Generation of single optical plasmons in metallic nanowires coupled to quantum dots

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Nature **450**, 402 (2007)

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LANP Seminar
January 22, 2008

Taking control of light

- Control over interactions between single photons and optical emitters
- Why is this important?
  - Photon collection
  - Single-photon transistors
  - High-resolution microscopy
  - Long-range quantum bit coupling

Single photon = single plasmon

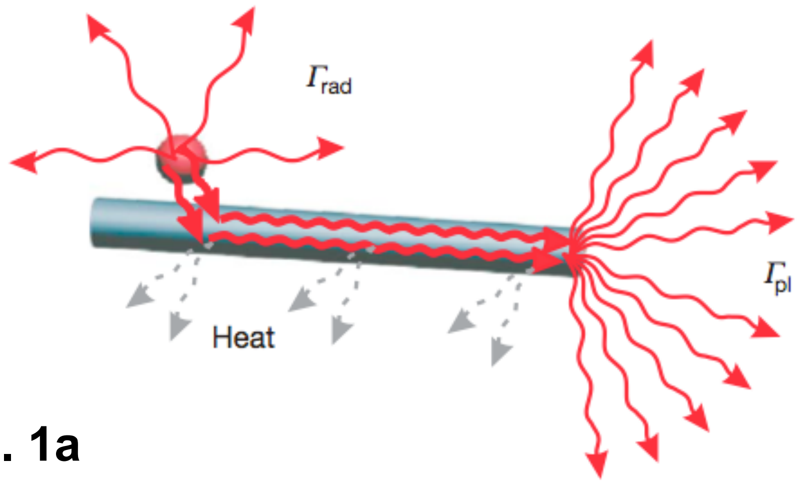
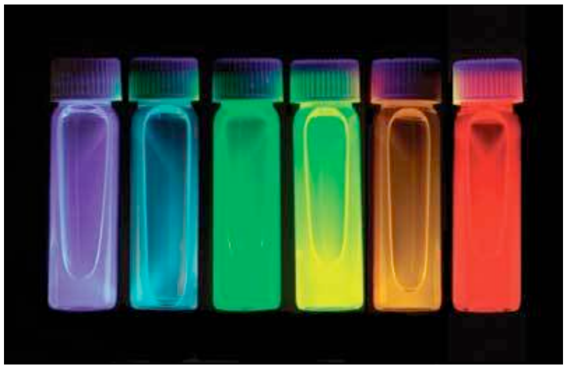


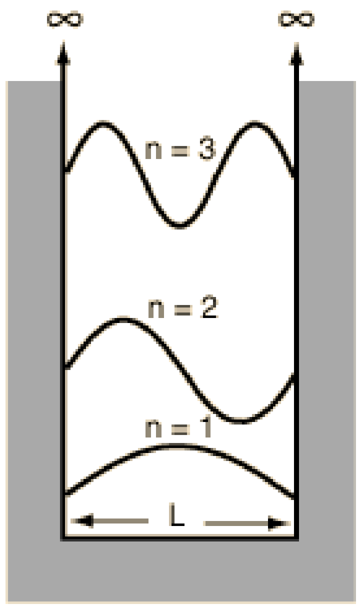
Fig. 1a

- Put a quantum dot (QD) next to a silver nanowire (NW)
- Zap the QD with a laser to emit <u>one photon at a time</u>
- Three ways for photon energy to decay:
  - Emission into free space ($\boldsymbol{\Gamma}_{rad}$)
  - Heat energy (ohmic losses)
  - NW captures radiation via surface plasmons, and energy is released at the end of the NW ($\boldsymbol{\Gamma}_{pl}$)

What is a quantum dot, anyway?

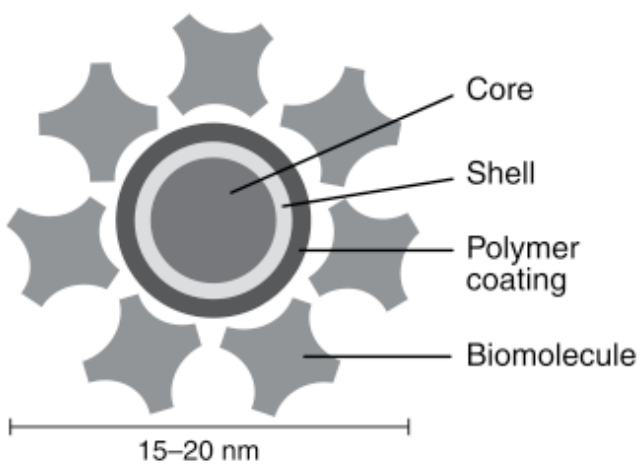
- Semiconductor nanoparticle that absorbs photons to release energy
- Quantum confinement - particle in a box
- Size-tunable to absorb/emit specific wavelengths of light
- Some have core-shell geometries
- Different QDs absorb different spectral ranges



