分子研研究会
赤外放射光の現状と将来計画
2002年11月13日

Osaka University

レーザーを用いた赤外時間分解分光

半導体量子ドットに閉じ込められた励起子の励起状態の研究

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Outline

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1. Introduction
   - Excited states of excitons confined in semiconductor quantum dots
   - CuCl quantum dots
2. Experimental procedure
   - Infrared transient absorption spectroscopy
   - ns and ps systems
3. Results
   - Excited-state absorption of confined excitons in CuCl quantum dots
   - Excited-state absorption of confined biexcitons
4. Summary

Semiconductor quantum structure

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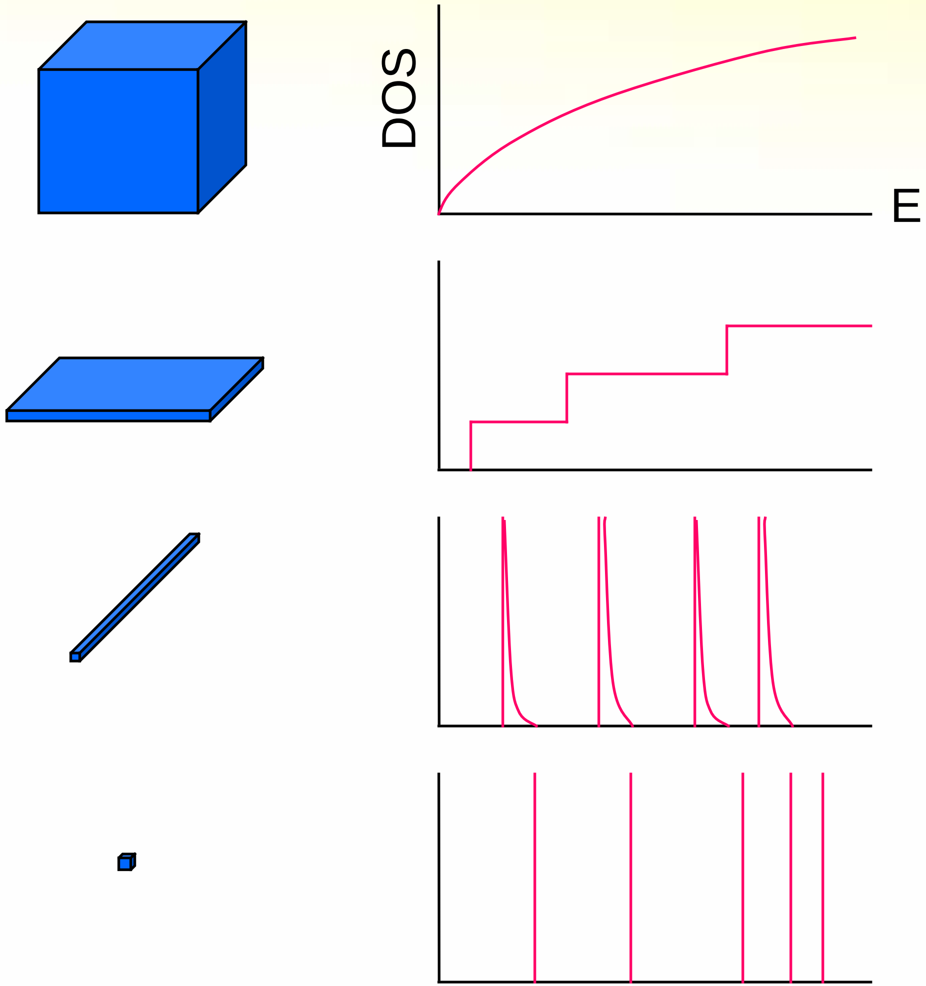
3D: Bulk crystal

2D: Quantum well

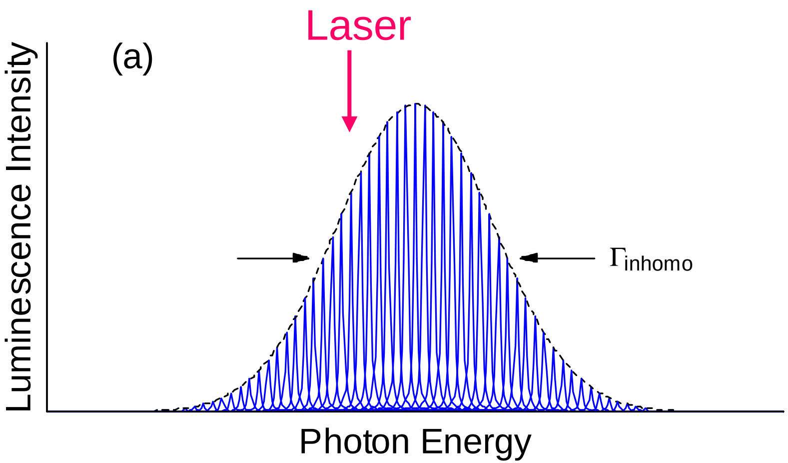
1D: Quantum wire

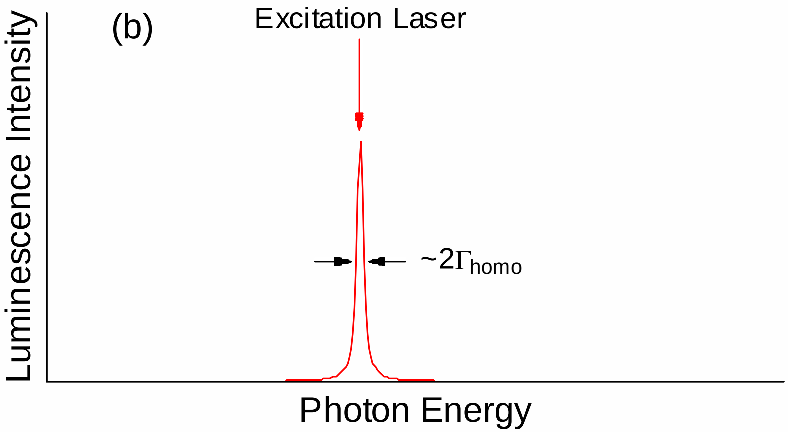
0D: Quantum dot (QD)
(or nanocrystal)



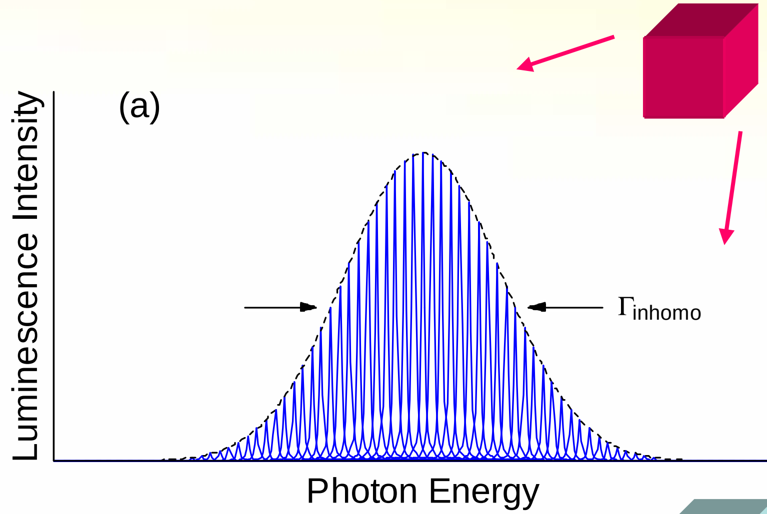
Size-selective observation of QDs

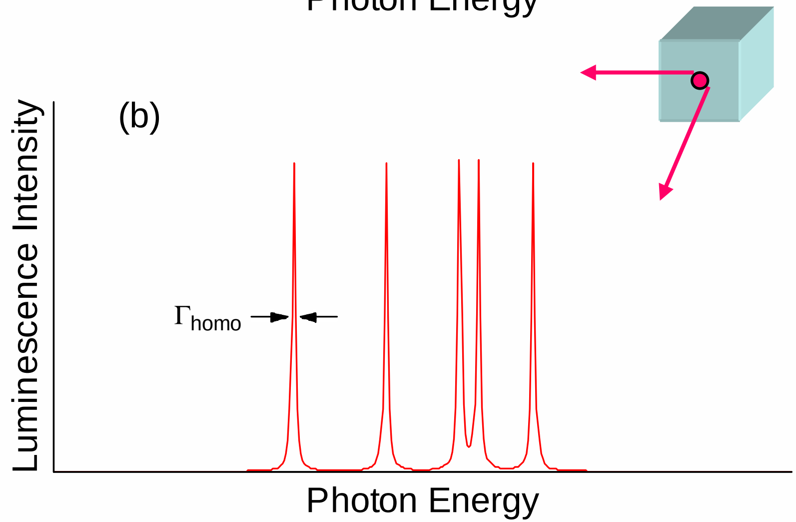
1. Resonant size-selective excitation





2. micro-photoluminescence





Quantum size effect in spherical QDs

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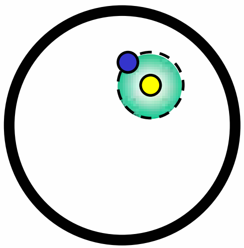
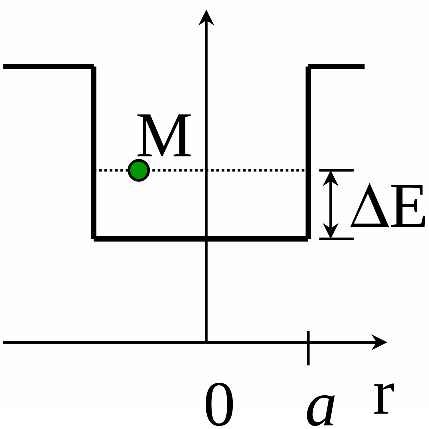
• Exciton confinement.
($a_B \ll a$)



