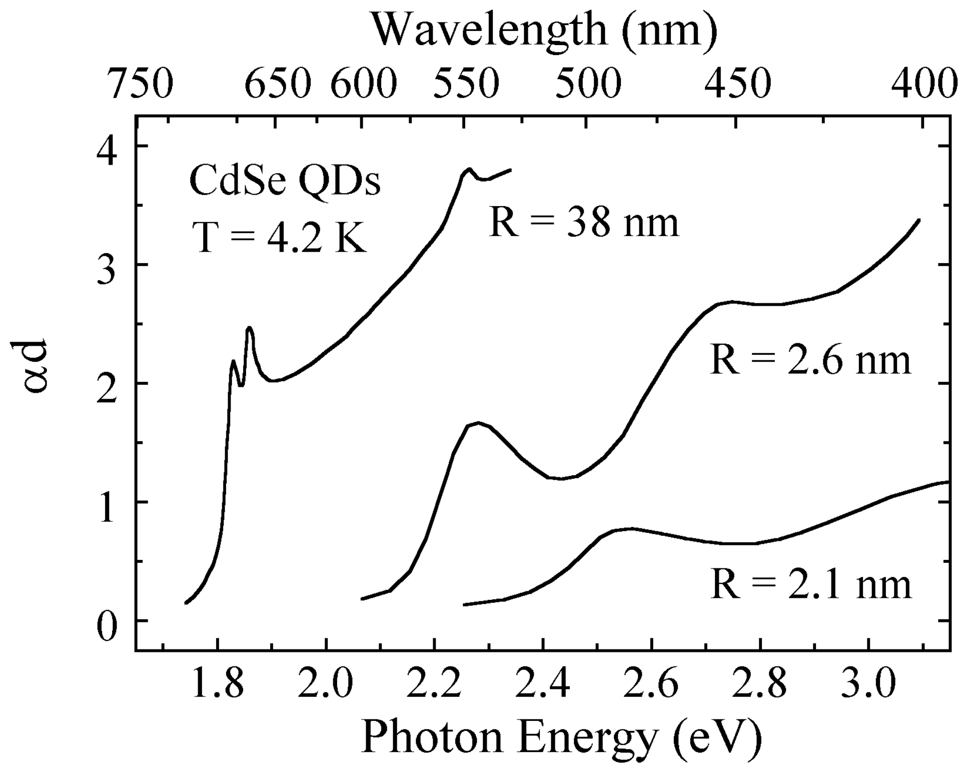


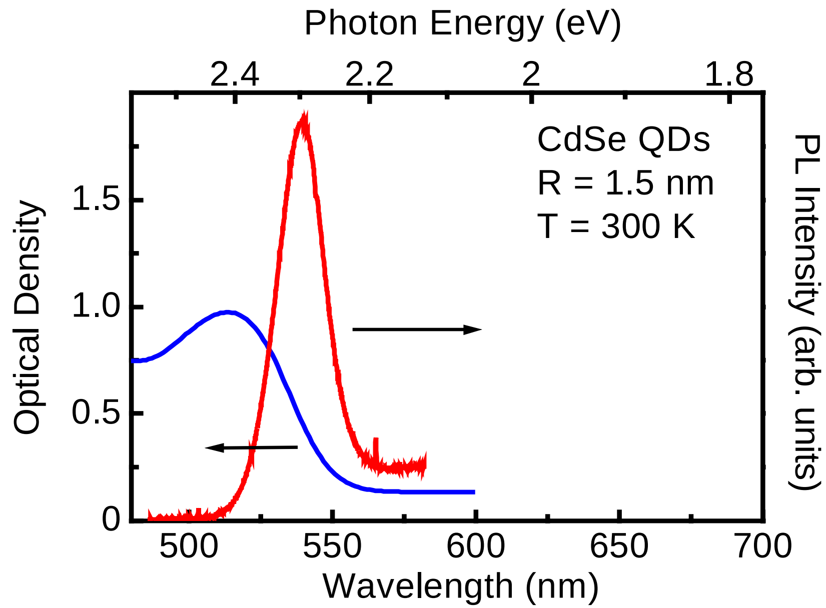
Precipitation of spherical nanocrystals in colloidal solution or glass, polymer etc. matrix



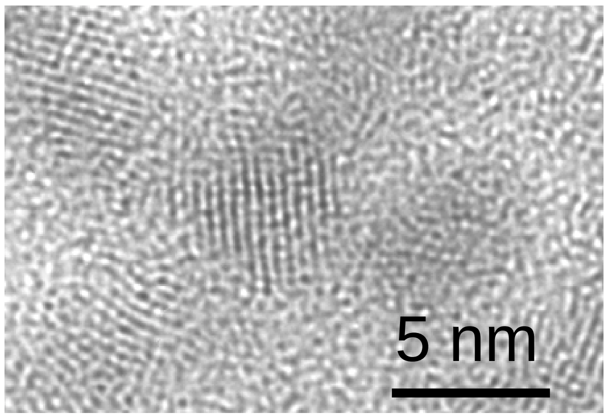
Lattice-mismatch induced island growth

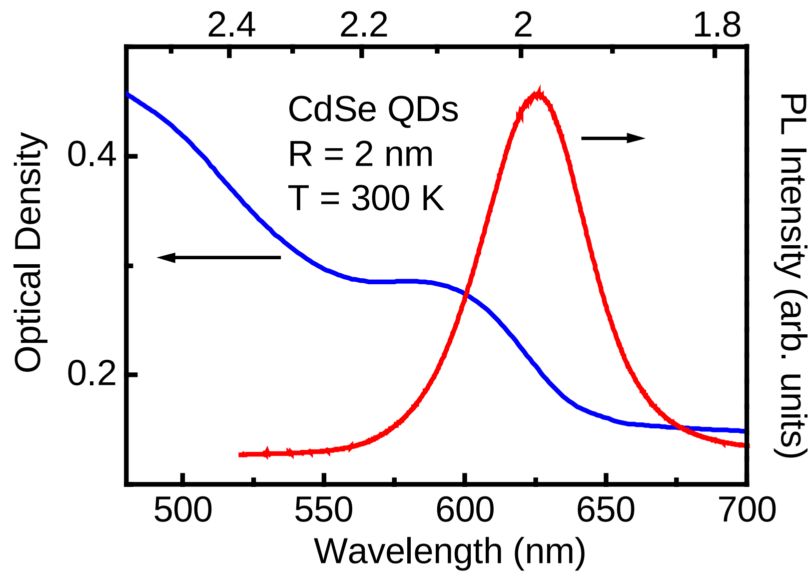
CdSe QDs emitting in the visible (nanocrystals)



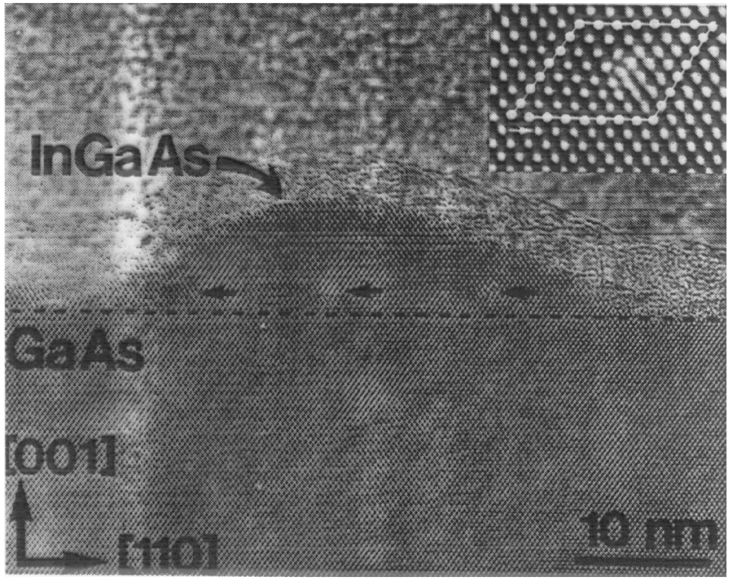


CdSe in glass

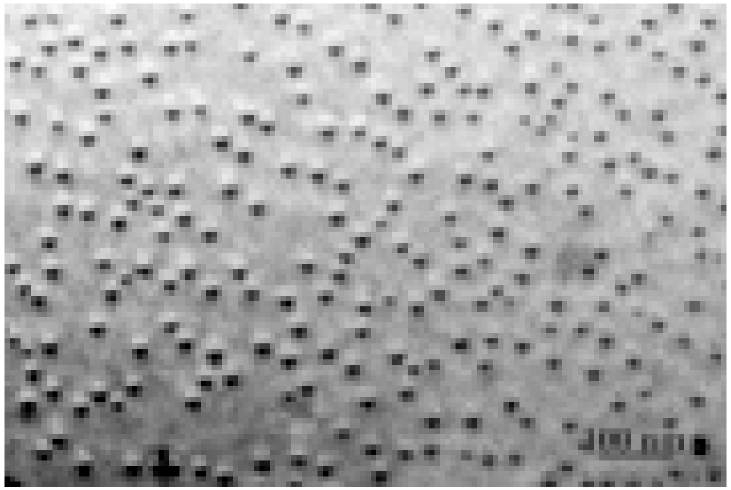




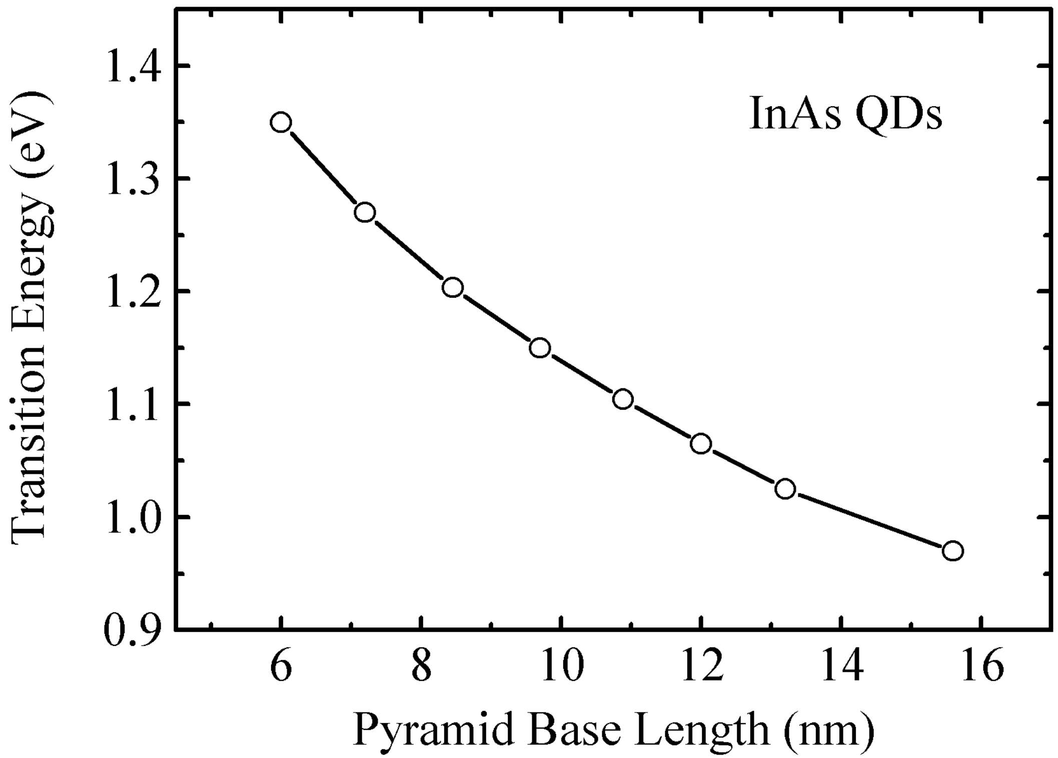
InGaAs self-assembled islands emitting in the NIR



D. Gerthsen et al., Karlsruhe



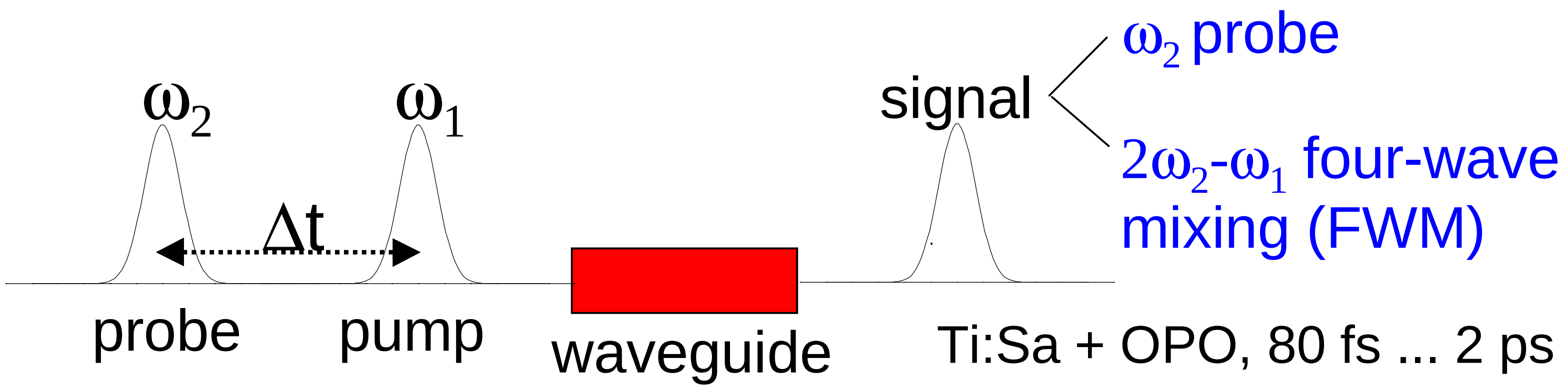
Calculated confined eh-pair energies for InAs assuming pyramidal shape



Grundmann, Bimberg et al., TU Berlin

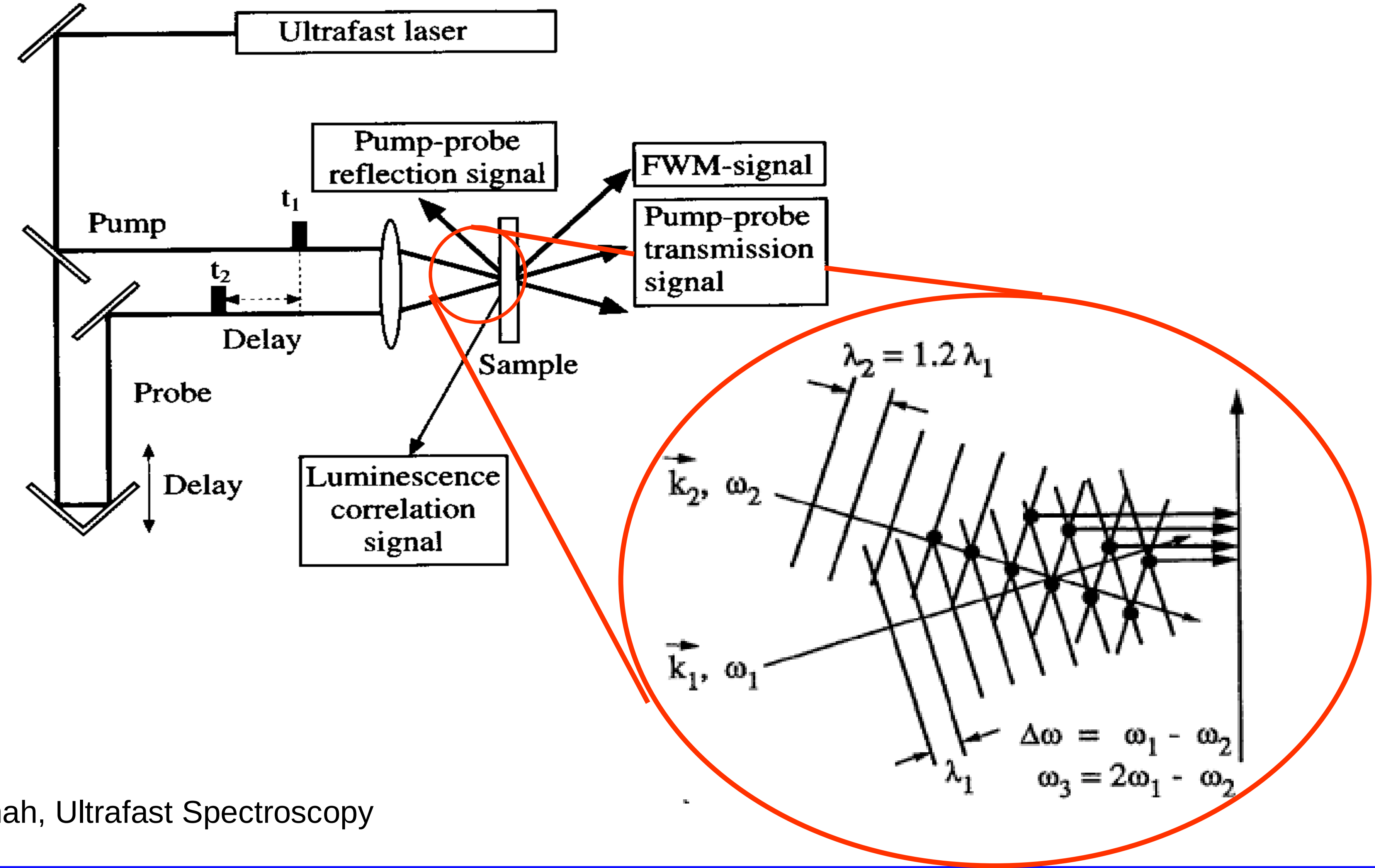
Part 1: *Types of QDs and Techniques...*

Femtosecond Heterodyne Technique



Femtosecond Ultrafast Spectroscopy

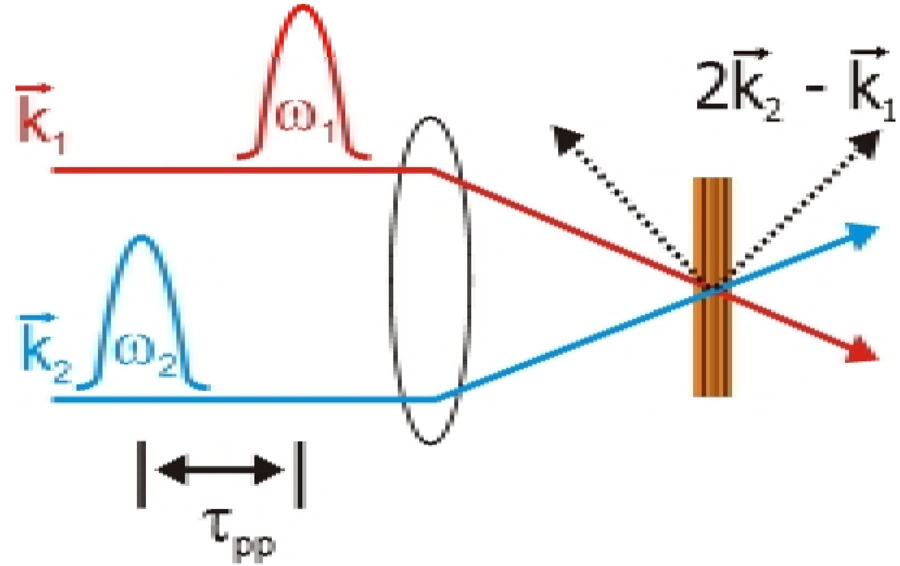
Usually:



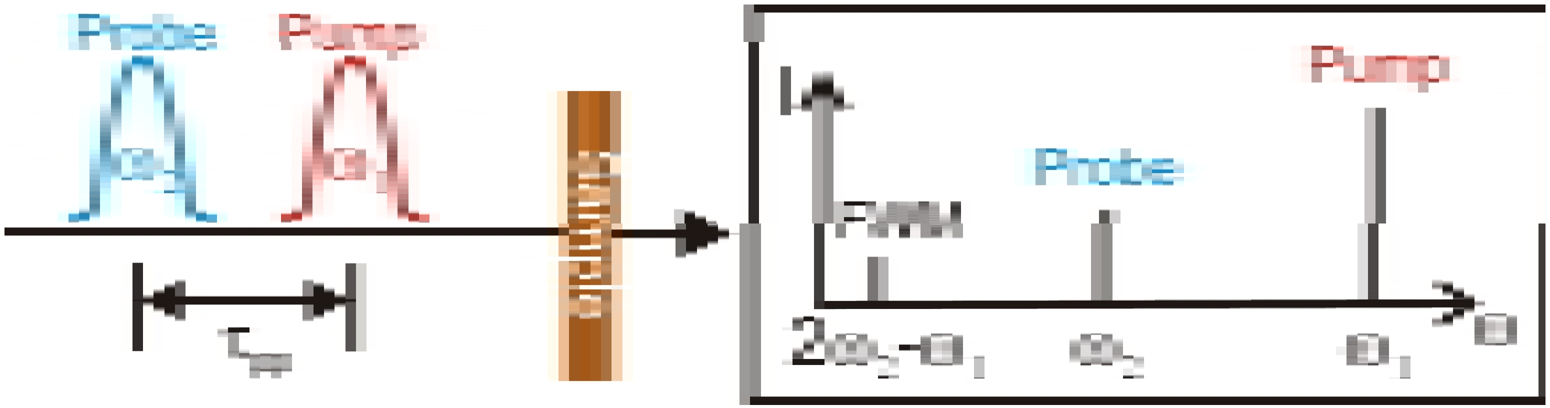
J. Shah, Ultrafast Spectroscopy

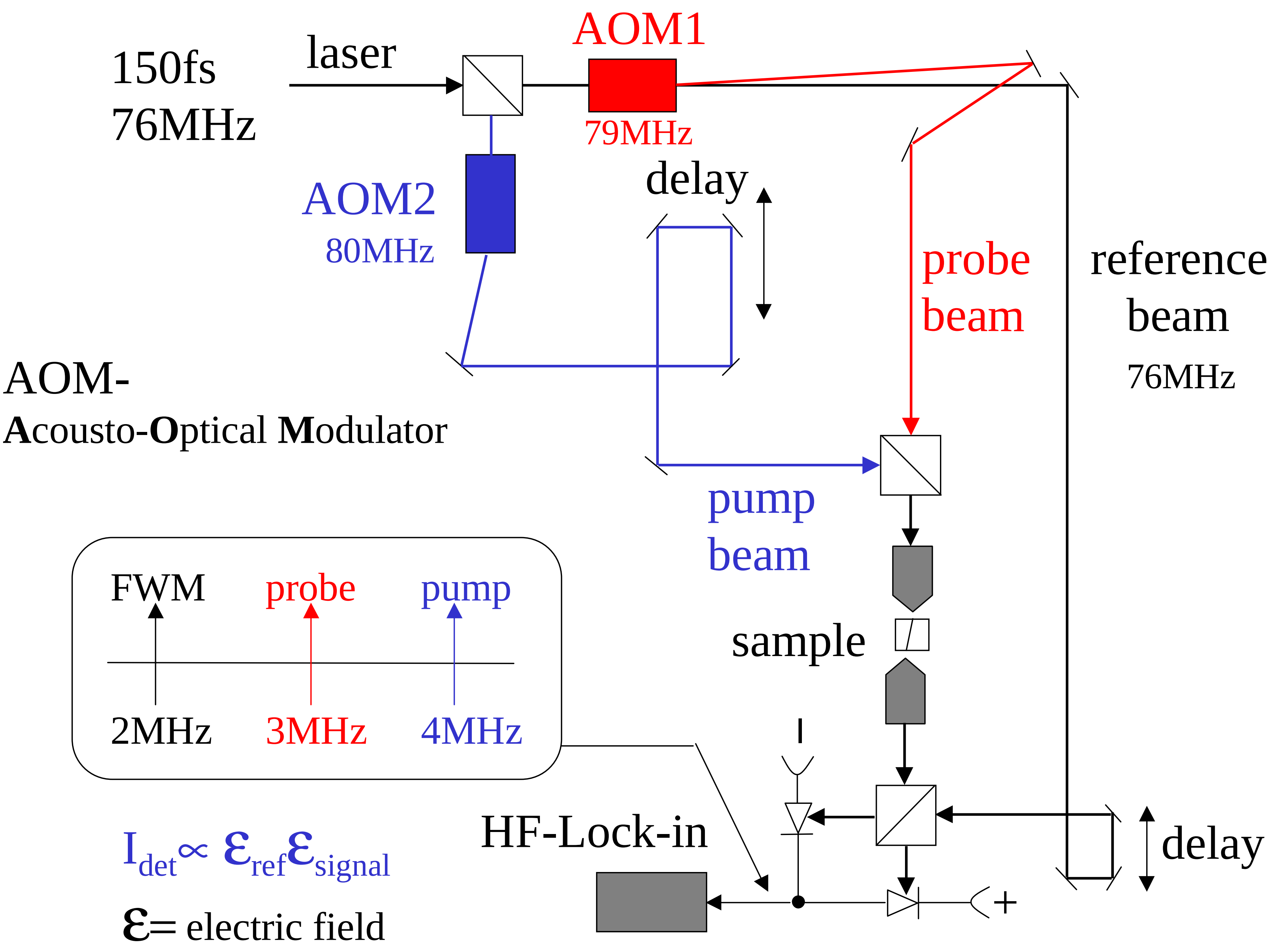
Femtosecond Heterodyne FWM- and PP-Spectroscopy

Usually:



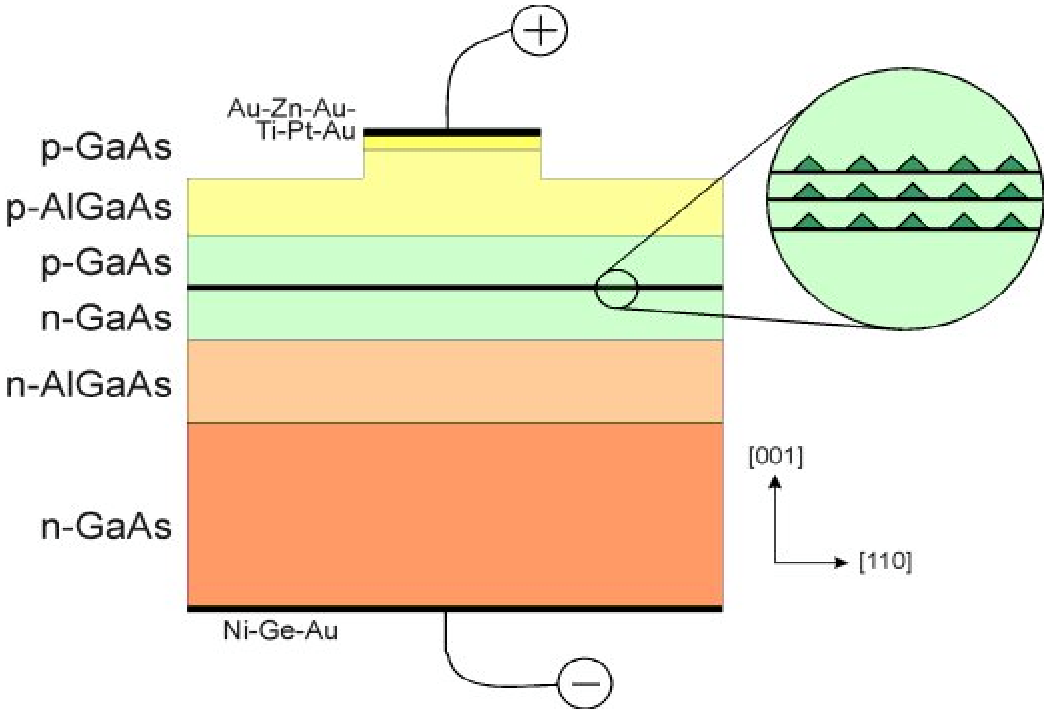
Here:




150fs
76MHz
laser
AOM1
79MHz
AOM2
80MHz
delay
probe beam
reference beam
76MHz
AOM-
Acousto-Optical Modulator
pump beam
FWM
probe
pump
2MHz
3MHz
4MHz
sample
HF-Lock-in
delay
$I_{det} \propto \varepsilon_{ref}\varepsilon_{signal}$
ε= electric field

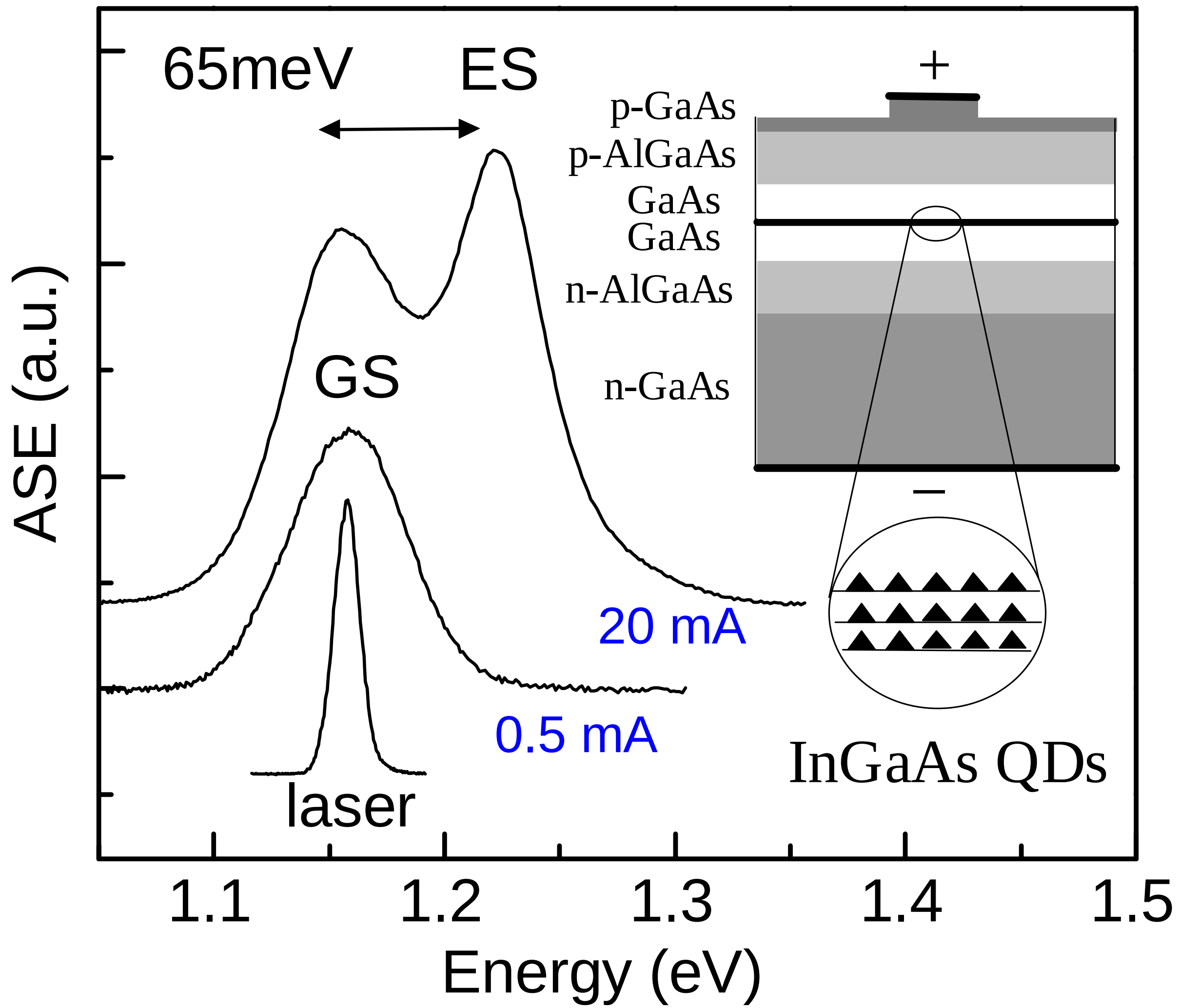
Part 2: *Applied aspects: QD-laser...*

Gain Dynamics in Quantum Dots



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

Gain Dynamics of InGaAs QDs



ridge waveguide
5x500µm,
3 stacked QD layers

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

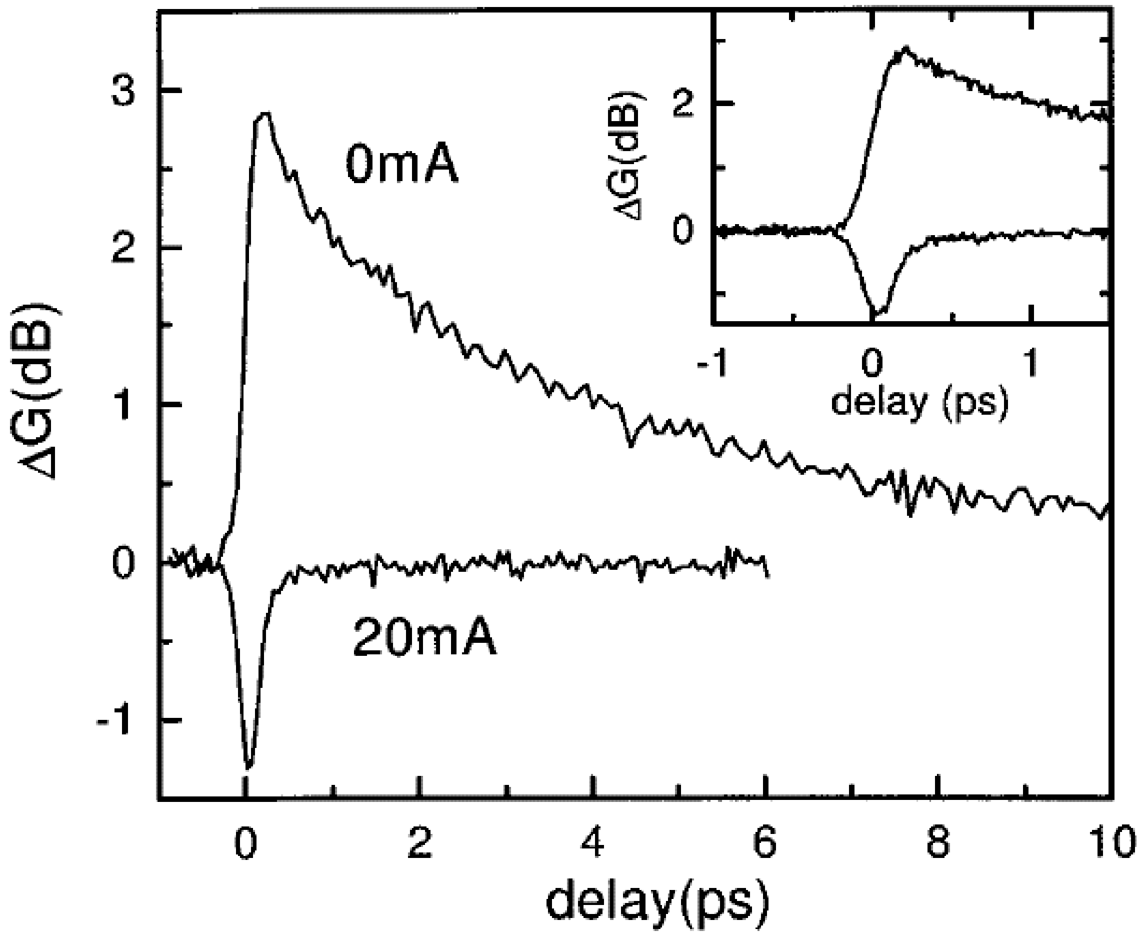
optical density ~ 1.5
(α~$30cm^{-1}$)

Carrier injection
electrically (0...20 mA)

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

Sample from TU Berlin, Prof. Bimberg

Gain Dynamics of InGaAs QDs

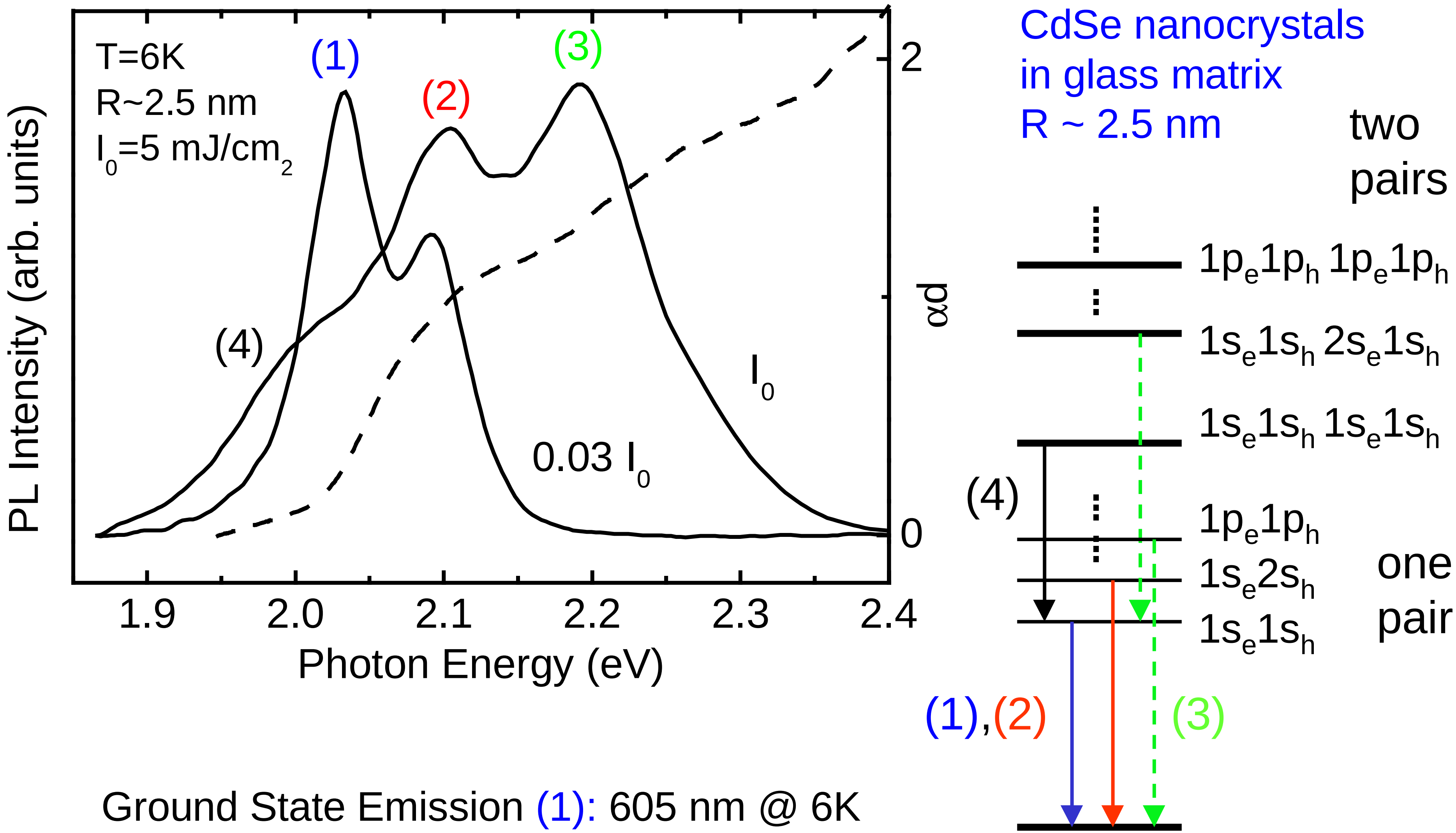


Pump-induced gain change in a heterodyne pump-probe experiment at maximum gain (20 mA) and without electrical injection (0 mA)

Gain recovery in < 100 fs at 300 K !