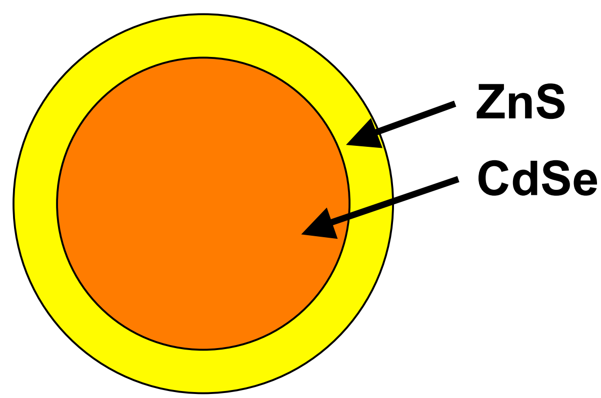


CdSe/ZnS QDs in buffer solution

- CdSe core with ZnS shell
- Coated with a polymer and streptavidin biomolecule to be soluble in a buffer solution ($Na_2B_4O_7$ & cysteine in water)
- Excitation wavelength 532 nm, Emission 655 nm



Invitrogen Q10121MP



Silver Nanowire Fabrication

- Chemically grown bicrystalline Ag NWs
- Solution phase polyol method
  - $AgNO_3$ + Fe-PVP soln at 160°C
- Dried in air on a poly(dimethylsiloxane) (PDMS) stamp and functionalized with 1-hexadecanethiol

