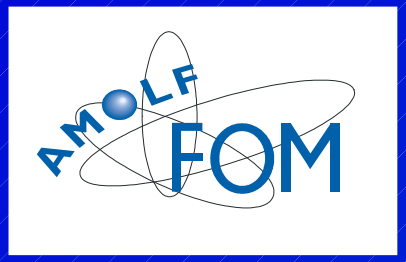
Strong Modification of Spontaneous Emission by Single Plasmonic Nanoantennas



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Outline

- Introduction
- Nanoantenna fabrication
- Darkfield characterization
- Fluorescence lifetime imaging

Plasmon-enhanced Luminescence

Propagating Surface Plasmon Polariton

Metal surface

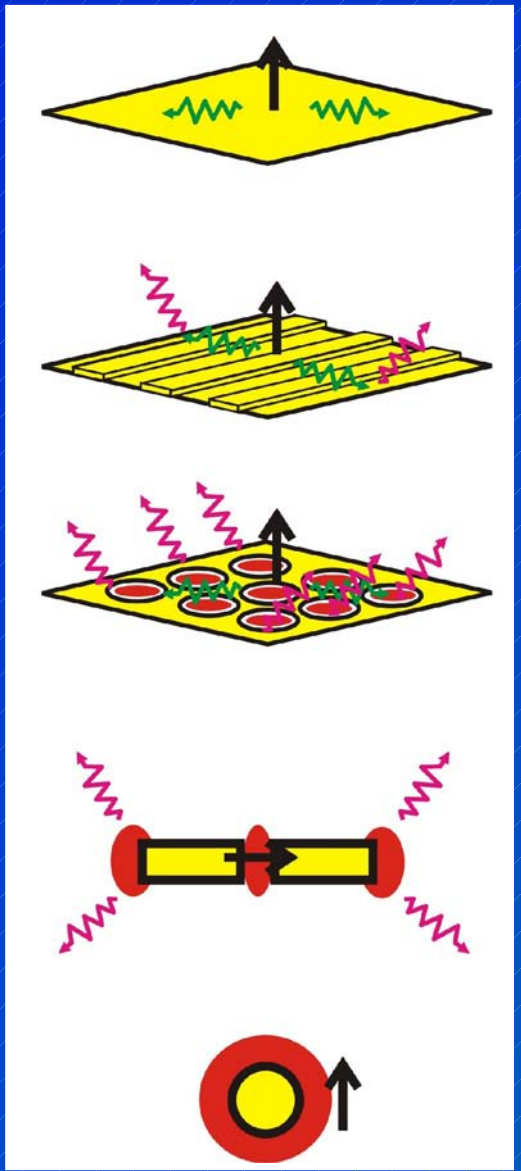
Periodic structures

Rough surface, hole array, plasmonic crystal, …

Nanoantenna, SPP cavity, …

Nanoparticle

Localized Surface Plasmon Resonance



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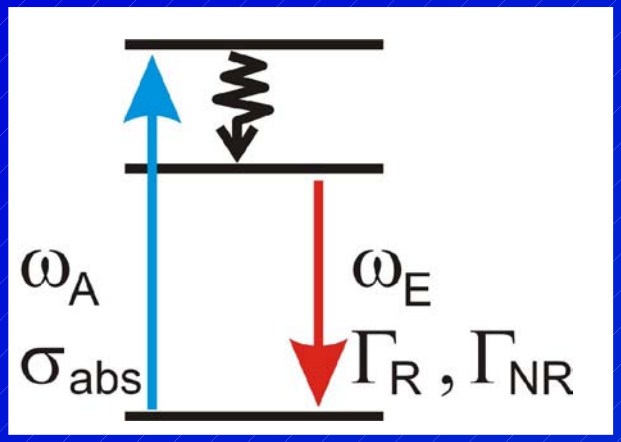
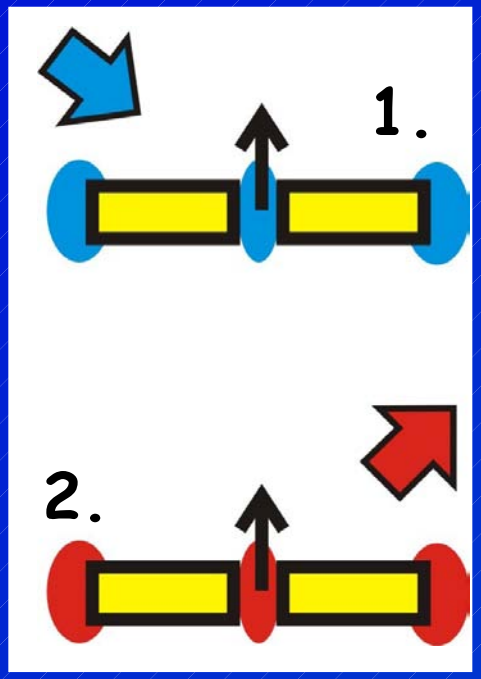
Fluorescence Enhancement



3-level system:

- Pump frequency ω_A
- Emission frequency $\omega_E \neq \omega_A$
- Decay rates γ_R , γ_{NR}



Interaction with surface plasmons:

- Excitation at ω_A

  ⇨ σ_{abs}

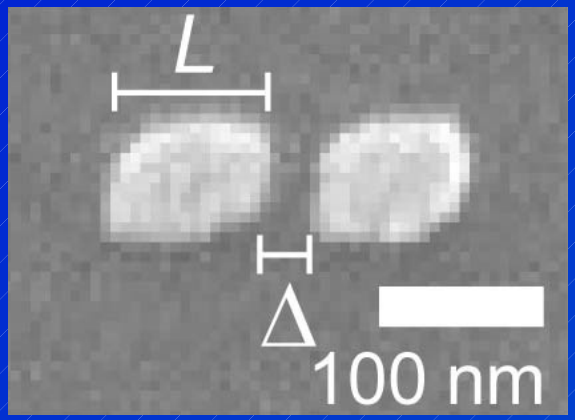
2. Decay rate at ω_E

   γ_R , γ_{NR}

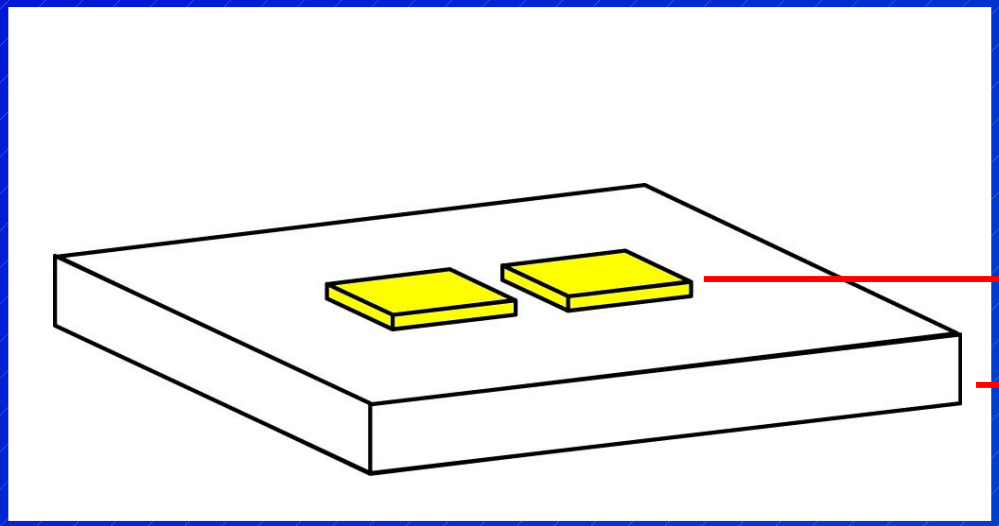
   → Modify Q.E. of high Efficiency emitters