P. Borri et al., J. Sel. Topics Q. El. 6, p. 544 (2000); Appl. Phys. Lett. 76, p.1380 (2000).

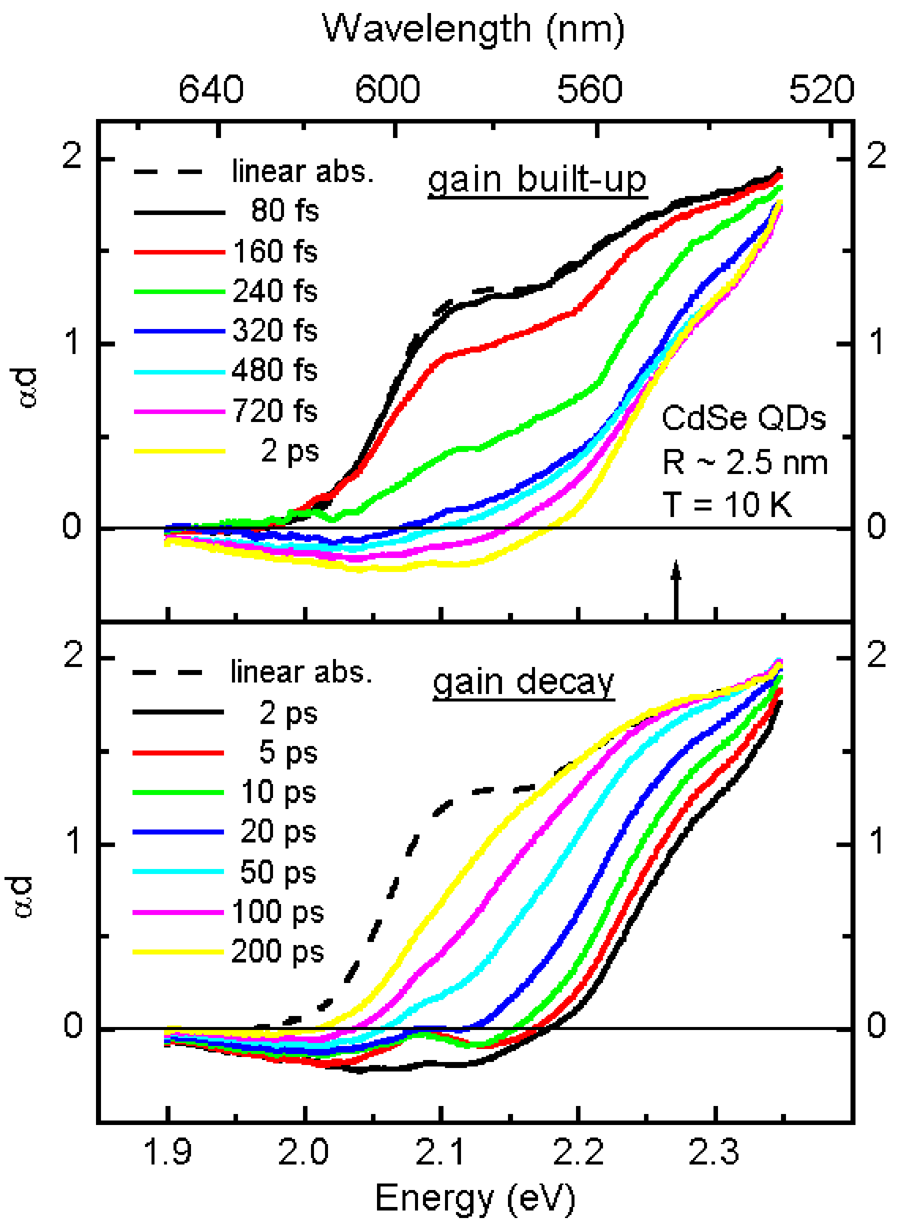
Gain Dynamics of CdSe QDs



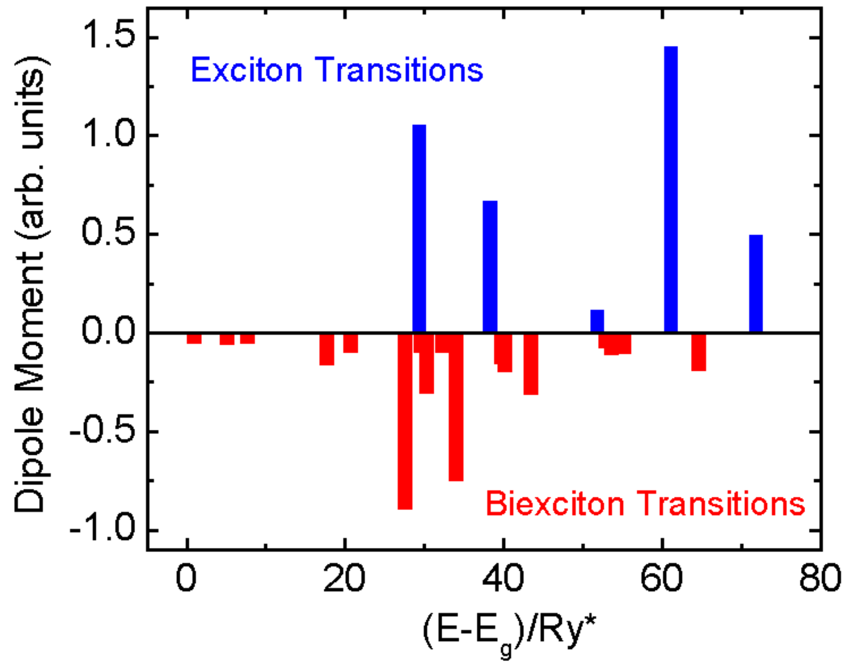
Ground State Emission (1): 605 nm @ 6K

Woggon et al., Phys. Rev. B 54, 17681 (1996), J. Lum. 70, 269 (1996).

Gain Dynamics of CdSe QDs



Excitonic and biexcitonic contributions to optical gain

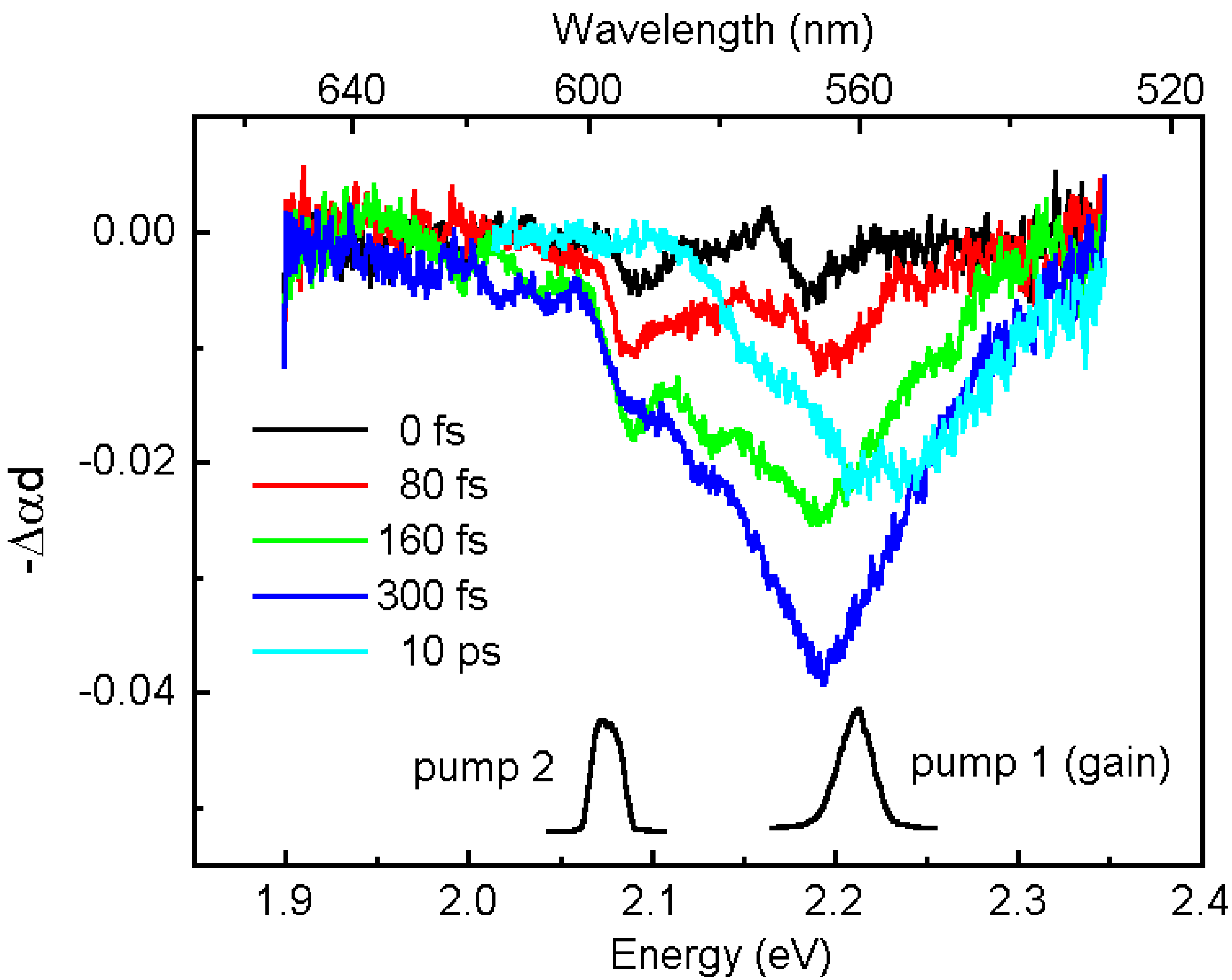


Gain recovery time spectrally varying, <1...100ps

Optics Lett. 21, 1043 (1996).

Gain Dynamics of CdSe QDs

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



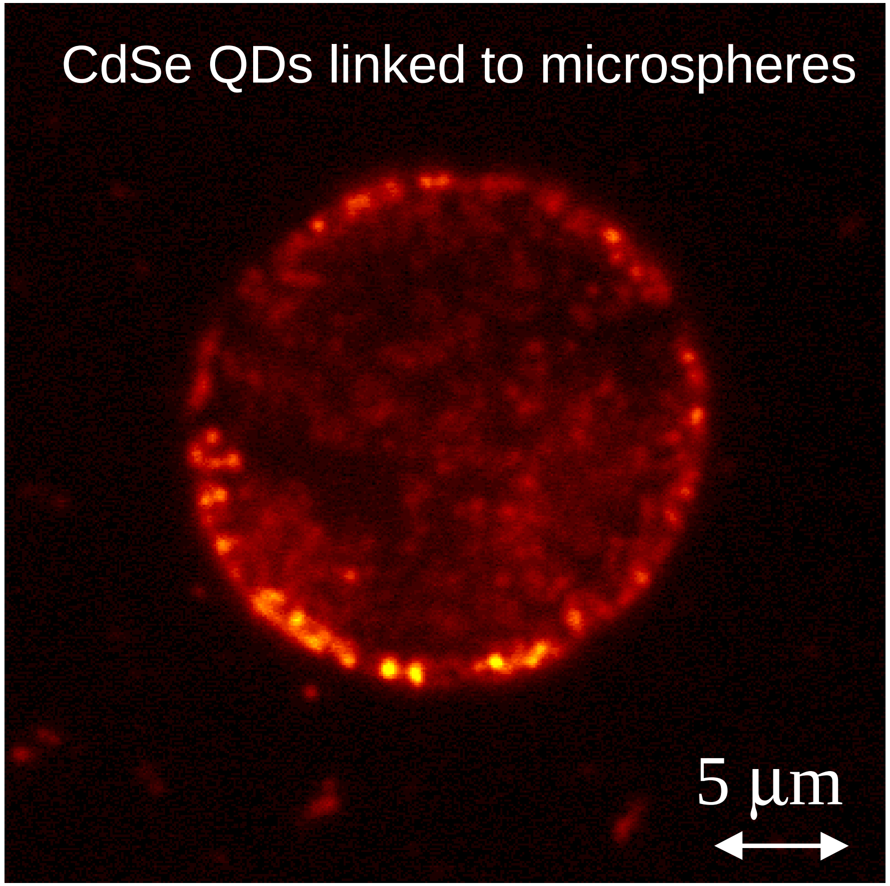
Spectral hole width of a single gain process ~20 meV

Intrinsic limit of gain recovery below 100 fs !

Chem. Phys. 210, 71 (1996)

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

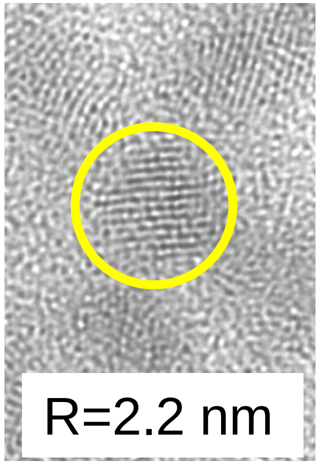
Quantum dots as active media in optical microcavities



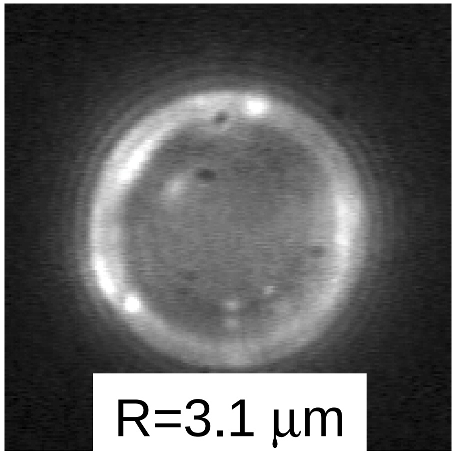
Picture: M.V.Artemyev, I. Nabiev

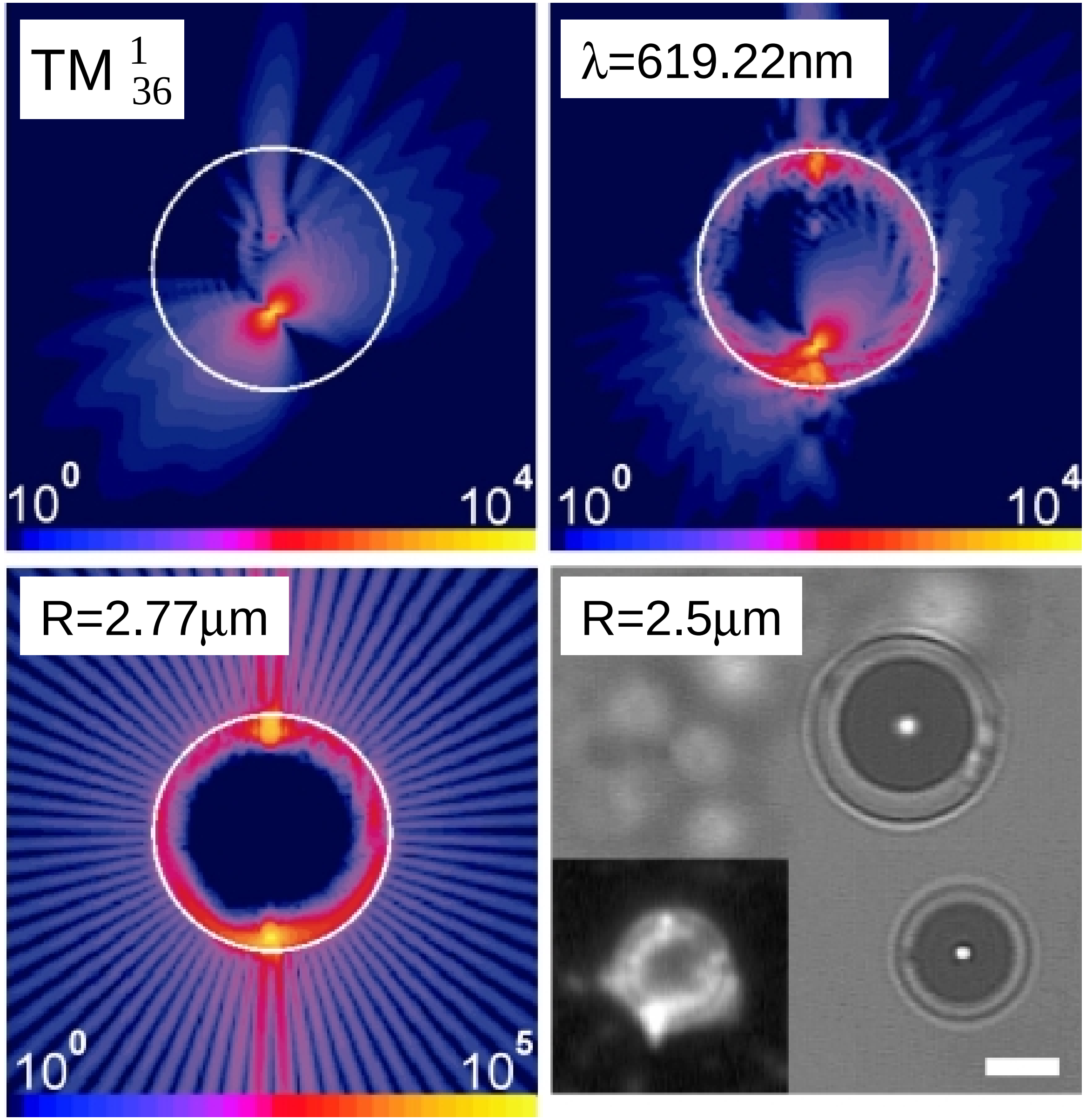
„Dot - in - a - Dot" - Structure

CdSe nanodot



Glass microsphere

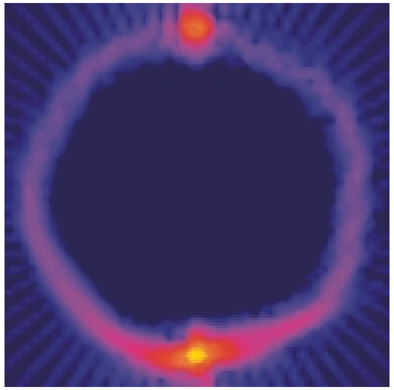




Artemyev et al., APL 78, p.1032 (2001), Nano Lett. 1, 309 (2001).