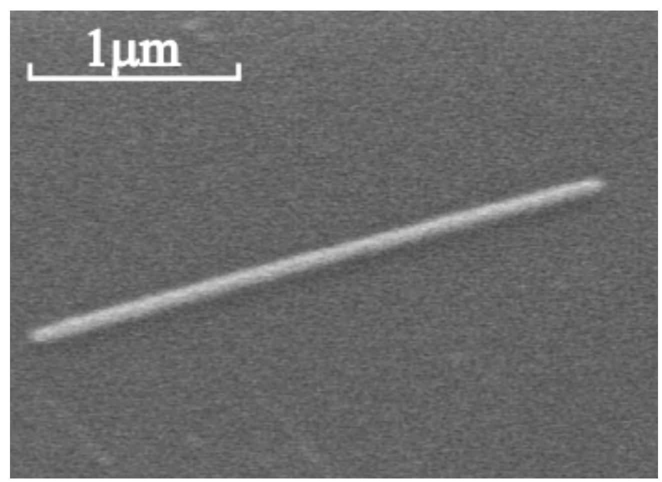
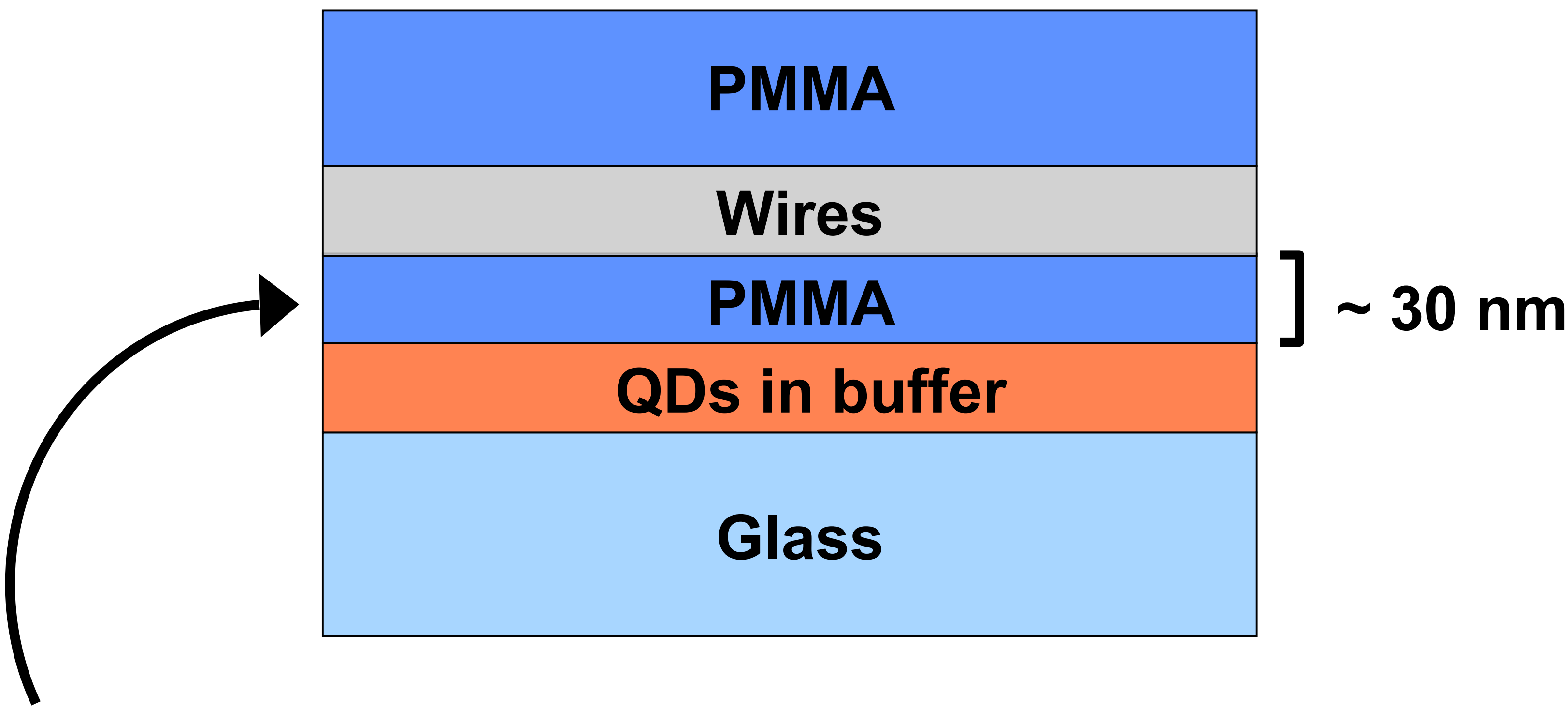


Fig. S1

Substrate Preparation

- Spin-coat QDs and ~ 30 nm polymethyl methacrylate (PMMA) on glass
- Deposit NWs via PDMS stamp
- Spin-coat thick layer of PMMA