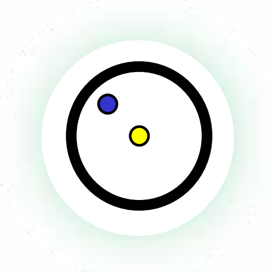
$$\Delta E = \frac{\hbar^2}{2M}\left(\frac{\pi}{a}\right)^2$$

$$M = m_e + m_h$$

M: translational mass



• Electron-hole individual confinement.
($a_B \gg a$)

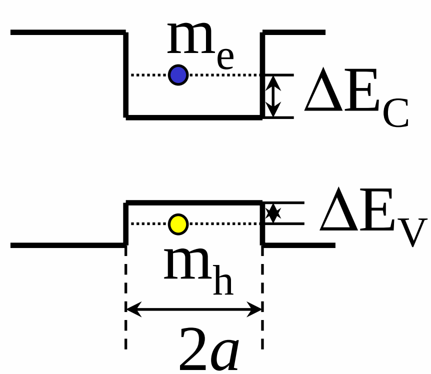


$$\Delta E = \Delta E_C + \Delta E_V$$

$$= \frac{\hbar^2}{2\mu}\left(\frac{\pi}{a}\right)^2$$

$$\mu^{-1} = m_e^{-1} + m_h^{-1}$$

μ: reduced mass



Confined excitons in CuCl QDs

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Excitons in CuCl quantum dots

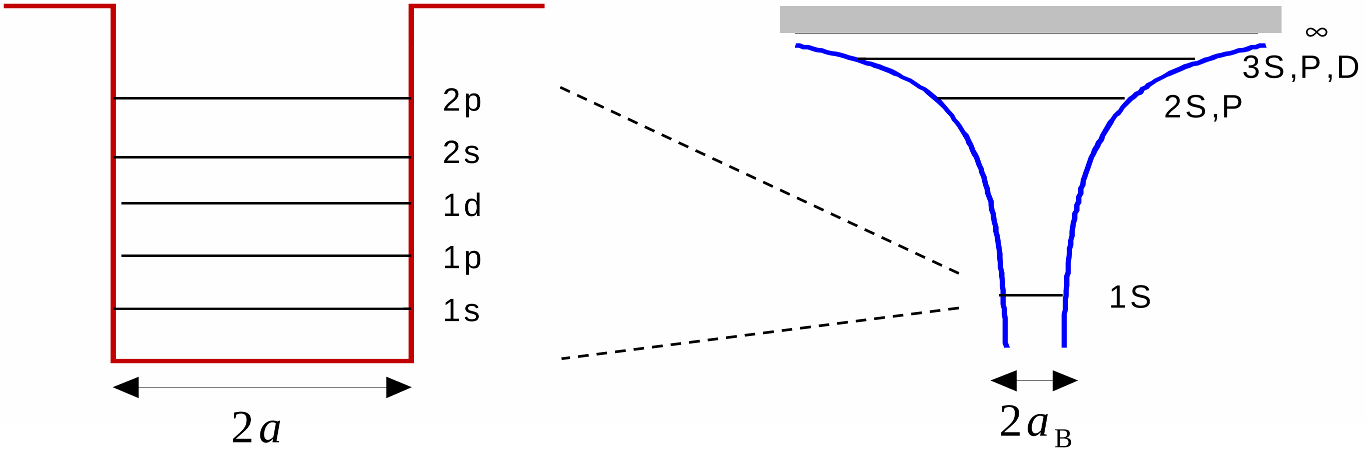
a_B=0.7 nm (1S); Ry*=197 meV
Confinement energy～ 10meV (a* = 4 nm)

⇒ Exciton confinement regime

Two quantum numbers of the confined exciton:

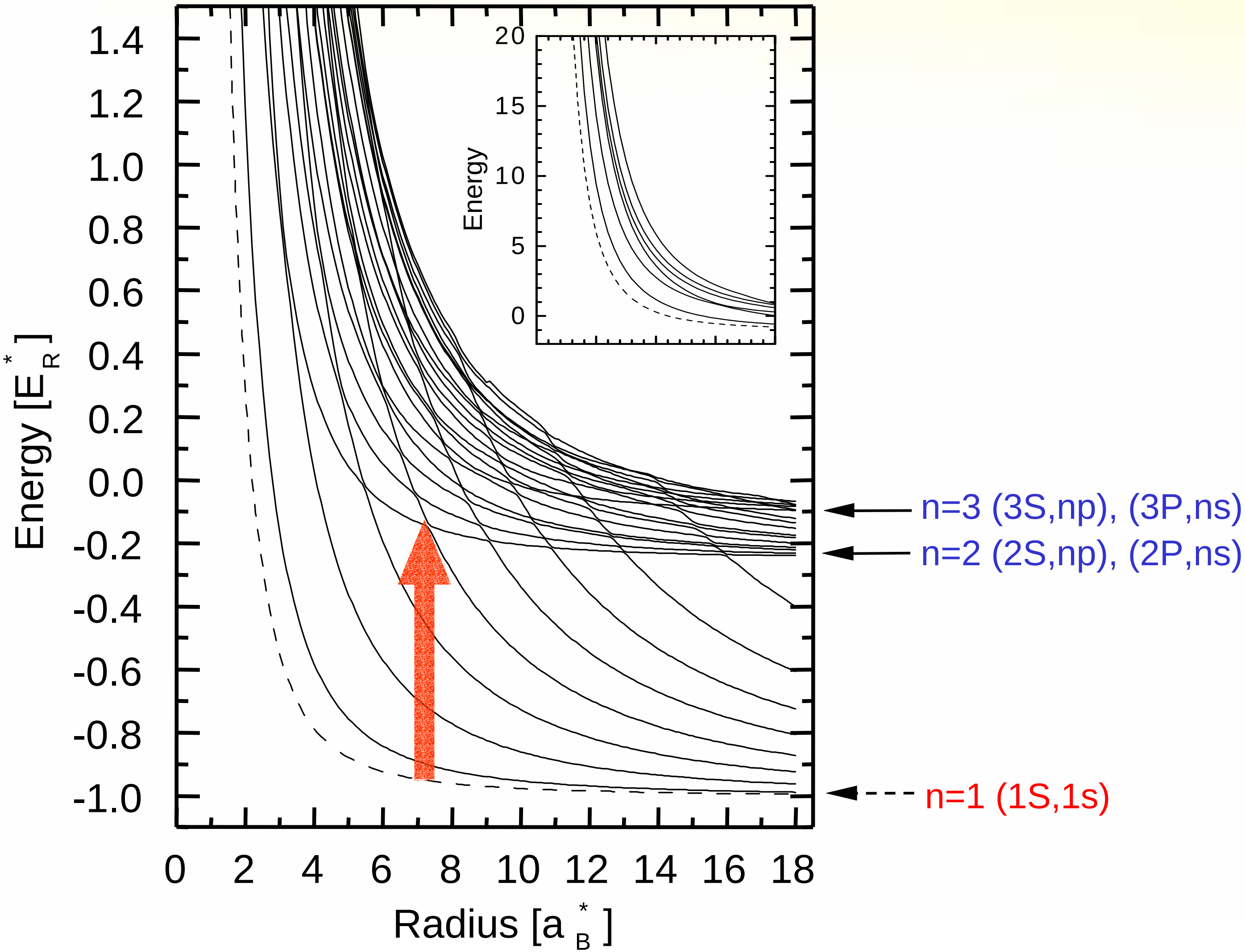
1) Confinement of translational motion (*nl*)

2) Coulombic states (*n'L*)



Energy levels of confined excitons

T. Uozumi, *et al*., Phys. Rev. B **59**, 9826 (1999)



Excited-state absorption of excitons confined in CuCl quantum dots

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(Infrared transient absorption)

- Mimura *et al*., J. Lumin. **66&67**, 401 (1996)
- Yamanaka *et al*., J. Lumin. **76&77**, 256 (1998)
- Yamanaka *et al*., J. Lumin. **87&89**, 312 (2000)
- Itoh *et al*., Int. J. Mod. Phys. **15**, 3569 (2001)
- Miyajima *et al*., Phys. Stat. Sol. (b), in press.

(Two-photon excitation)

- Edamatsu *et al*., Phys. Rev. B **59**, 15868 (1999)

(Theoretical works)

- Kayanuma, Phys.Rev. B **38**, 9797 (1988)
- Uozumi *et al*., Phys. Rev. B **59**, 9826 (1999)
- Uozumi *et al*., Phys. Rev. B **65**, 165318 (2002)