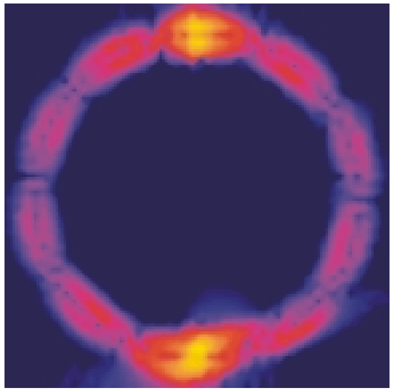
Cavity Modes of a CdSe-doped Microsphere

WGM

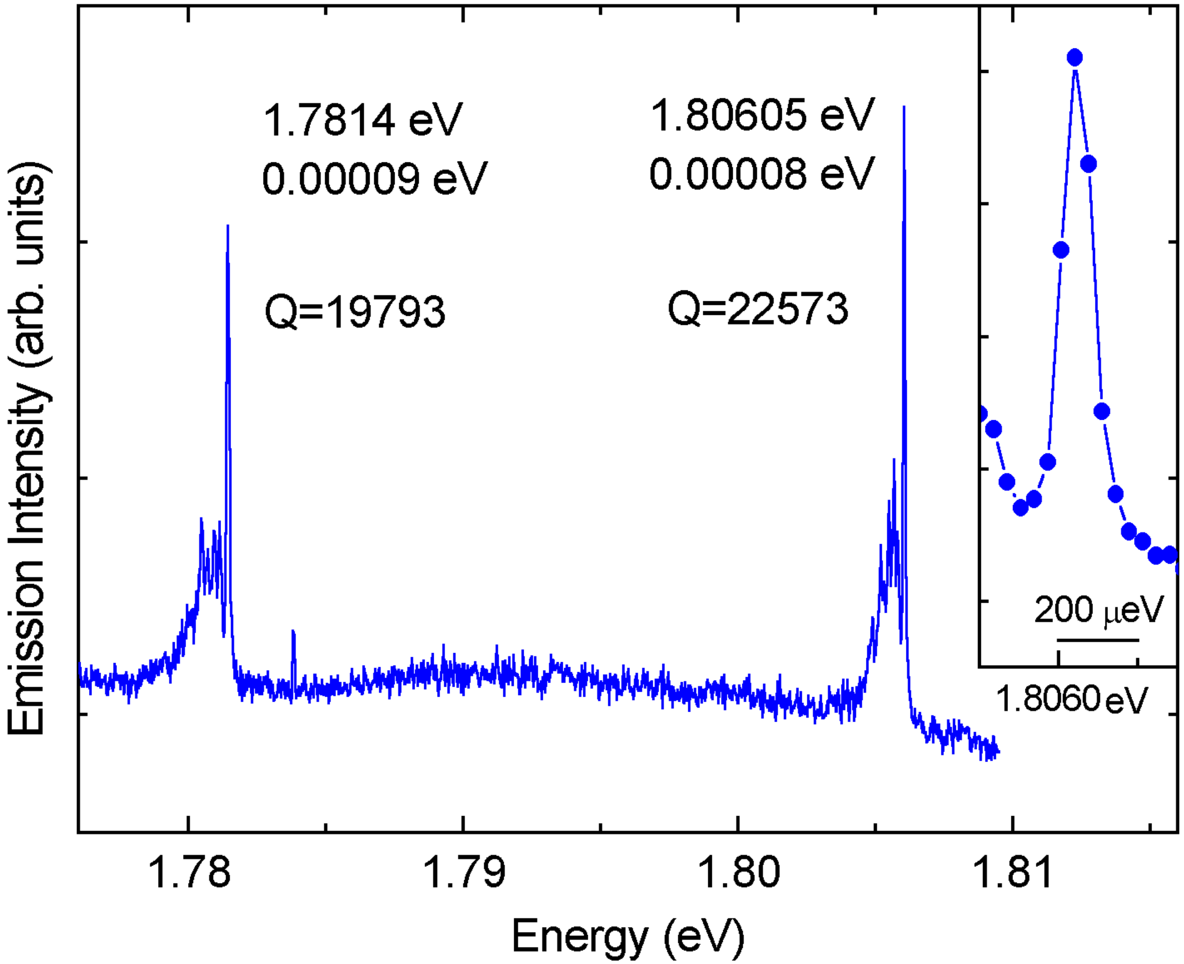


TM, ℓ=36, n=1

TM, ℓ=36, n=2

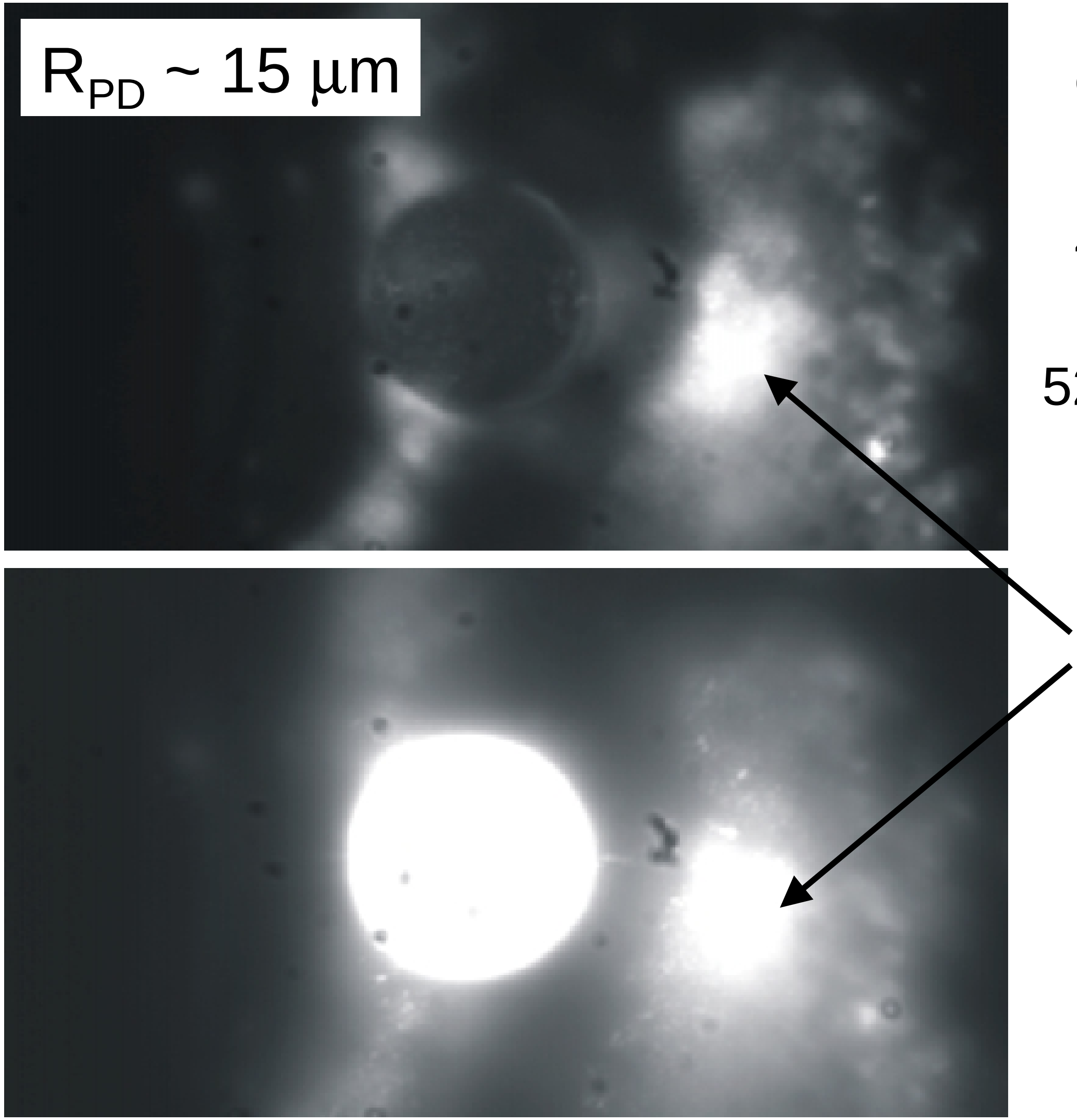


R_{PD} = 2.5 µm R_{QD} = 2.5 nm



Nano Lett. 1, p. 309 (2001), Appl. Phys. Lett. 80, p.3253 (2002)

Optical Pumping of a CdSe-doped Microsphere



cw-Ar laser, 488 nm

Excitation spot size
40 μm^2

T = 300 K
520 nm < λ_{em} < 640 nm

10 mW

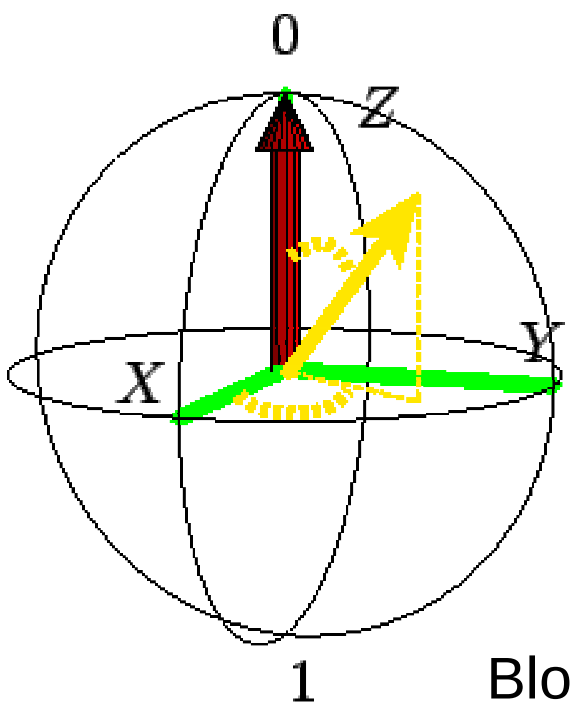
CdSe nanocrystals
(not on microsphere)

14 mW

See also:
Artemyev, Woggon et al.
Nano Letters 1, 309 (2001)

Part 3: *Fundamental aspects: Artficial atoms...*

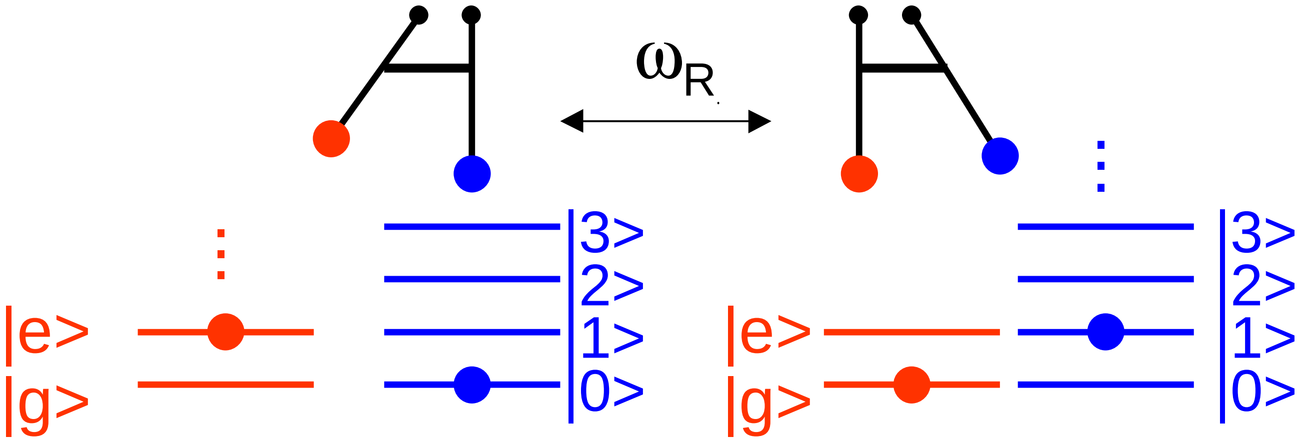
Rabi Oscillations in Quantum Dots



Bloch-sphere:
population oscillation

Rabi-Oscillations in Atoms

Simple model: two coupled oscillators



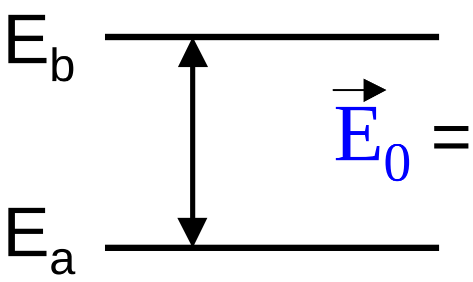
photon field

atom states

Rabi frequency

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

Two-level system in resonance with photon field



$$\vec{E}_0 = \hbar\omega_0 = E_b - E_a$$

E_0 : electromagn. field vector

$\hbar\omega_0$: transition energy

$\vec{\mu}$: transition dipole moment

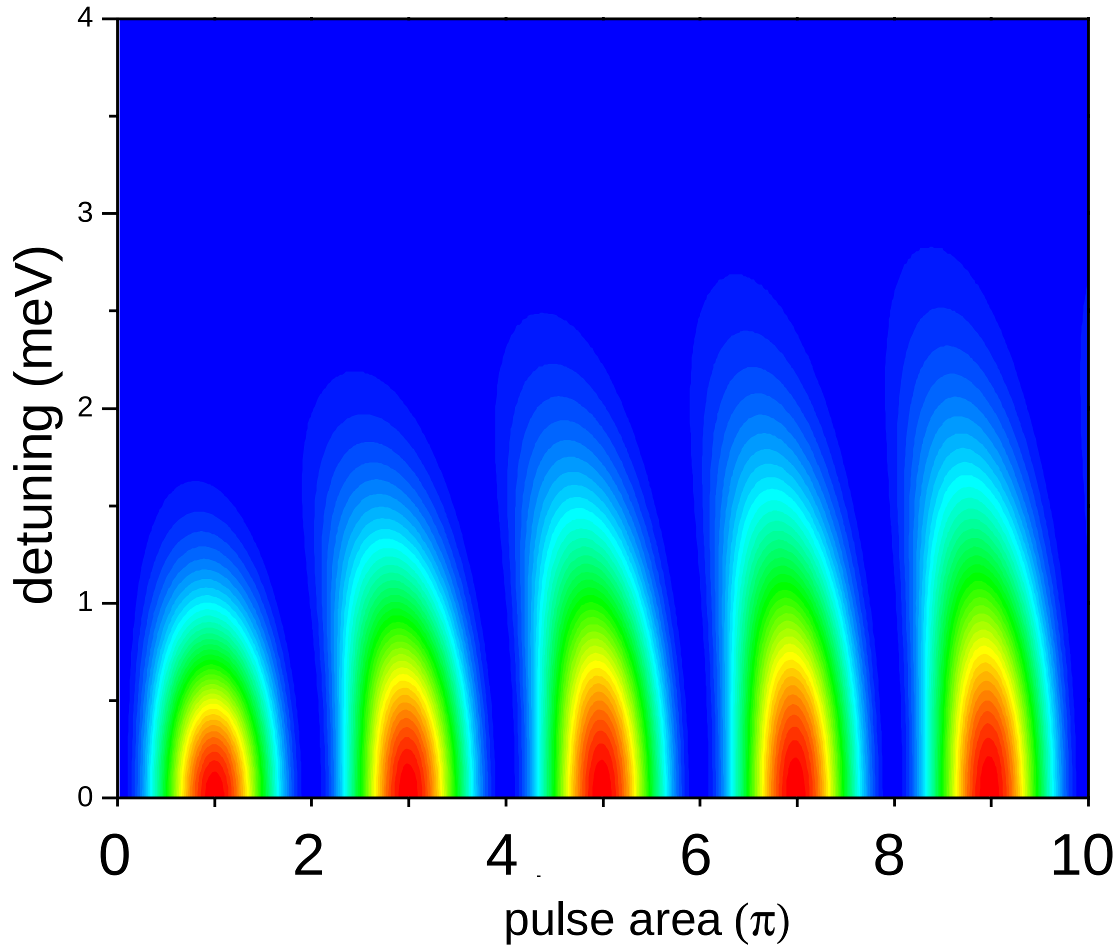
Rabi Oscillations versus Pulse Area

Here pulsed excitation !