QD-NW separation is determined by PMMA thickness

Optical Analysis

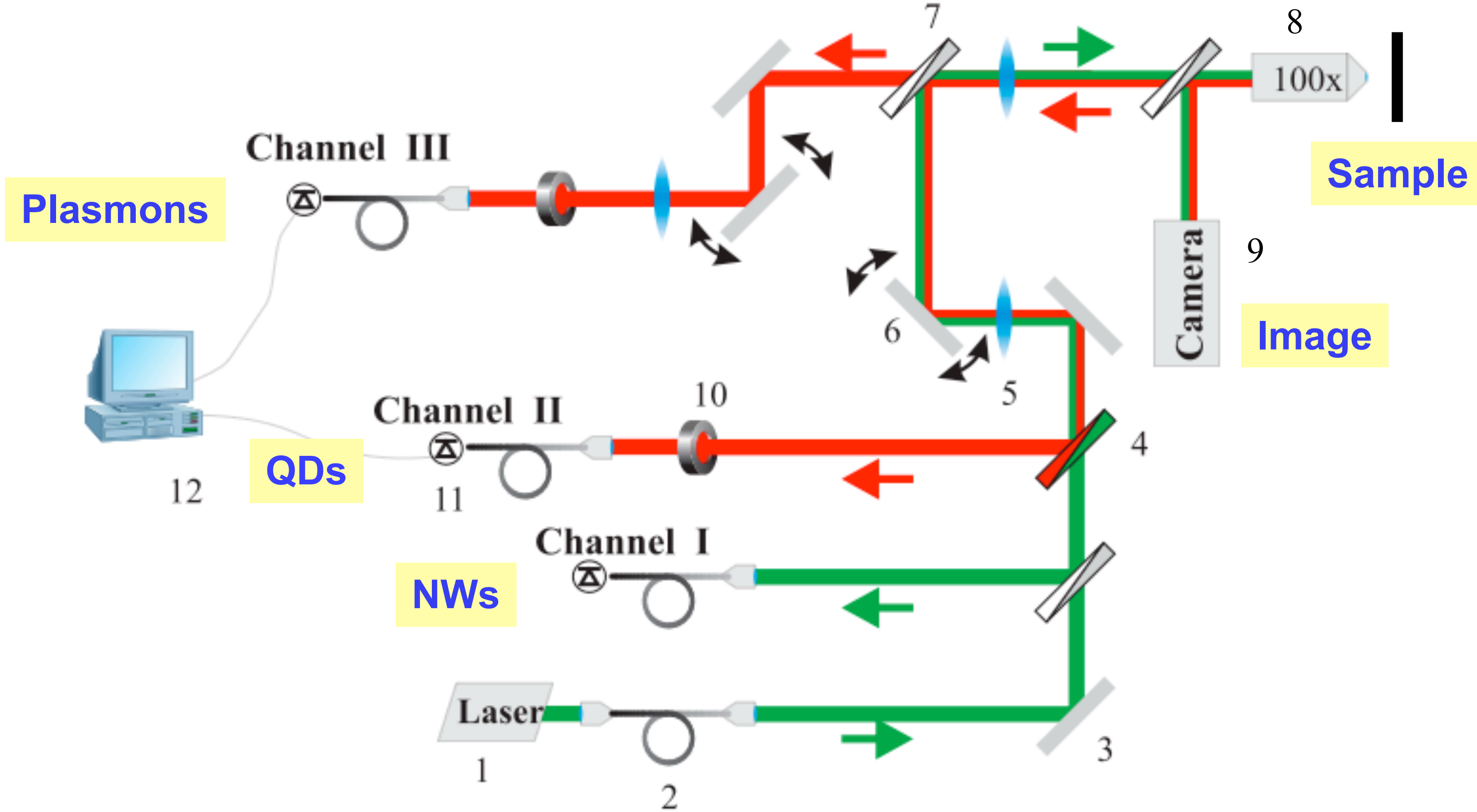


Figure S3 Scheme of experimental setup. 1 – excitation laser, 2 – single mode fiber, 3 – mirror, 4 – dichroic mirror, 5 – lens, 6 – mirror, mounted on galvanometer, 7 – beamsplitter, 8 – Nikon CFI Plan Fluor 100x oil immersion objective NA1.3, 9 – CCD camera Starlight Xpress SXVF-H9, 10 – red filter, 11 –avalanche photodiode, 12 – Computer with installed Becker & Hickl GmbH SPC-630 counter board.

Fig. S3

Nanowire Out-coupling

- CCD camera image
- Laser (large spot) directly excites surface plasmons on the NW
- Plasmons travel to and scatter from the NW end (small spot)
- 100 nm NWs exhibited 80% out-coupling