Sample preparation

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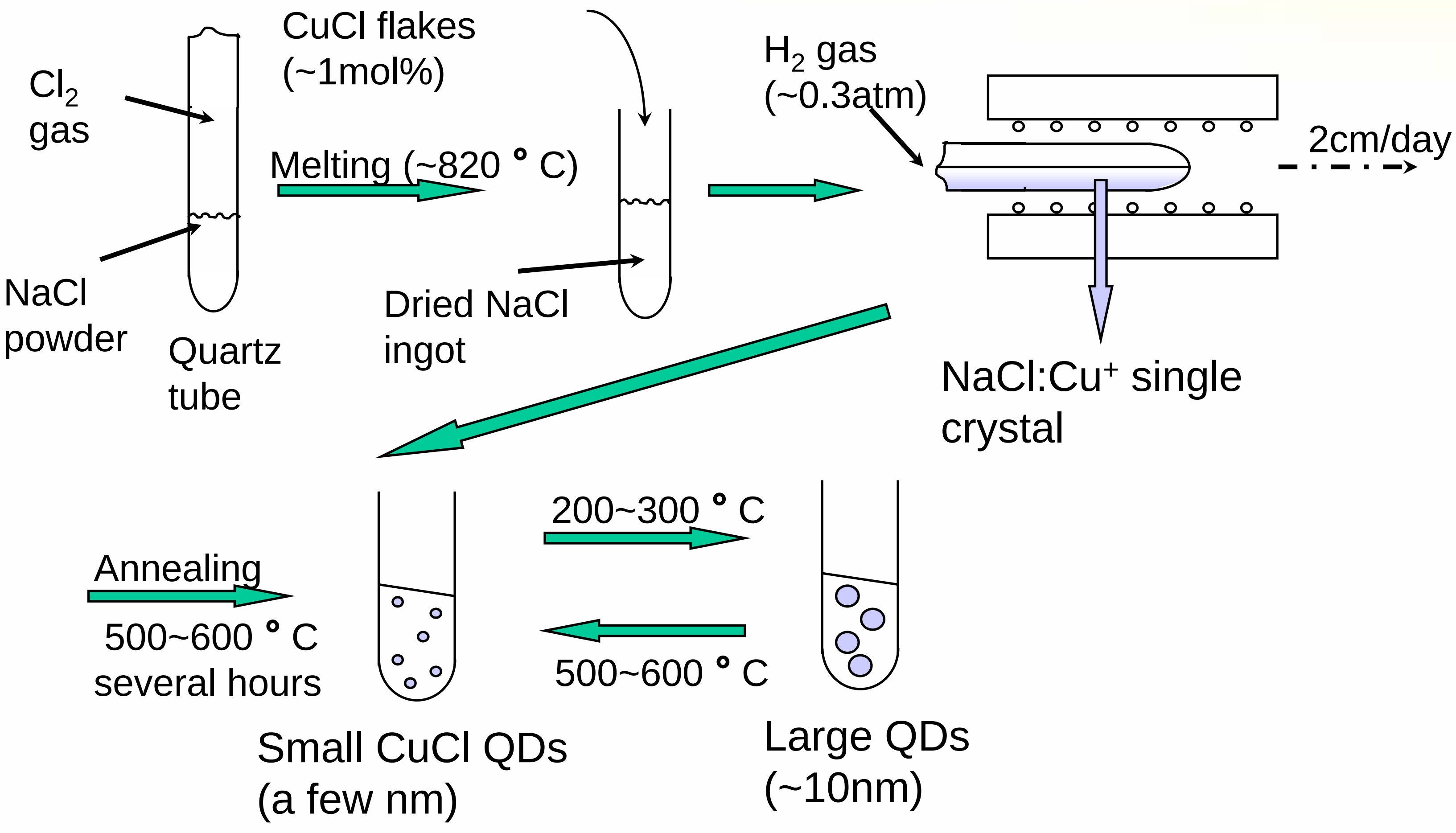
CuCl QDs (or nanocrystals) embedded in NaCl matrices



Absorption and luminescence spectra of CuCl QDs in NaCl matrices

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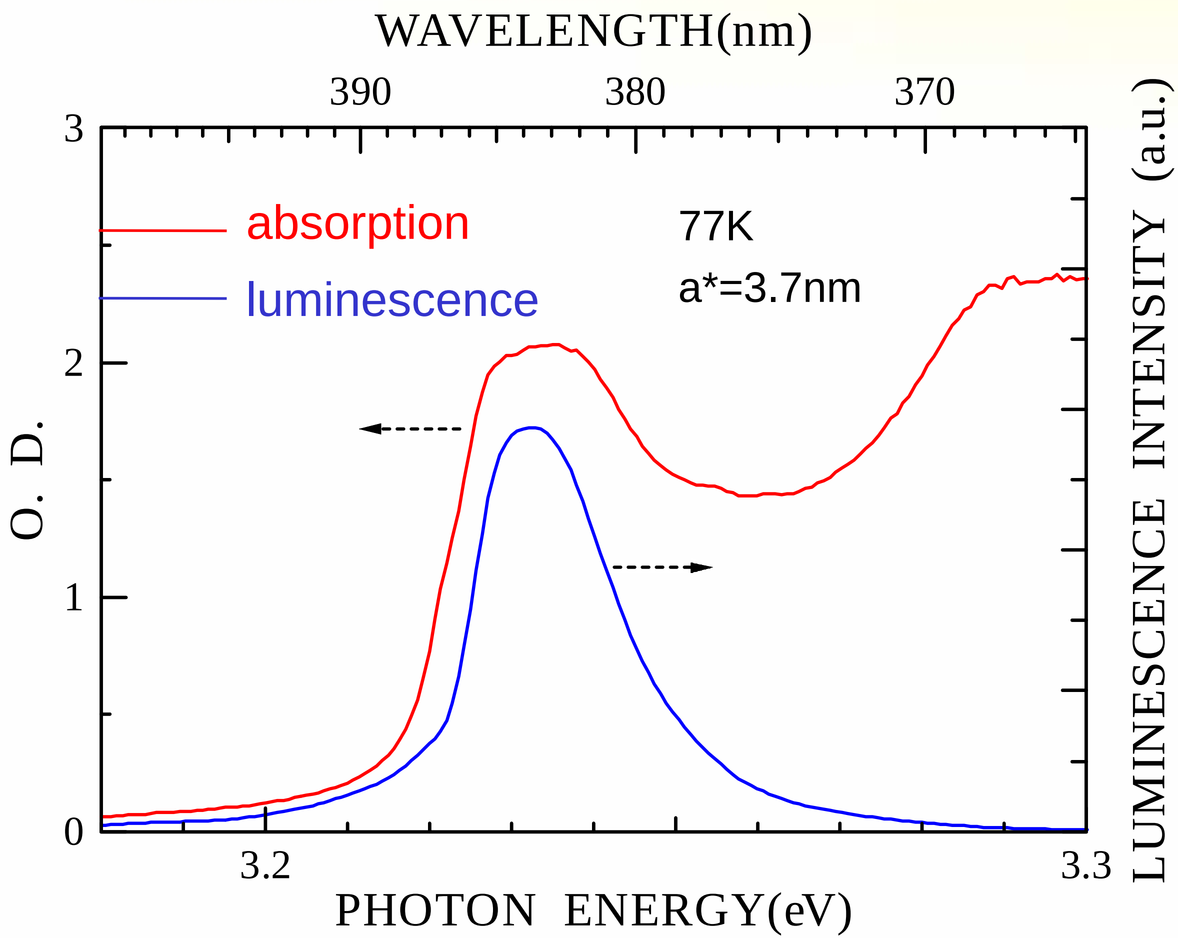

Transient absorption spectroscopy under size-selective excitation

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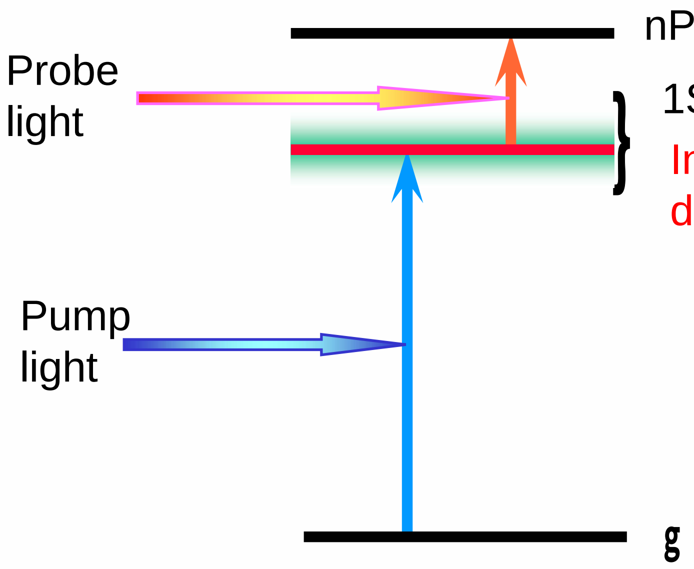
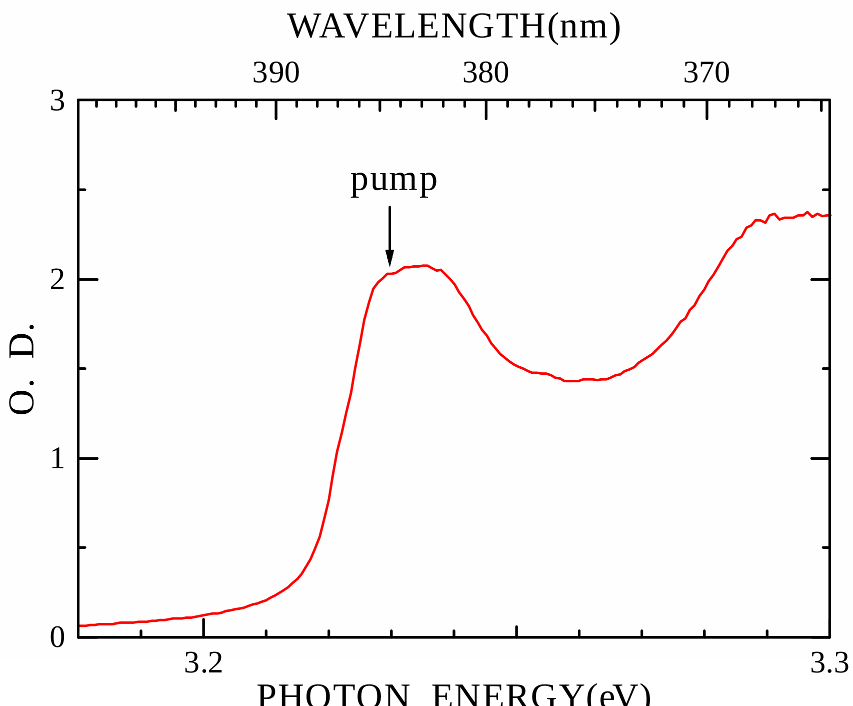


Inhomogeneous broadening due to size distribution.