Fabrication: Gold Nanoantennas

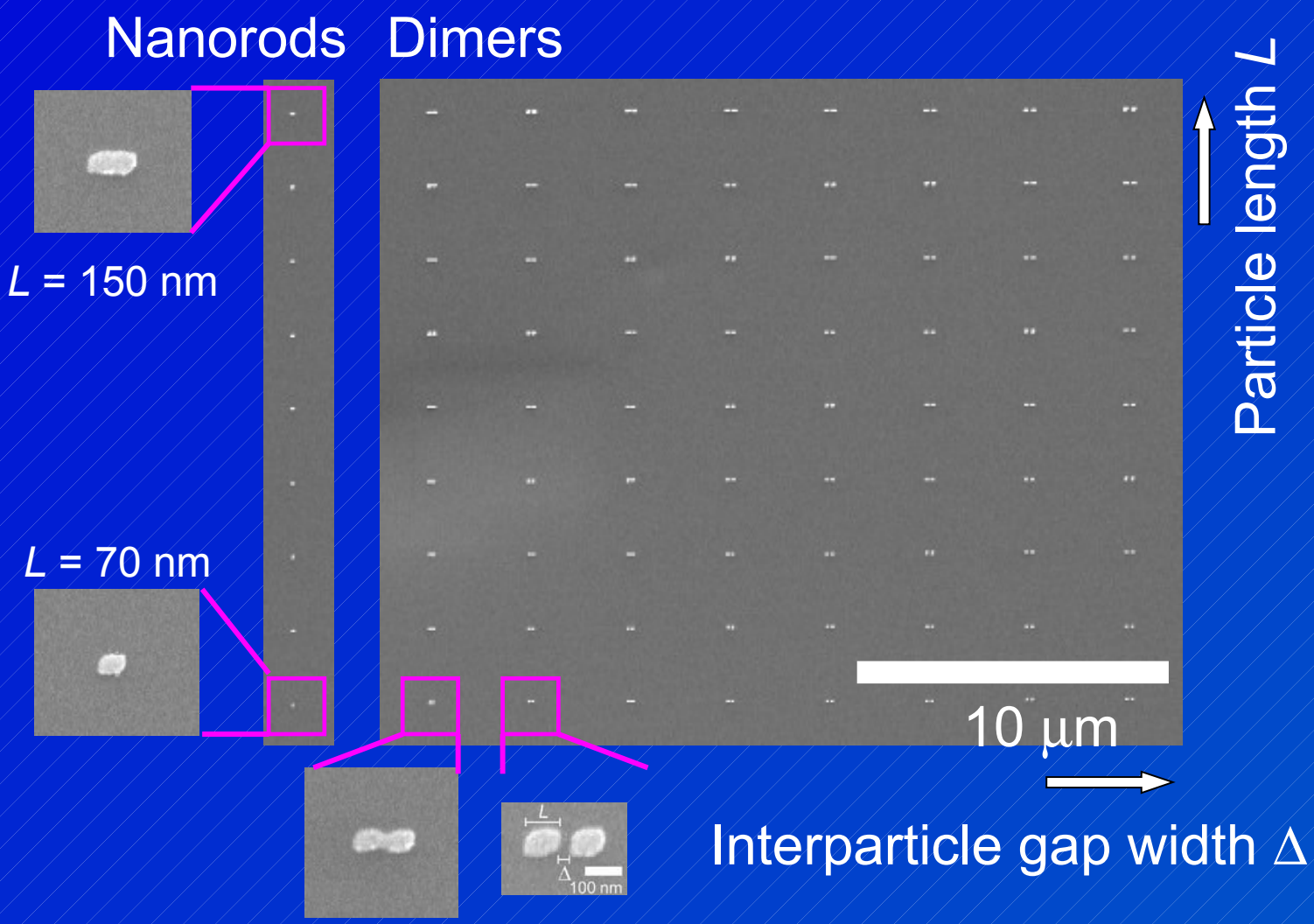
Antenna parameters: Length L, gap Δ



E-beam lithography:



Dimer Nanoantenna Array (SEM)



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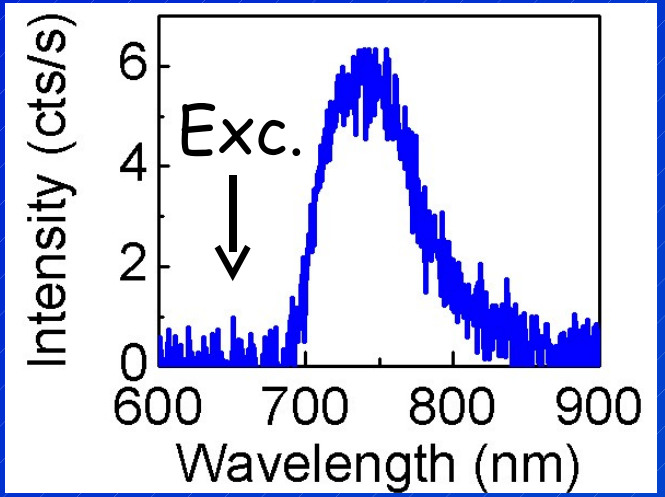
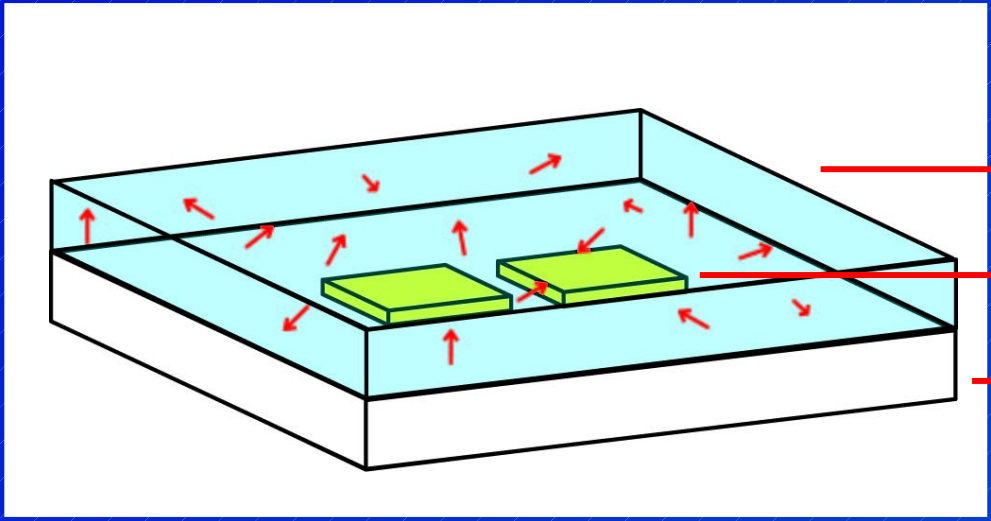
Fabrication: Active Layer