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

$$\theta = \int_{-\infty}^{+\infty} \frac{\vec{\mu} \cdot \vec{E}_0}{\hbar} dt$$

t_0=1ps (ΔE_{HWHM}=0.75meV)

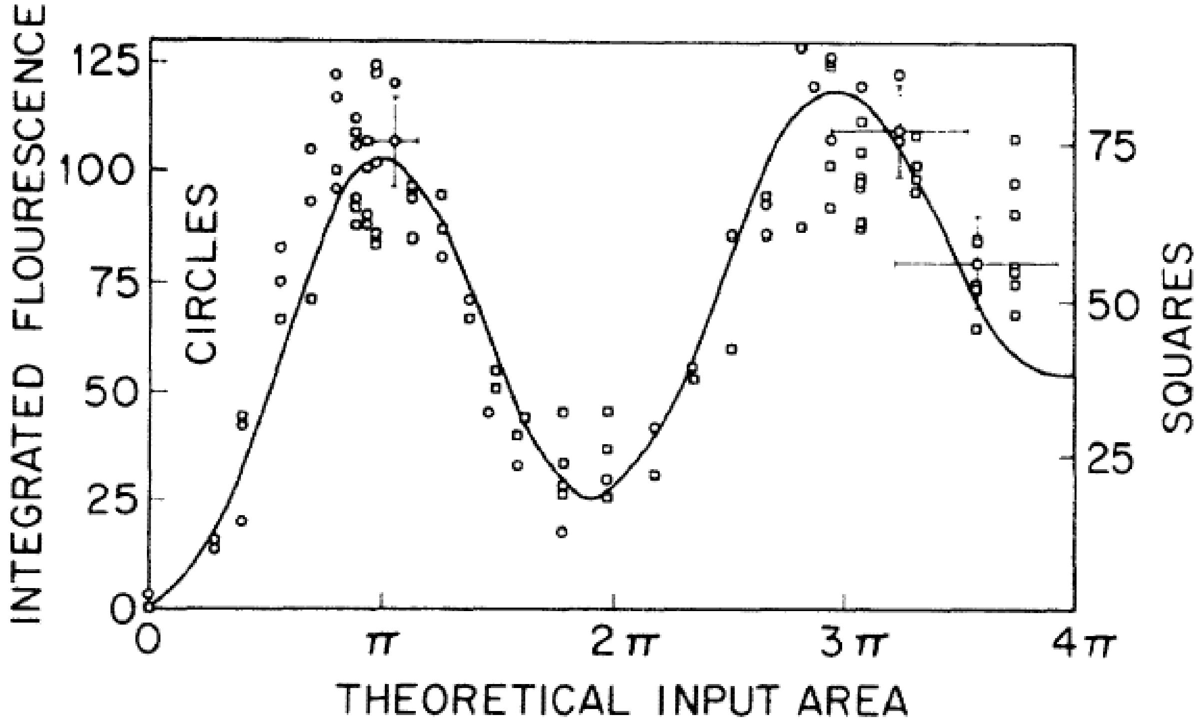


Occupation probability of the ground (excited) state

Population oscillation
blue = -1
red = +1

Initial conditions:
for t << -t_0 in ground state
No dephasing!

Rabi Oscillations in Rb-Atoms



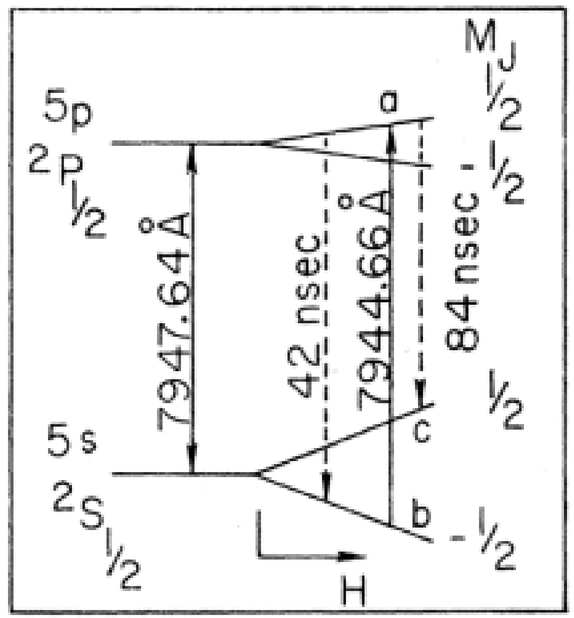
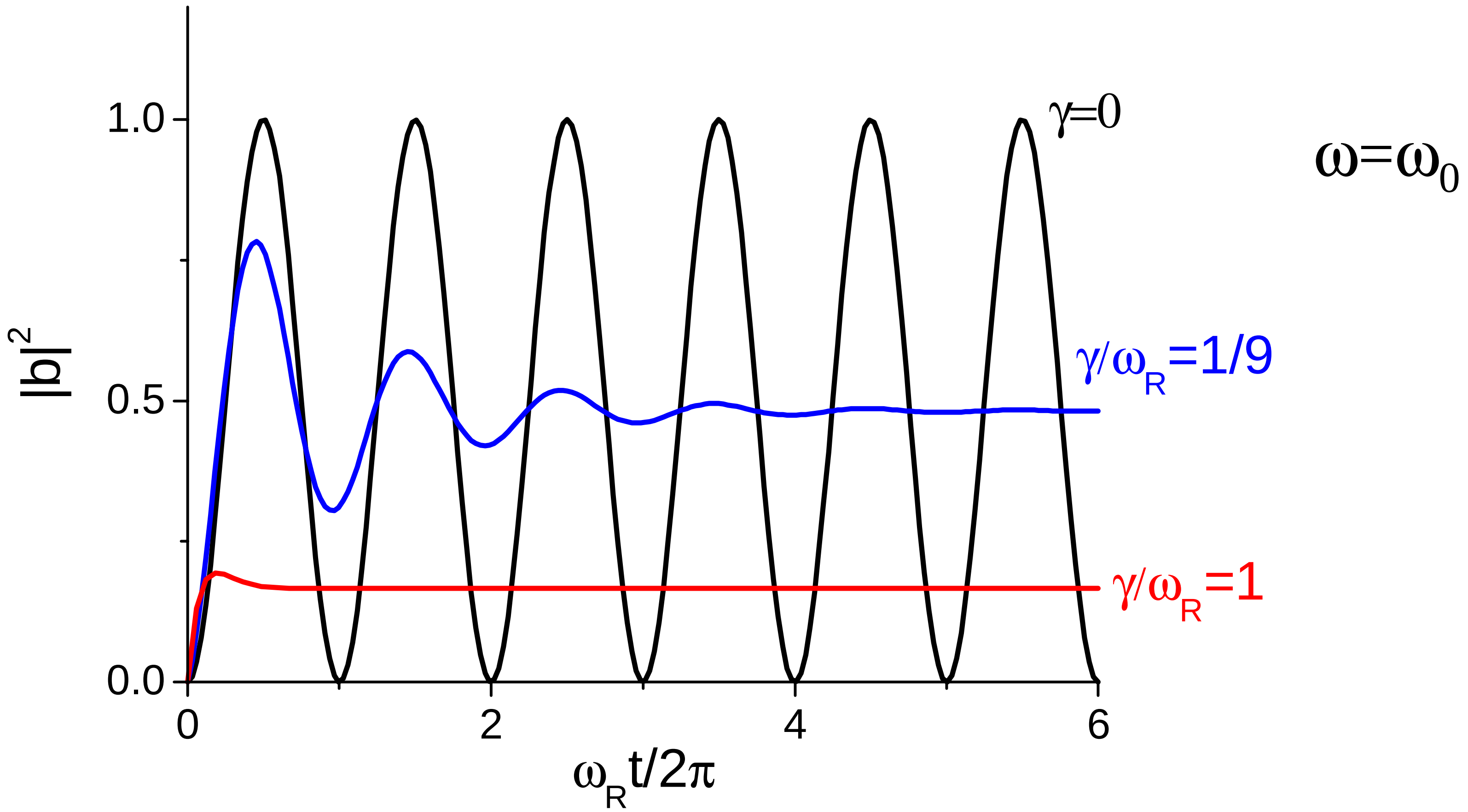


FIG. 2. Simplified Rb energy-level diagram.

H.M. Gibbs, Phys. Rev. Lett. 29, 495 (1972),
Phys. Rev. A8, 446 (1973)

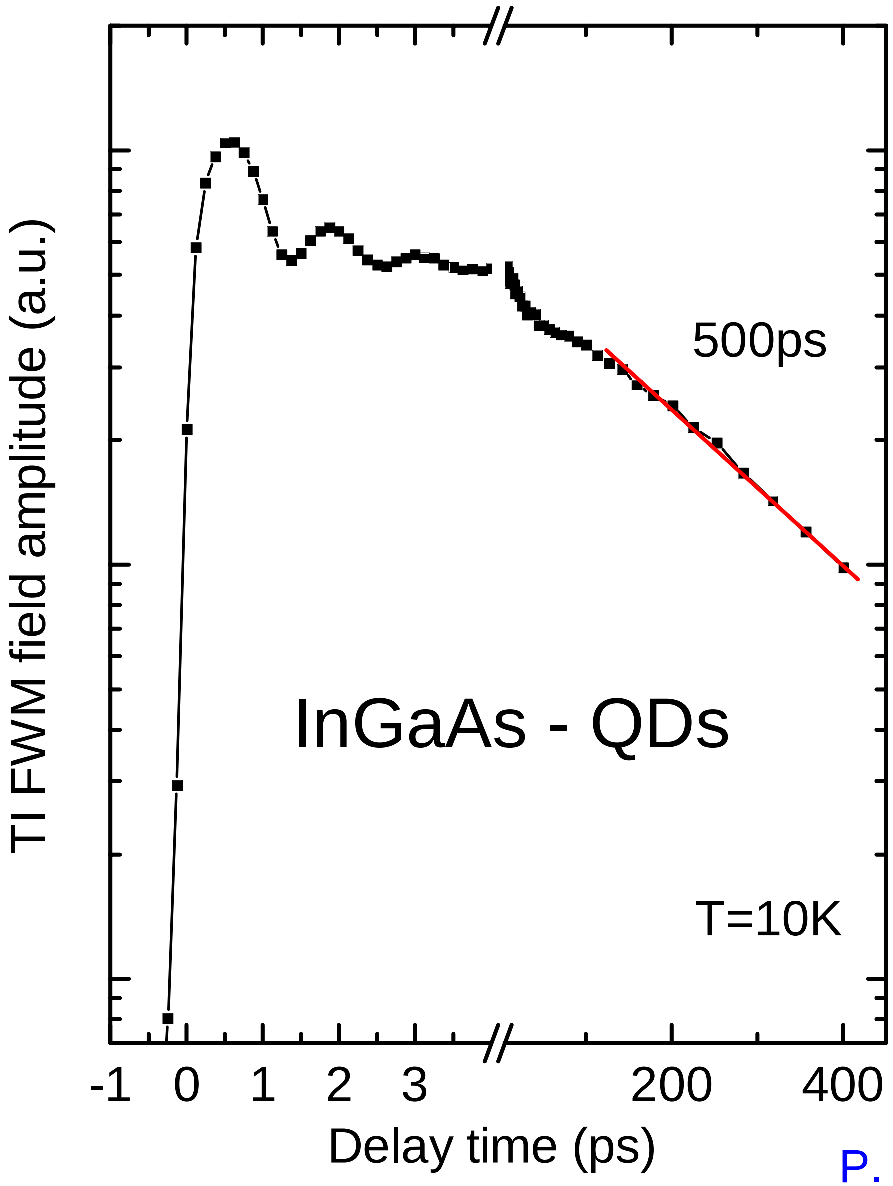
Effect of Dephasing T_2 on Rabi oscillations

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



Population flopping over many periods is possible in systems with long dephasing times and large transition dipole moments: $\gamma / \omega_R \ll 1$.

Dephasing time T_2 of InGaAs quantum dots



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

Below T=10 K a slow dephasing time > 500 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 ?????

P. Borri *et al.*, Phys. Rev. Lett. **87**, 157401 (2001)

Rabi Oscillations in InGaAs Quantum Dots

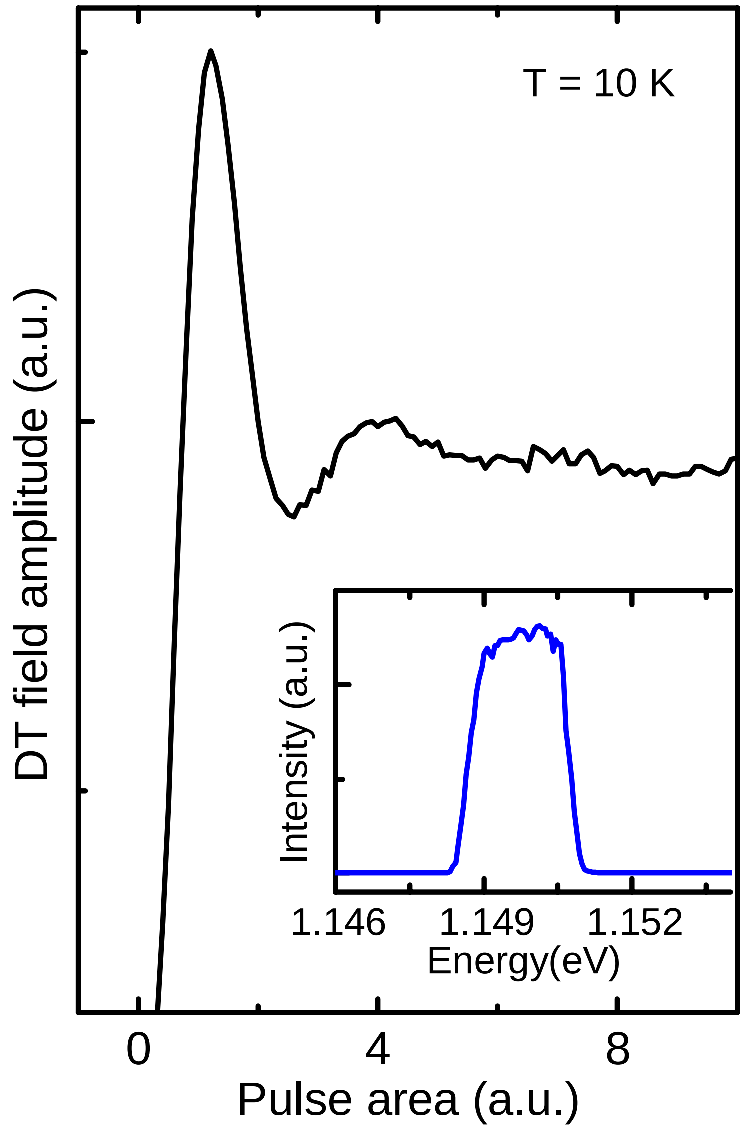
Experiment

Use of spectrally shaped ps-pulses

$\Rightarrow$ a sharpened distribution of the spectral intensity improves the visibility of the oscillations.

<u>Rabi oscillation</u>:
two oscillation maxima can be clearly distinguished

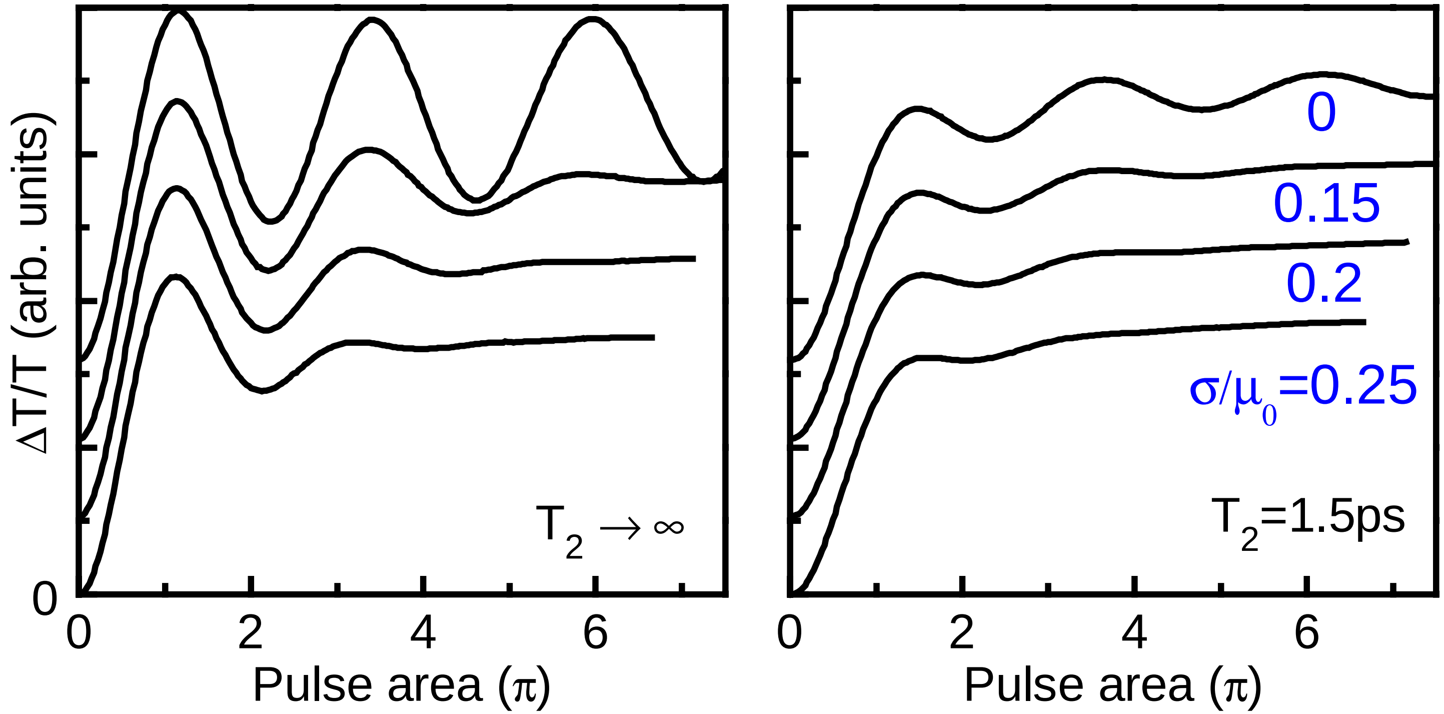
Borri et al., Phys. Rev. B (Rapid Comm.), in press



Distribution in Transition Dipole Moments μ

$$\frac{\Delta T}{T} \propto \int_{-\infty}^{+\infty} d\omega \left| E_{probe}(\omega) \right|^2 \int_{0}^{+\infty} \Psi^2_{TE_0}(r) dr \int_{-\infty}^{+\infty} P(\mu) d\mu \int_{-\infty}^{+\infty} \alpha\left(\omega, \omega_\xi, \mu, \theta\left(E_{pump}, \mu\right)\right) f(\omega_\xi) d\omega_\xi$$