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Experimental setup (ns system)

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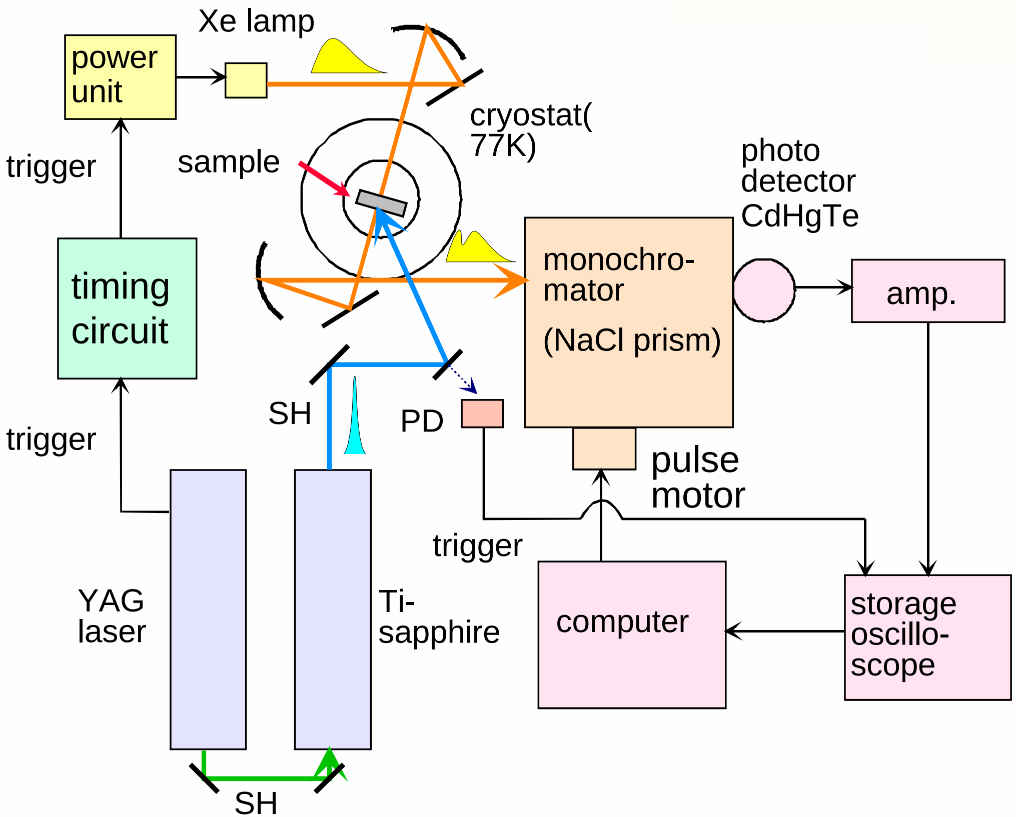
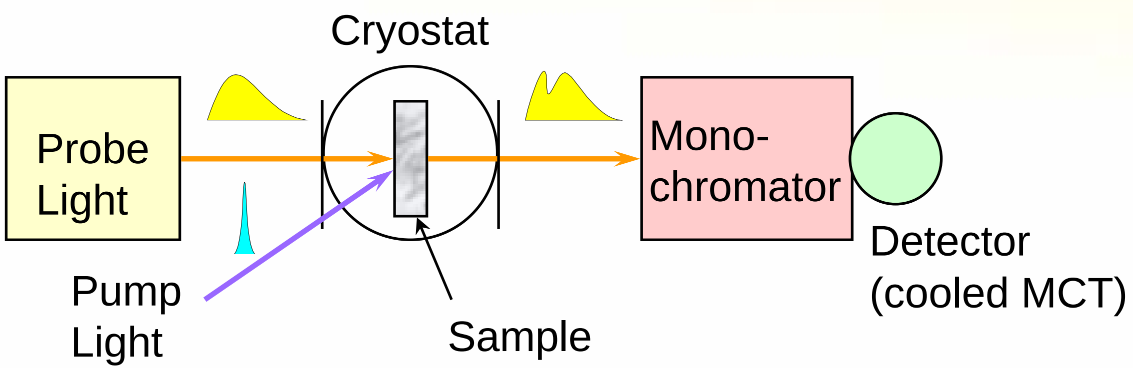
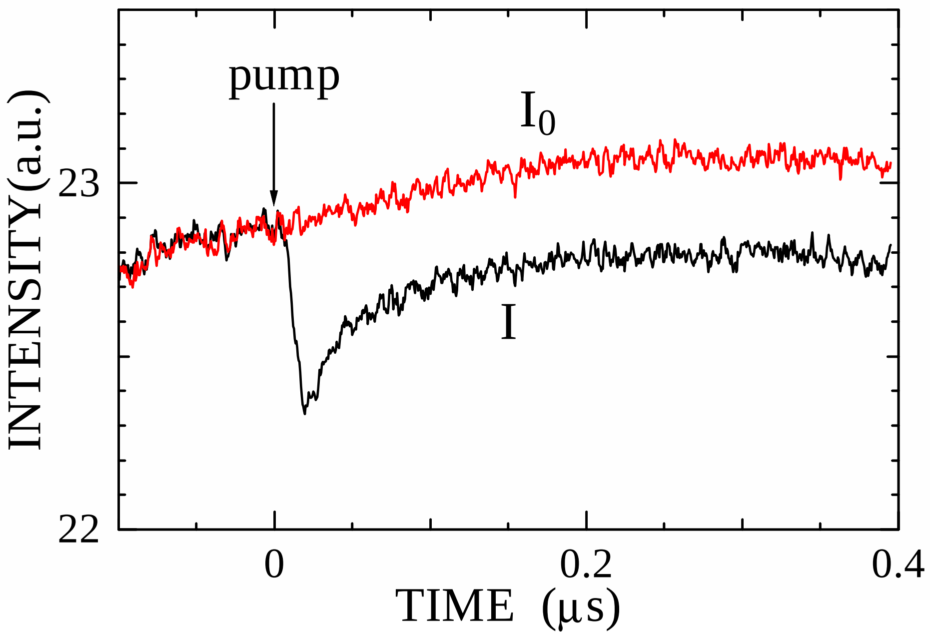


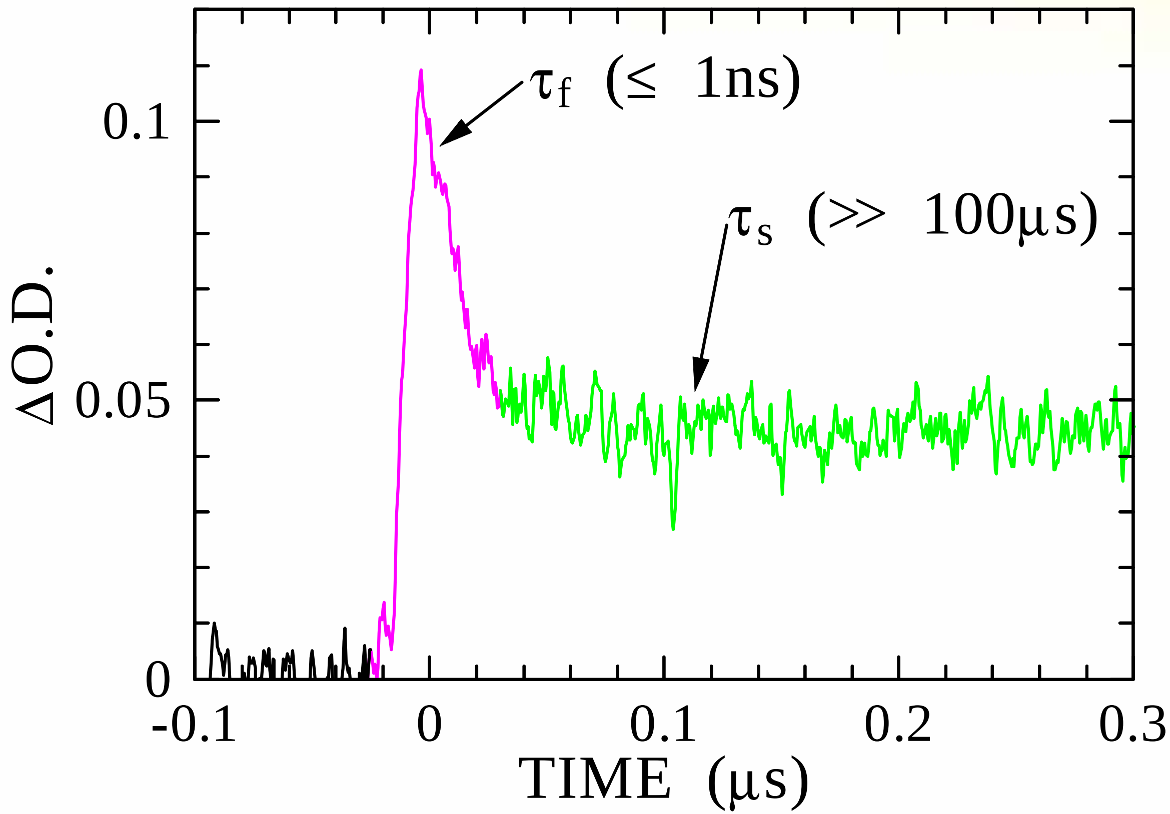
- Probe light source
  - Xe flash lamp
    - Pulse duration: ~8μs
    - 1~8μm (160~1200meV)
- Pump light source
  - Second harmonic of Ti:Sa laser
    - Tunability (SH)
      350~470nm(2.64~3.54eV)
    - Pulse energy : ~1mJ
    - Pulse duration: ~15ns
    - Excitation power : ~70MW/cm²
    - Line width <0.1nm(1meV)