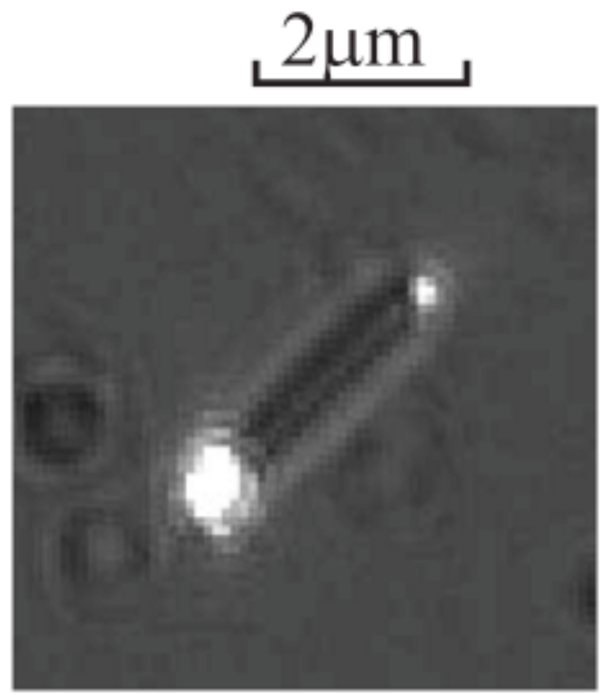
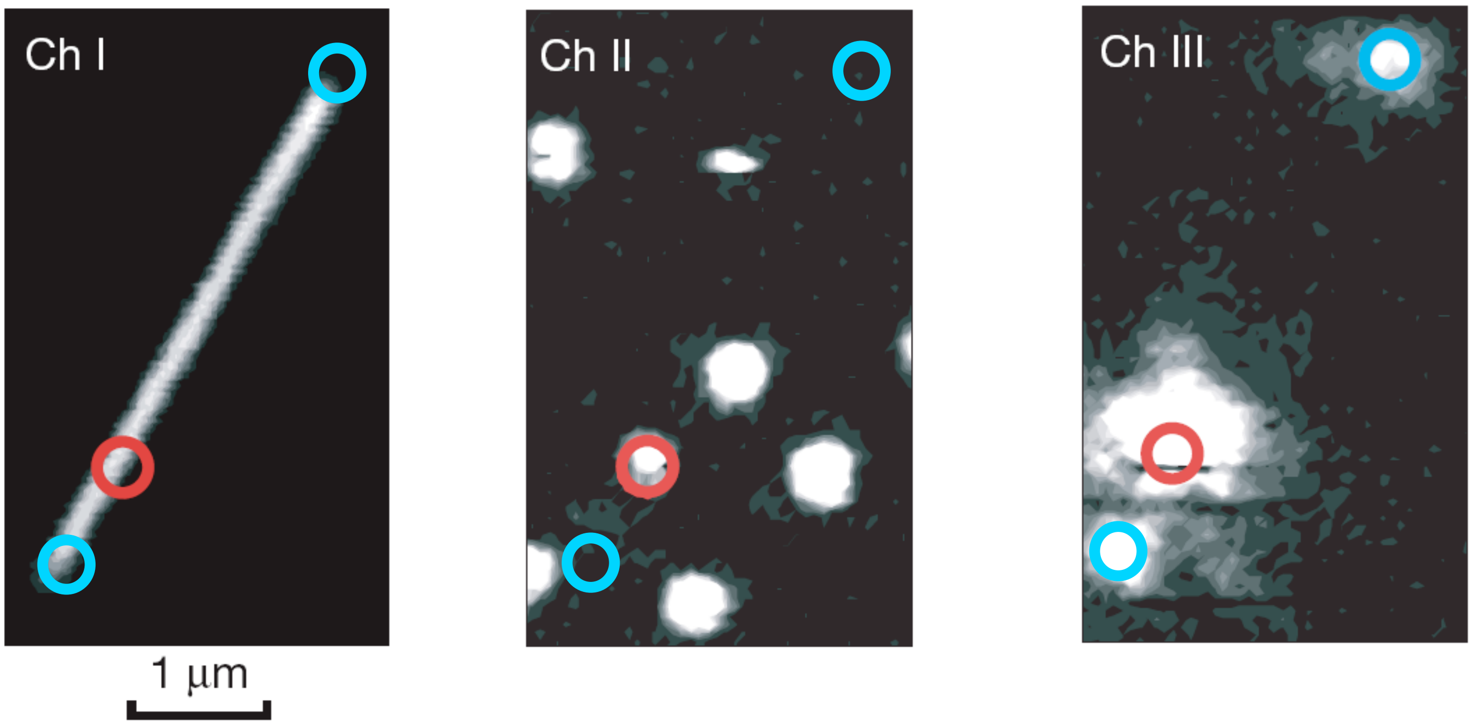


Fig. S4

QD Radiative Coupling



NW

QDs

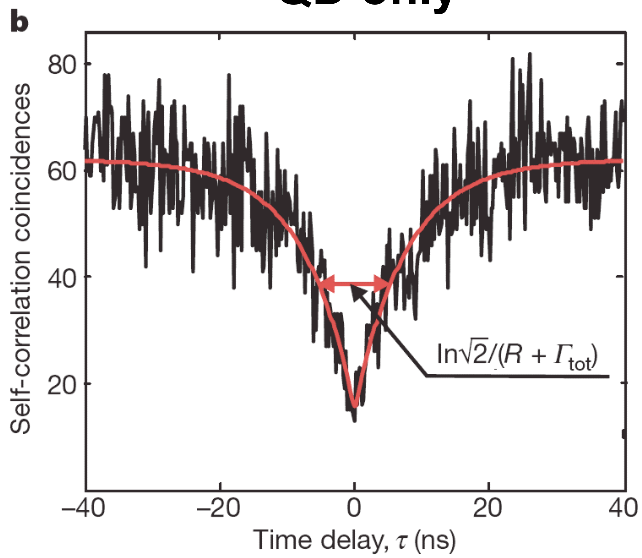
Scattered surface plasmons (and QD fluorescence)