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



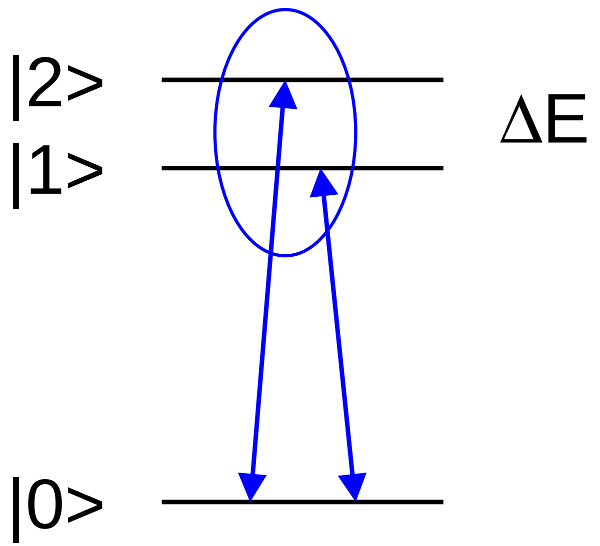
in average
μ = 35 D
σ = 20%

Borri et al.,
Phys. Rev. B
(Rapid Comm.),
in press

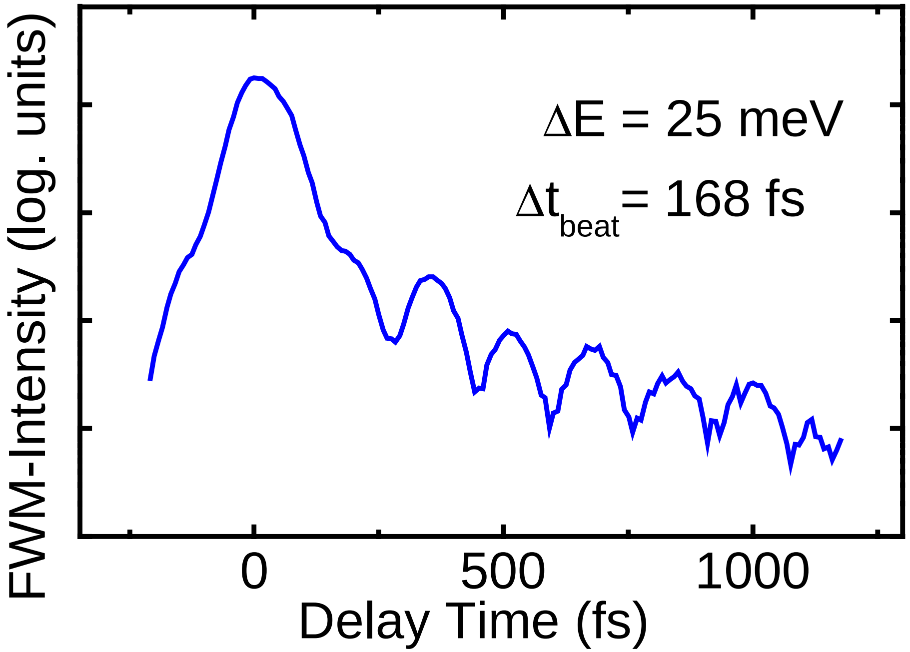
Part 3: *Fundamental aspects: Artficial atoms...*

Quantum Beats in Quantum Dots

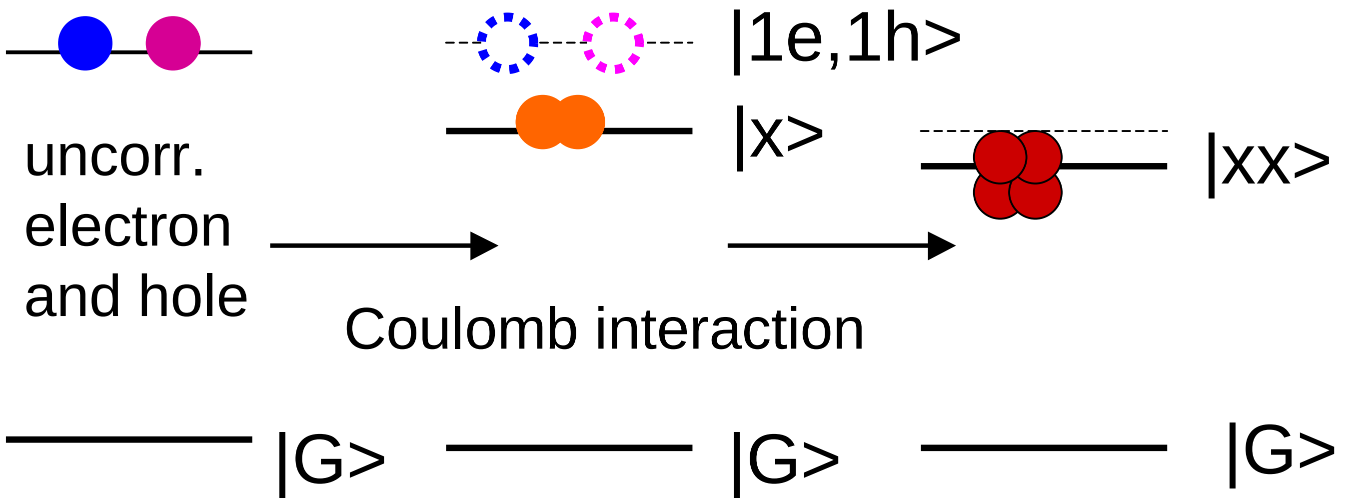
Discrete Level-System



ΔE can be derived from beat period



Exciton-Biexciton Quantum Beats in QDs

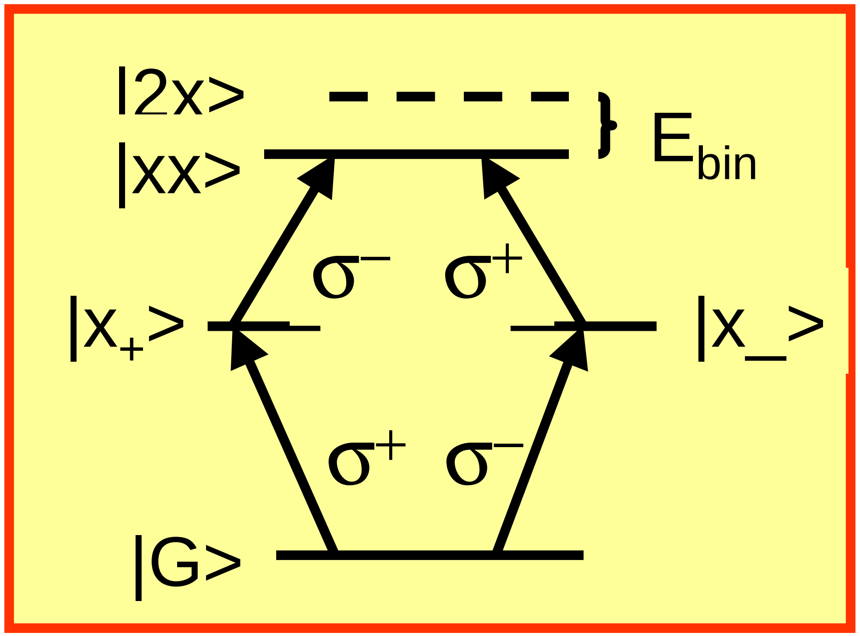


exciton |x>
biexciton |xx>
formation

Quantum Beats between
two optical transitions:

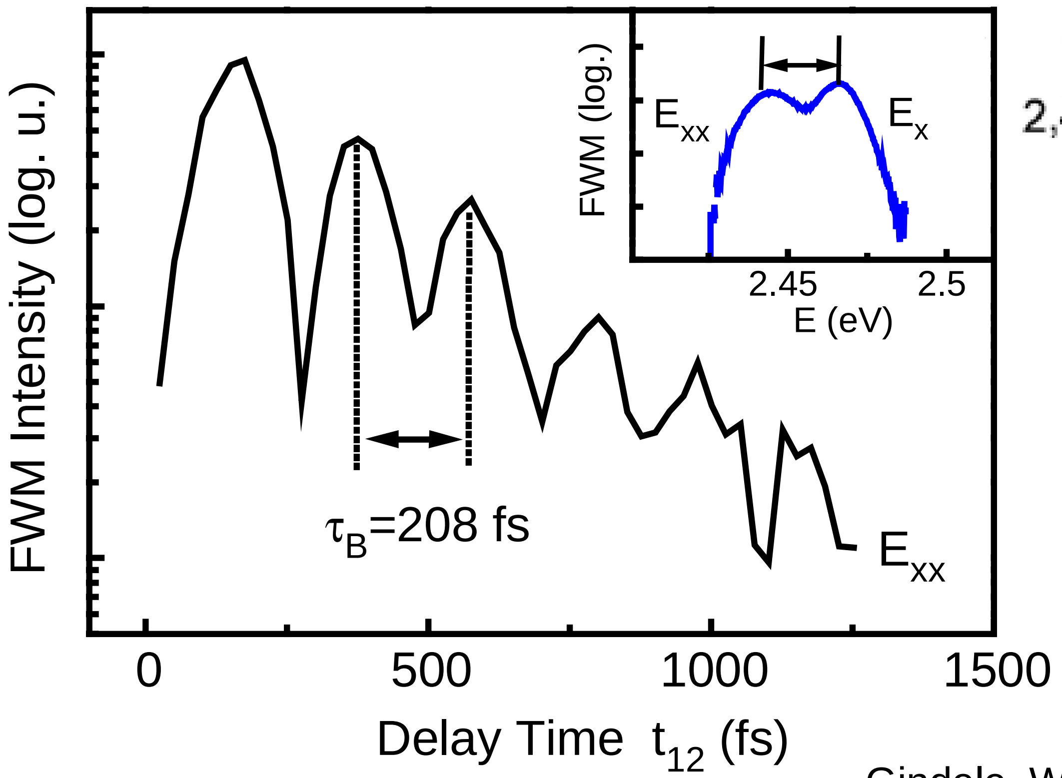
|G> ⟶ |x> with E_X
|x> ⟶ |xx> with E_{XX}

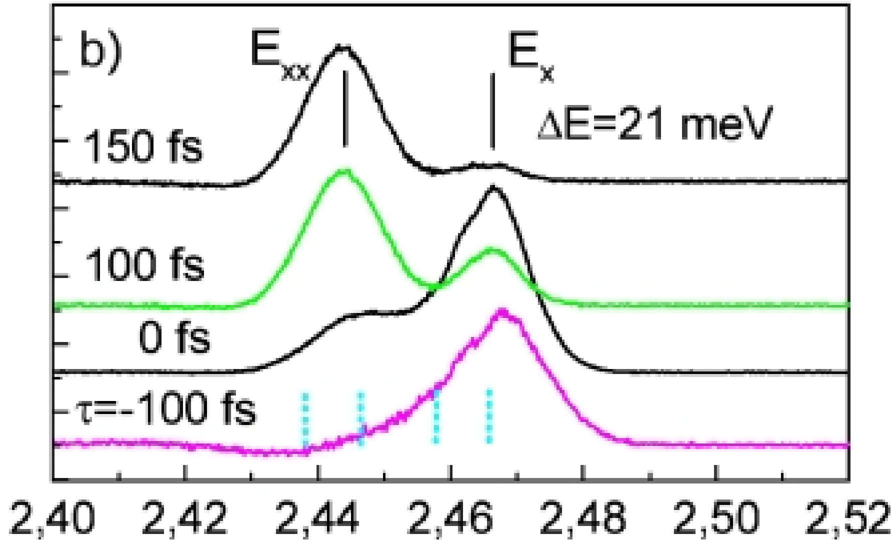
$E_X - E_{XX} = E_{bin}$ (biexciton binding)



Exciton-Biexciton Quantum Beats in QDs

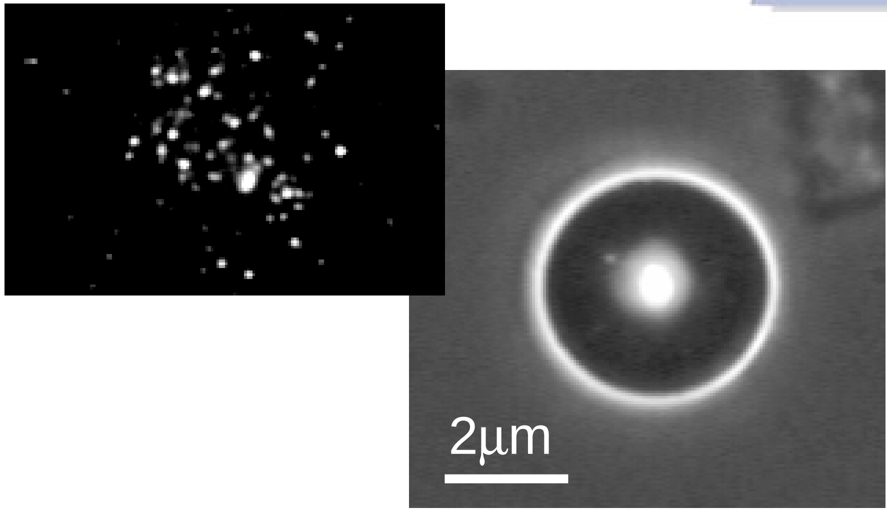
Determination of biexciton binding energy in CdSe/ZnSe QDs by femtosecond quantum beat spectroscopy

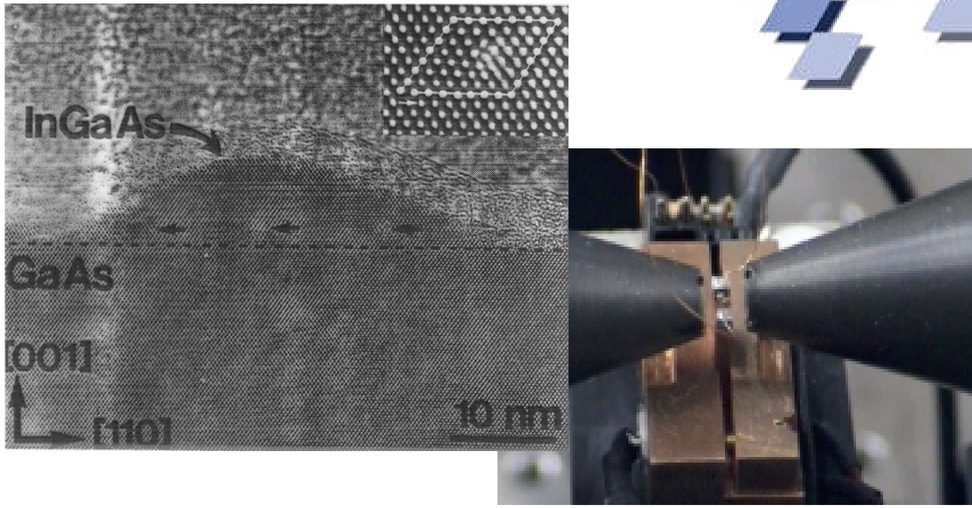




Biexciton binding energy ΔE = 21 meV

Gindele, Woggon et al., Phys. Rev. B 60, p. 8773 (1999).





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 Artficial Atoms*

For discussion

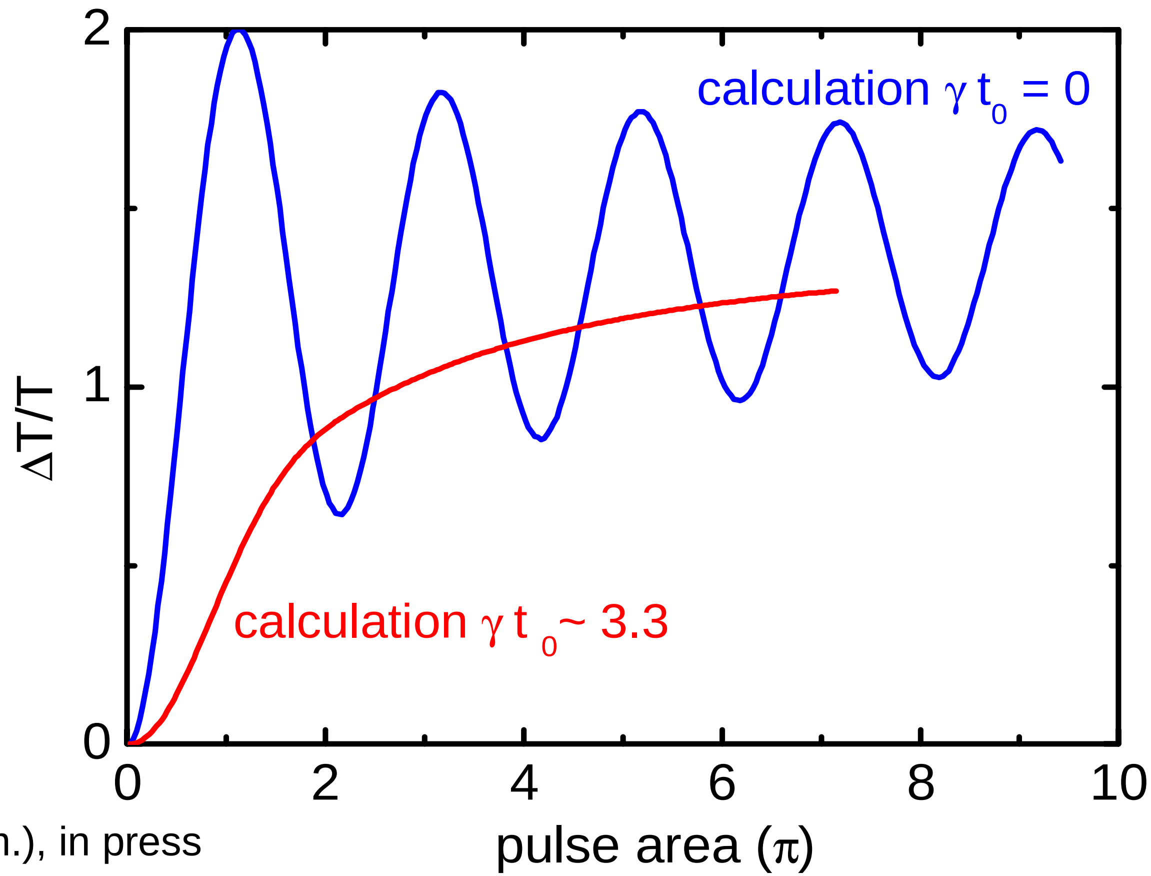
Rabi oscillations in differential transmission

