Efficient interfacing of single photons and single molecules

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Outline

Introductory discussion of light-matter interaction

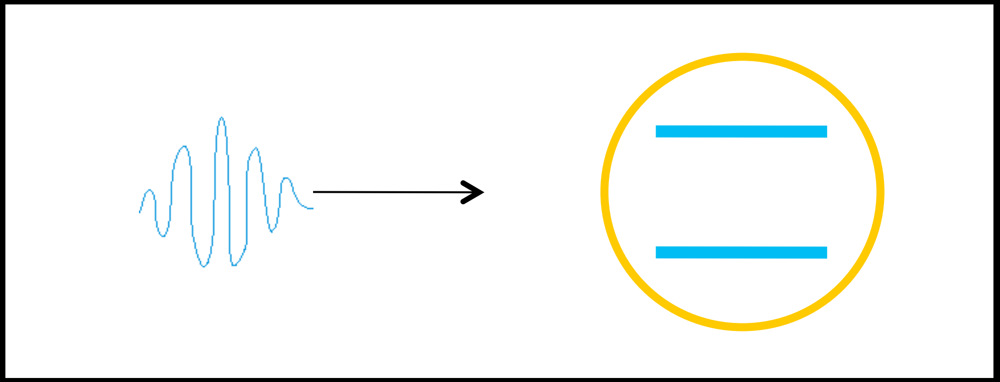
High resolution single molecule spectroscopy

Strong interaction of light and a single molecule

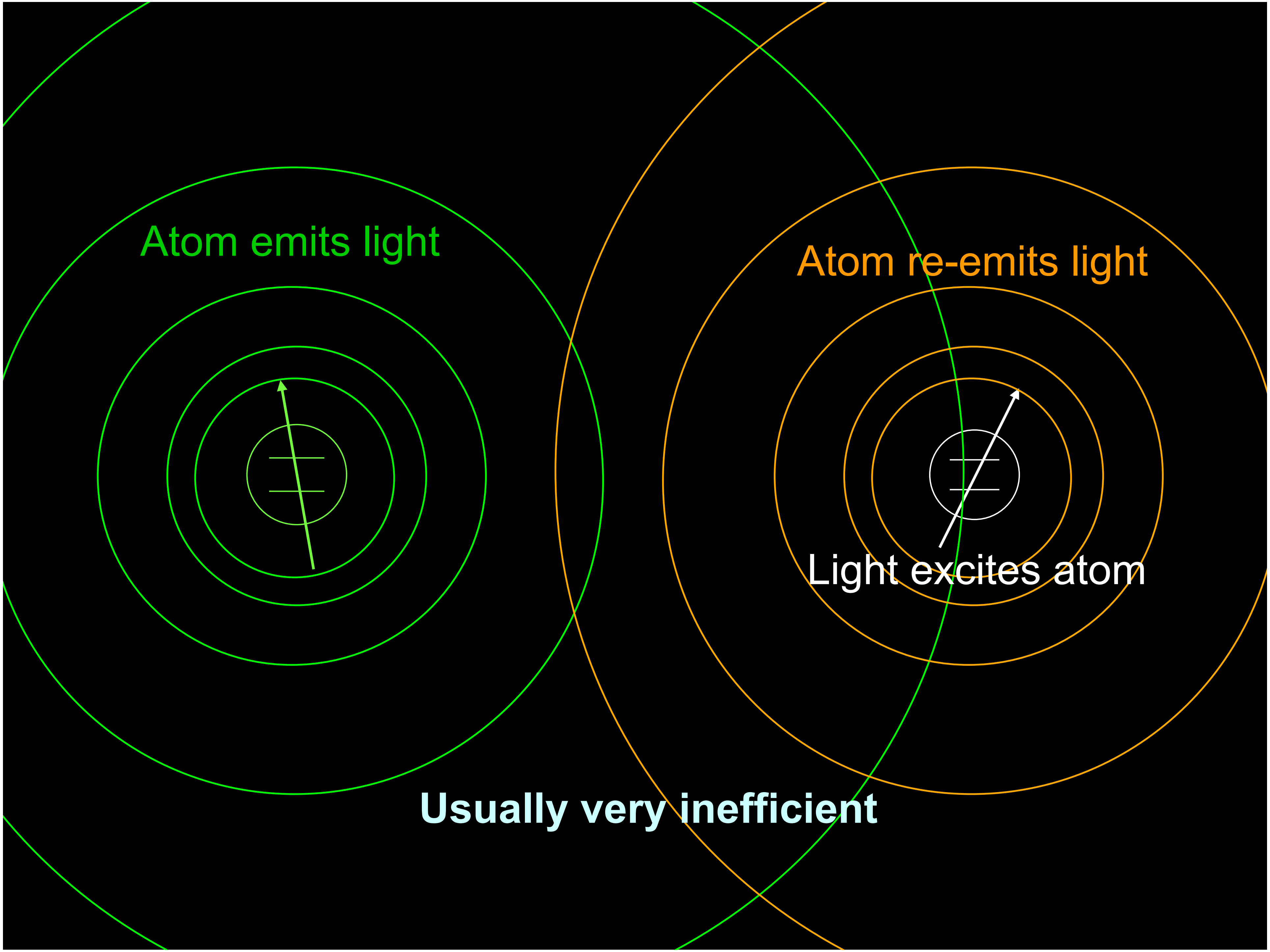
Generation of tunable narrow-band single photons

Tailoring light emission with optical antennas

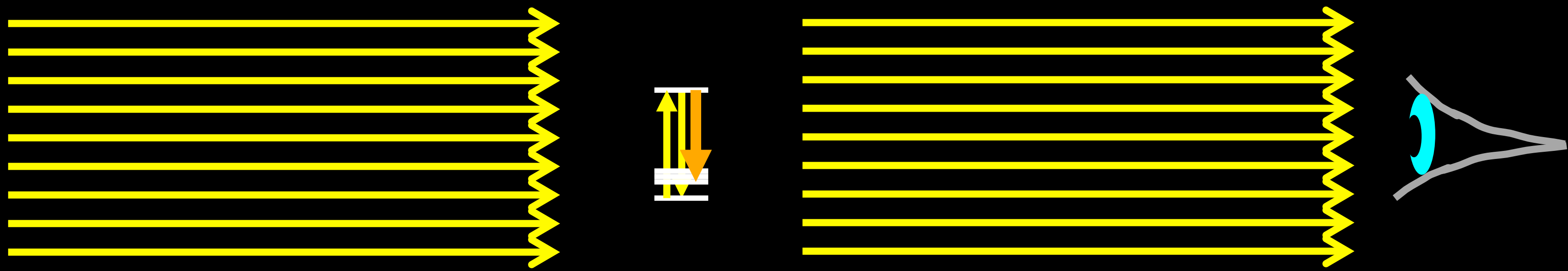
A single photon meets a single atom



The probability that a photon meets the emitter?


Atom emits light
Atom re-emits light
Light excites atom
Usually very inefficient

Simple absorption measurement



$$I_{int} = I_0 (\sigma_{abs}/A)$$

Ideal case: σ ~(500 nm)^2 ; typical A~ (10-100 μm)^2

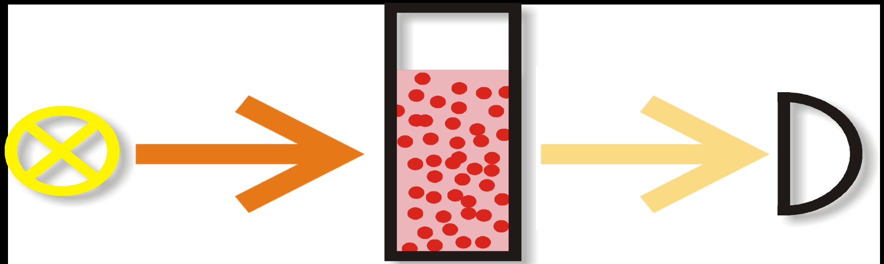
Experimental challenge:

See the effect of one atom on the intensity of a laser beam

As a result, single molecule detection is commonly done in fluorescence

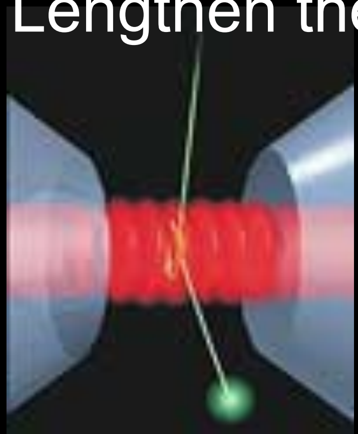
Imrpoving the efficiency

Easiest solution:
Strengthen the signal by using many atoms



$$I = I_0 \cdot \exp\left(-\sigma_{\mathrm{abs}} \cdot N \cdot l\right)$$

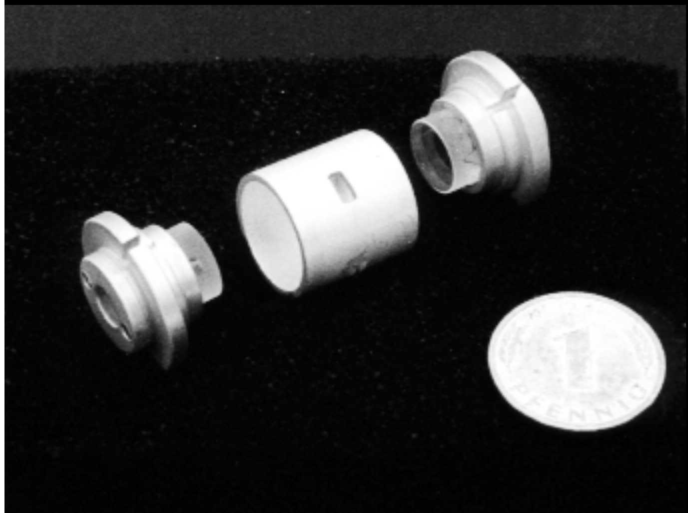
Another trick:
Lengthen the interaction time in a cavity

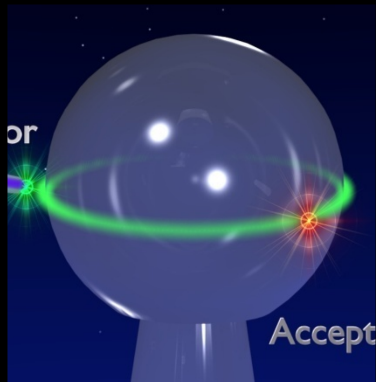


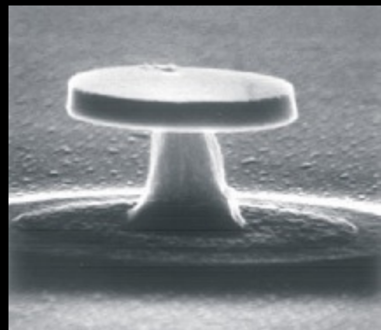
Make the Photon Stronger!

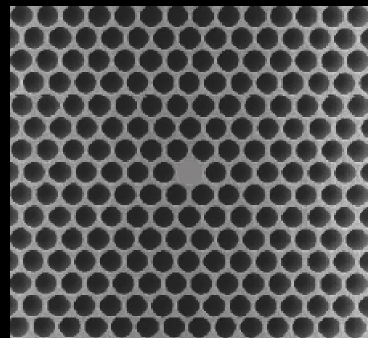
Confinement in a Microcavity

$$E_{ph} \sim \sqrt{\frac{h\omega}{\varepsilon V}}$$







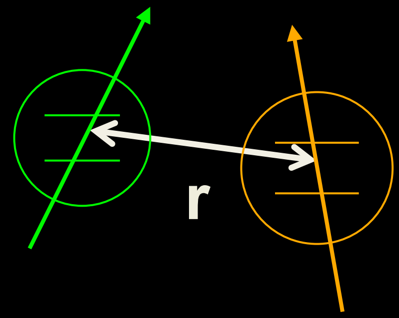


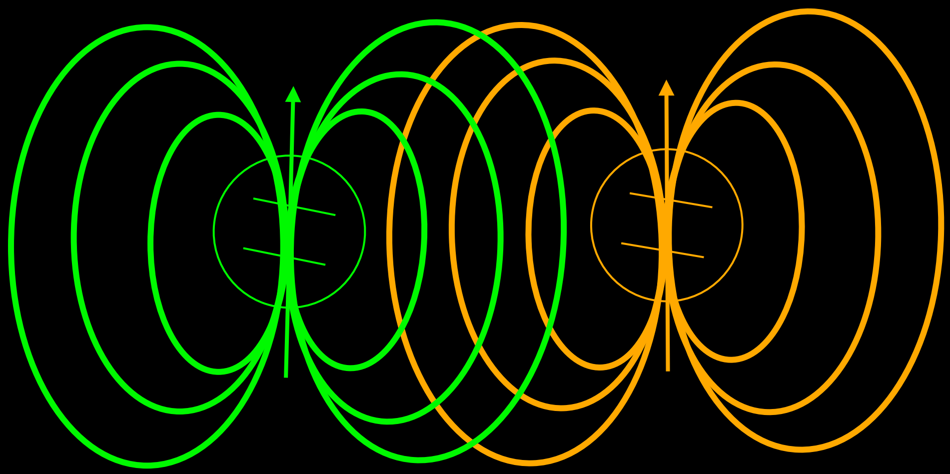
An atom also gets manipulated:

Change of the spontaneous emission rate

Change of the emission pattern

near-field dipole-dipole interaction

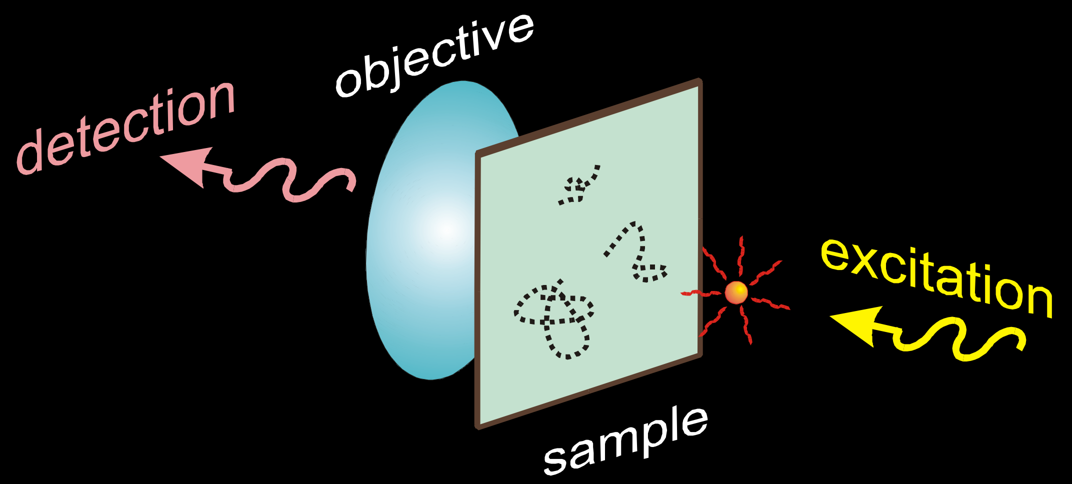


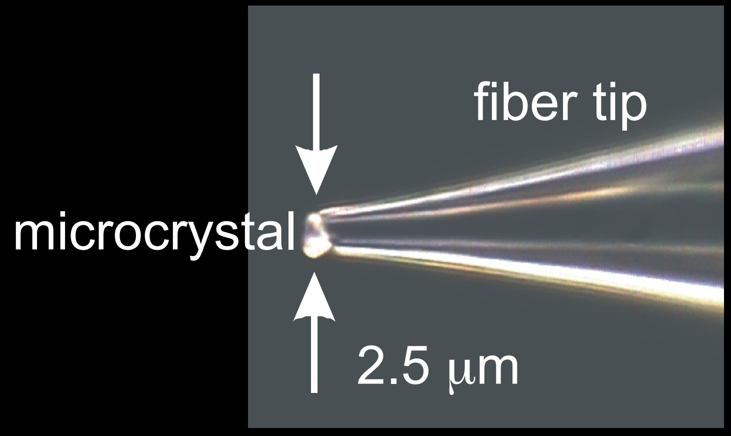


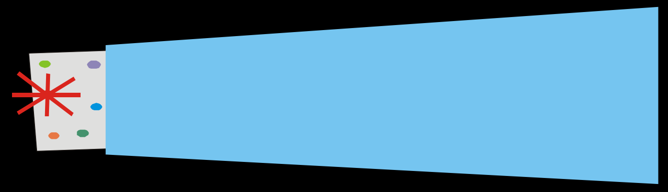
Variations:

- dissipative → Fluorescence (Förster) resonant energy transfer (FRET)
- coherent → Dicke sub and superradiance

A positionable single-molecule probe: realization of a point-like light source

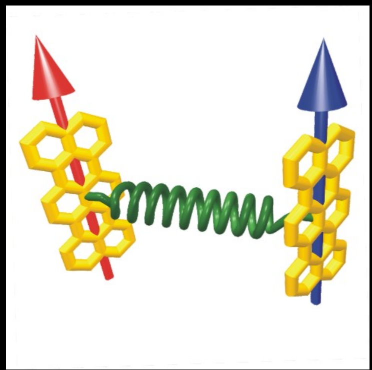


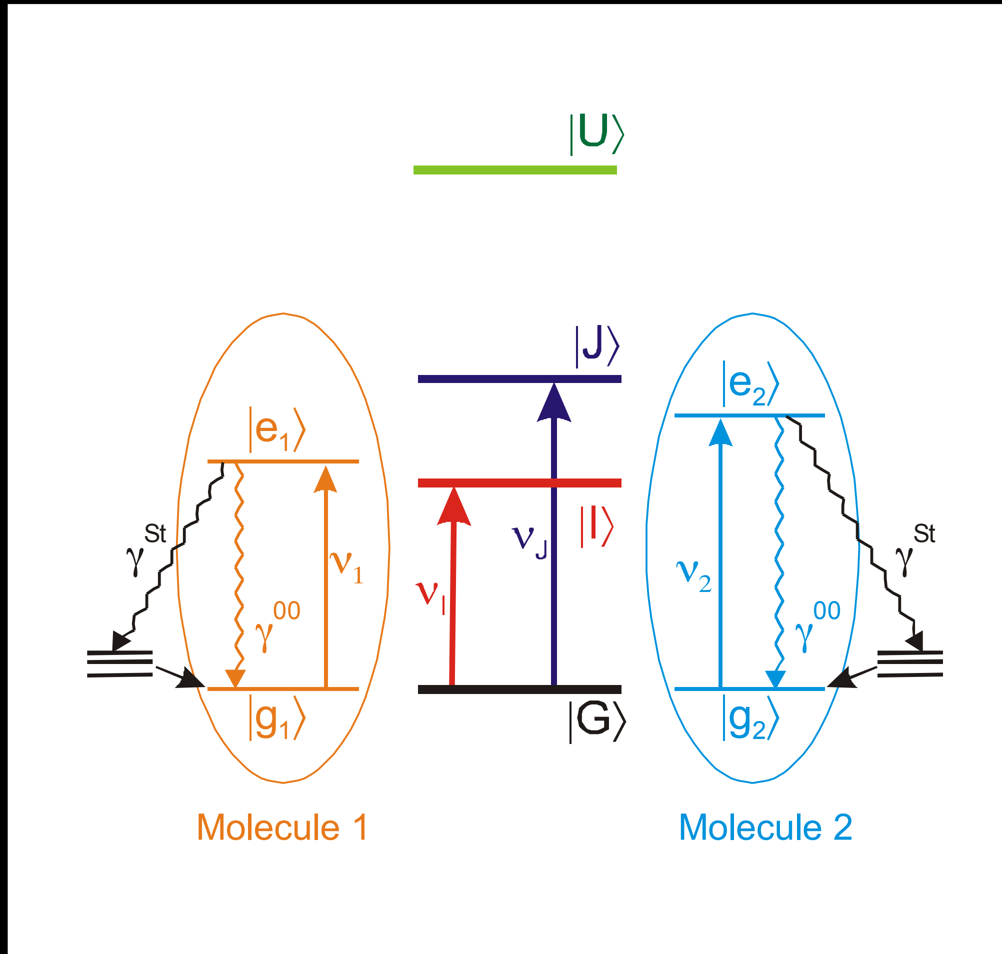




J. Michaelis, C. Hettich, J. Mlynek & V. Sandoghdar, *Nature* **405**, 325 (2000).

Coherent dipole-dipole coupling between two individual molecules



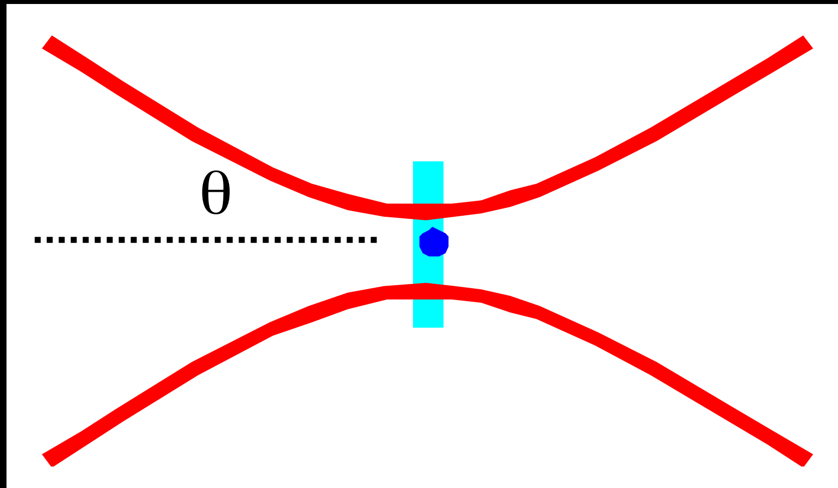


C. Hettich, C. Schmitt, J. Zitzmann, S. Kühn, I. Gerhardt & V. Sandoghdar, *Science* **298**, 385-389 (2002).

What if we simply confine a propagating wave?

Reduce A

$$I = I_0\,(1-\sigma_{abs}/A)$$



FWHM of a focused beam ~ $\lambda/2NA$; $NA = n\sin\theta$

$A \sim \lambda^2/4$ for $NA \sim 1$

$\sigma_{abs} = 3\lambda^2/2\pi \sim \lambda^2/2$

Scattered power = Incident intensity . cross section

Probability of being in the excited state

Spontaneous emission rate (natural linewidth)

Scattered power= $\rho_{22}.\gamma_0.h\nu$ — Energy per emitted photon

$$\rho_{22} = \frac{\frac{1}{4}\left(\frac{\gamma}{\gamma_0}\right)|\mathcal{V}|^2}{(\omega_0-\omega)^2 + \frac{1}{2}\left(\frac{\gamma}{\gamma_0}\right)|\mathcal{V}|^2 + \gamma^2}$$

$$\gamma = \gamma_0 + \gamma_{coll}$$

On resonance and in the weak excitation limit,

$$|\mathcal{V}|^2 \propto |E|^2 |D_{eg}|^2$$

Scattered power $\propto \gamma_0.h\nu.\frac{|\mathcal{V}|^2}{\gamma_0\gamma} = \left(\frac{\gamma_0}{\gamma}\right)h\nu\frac{|\mathcal{V}|^2}{\gamma_0}$

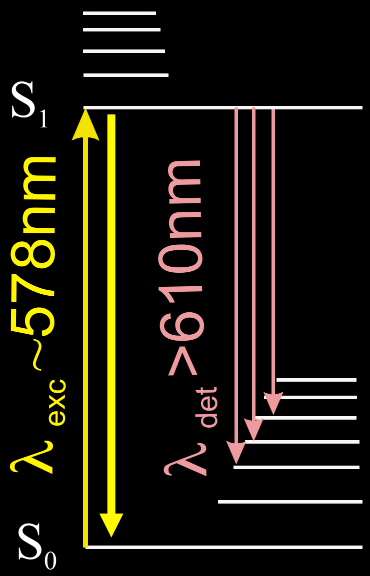
$$\gamma_0 \propto |D_{eg}|^2 \nu^3$$

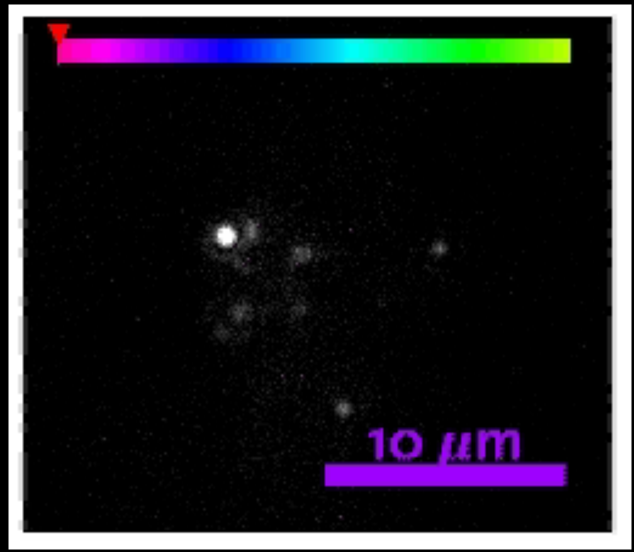
$$\sigma_{abs} = \frac{3\lambda^2}{2\pi}\frac{\gamma_0}{\gamma}\ \alpha$$