Fig. 2c

One photon at a time

- Two detectors measured time delay (τ) between photon coincidence measurements
- Zero coincidences at τ = 0 confirmed that
  - QD is a single photon source
  - NW emission results from single, quantized surface plasmons
- Offset from zero is due to stray light, resolution limit, etc.

QD only



QD & NW

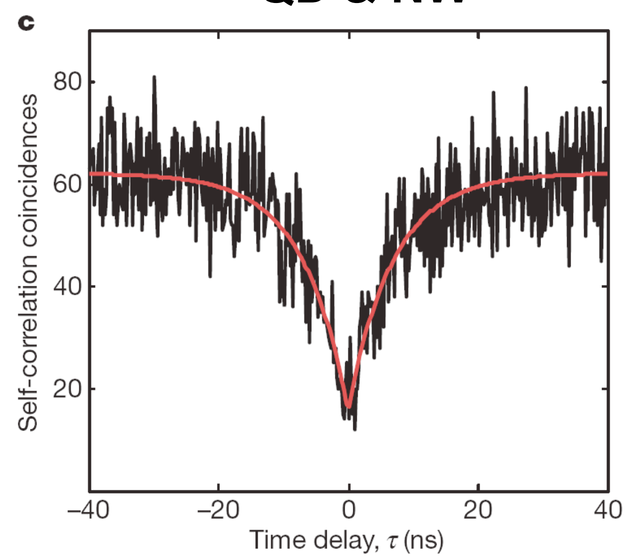
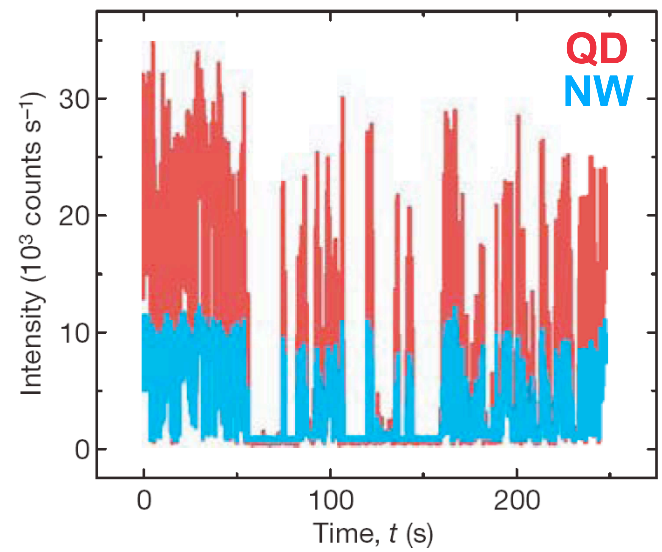


Fig. 3b, 3c

Photon Tracking

- High correlation between:
  - Time trace of fluorescence counts
  - Fluorescence wavelength
- NW fluorescence is due to QD photon emission
- Fluorescence spectrum is not affected by the NW