Observation of the infrared transient absorption





Infrared transient absorption: temporal profile



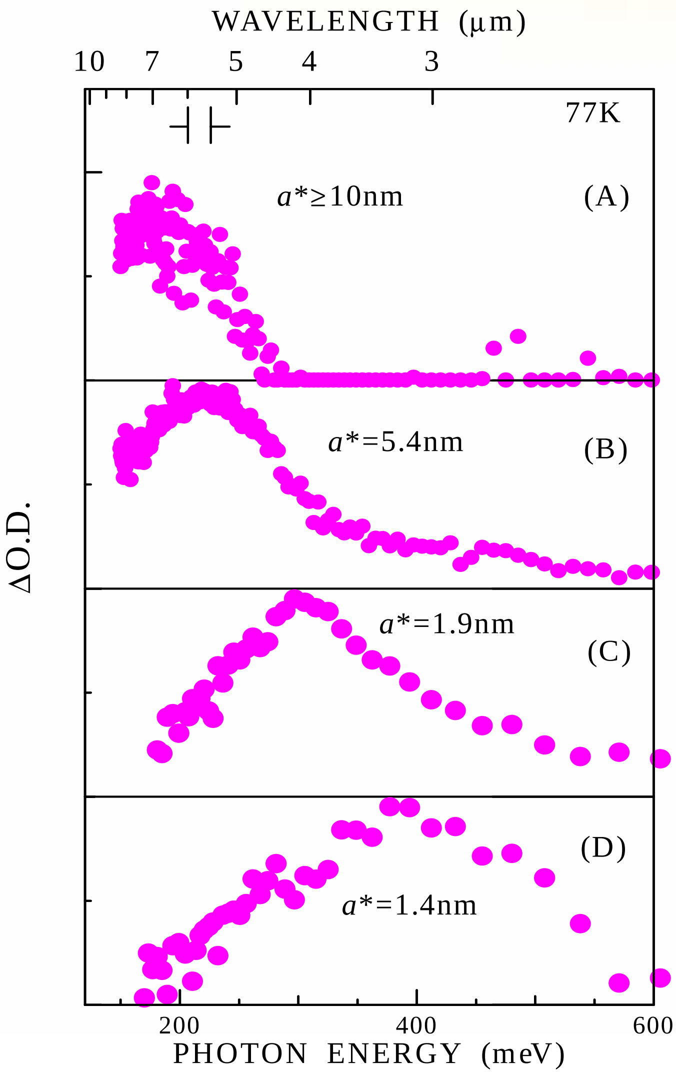
Excitation: 3.224 eV
(a^*~6 nm)

Probe: 246 meV
(λ=5.03 μm)

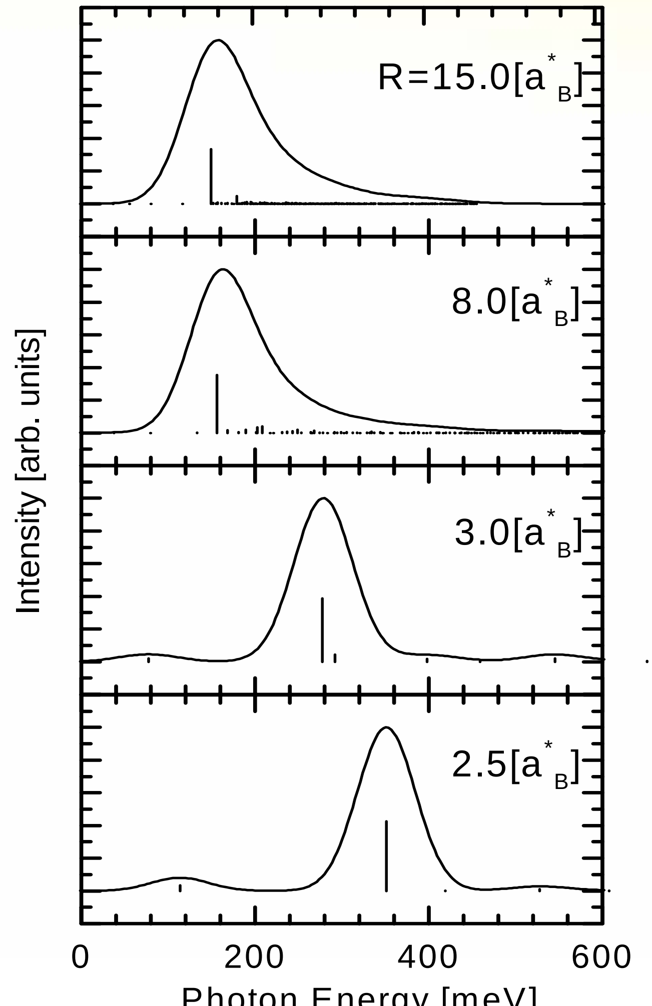
Fast decay component (τ_f):
originates from the confined exciton (1S → 2P)

Transient absorption spectra

Experiment



Theory (Uozumi and Kayanuma)



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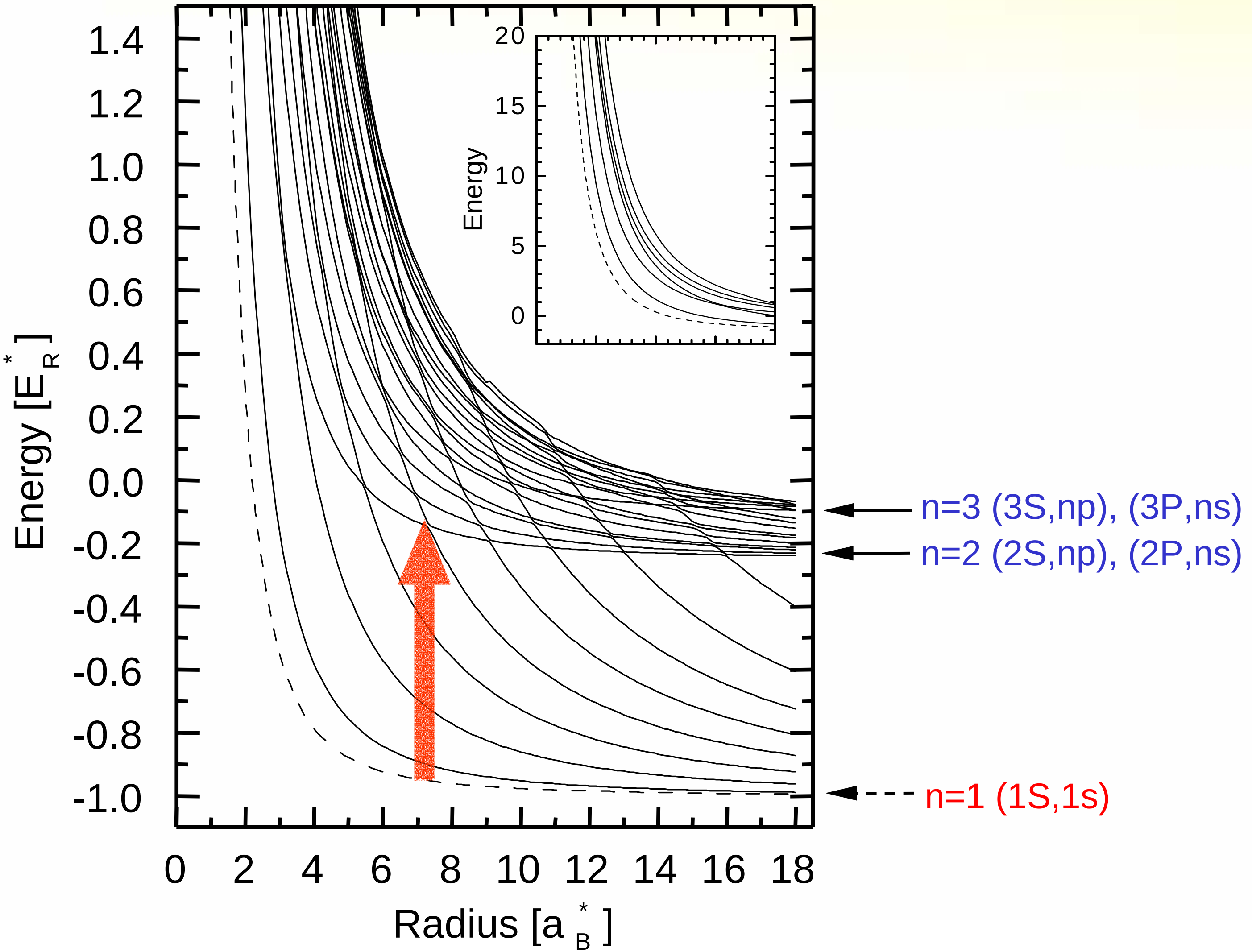

1S → P-like state transition energy: comparison with theory

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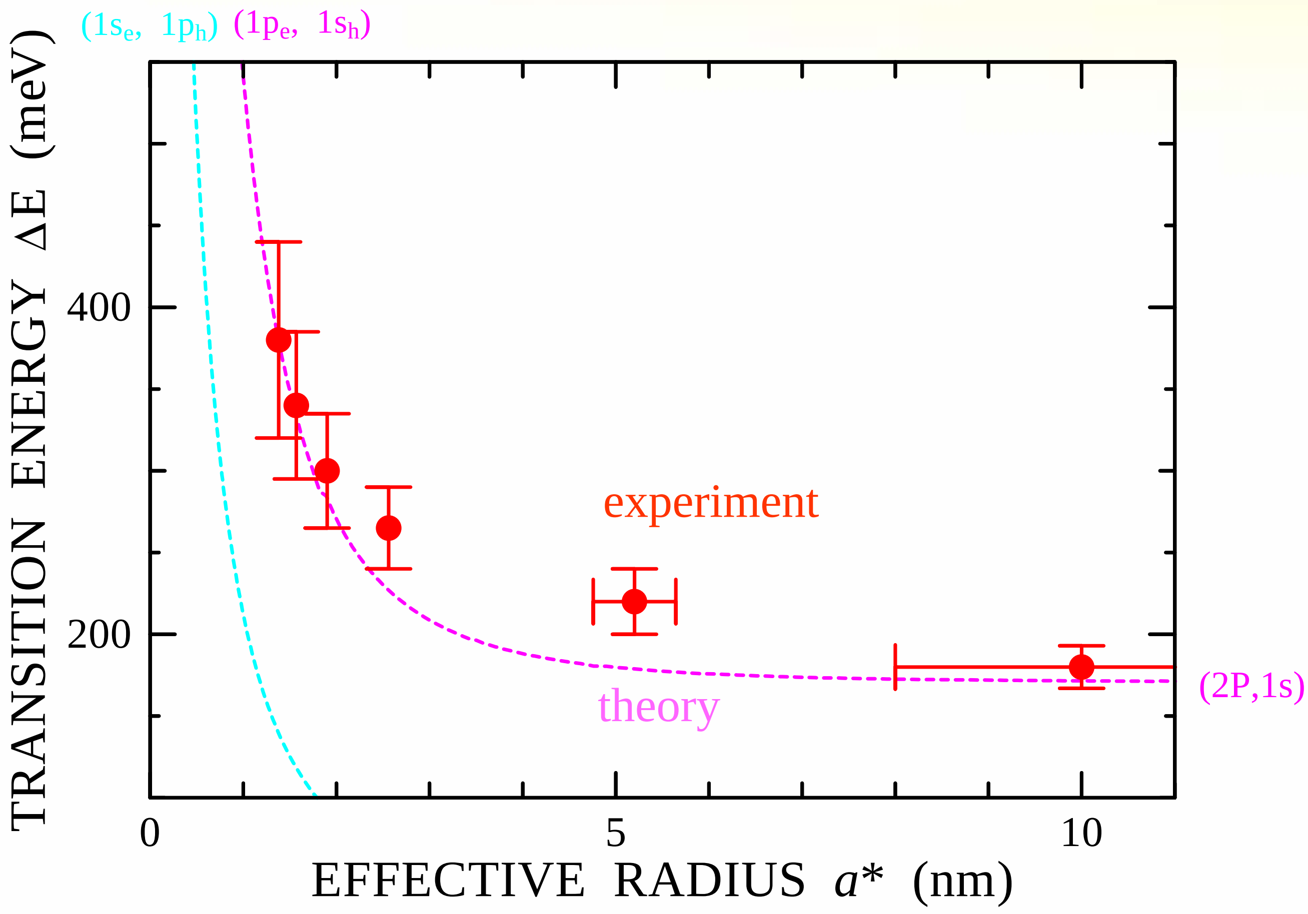