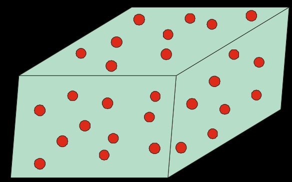
γ_0 → natural linewidth

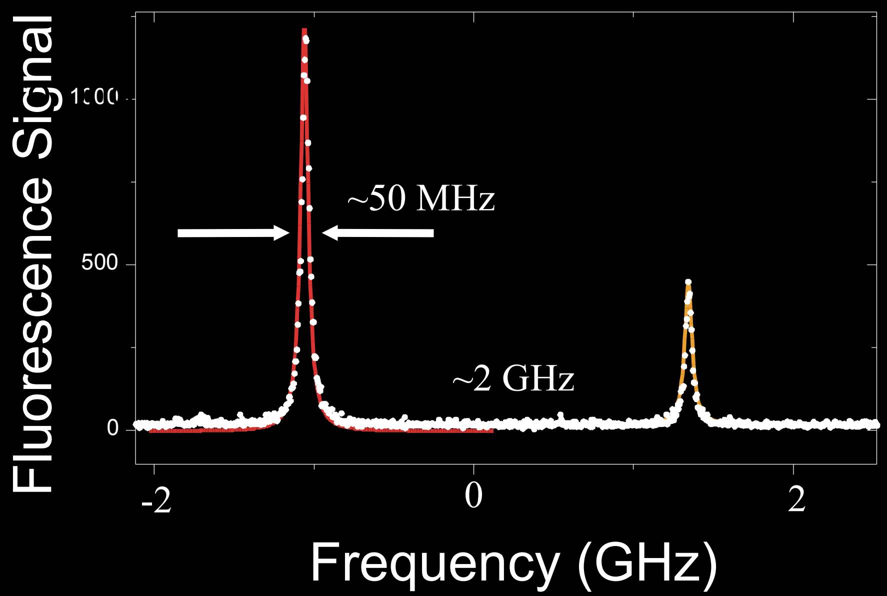
γ → homogeneous linewidth

Single molecule spectroscopy in solid matrices



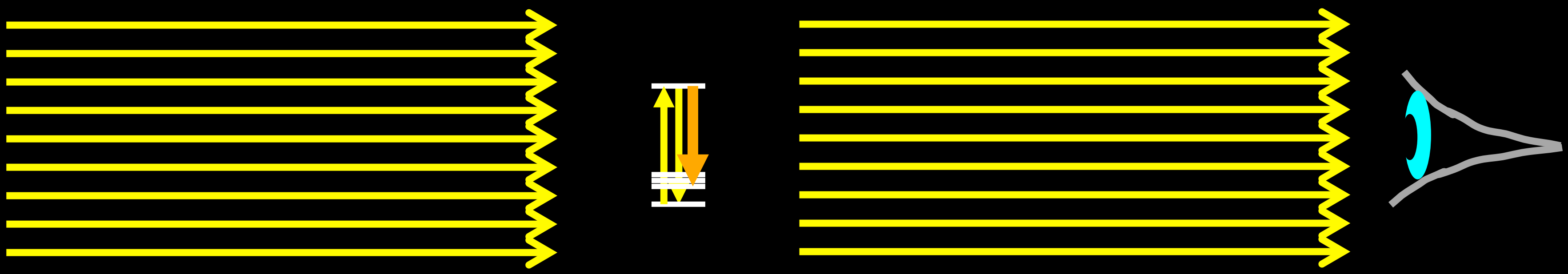


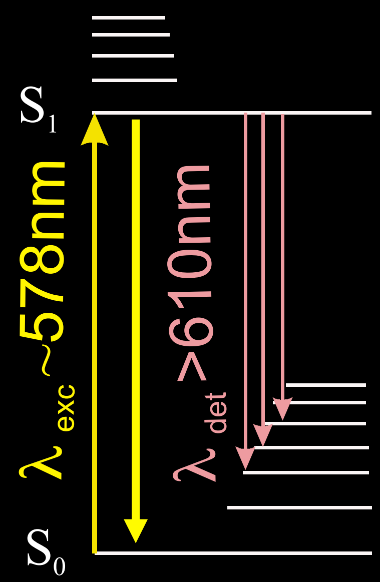




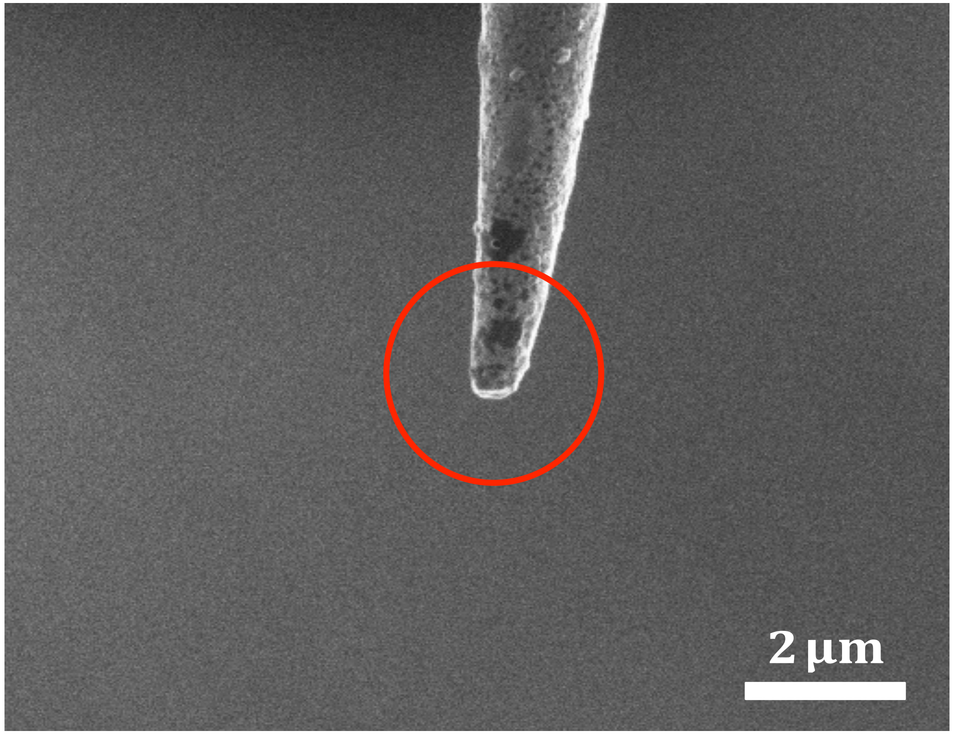
M. Orrit and J. Bernard, *Phys. Rev. Lett*. **65**, 2716 (1990).

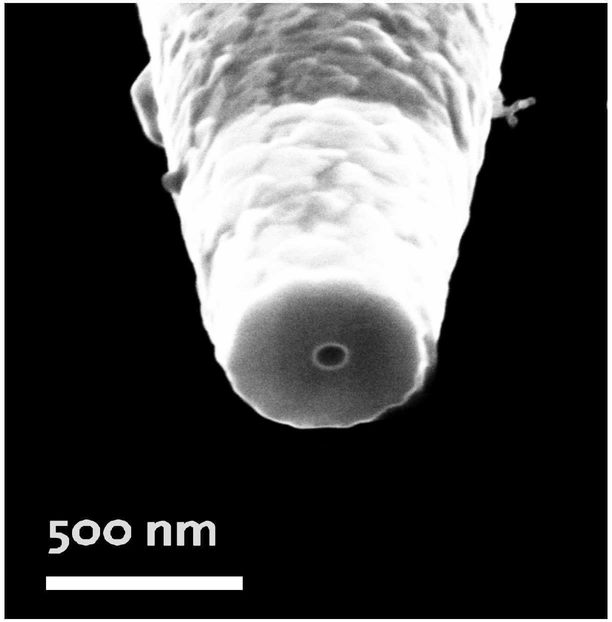
Coherent detection of single molecules:
Extinction spectroscopy





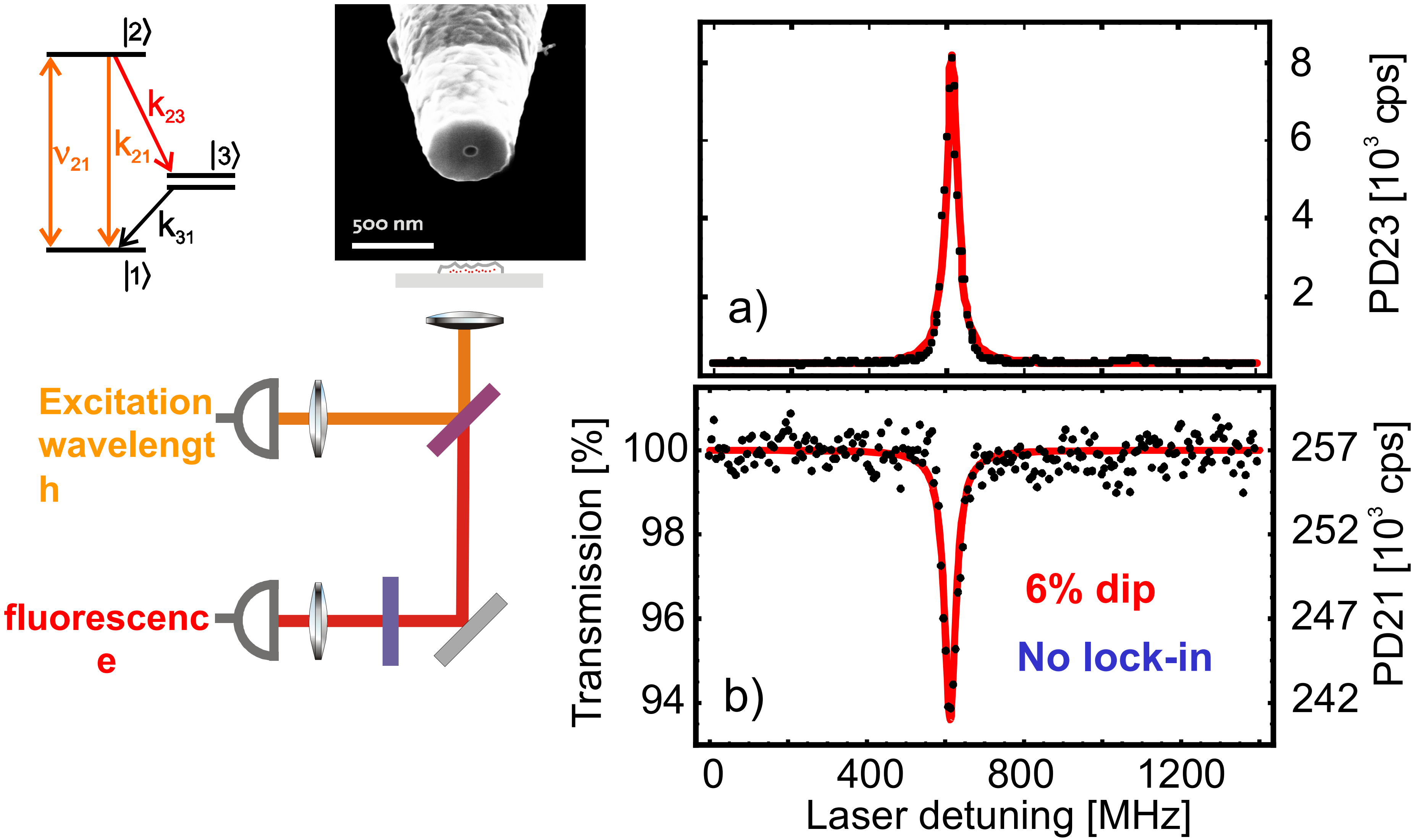
Confine light by a subwavelength aperture





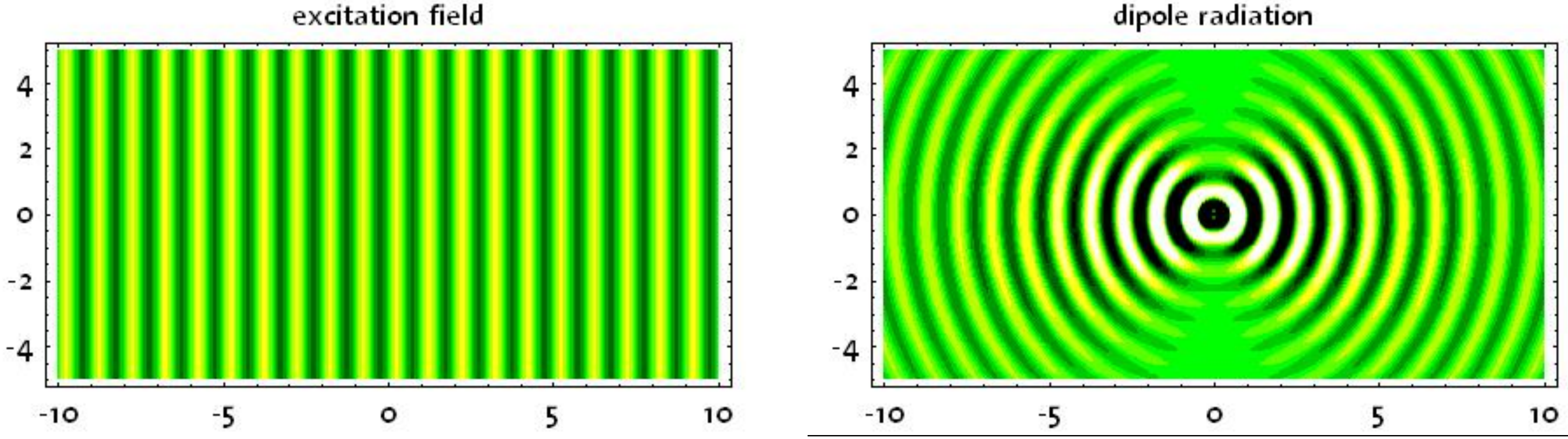
Focused ion beam (FIB) milled

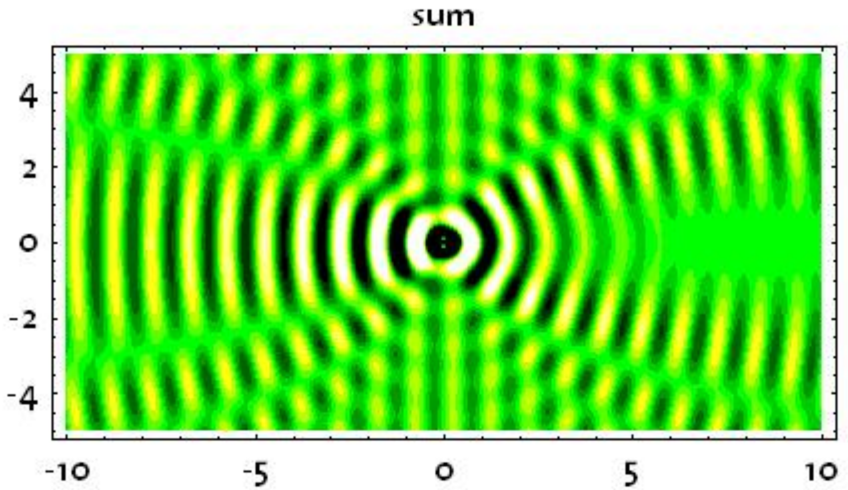
Physical confinement of light to a subwavelength area



I. Gerhardt, G. Wrigge, P. Bushev, G. Zumofen, M. Agio, R. Pfab, V. Sandoghdar, *Phys. Rev. Lett*. **98**, 033601 (2007).