Fig. 3a



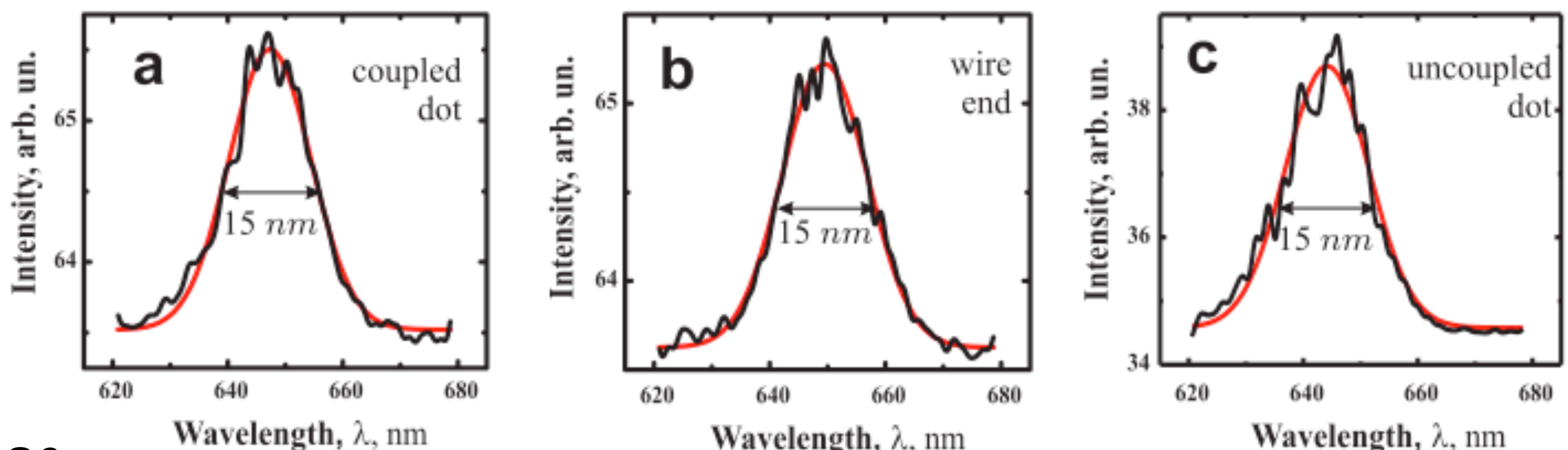


Fig. S6

Life is Short

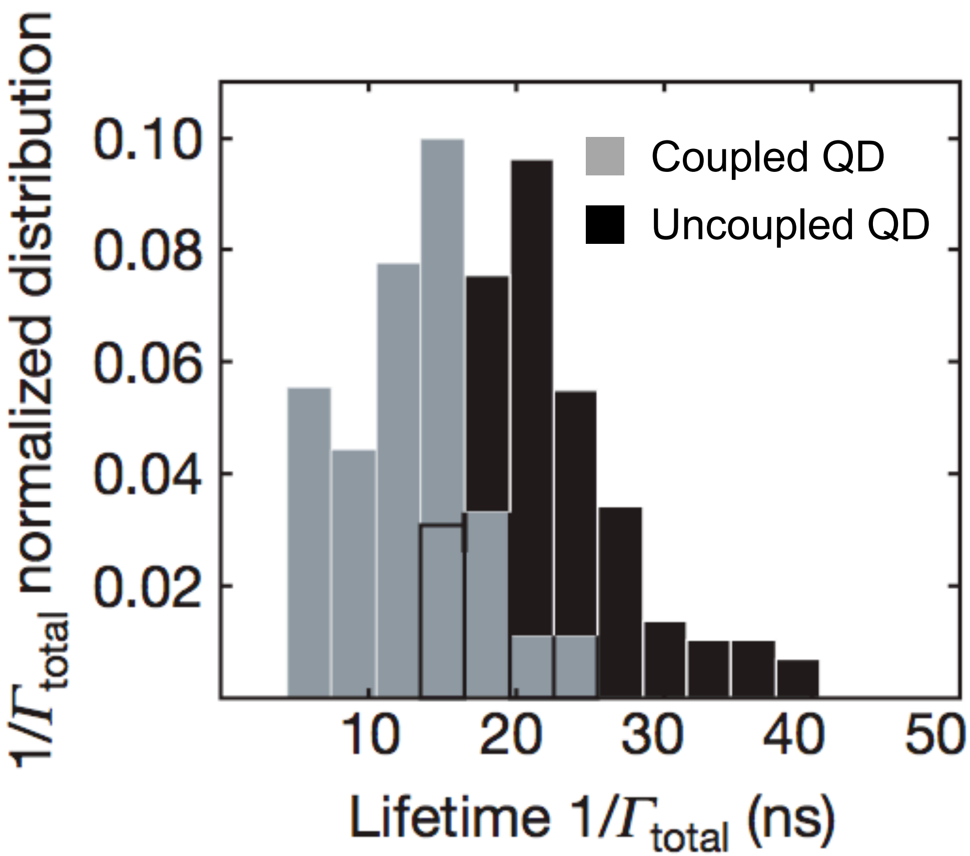


Fig. 4b

- Coupled QDs have shorter lifetimes
- Evidence of strong QD-NW coupling

Incredibly Efficient

$I_{1,2}$ = intensity at NW ends

I_{dot} = intensity of QD

η_m = apparent efficiency

η = actual efficiency (accounts for dissipation of SPs along NW)