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Motivation of the ps experiment

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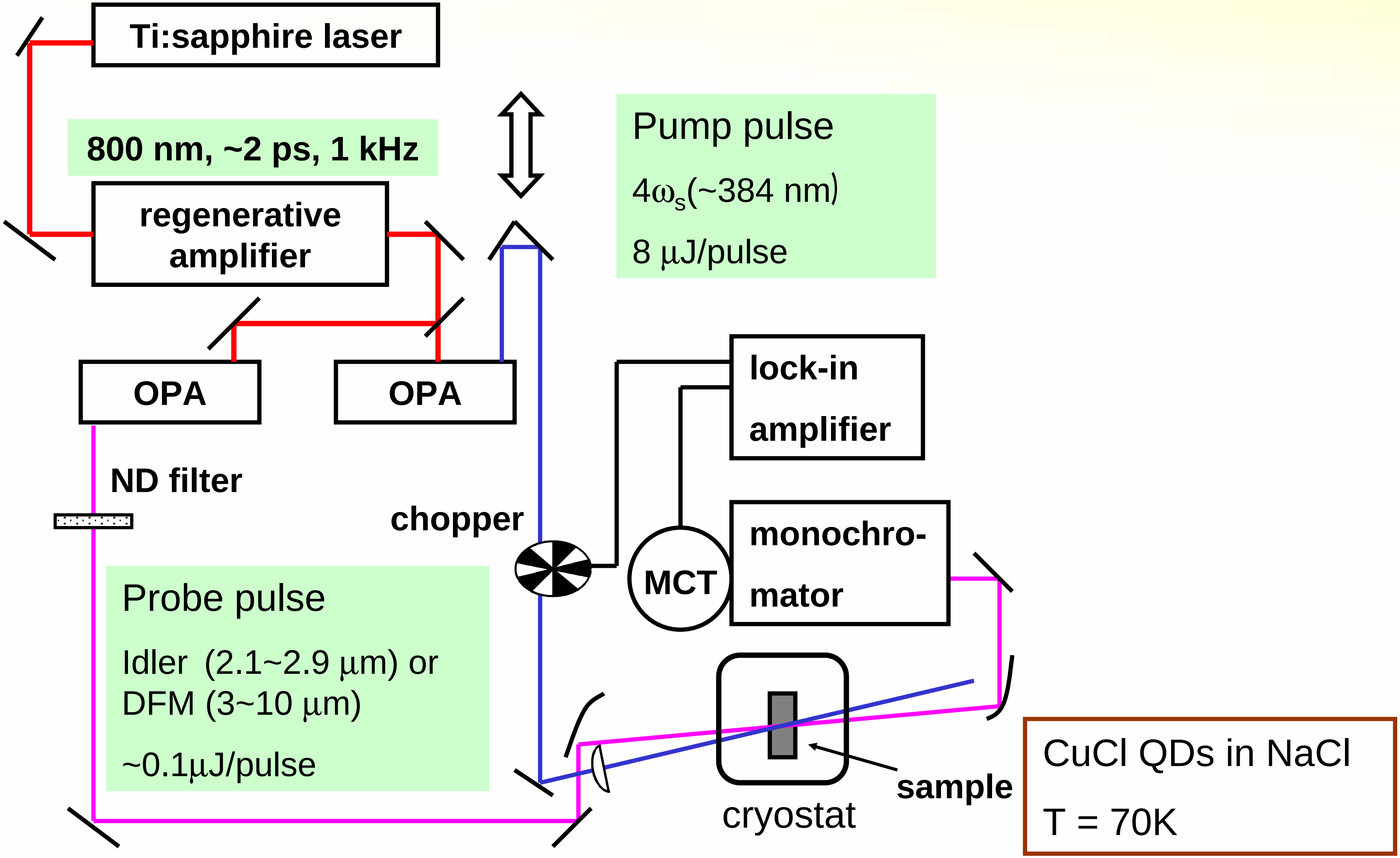
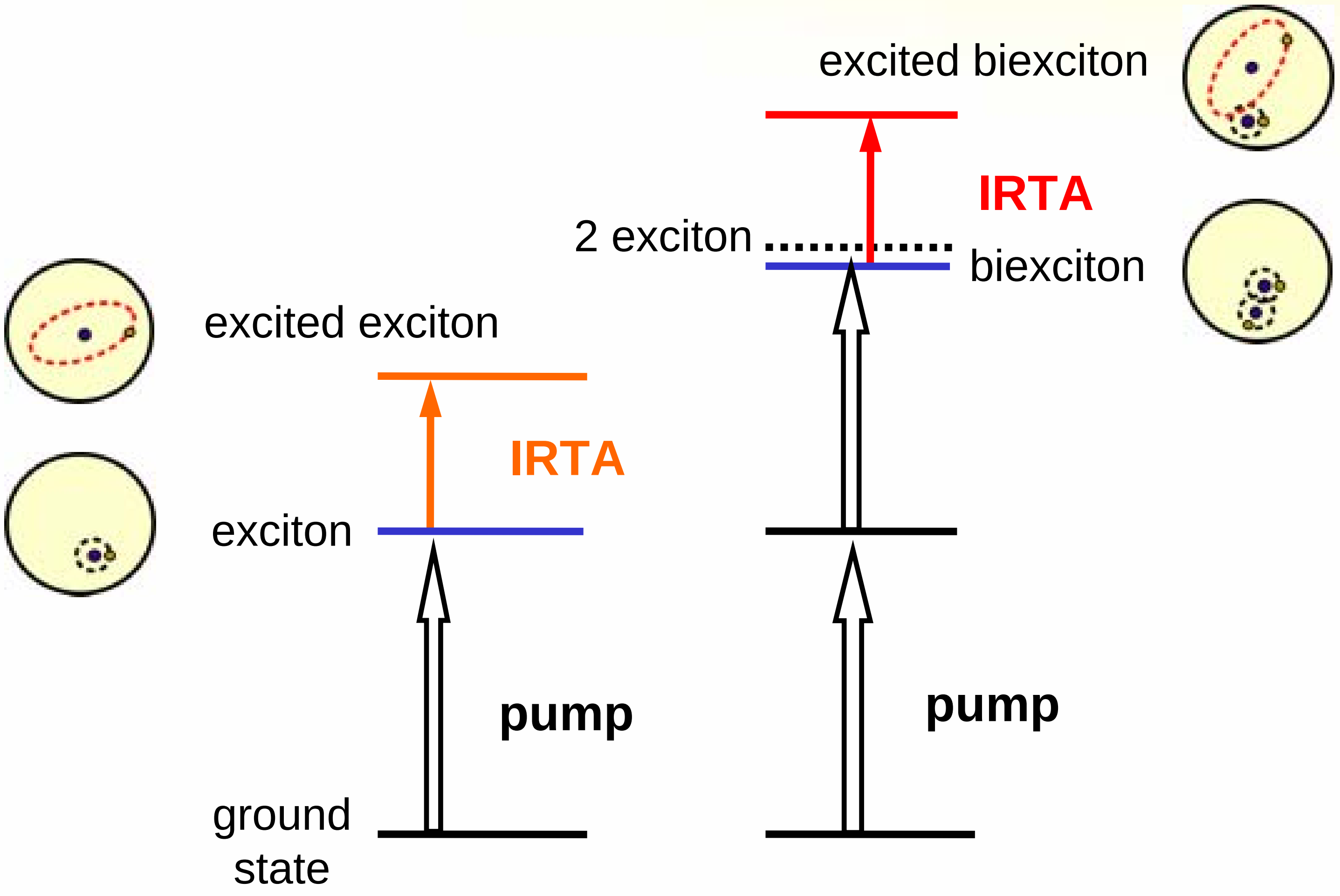
- Insufficient resolution (in energy and time) of the ns experiment
  $\Rightarrow$ ambiguous attribution of the observed transient absorption
- Excited-state absorption of confined biexcitons?
- Modification of the confined exciton states induced by strong IR pulses?