Absorption coefficient of an inhomogeneously broadened ensemble:

$$\alpha(\omega) = \int_{-\infty}^{+\infty} \sigma_0 \frac{1/T_2}{(\omega-\omega_\xi)^2 + (1/T_2)^2} \Delta N \; f(\omega_\xi) d\omega_\xi$$

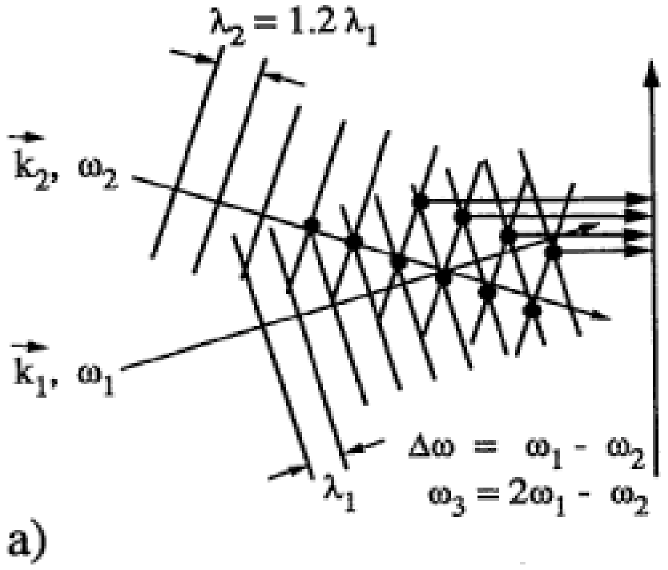
α is probed by a weak probe pulse after the pump: differential transmission intensity of the probe

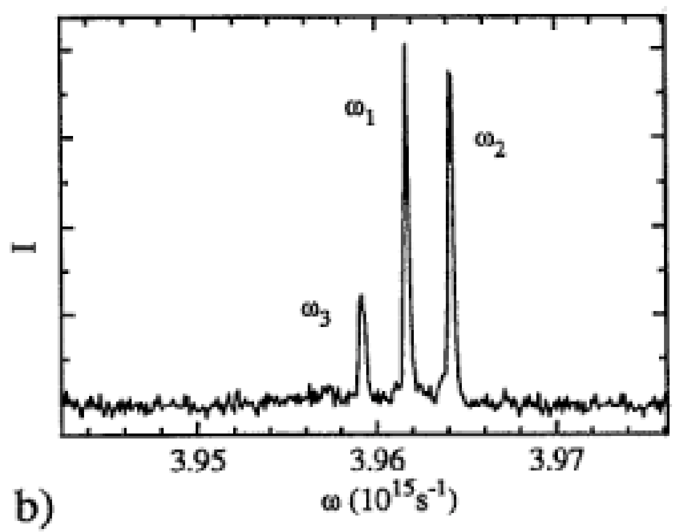
dephasing time T_2= 1.5 ps
(T_2= 0 ps),
pulse length t_0 = 5 ps



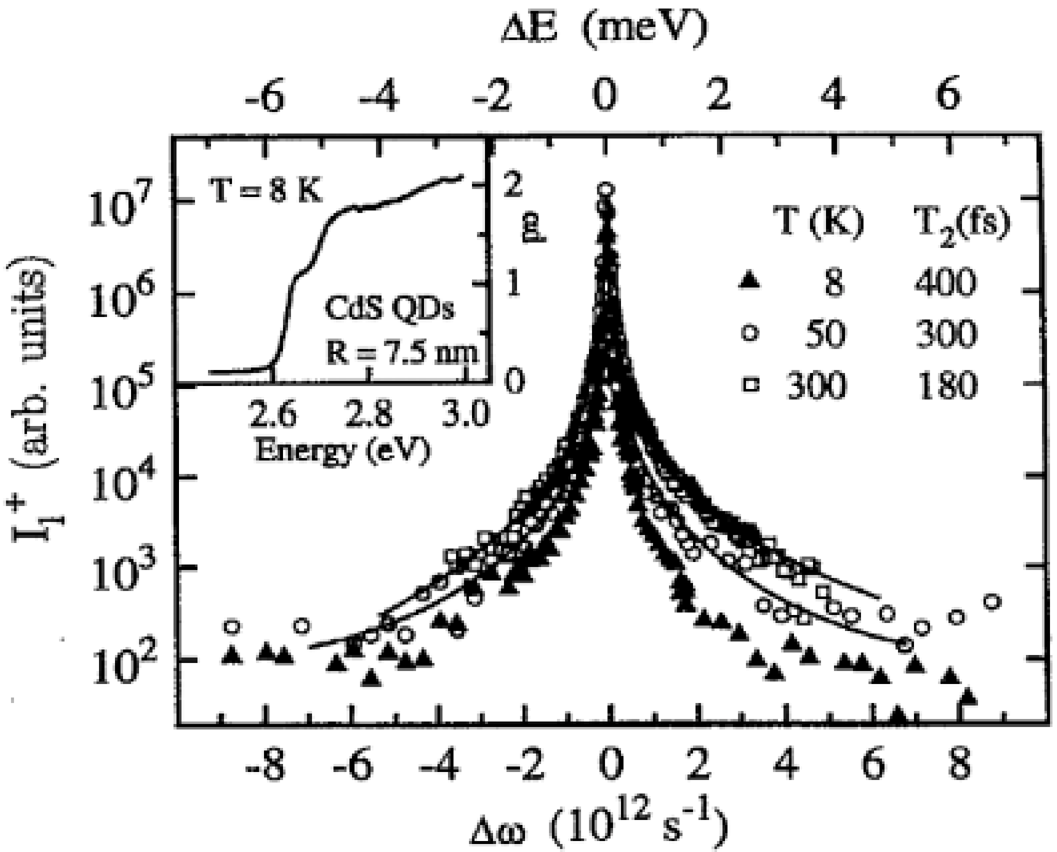
Borri et al., Phys. Rev. B (Rapid Comm.), in press

Non-degenerate FWM with ns-pulses





asymmetric line shape at T < 10 K



*U. Woggon and M. Portune, Phys. Rev. B **51**, 4719 (1995).*

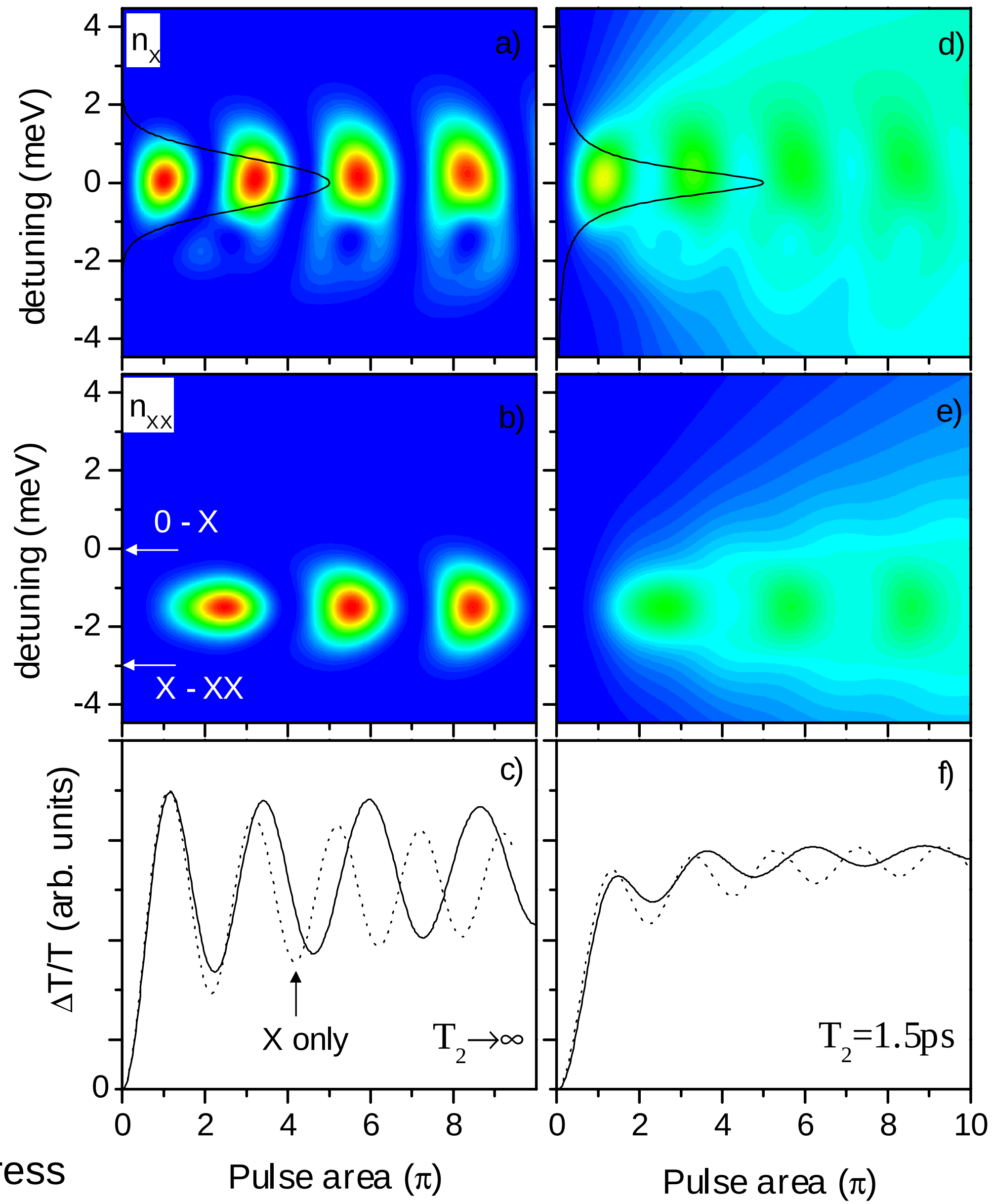
Rabi oscillations in InGaAs quantum dots

Example: t_0=1ps

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

The dephasing reduces the amplitude of the oscillations.

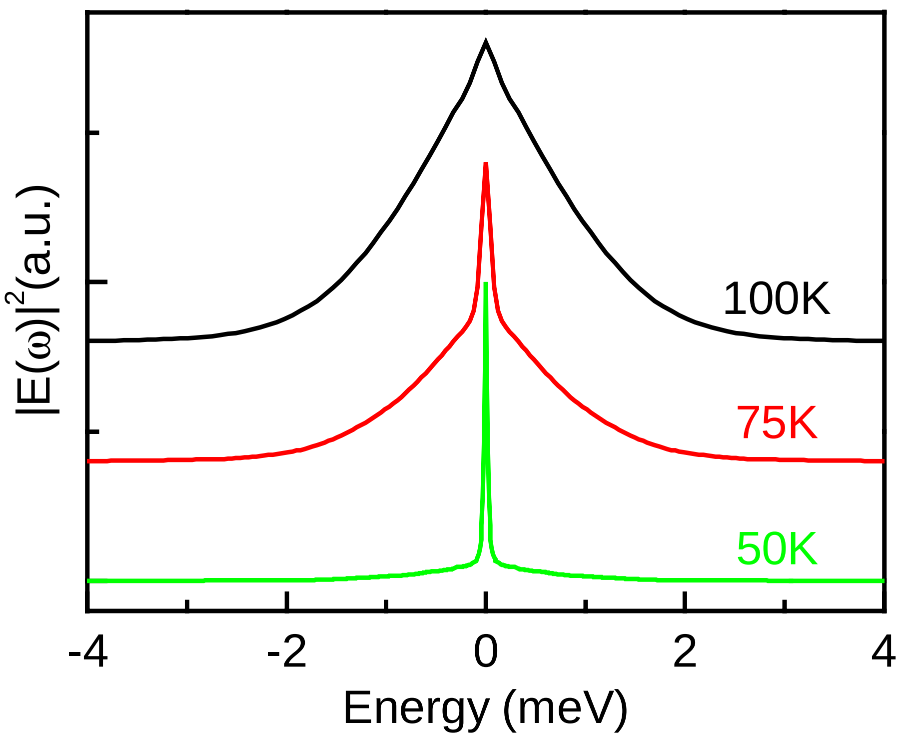
Many oscillation periods are present, even when the averaging over the inhomogeneous broadening and the spatial mode profile are included.

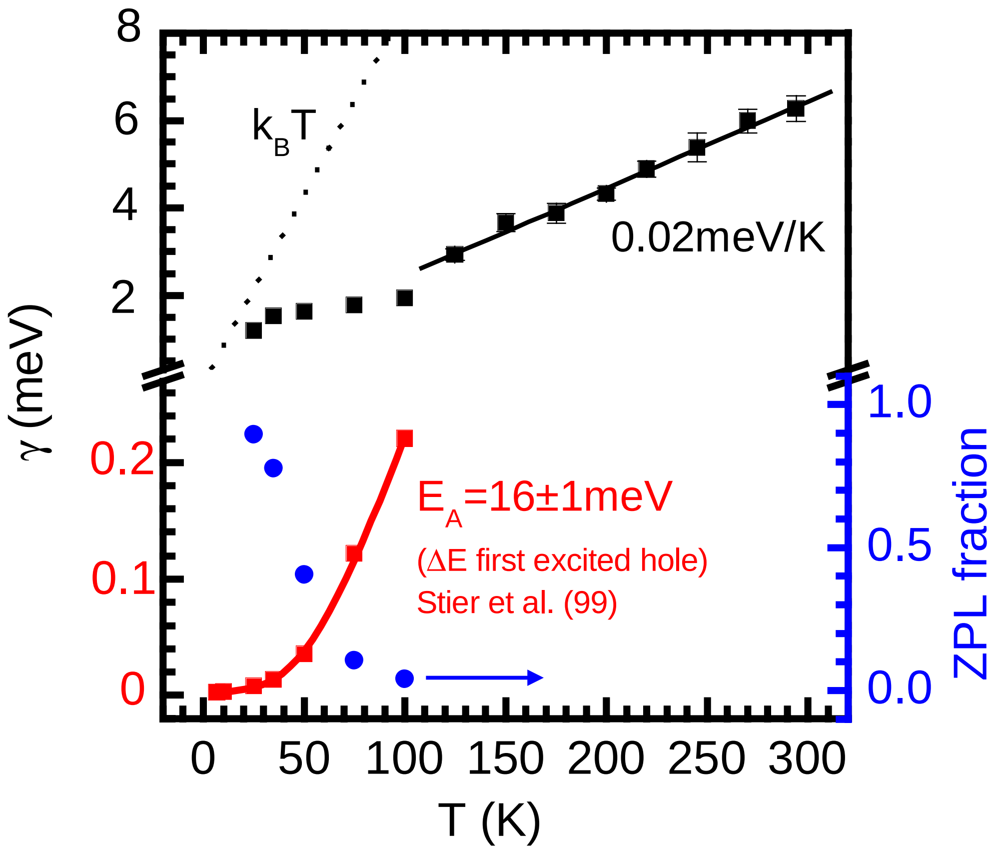


Borri et al., Phys. Rev. B (Rapid Comm.), in press

Analysis of homogeneous line broadening - InGaAs

Fourier transform of the TI-FWM signal





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

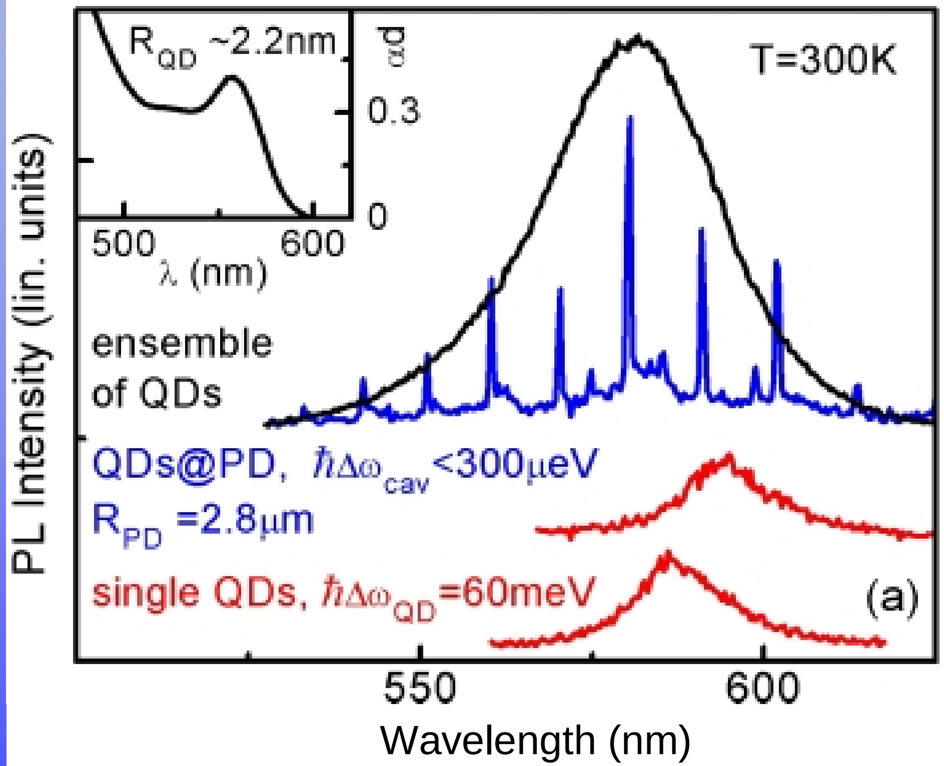
Extrapolation to T = 0K:

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

Dephasing close to the radiative limit !