Dye (Atto680) fluorescence spectrum:





Polymer/Dye 10nm

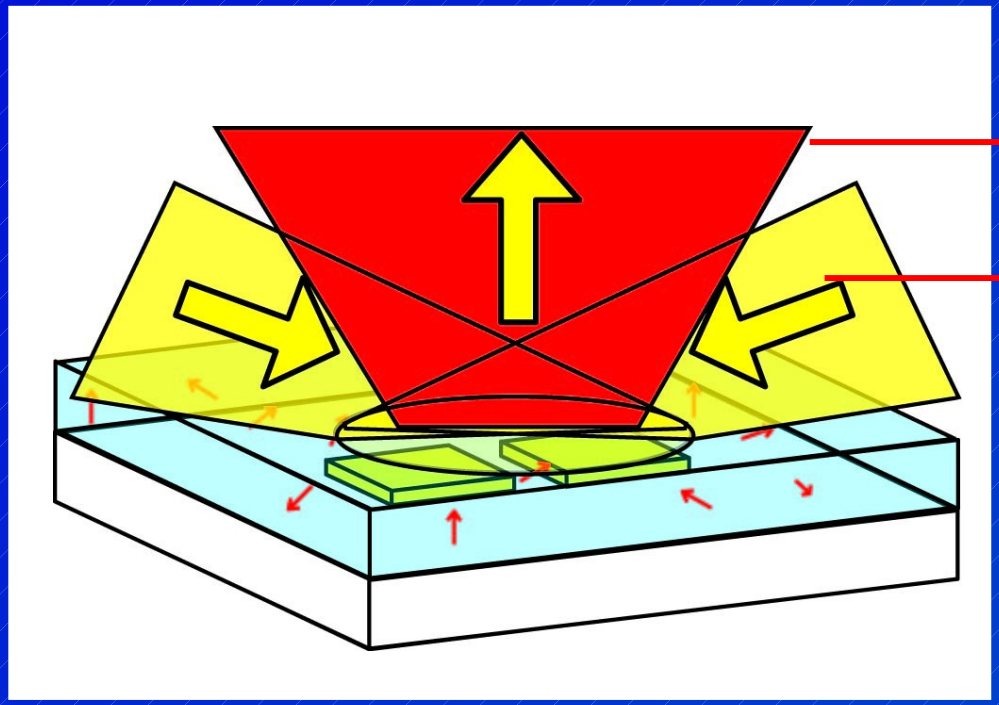
Sol-gel silica 10nm

Gold antenna

Si/SiO_2 Substrate

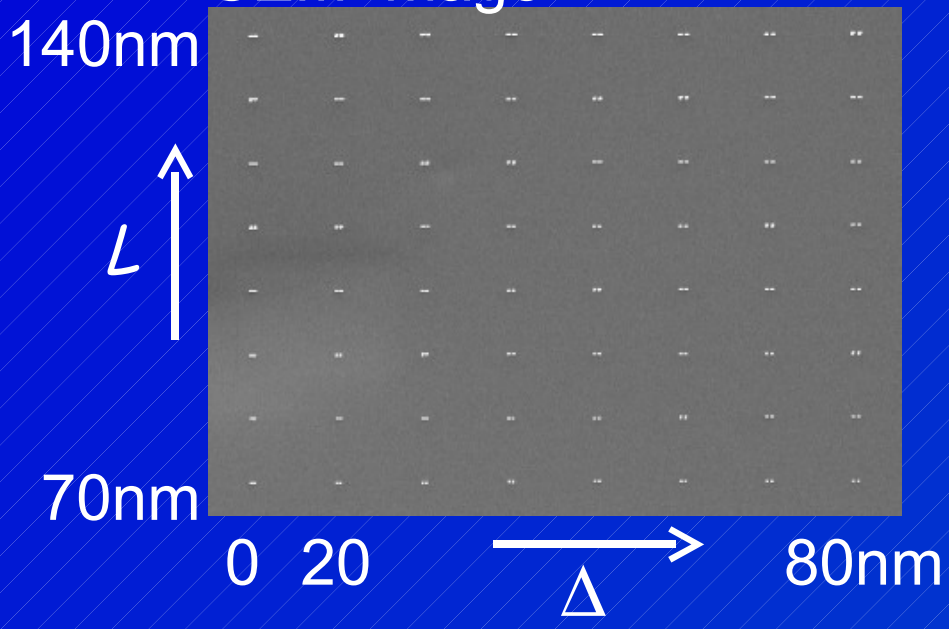
Antenna Resonance Characterization

Optical darkfield microscopy