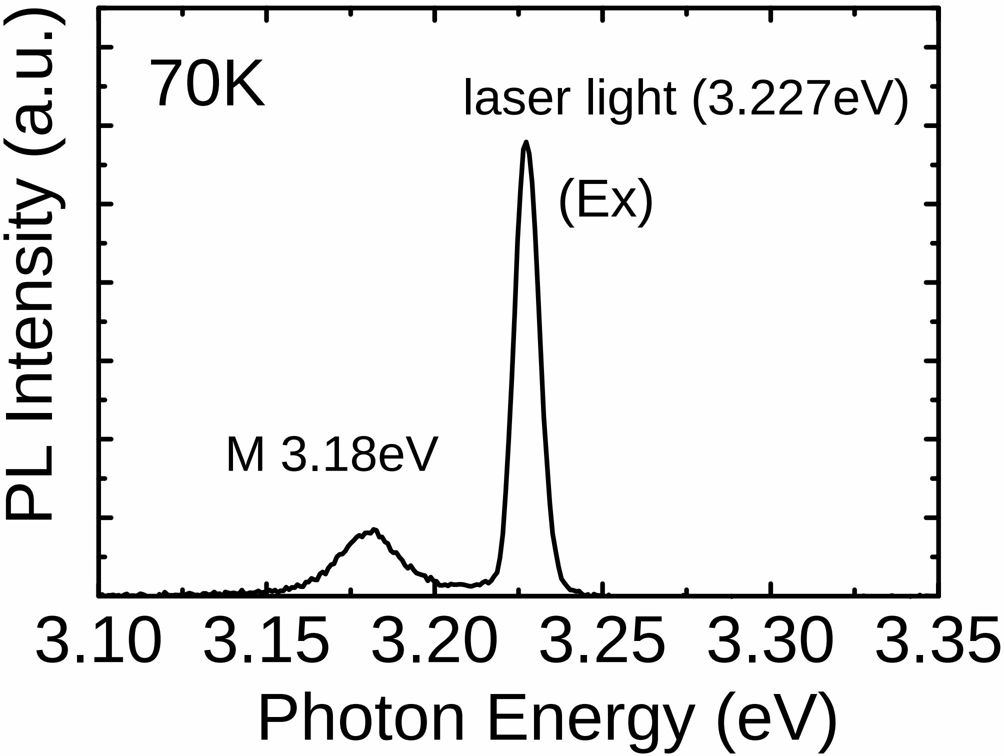
Experimental setup (ps system)



Excited-state absorption of confined biexcitons ?

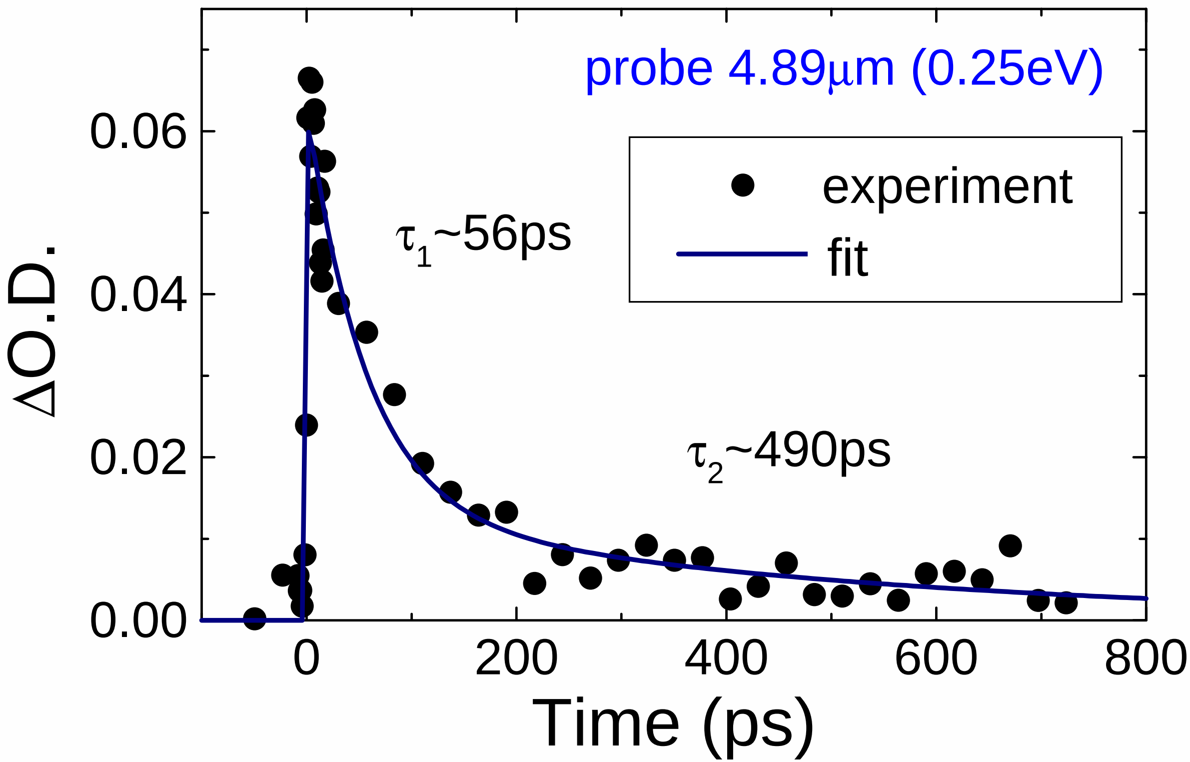


PL spectrum: exhibiting biexciton luminescence



- Excitation : Resonant to the confined excitons with the effective radius ~4.2 nm (3.227eV).
- PL from the biexcitons (M band) at ~3.18eV.

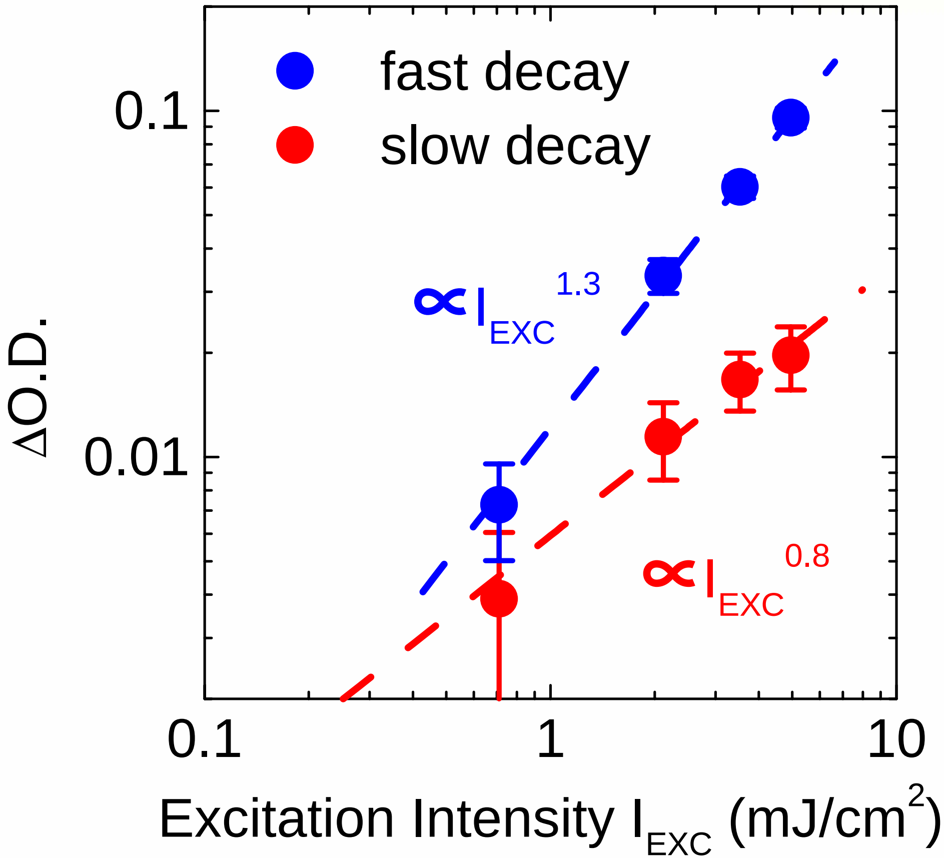
Infrared transient absorption: ps decay profile



Two decay components

- Fast decay
  τ_1 ~56±15ps
  ⇒ Biexciton
- Slow decay
  τ_2 ~490±290ps
  ⇒ Exciton

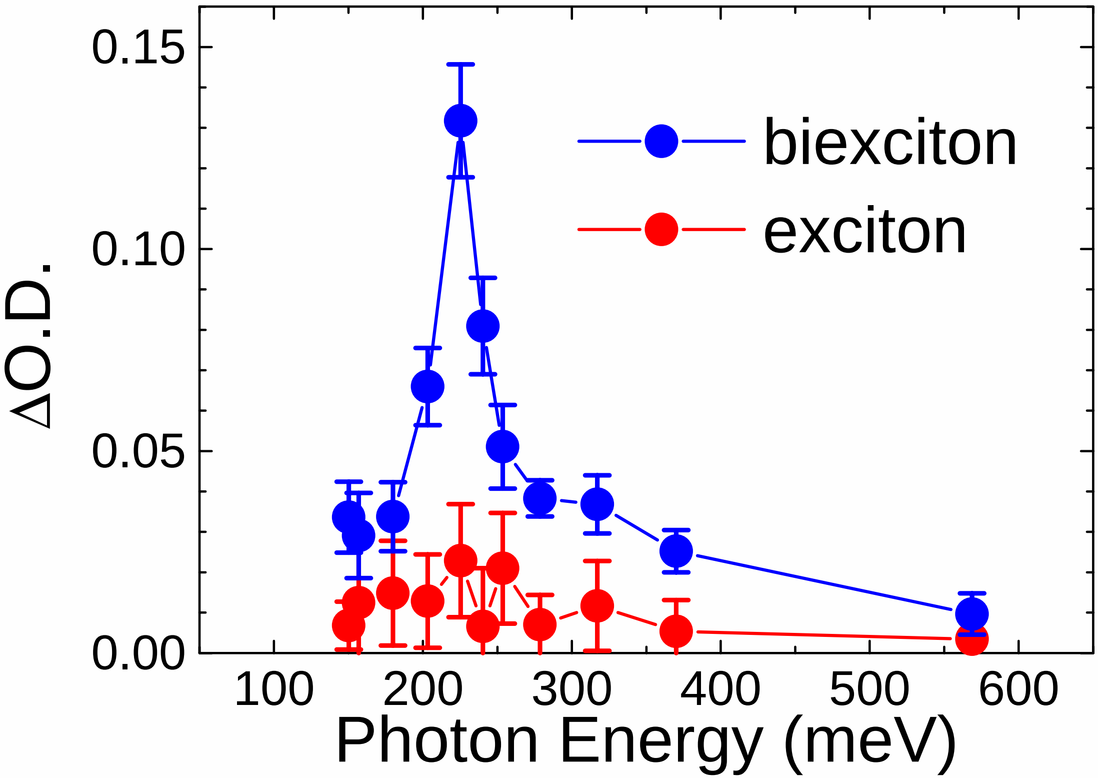
Infrared transient absorption: excitation power dependence