The extinction signal is due to interference





$$I_{total} = |E_{inc}|^2 + |E_{sca}|^2 + 2Re(E_{inc}E_{sca}^*)$$

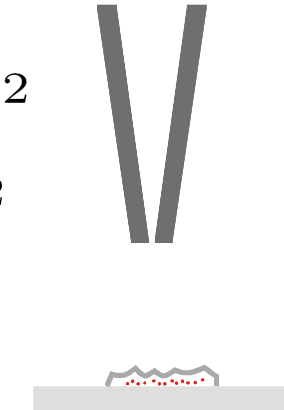
↓

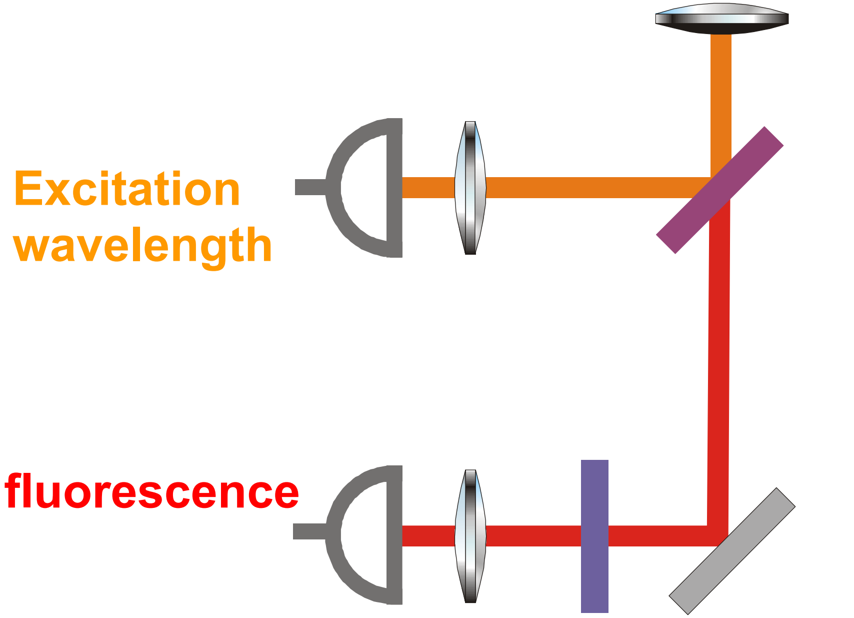
extinction

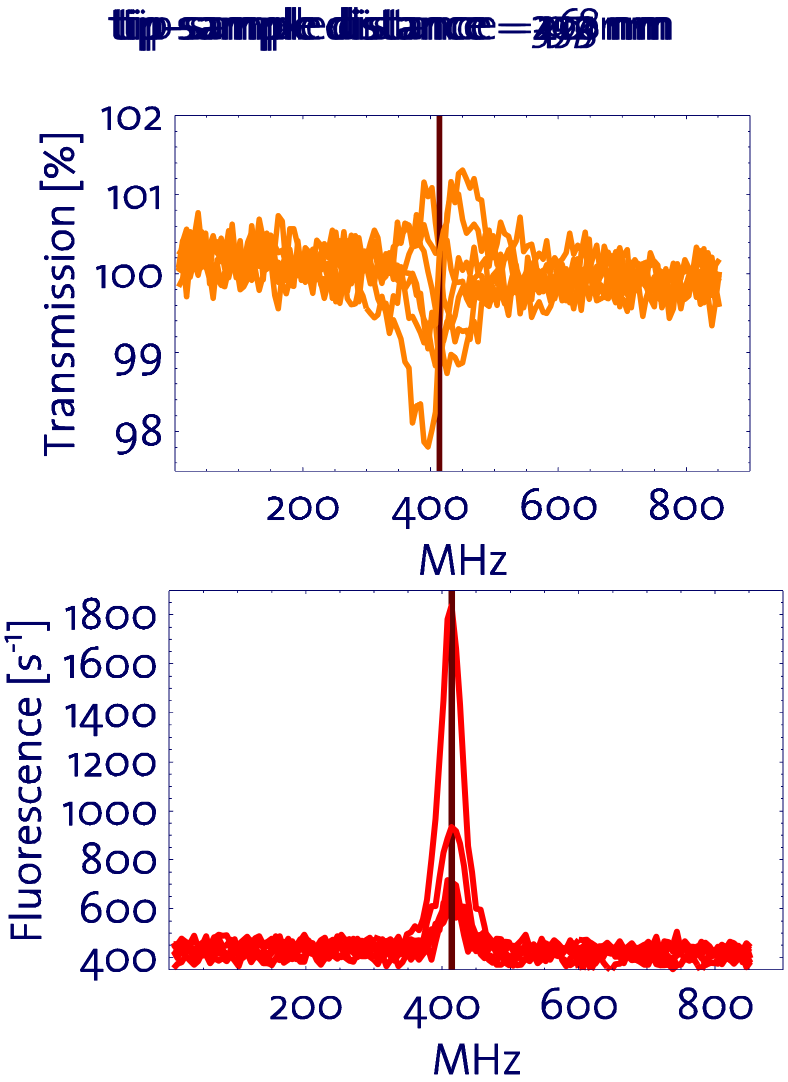
Complete extinction in the forward direction, hence the definition of the absorption cross section (optical theorem)

The signal originates from interference

$$I_{\mathrm{total}} = |E_{\mathrm{inc}} + E_{\mathrm{sca}}|^2 = |E_{\mathrm{inc}}|^2 + |E_{\mathrm{sca}}|^2 + 2\,\mathrm{Re}\,(E_{\mathrm{inc}}\,E^*_{\mathrm{sca}})$$







I. Gerhardt, G. Wrigge, M. Agio, P. Bushev, G. Zumofen, V. Sandoghdar, *Opt. Lett.* **32**, 1420 (2007).

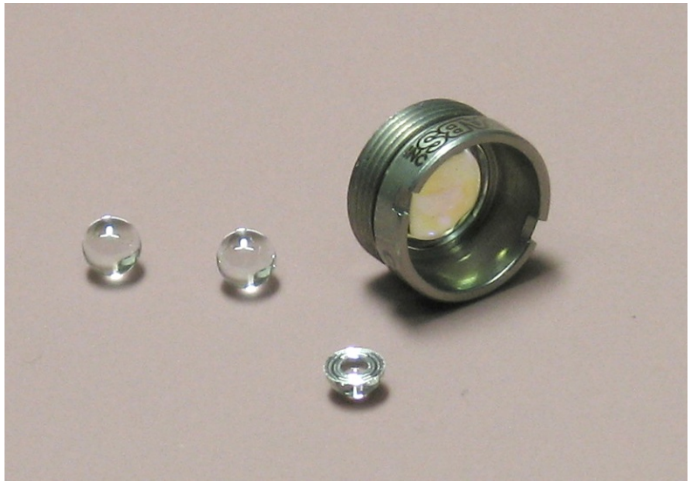
Disadvantages of near-field coupling

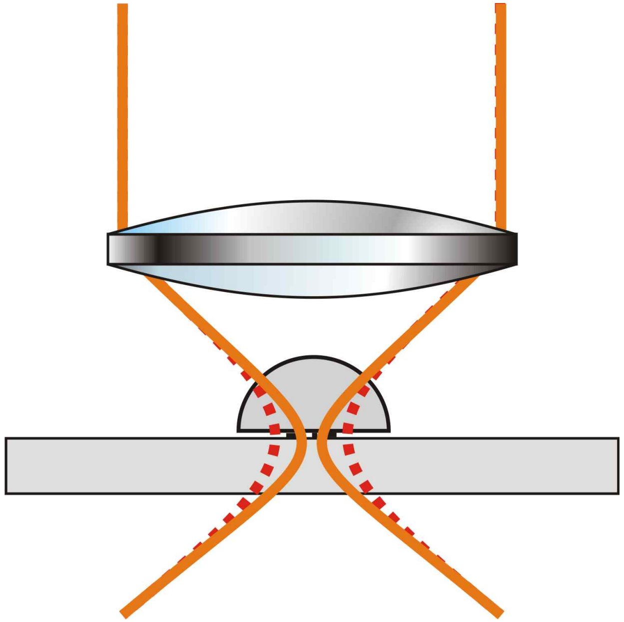
Very small through-put of about 10^{-4}-10^{-5}

Fragile tips

Needs very thin samples

Solid immersion lenses

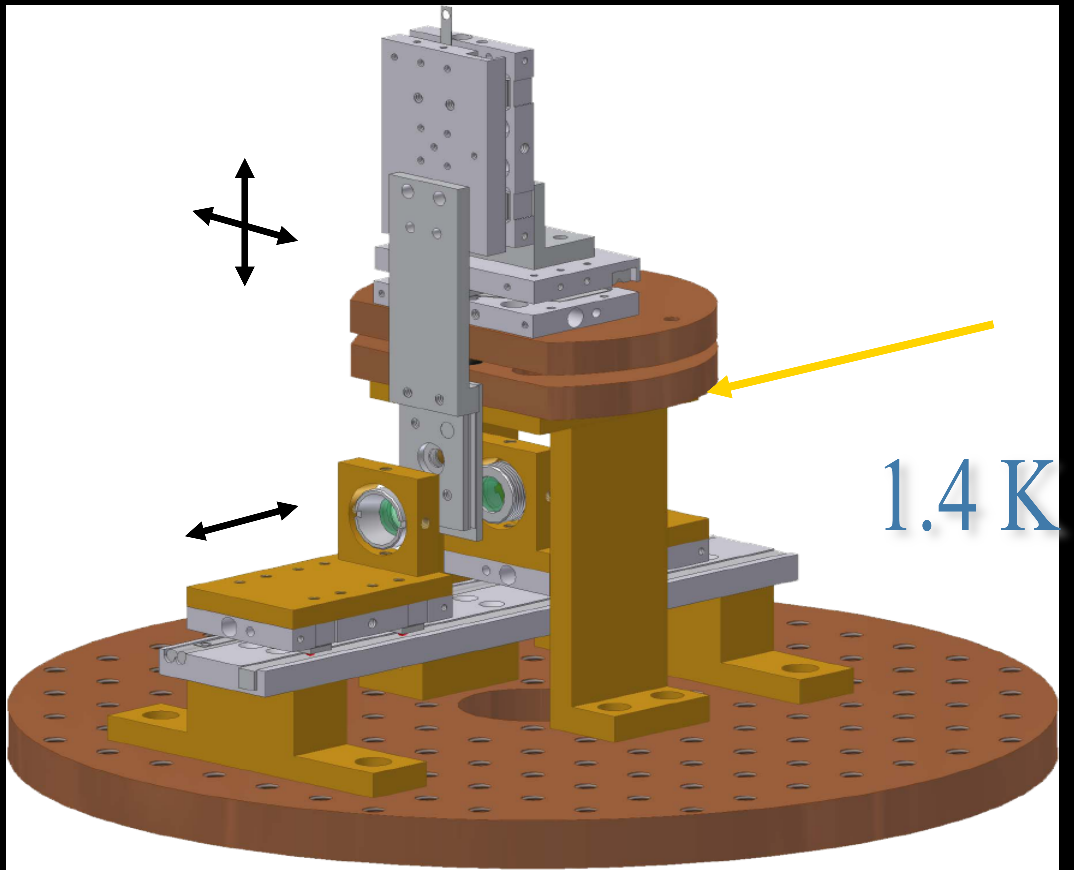




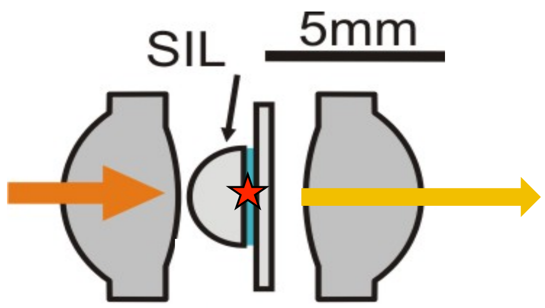
$$\lambda = \frac{\lambda_{\mathrm{vac}}}{n}$$

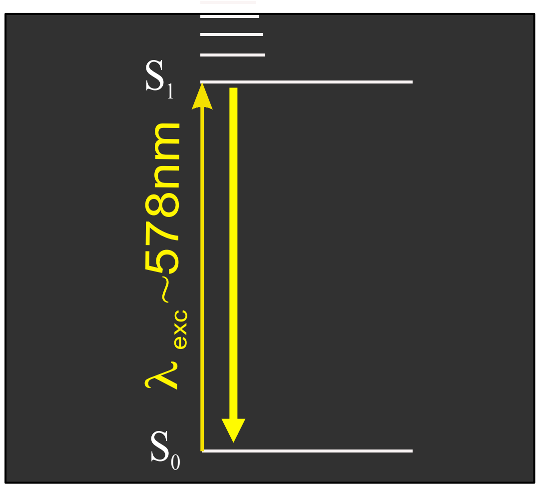
Mansfield & Kino, *Appl. Phys. Lett.* 57 (1990).