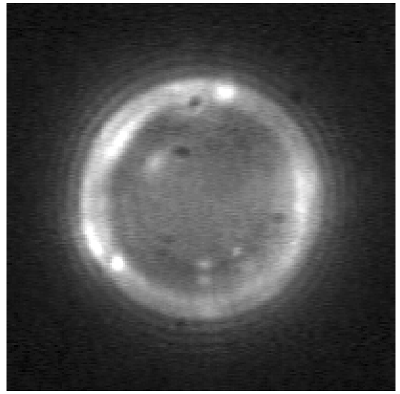
P. Borri *et al.*, Phys. Rev. Lett. **87**, 157401 (2001)

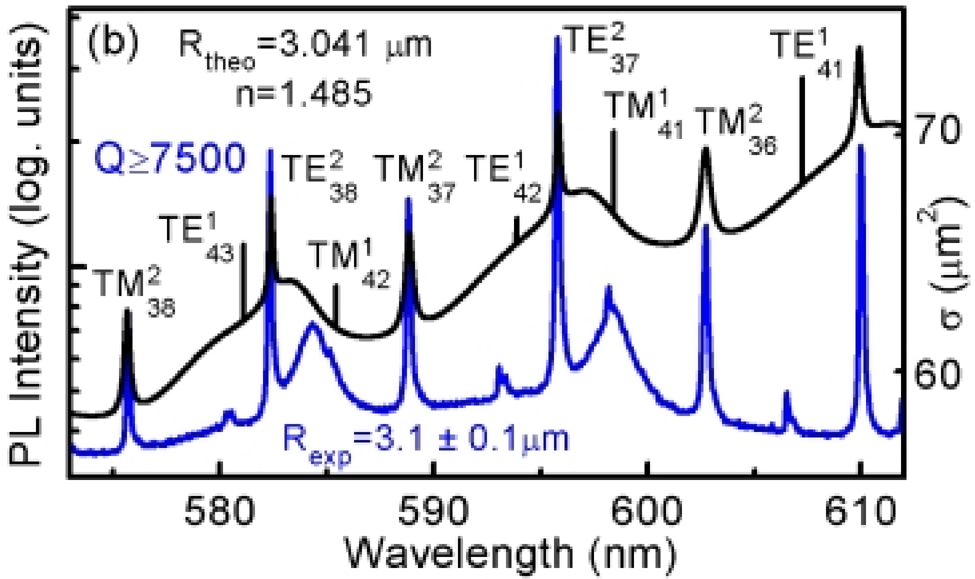
CdSe QDs attached to a glass μ-sphere



Here: CdSe-shell
on glass μ-sphere

R=3.1 μm

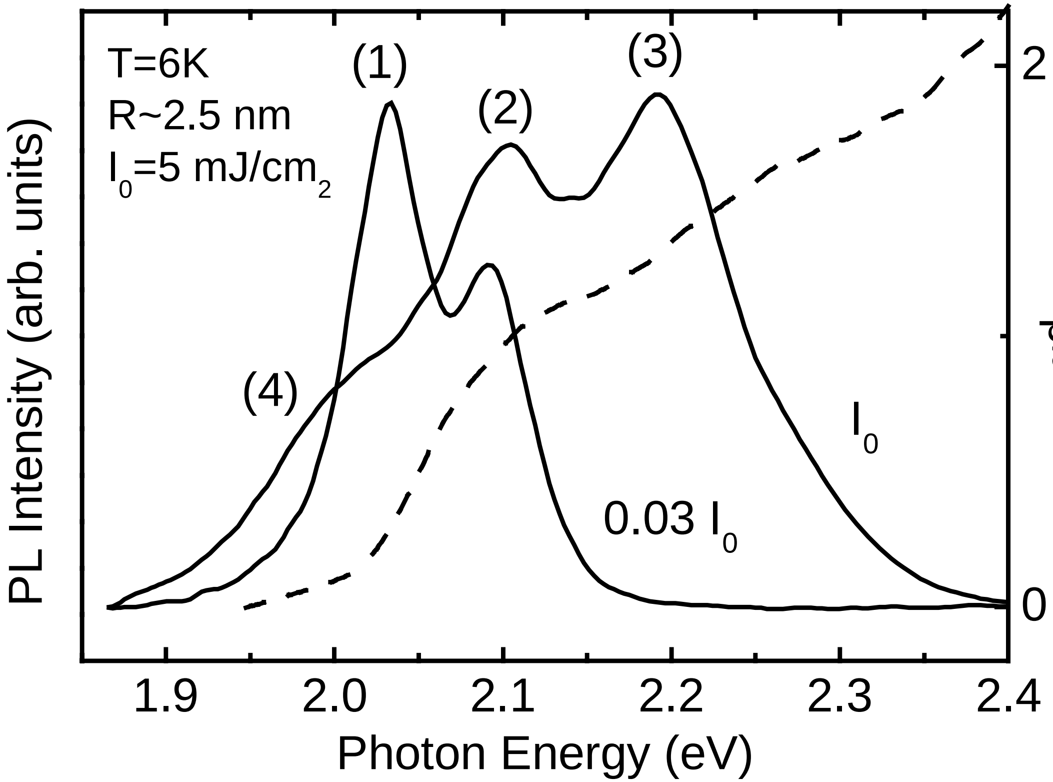


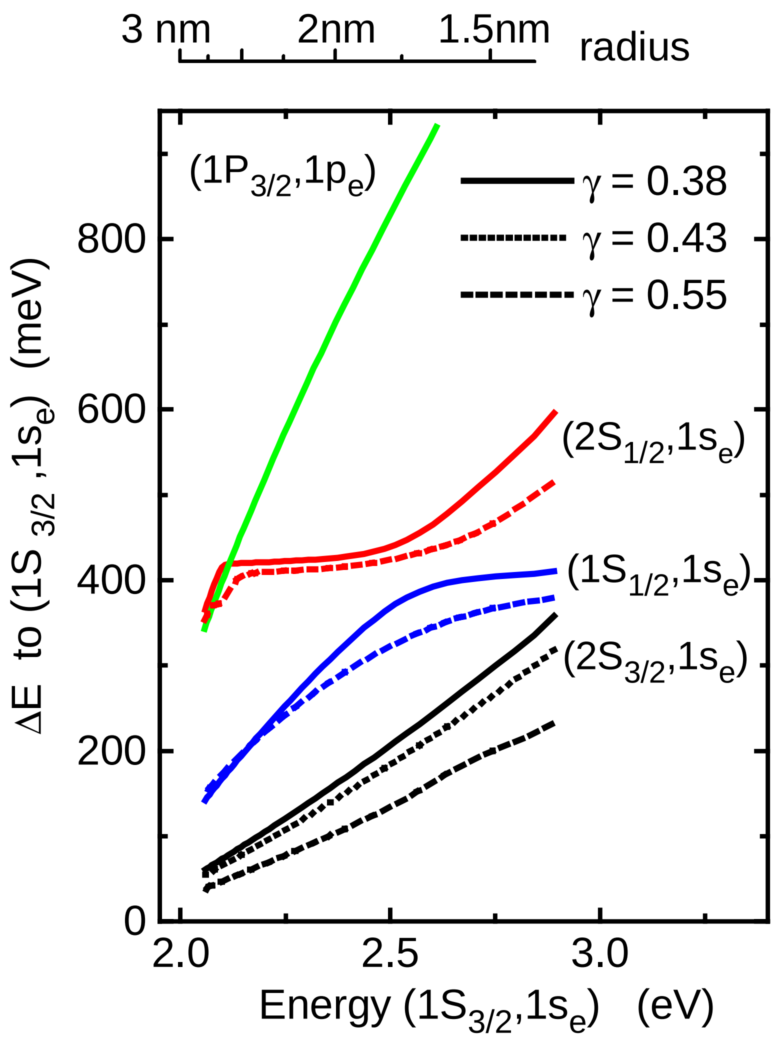


mode separation > Γ_{QD}
room temperature emission

Nano Lett. 1, 309 (2001)

CdSe Quantum Dots

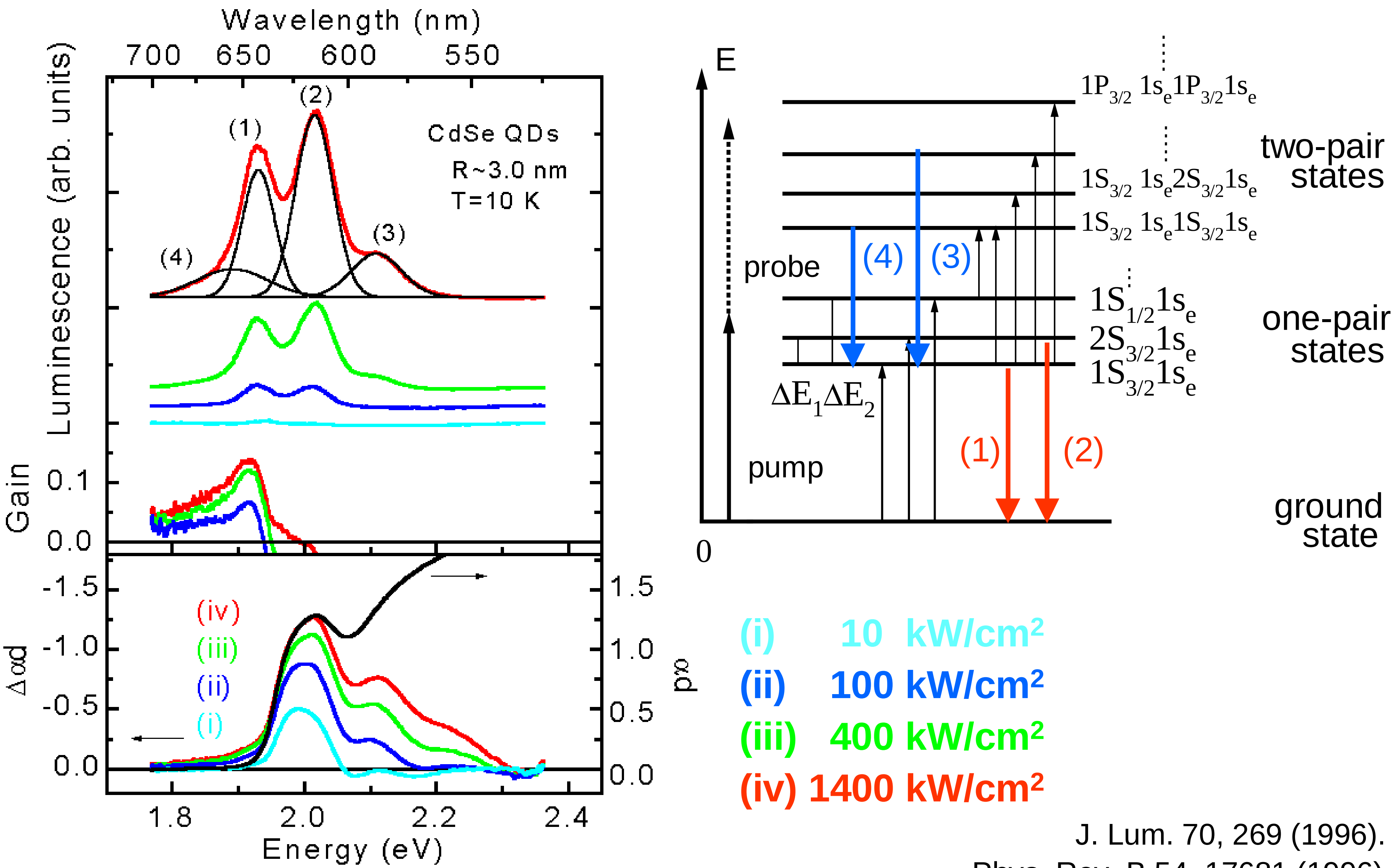




excitons	biexcitons
(1) $1S_{3/2}\,1s_e$	**(3) $1s_e,1S_{3/2},1s_e,2S_{3/2}$**
(2) $2S_{3/2}\,1s_e$	**(4) $1s_e,1S_{3/2},1s_e,1S_{3/2}$**

J. Cryst. Growth 159, 867 (1996)

Spectra of PL and optical gain at high excitation



(i) 10 kW/cm²

(ii) 100 kW/cm²

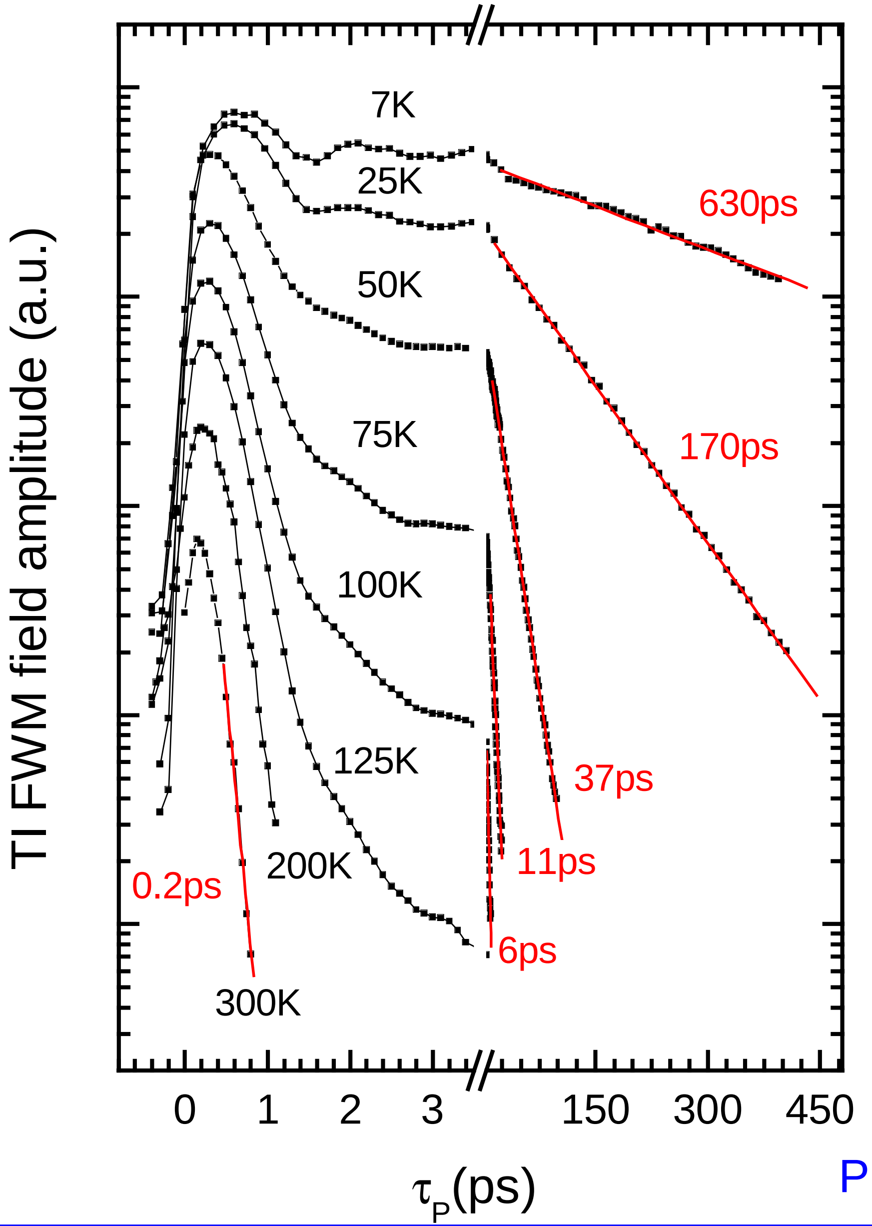
(iii) 400 kW/cm²

(iv) 1400 kW/cm²

J. Lum. 70, 269 (1996).

Phys. Rev. B 54, 17681 (1996).

Temperature-dependent dephasing time T_2



Temperature-dependent FWM

InGaAs SA-QDs

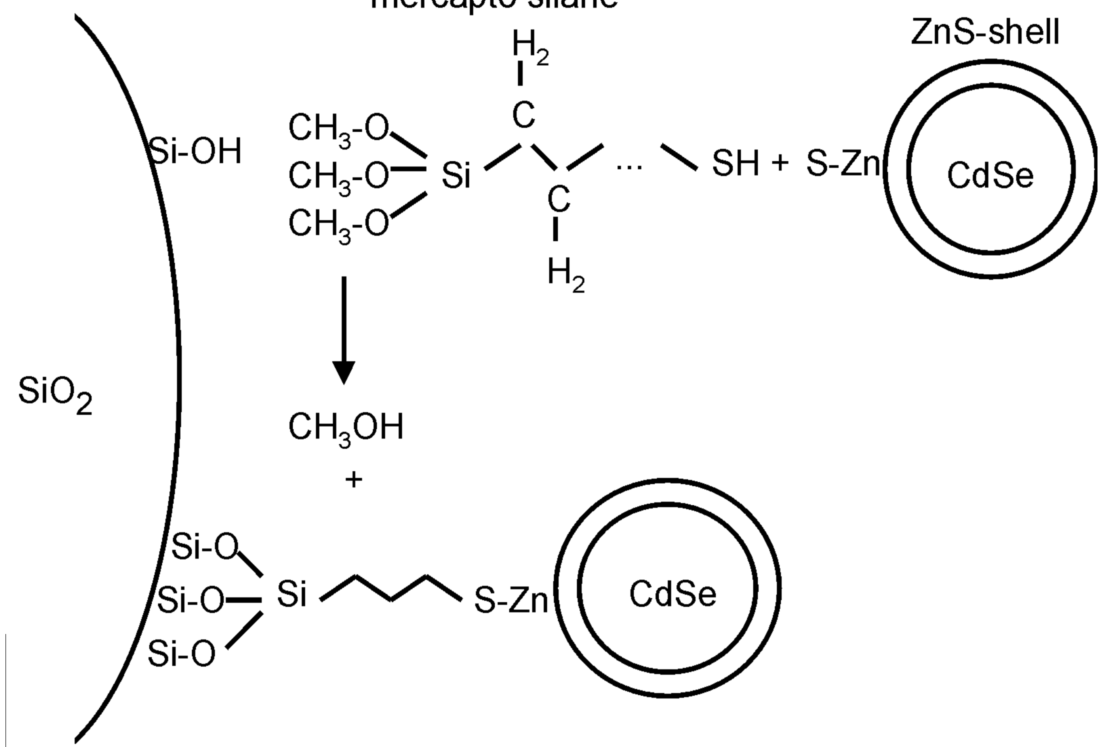
From 300K to 100K the FWM decay is dominated by a dephasing time below 1ps

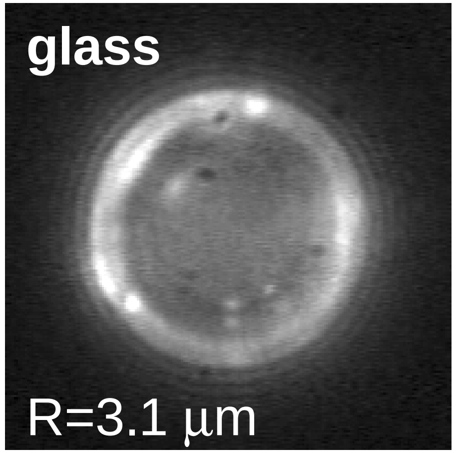
Below 100K a *slow component* appears with an exponential decay time that increases with decreasing temperature.

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

P. Borri *et al.*, Phys. Rev. Lett. **87**, 157401 (2001)

Spherical Microcavities - Photonic Dots





Artemyev et al., APL 78, p.1032 (2001),
Nano Lett. 1, 309 (2001).