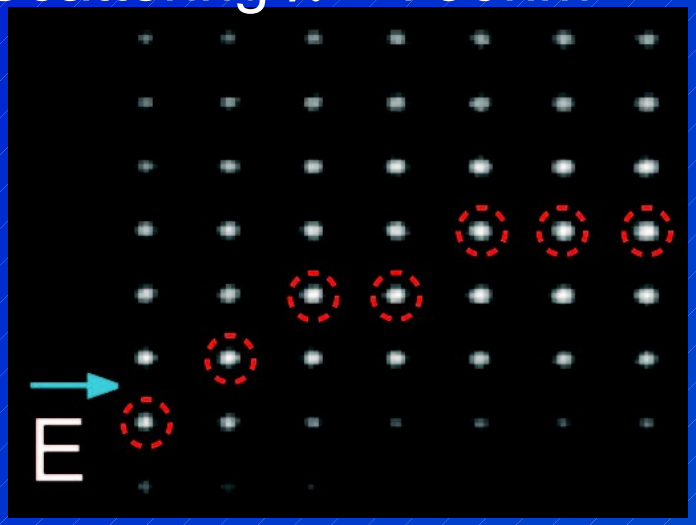


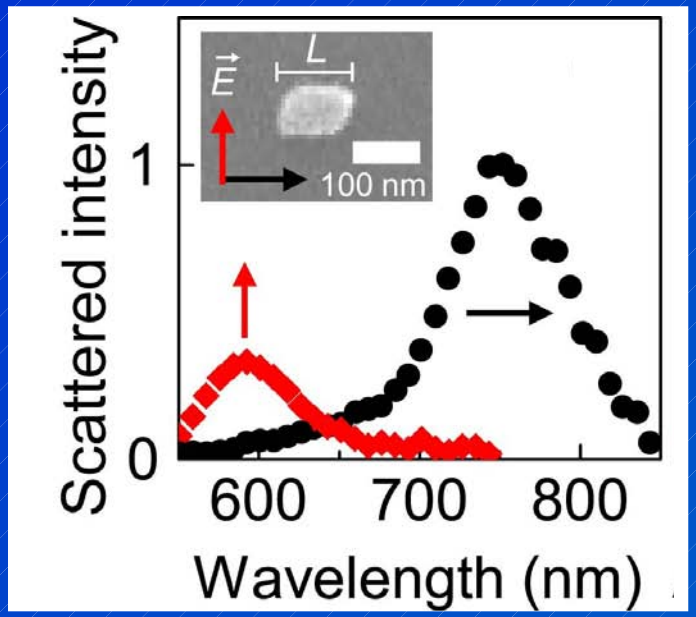
Antenna Resonance Characterization

SEM Image



Scattering λ = 730nm

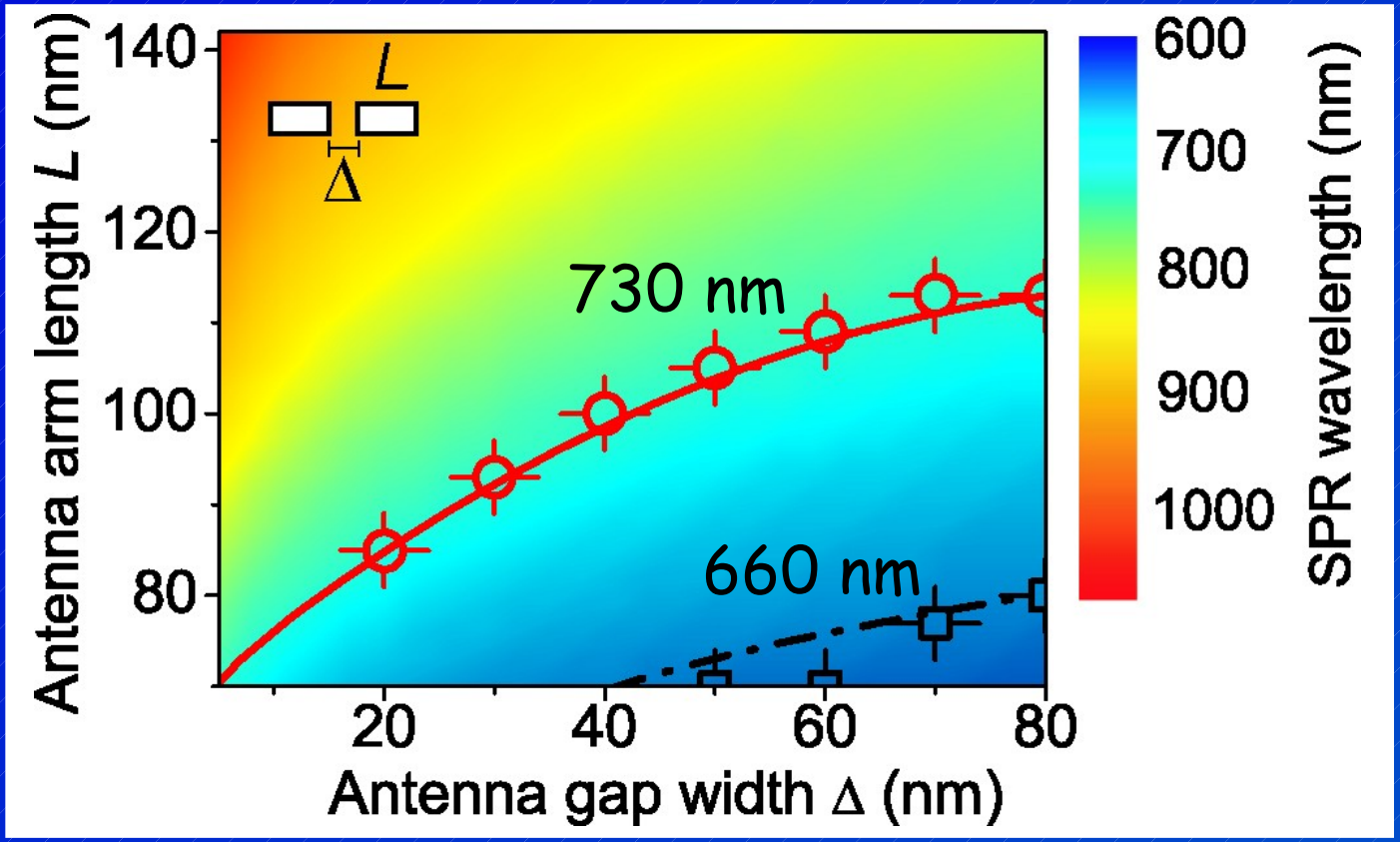




- Longitudinal resonance
  - ➡ Resonant *L* at fixed λ, Δ
  - ➡ Capacitive arm coupling