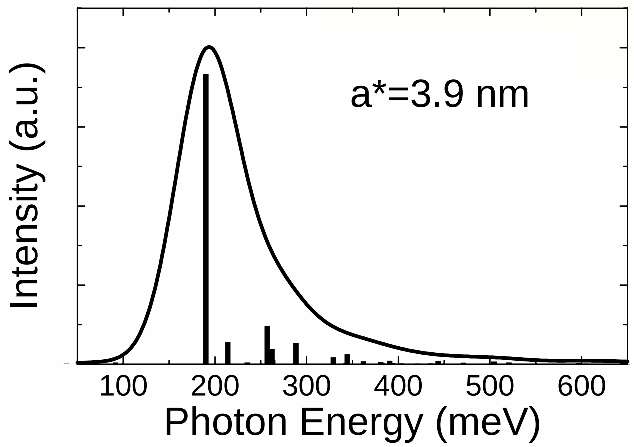
- Fast decay
  superlinear
  ⇒ Biexciton
- Slow decay
  linear~sublinear
  ⇒ Exciton

Infrared transient absorption spectra: exciton and biexciton components

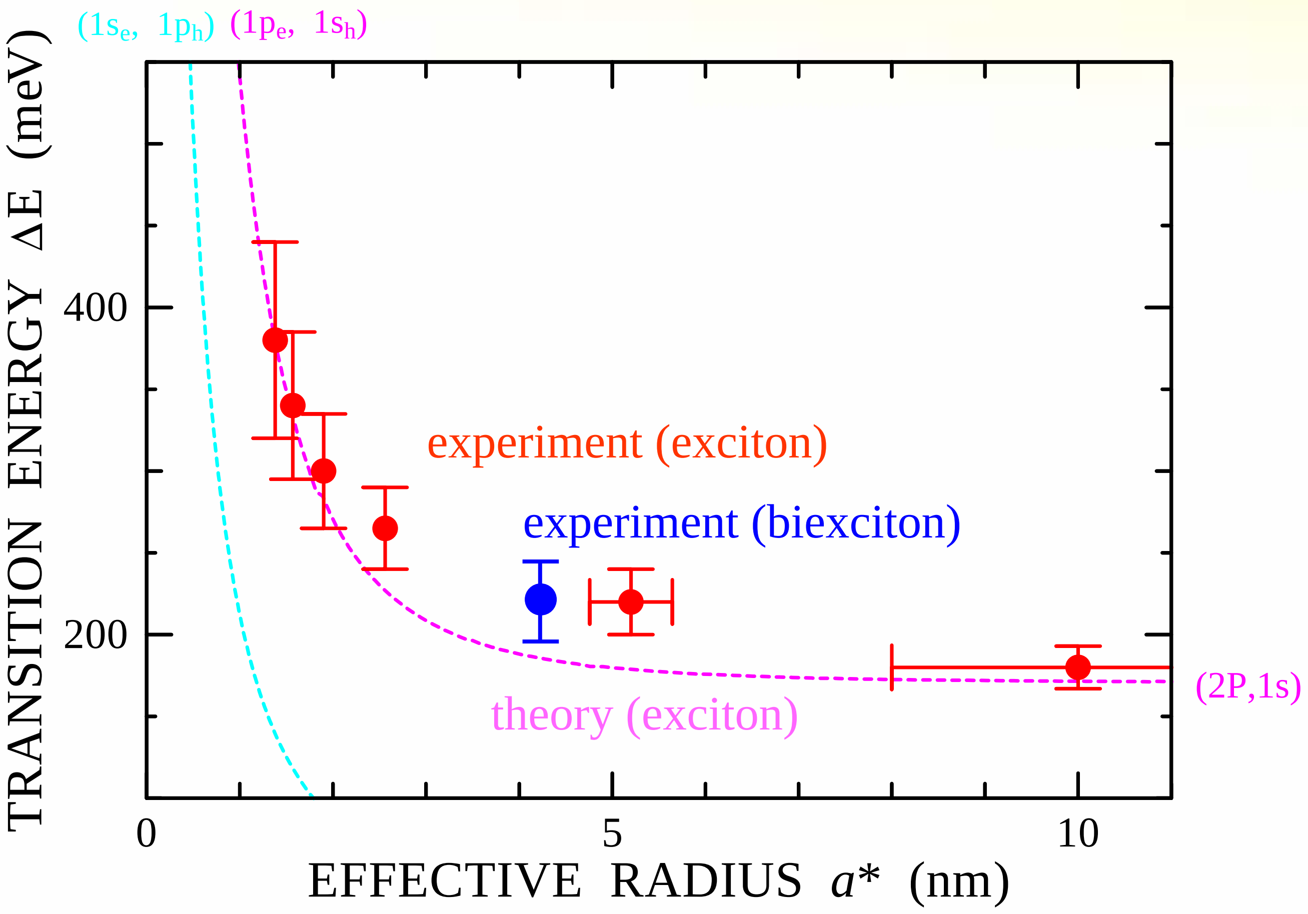
Experiment



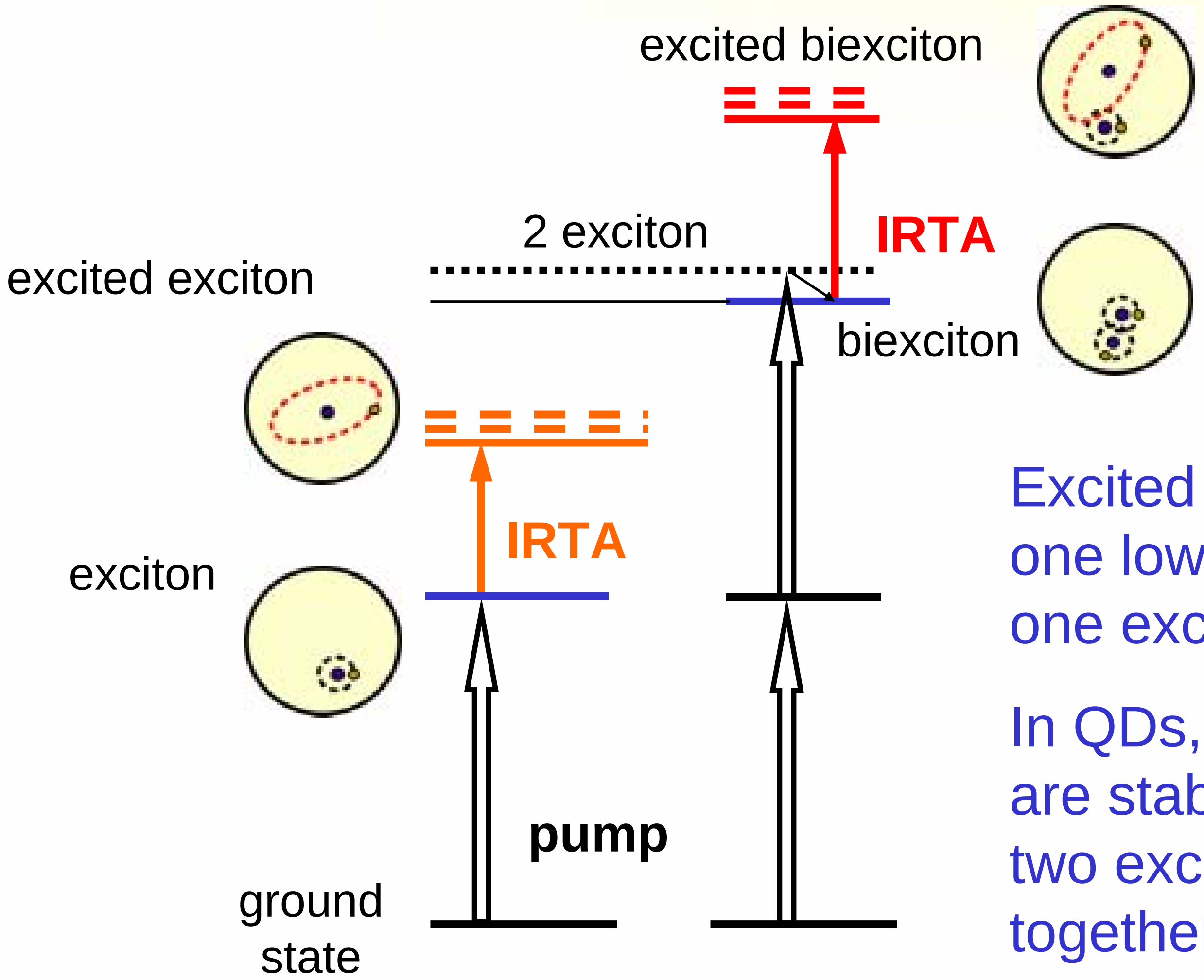
Theory (exciton)



Transition energy: comparison with theory



Excited-state absorption of the confined biexciton



Excited biexcitons:
one lowest exciton and
one excited exciton.

In QDs, excited biexcitons are stable because the two excitons are confined together in a QD.

Summary

We have investigated the infrared transient absorption spectra of CuCl QDs under size-selective excitation.

- Direct observation of the Rydberg 1S-2P transition of the confined exciton.
- The transition energy depends on the dot size, in agreement with the theoretical calculation.
  $\Rightarrow$ Deviation from the "exciton confinement" regime
- Finding of the excited-state absorption of the confined biexciton.

赤外時間分解分光：放射光の利用

- ◎ エネルギー範囲：　遠赤外まで連続
- ○ エネルギー分解能：　分光器，強度に依存
- ○ 時間分解能：ns~ps？
- △ ポンプ･プローブ分光：　可視～紫外の強度に依存

⇩

放射光とレーザー光源との組み合わせ（同期）