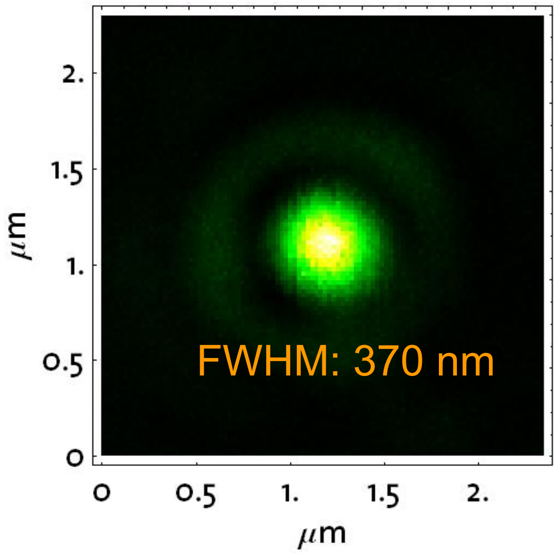
Inside the cryostat

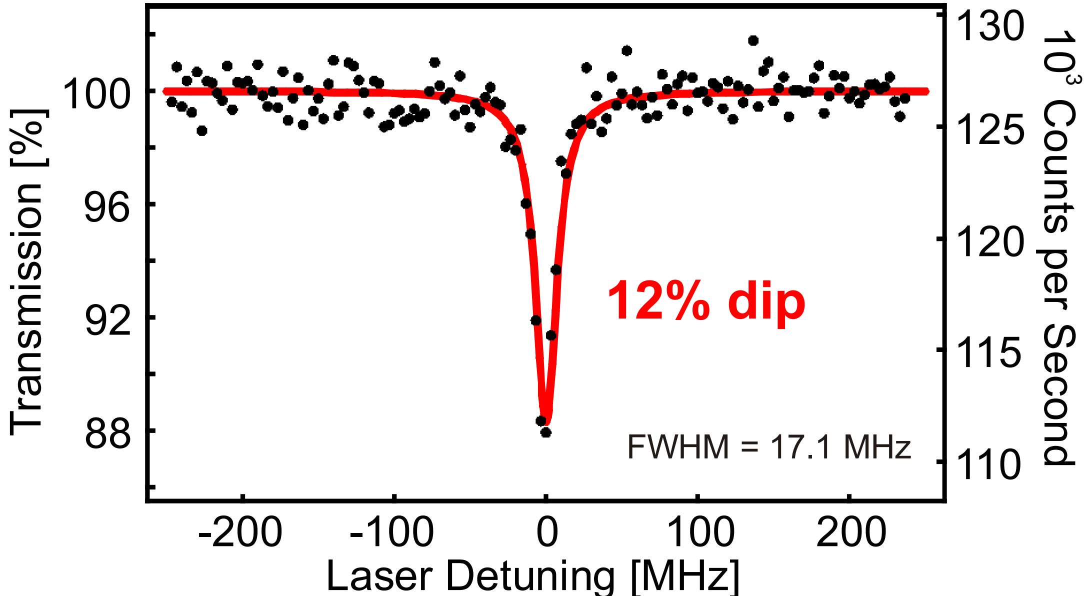


Far-field extinction spectroscopy on a single molecule









G. Wrigge, I. Gerhardt, J. Hwang, G. Zumofen, V. Sandoghdar, *Nature Phys.* **4**, 60 (2008).

Absorption spectroscopy of a single molecule (1989)

VOLUME 62, NUMBER 21 PHYSICAL REVIEW LETTERS 22 MAY 1989

Optical Detection and Spectroscopy of Single Molecules in a Solid

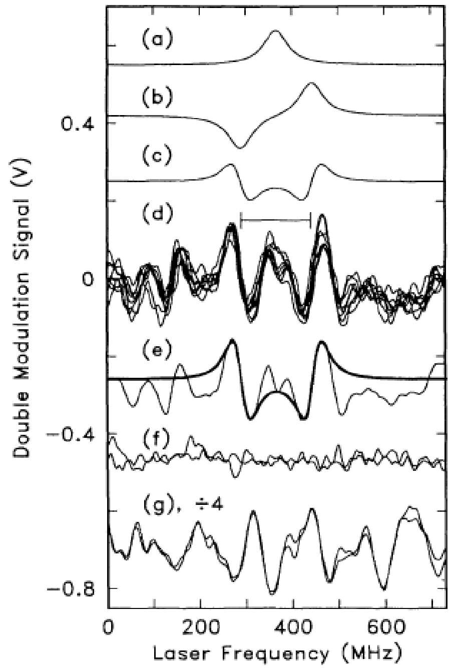


FIG. 1. Illustration of single-molecule spectra using FMS technique. (a) Simulation of absorption line, $\gamma = 65$ MHz. (b) Simulation of FM spectrum for (a), $\nu_m = 75$ MHz. (c) Simulation of FMS line shape. (d) SMD spectra at 592.423 nm, 512 averages, 8 traces overlaid, bar shows value of $2\nu_m = 150$ MHz. (e) Average of traces in (d) (S_2 removed) with fit to the in-focus molecule (smooth curve). (f) Signal far off line at 597.514 nm. (g) Traces of SFS at the O_2 line center, 592.186 nm.

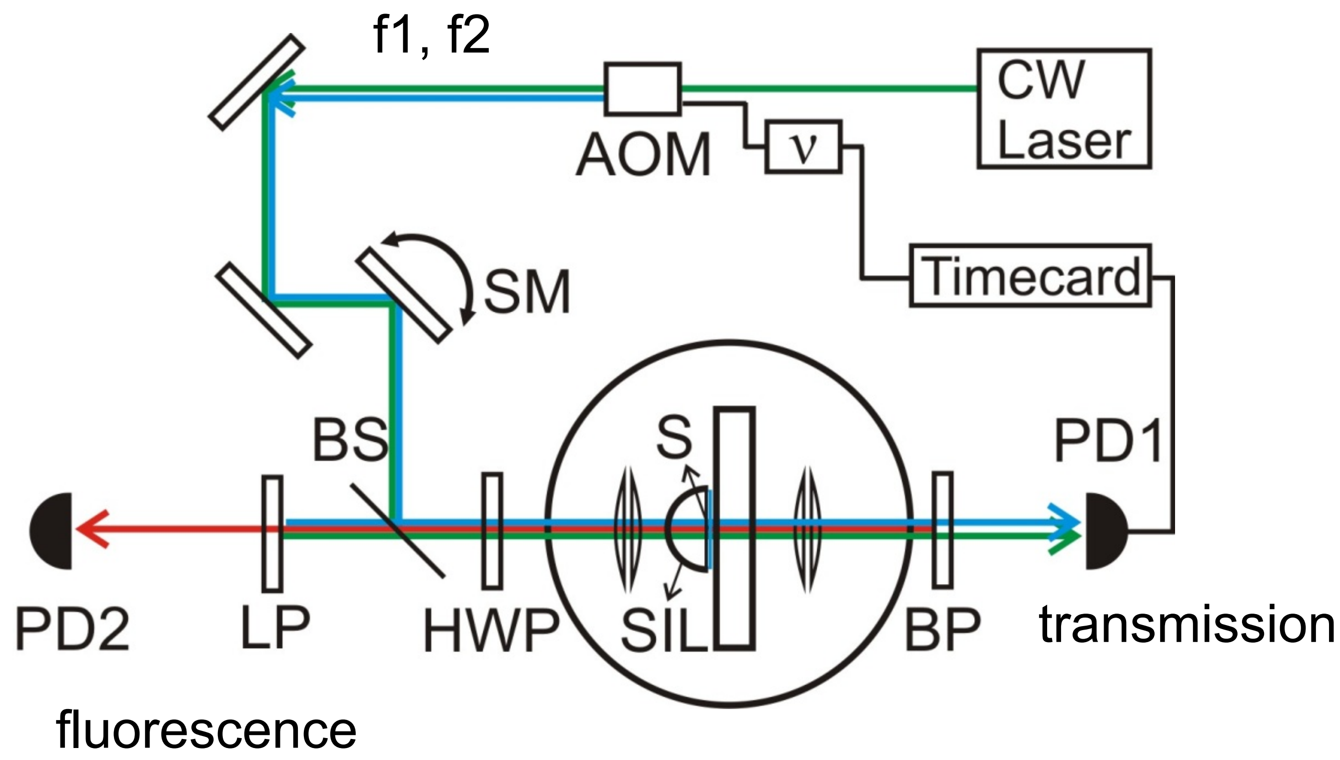
Double FM detection

A ~ 3μm

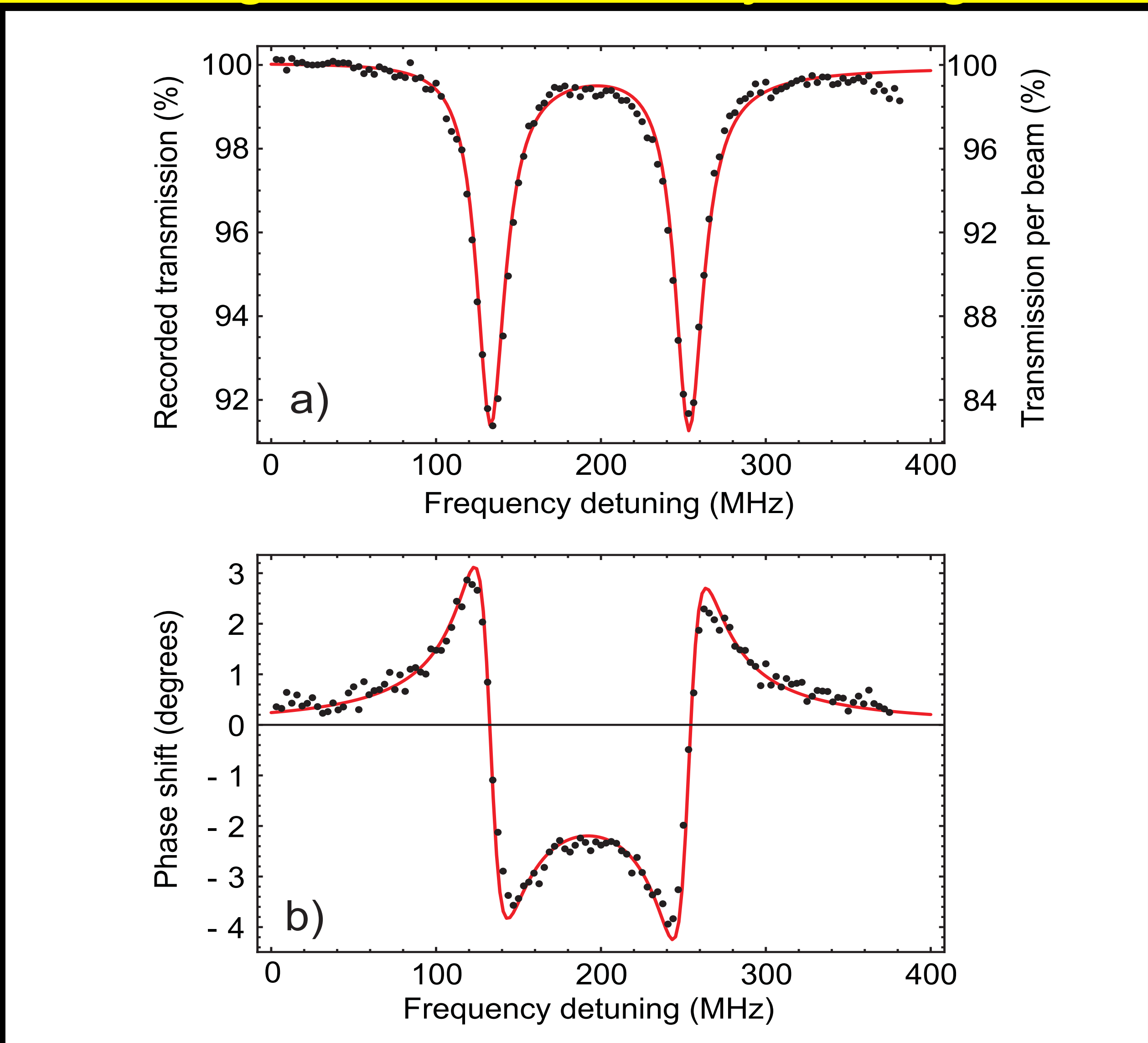
Phase shifting a laser beam by a single molecule