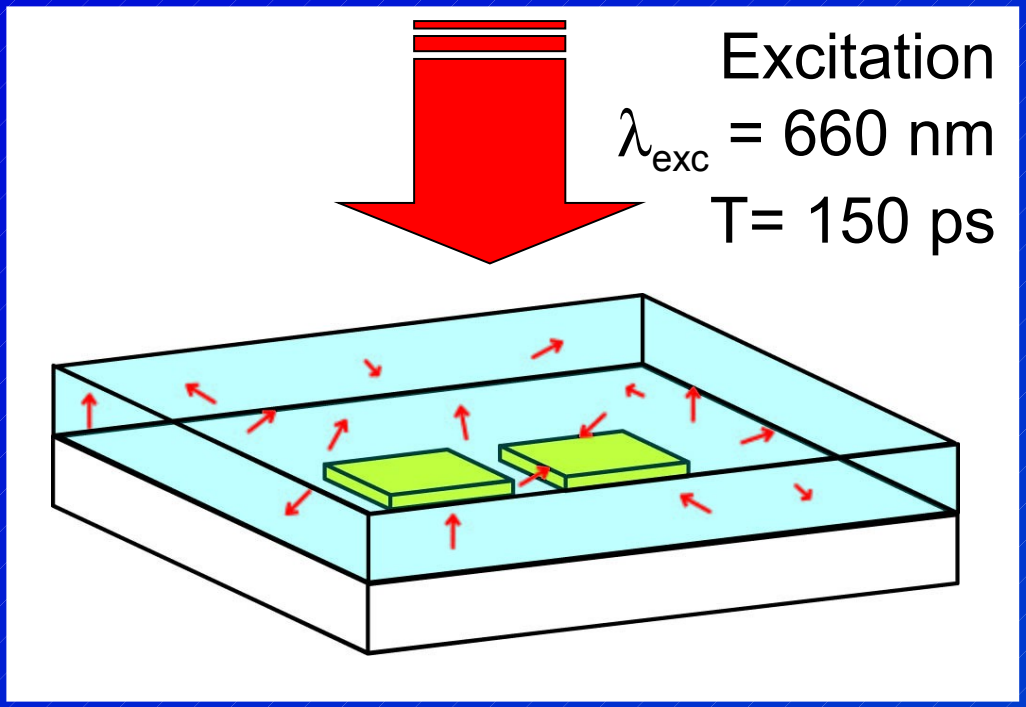
Antenna Resonance Characterization

- Tuning of antenna mode via arm length and antenna gap



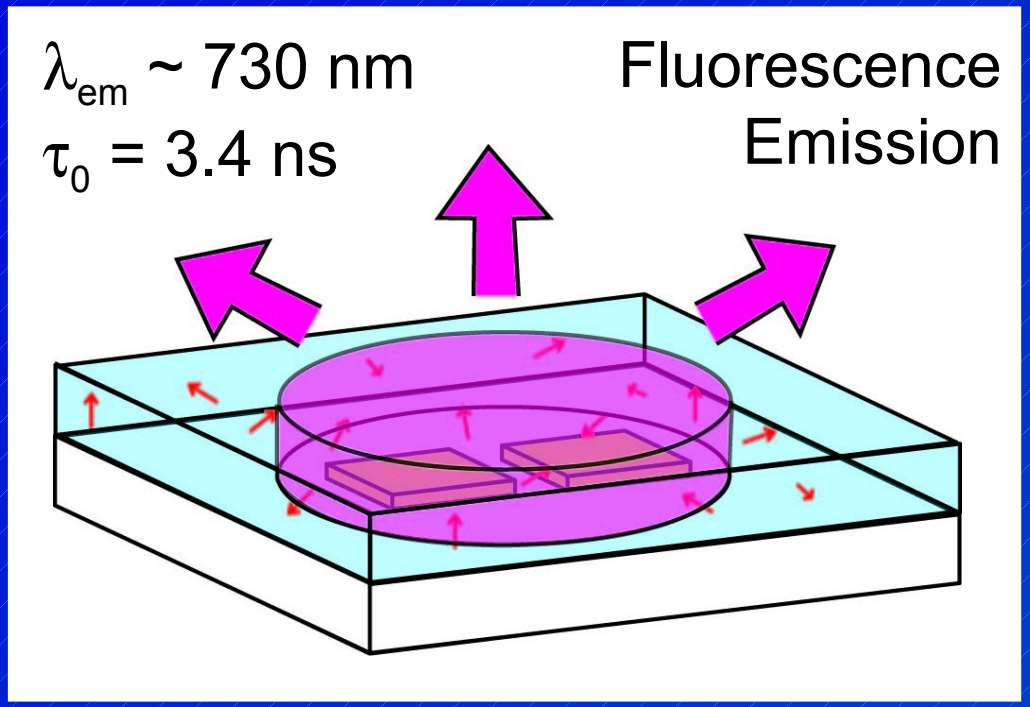
OLM et al., cond-mat/0612689

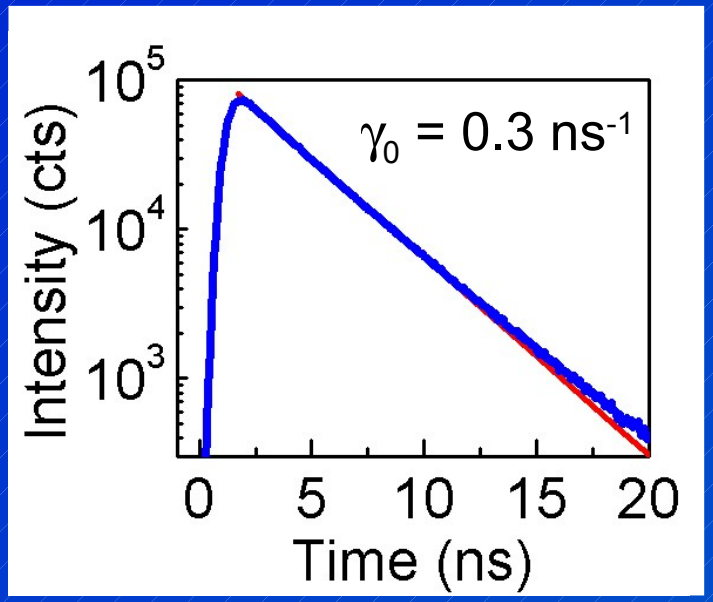
Time-resolved Fluorescence Imaging



Time-resolved Fluorescence Imaging

Fluorescence spectrum:

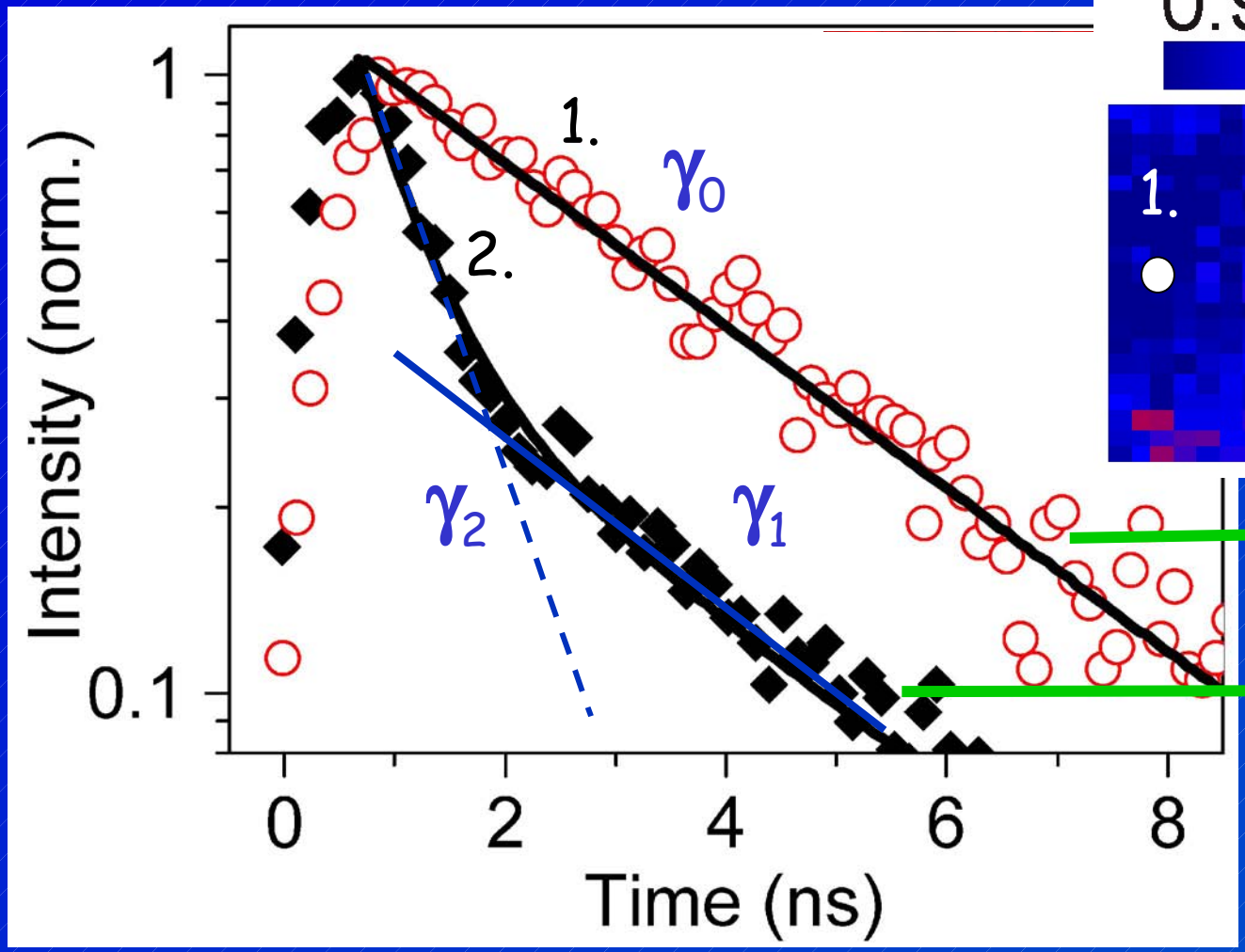


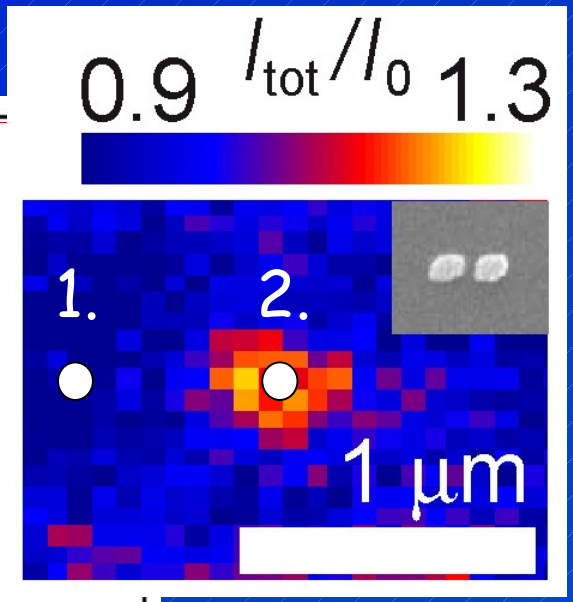


Quantum Efficiency: ~ 0.4

Fluorescence decay time: 3.4 ns

Time-resolved Fluorescence Imaging





Next to antenna

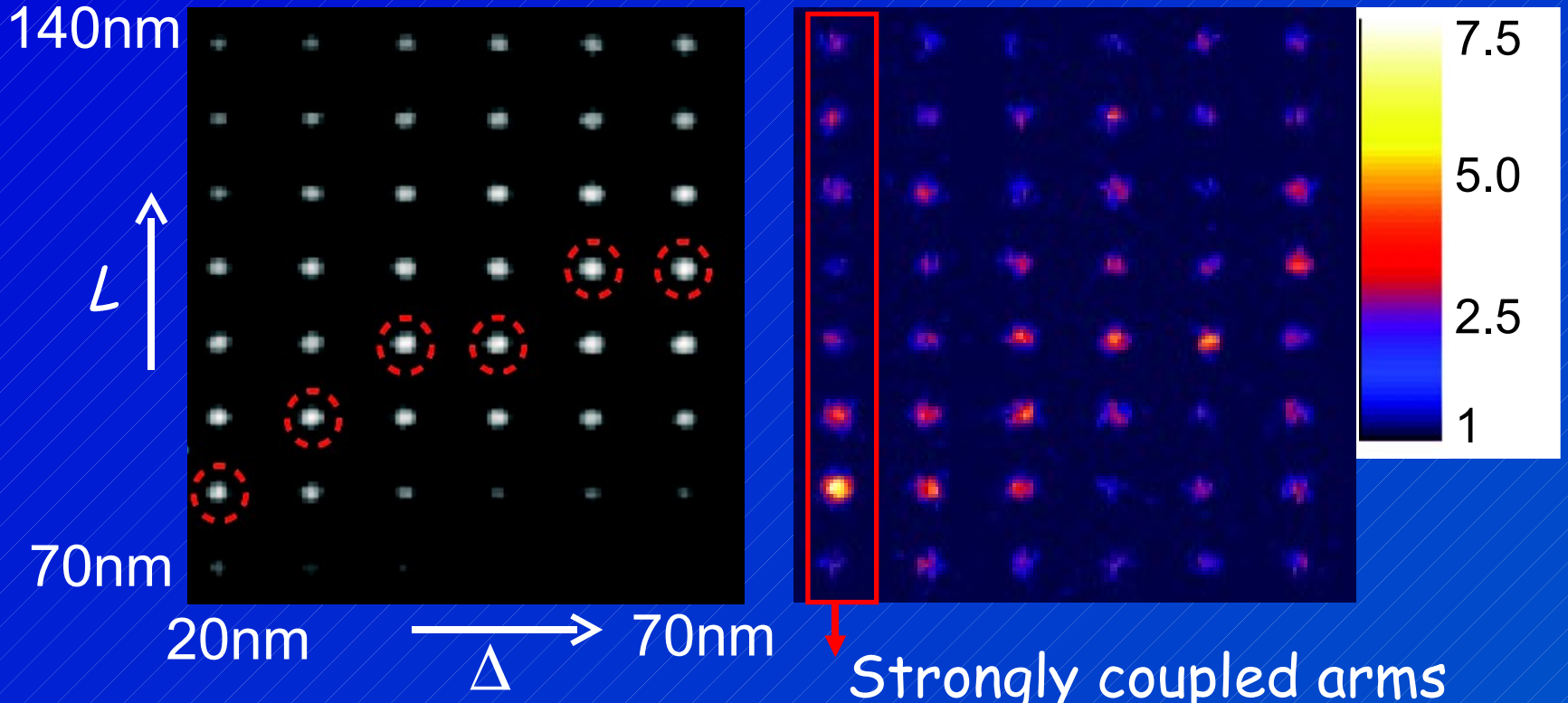
At antenna

- $\gamma_2 = 5\ \gamma_0$
- $\gamma_1 = \gamma_0$

Time-resolved Fluorescence Imaging

Scattering λ_E = 730nm

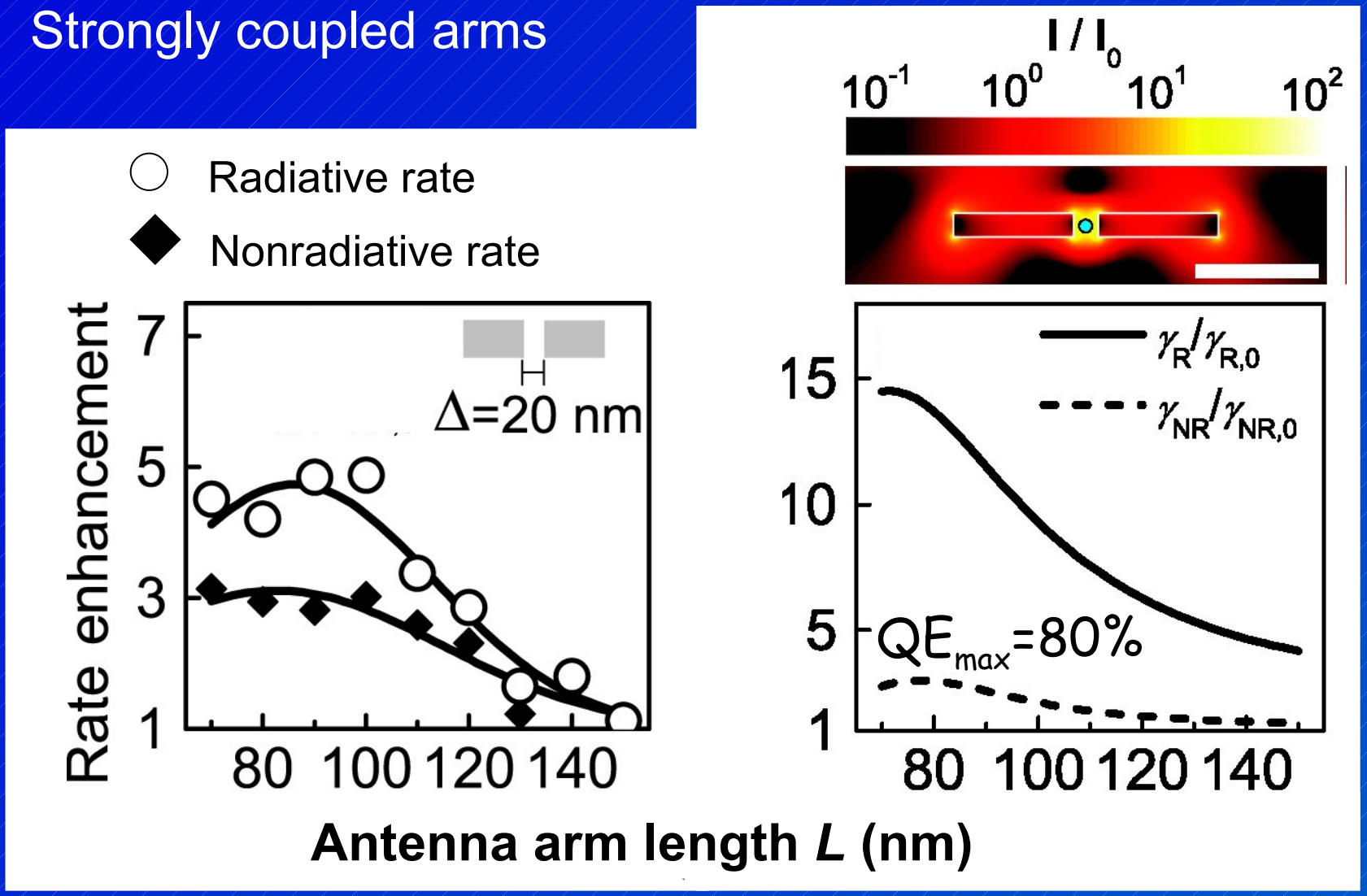