SPs dissipate exponentially

β = absorption coefficient

l = length of NW

For 100 nm NW with QDs 35 nm away,

Calculated η = 50%

Actual η = 60 ± 10%

$$\eta_m = \frac{I_1 + I_2}{I_{dot} + I_1 + I_2},$$

$$\eta = \frac{I_2 e^{\beta l_2} + I_1 e^{\beta l_1}}{I_{dot} + I_2 e^{\beta l_2} + I_1 e^{\beta l_1}}.$$

$$I = I_0 e^{-\beta l}$$

$$\beta = \frac{1}{(l_2 - l_1)} \ln(\frac{I_1}{I_2})$$

Just the right thickness

Max efficency ~60% with ~30 nm PMMA thickness

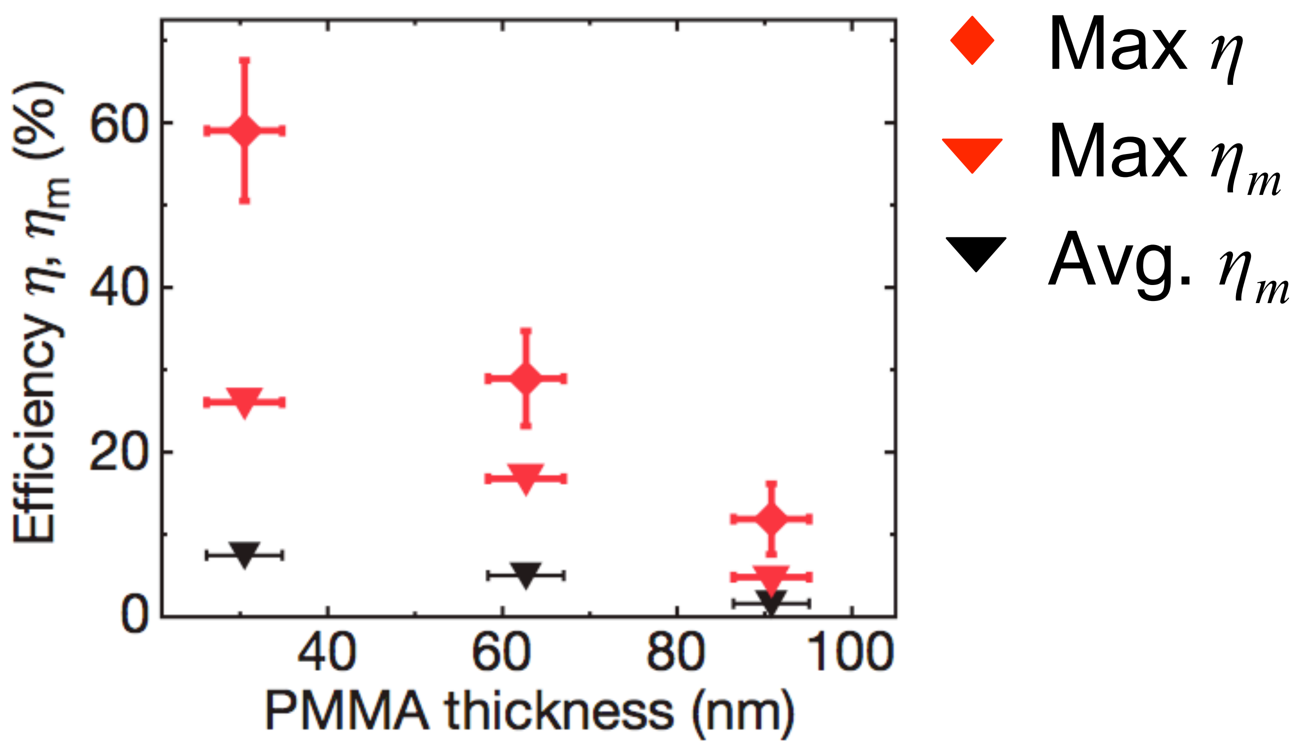


Fig. 4d

Conclusions

- Photons emitted from QDs can couple to SPs in metal NWs
- Energy is released at NW ends
- QDs release photons one at a time
- Coupling efficiency changes with QD-NW separation
- Maximum efficiency ~60% vs. typical ~1%