Need an interferometer to measure

A robust heterodyne interferometer

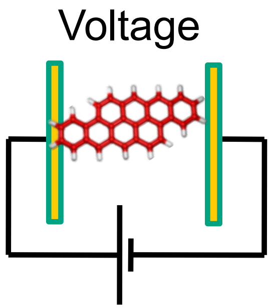


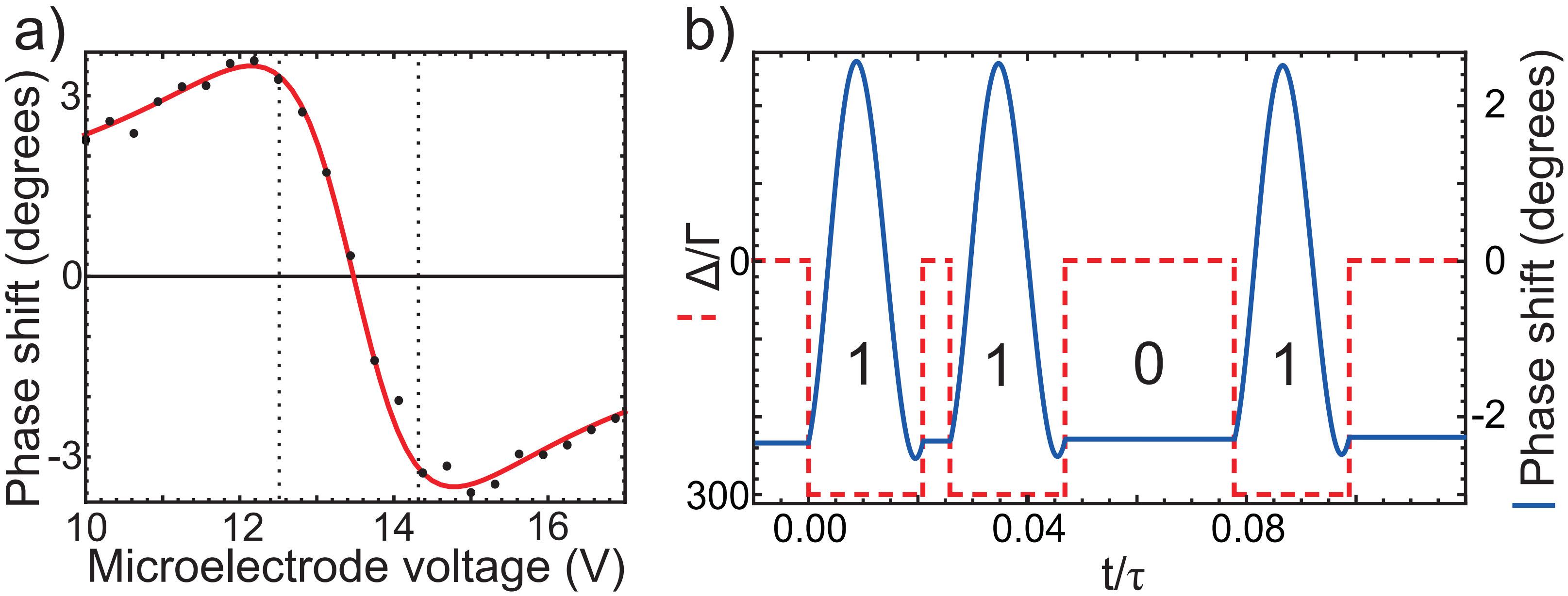
Phase shifting a laser beam by a single molecule



M. Pototschnig, Y. Chassagneux, J. Hwang, G. Zumofen, A. Renn, V. Sandoghdar, *Phys. Rev. Lett.* **107**, 063001 (2011).

Electric-field controlled fast phase switch

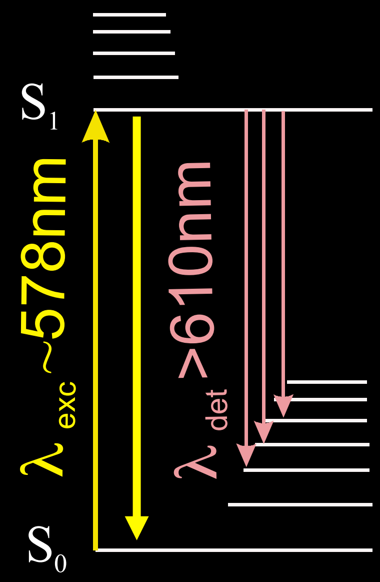


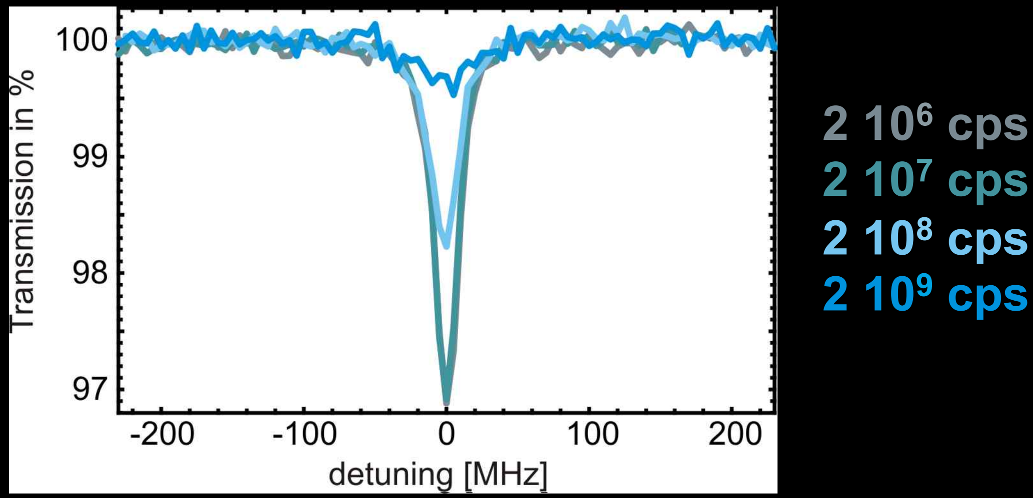


Exploring nonlinearity

Intensity dependence of the extinction dip

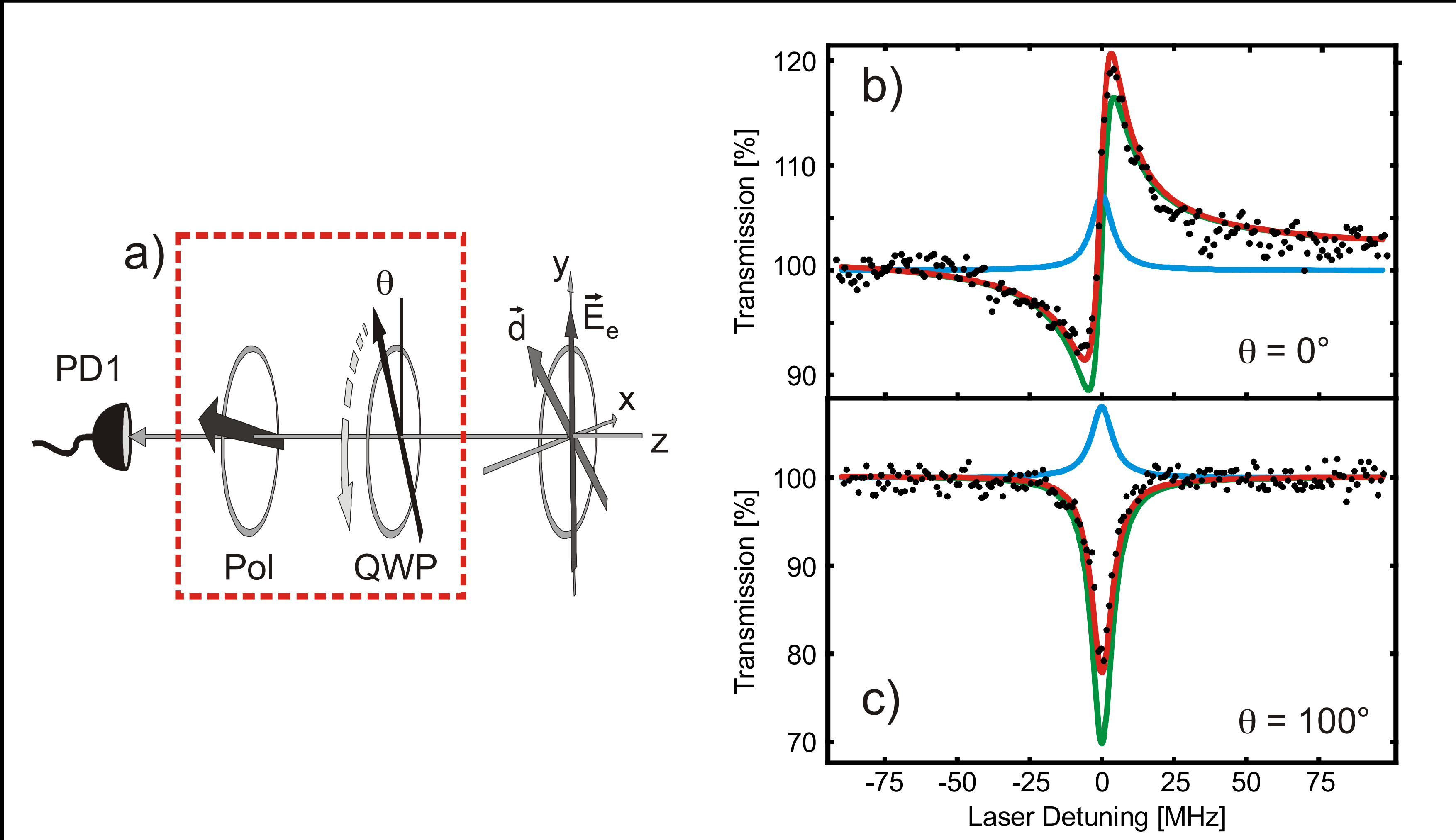
Transition from coherent to incoherent scattering





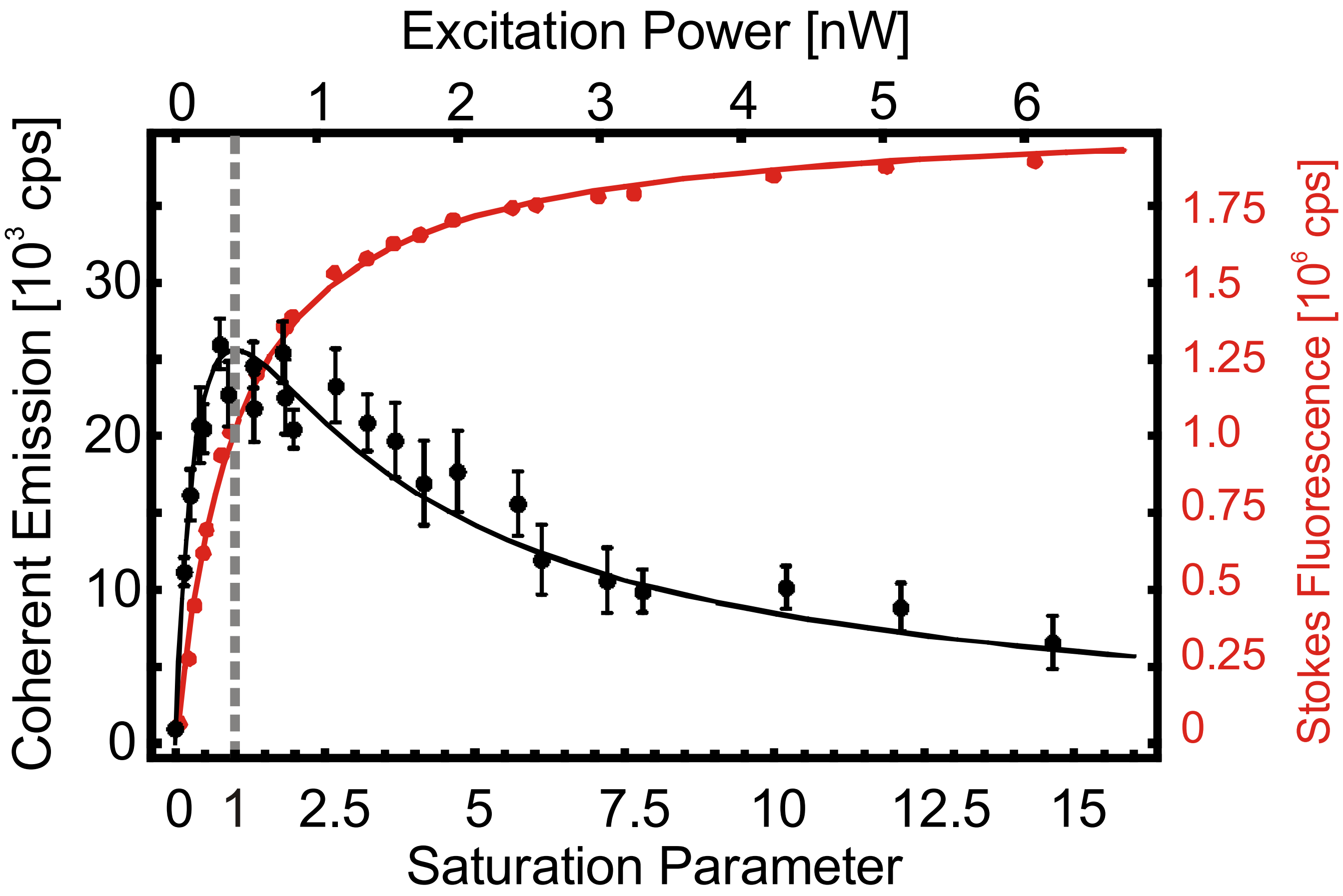
The molecule can maximally deal with one photon per two lifetimes (10 ns). At higher excitation intensity the extinction signal saturates.