Fast decay rate γ_2



- γ_2 maxima correspond to antenna resonances at λ_E

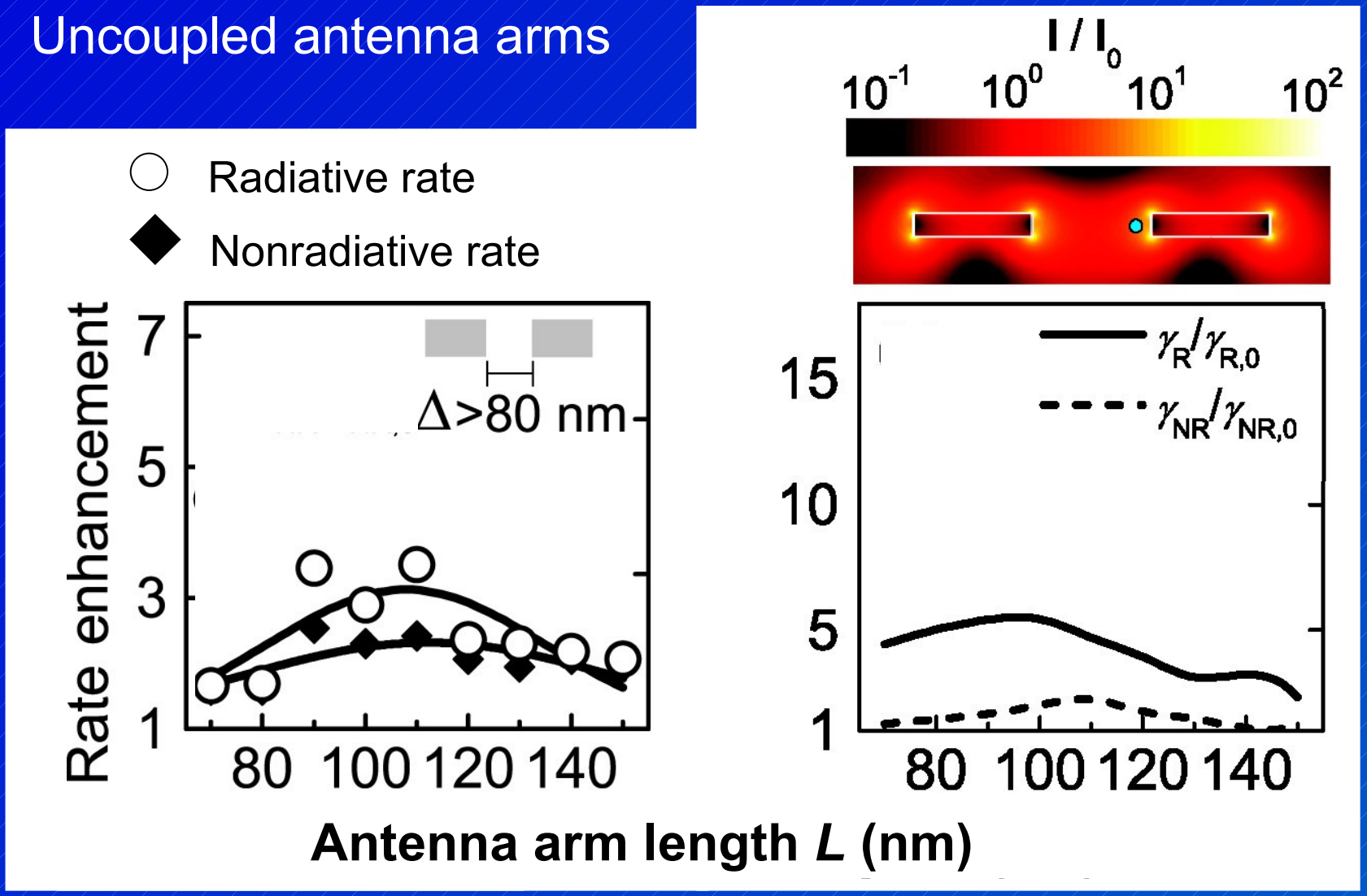
Decay Rates: experiment and calculated

- Strongly coupled arms

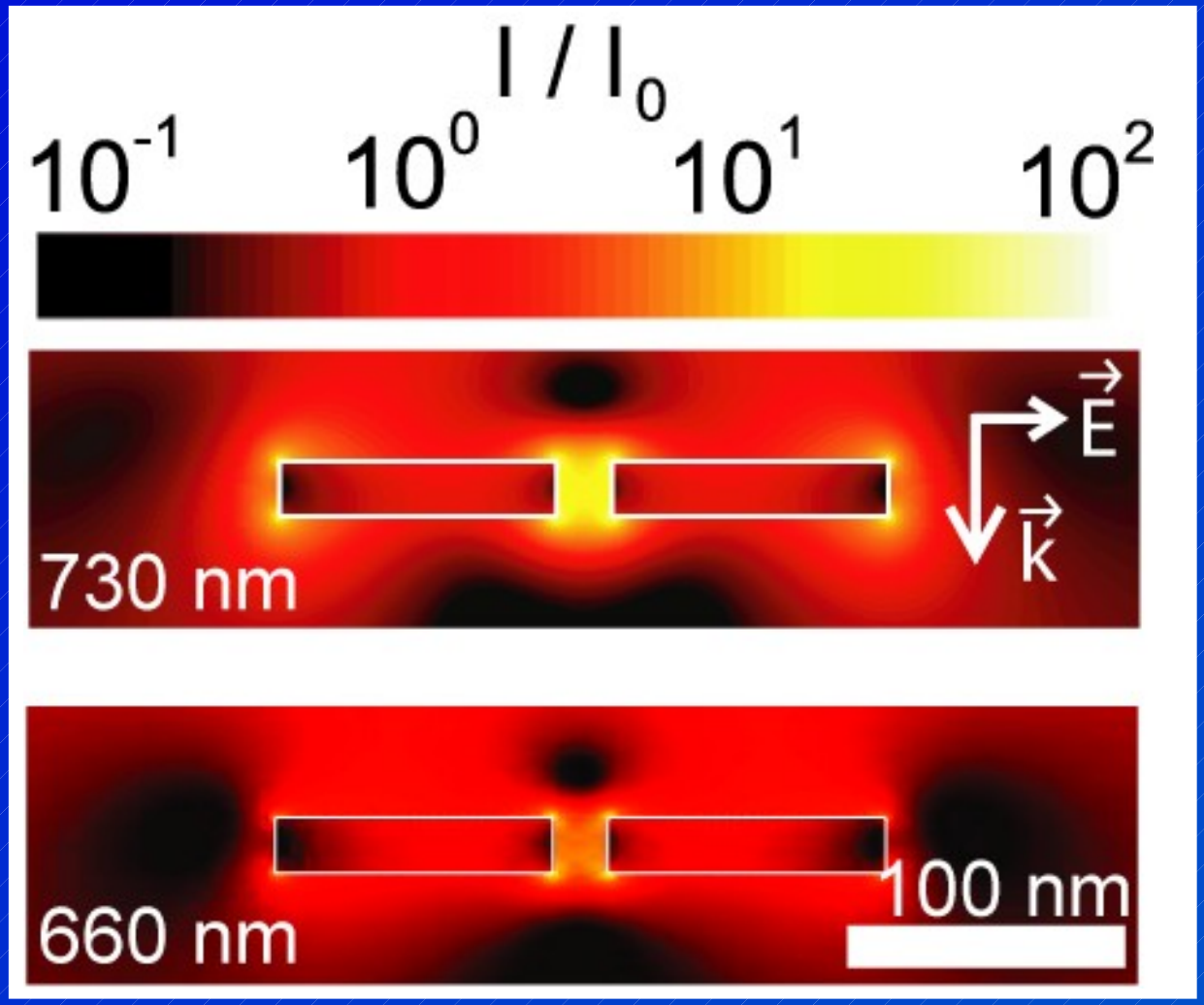


Decay Rates: experiment and calculated

- Uncoupled antenna arms



Excitation and Emission Enhancement



Calculation by V. Giannini