Separating the coherent and incoherent parts



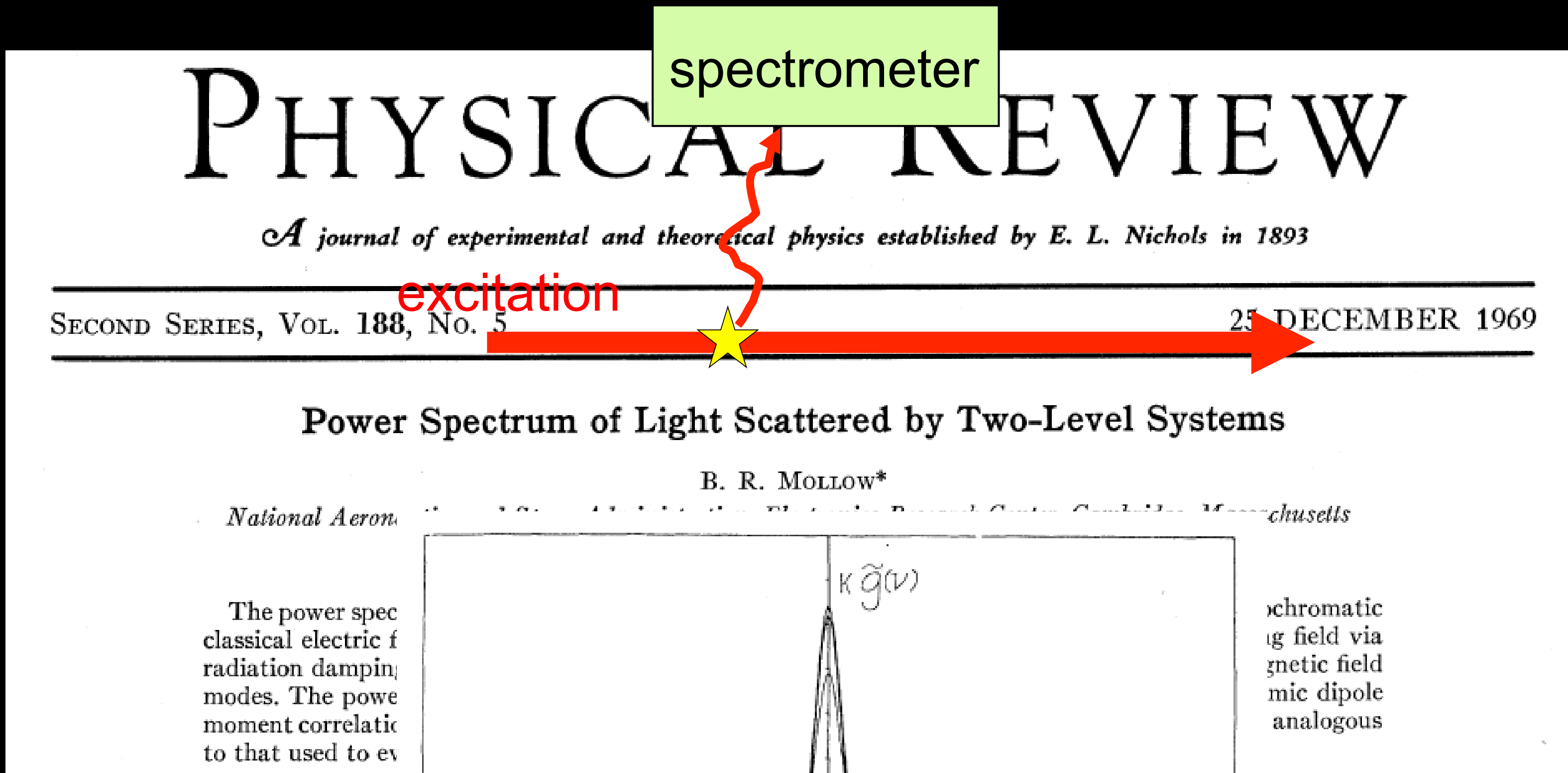
G. Wrigge, I. Gerhardt, J. Hwang, G. Zumofen, V. Sandoghdar, *Nature Physics* **4**, 60 (2008).

Transition from coherent to incoherent scattering



G. Wrigge, I. Gerhardt, J. Hwang, G. Zumofen, V. Sandoghdar, *Nature Physics* **4**, 60 (2008).

Resonance fluorescence and Mollow Triplet



PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 188, NO. 5 — 25 DECEMBER 1969

Power Spectrum of Light Scattered by Two-Level Systems

B. R. MOLLOW*

National Aeron... ...chusetts

The power spec... ...ochromatic
classical electric f... ...g field via
radiation dampin... ...gnetic field
modes. The powe... ...mic dipole
moment correlatio... ...analogous
to that used to ev...

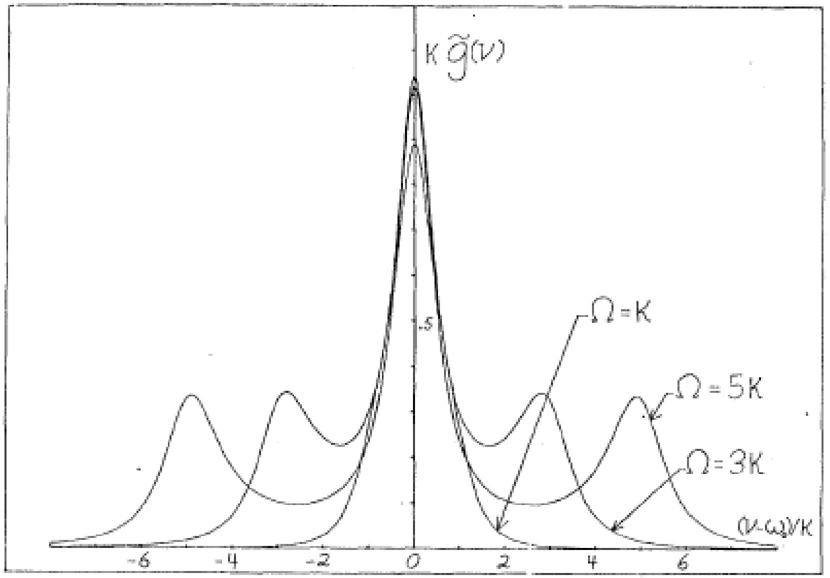
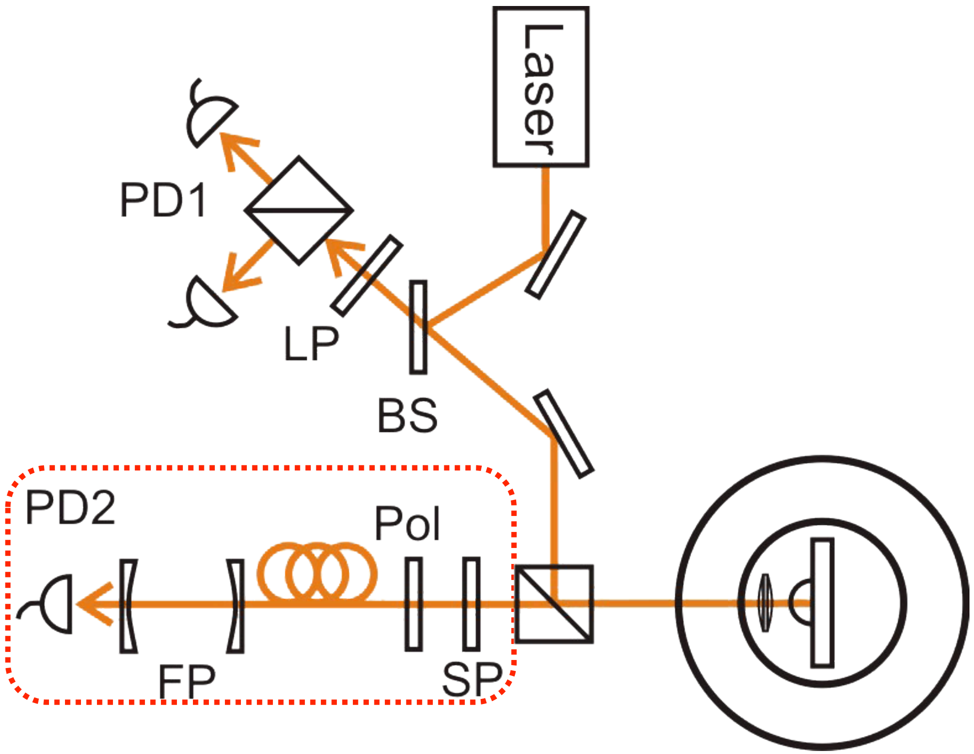


FIG. 1. Spectral density $\bar{g}(\nu)$ for a two-level atom driven exactly on resonance.