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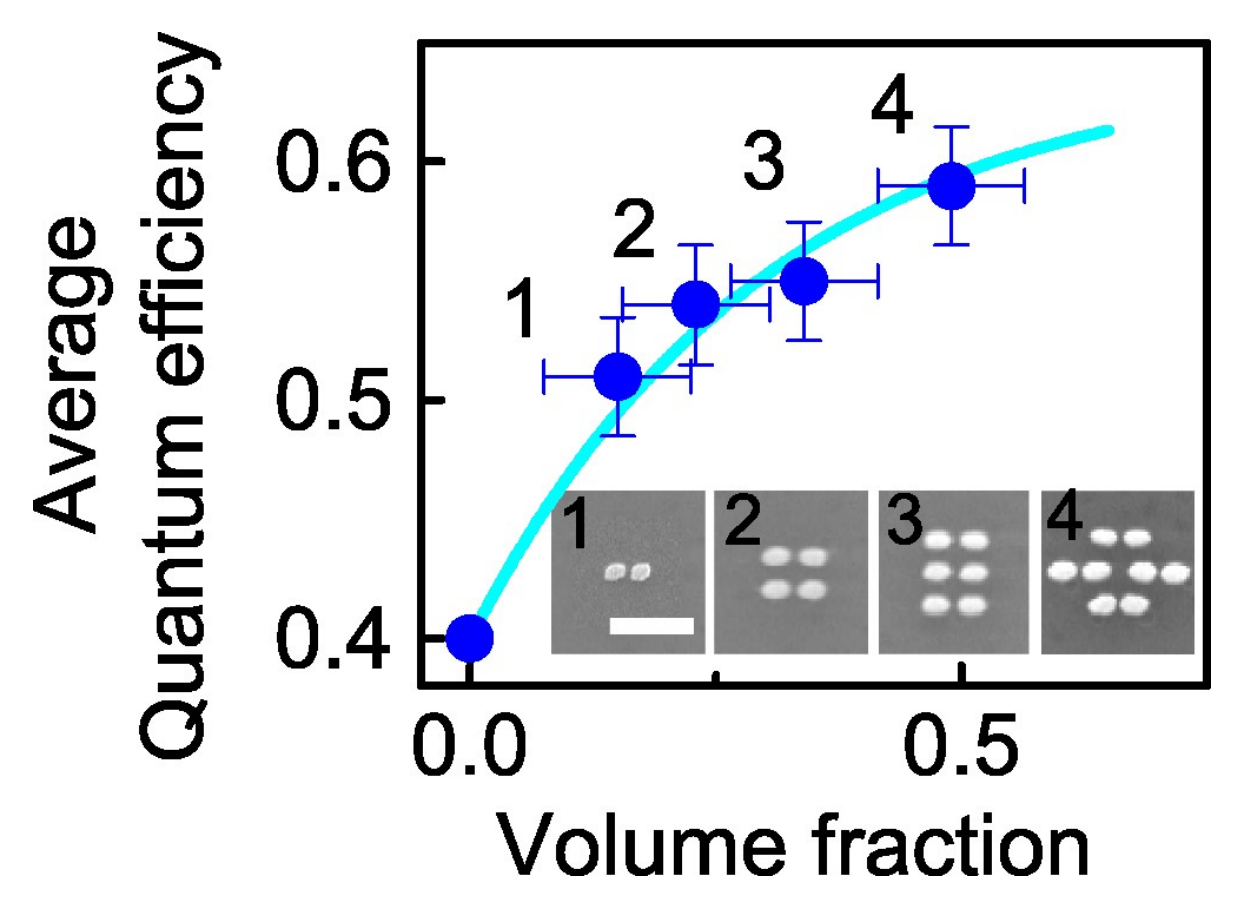
Effect of antenna number: clusters

- Increase antenna-coupled dye fraction to 50%

  ⇒ QE enhancement up to 59%



Conclusions

- 6x enhancement of spontaneous emission by nanoantennas
  - ➡ Resonant dependence on *L*
  - ➡ Effect of arm coupling
- Quantum efficiency:
  - ➡ Up to 150% QE enhancement
  - ➡ Theory predicts a maximum by nonradiative losses