Direct detection of the Mollow triplet



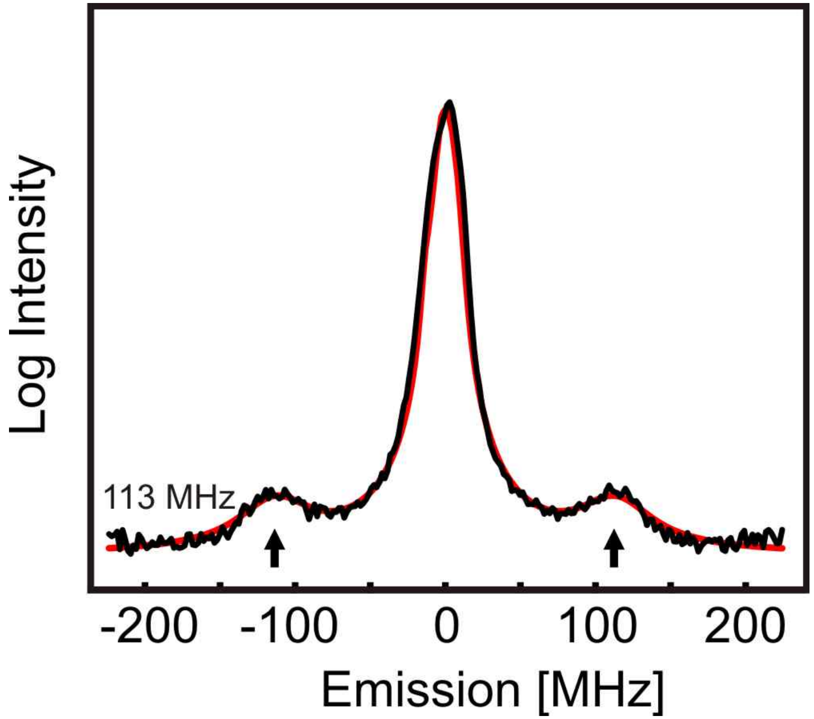
PD: photodetector

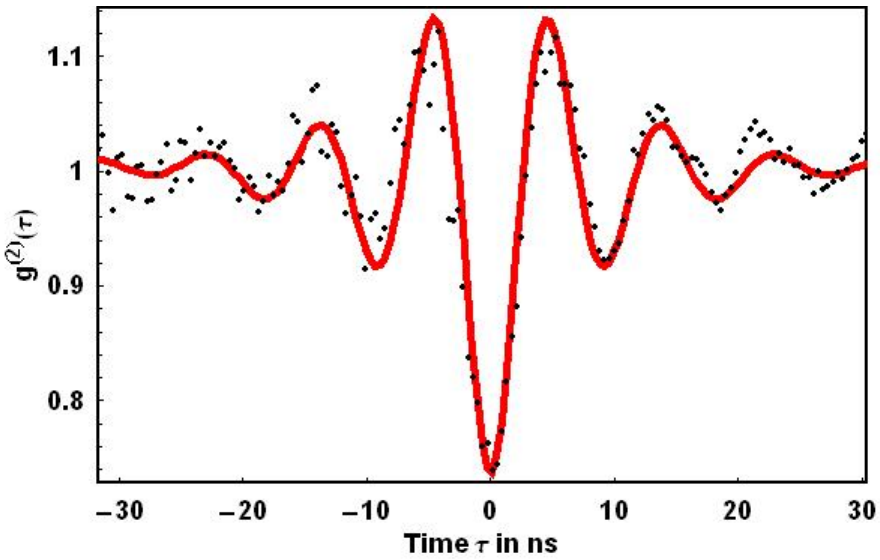
SP/LP: short/long pass

Pol: polarizer

FP: Fabry-Perot cavity

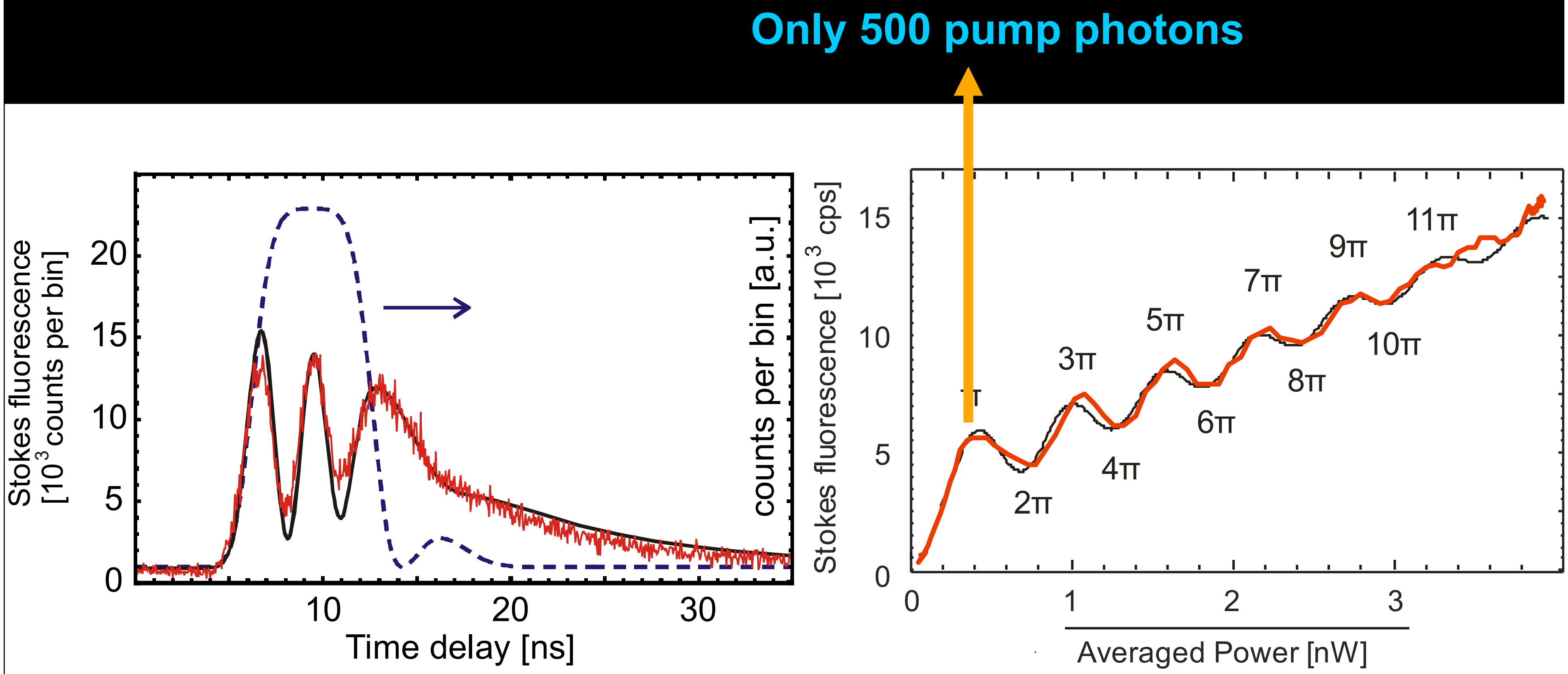
The first observation of resonance fluorescence and the Mollow triplet in the solid-state





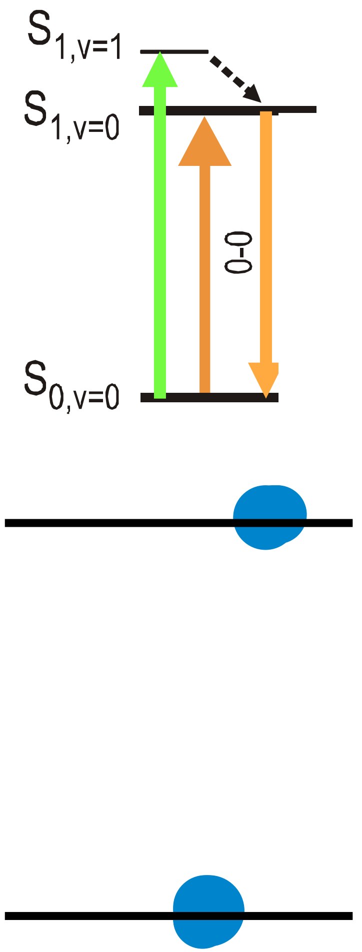
G. Wrigge, I. Gerhardt, J. Hwang, G. Zumofen, V. Sandoghdar, *Nature Physics* **4**, 60 (2008).

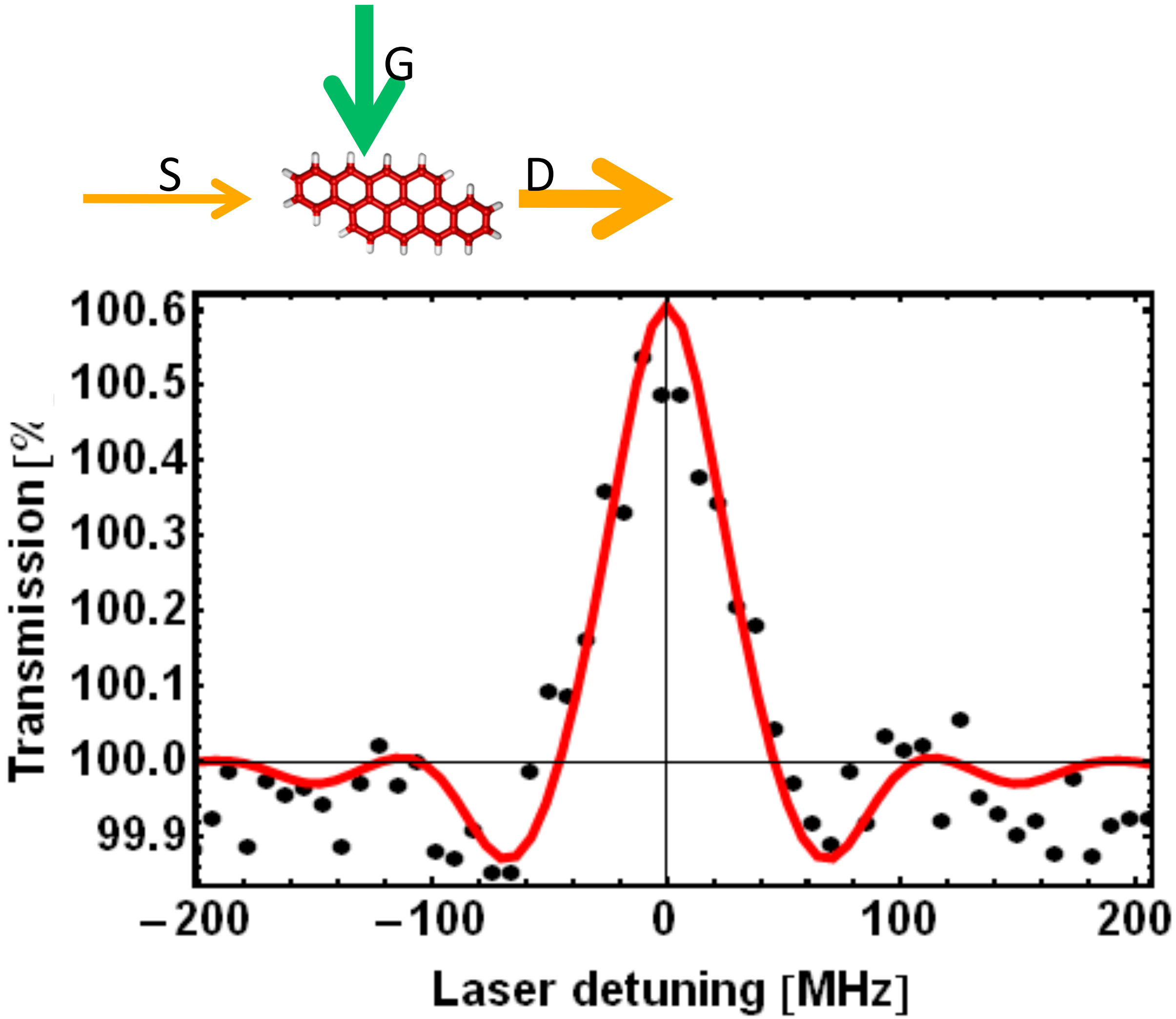
Pulsed excitation of single molecules and Rabi oscillations



I. Gerhardt, G. Wrigge, G. Zumofen, J. Hwang, A. Renn, V. Sandoghdar, *Phys. Rev. A*, **79**, 011402(R) (2009).

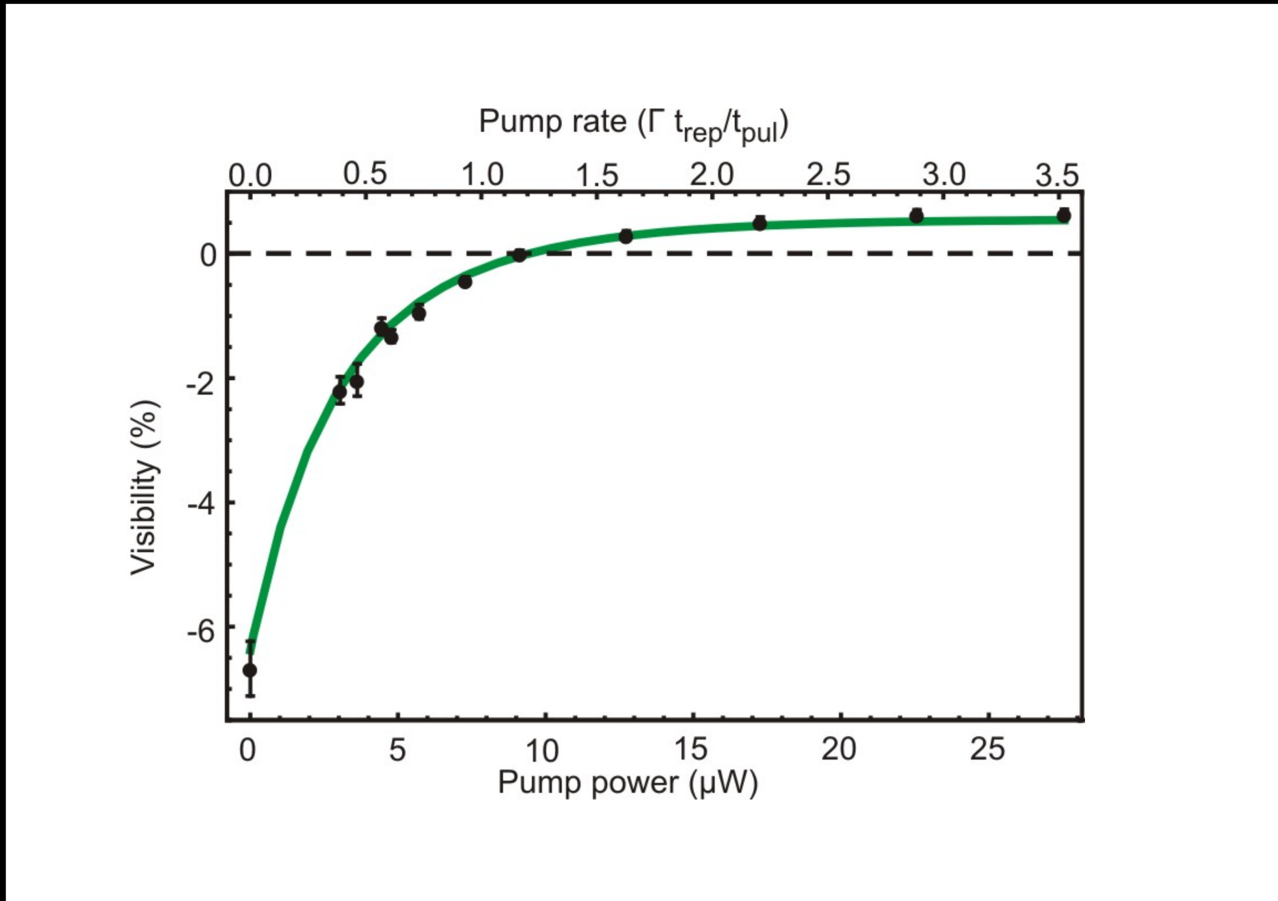
Amplification of light by a single molecule





J. Hwang, M. Pototschnig, R. Lettow, G. Zumofen, A. Renn, S. Götzinger, V. Sandoghdar, *Nature* **460**, 76 (2009).

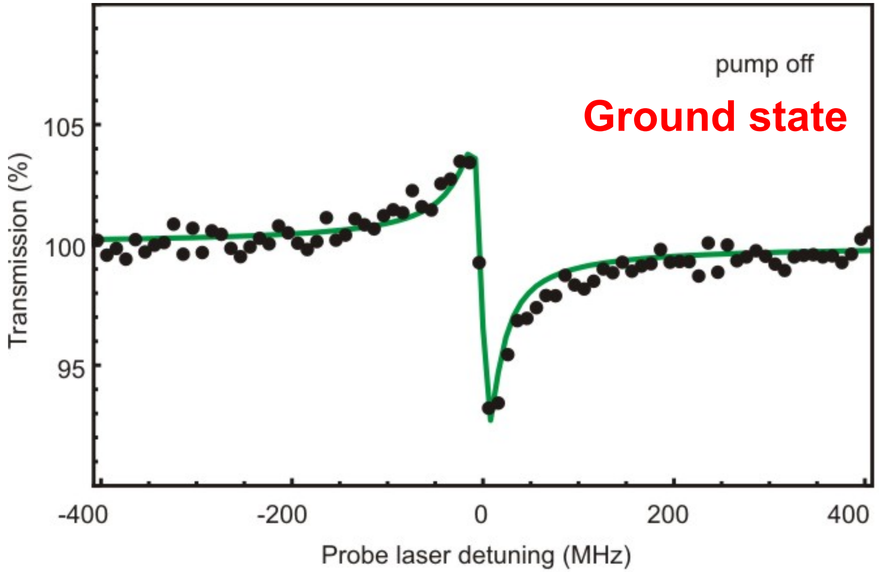
A single molecule as an optical transistor



J. Hwang, M. Pototschnig, R. Lettow, G. Zumofen, A. Renn, S. Götzinger, V. Sandoghdar, *Nature* **460**, 76 (2009).

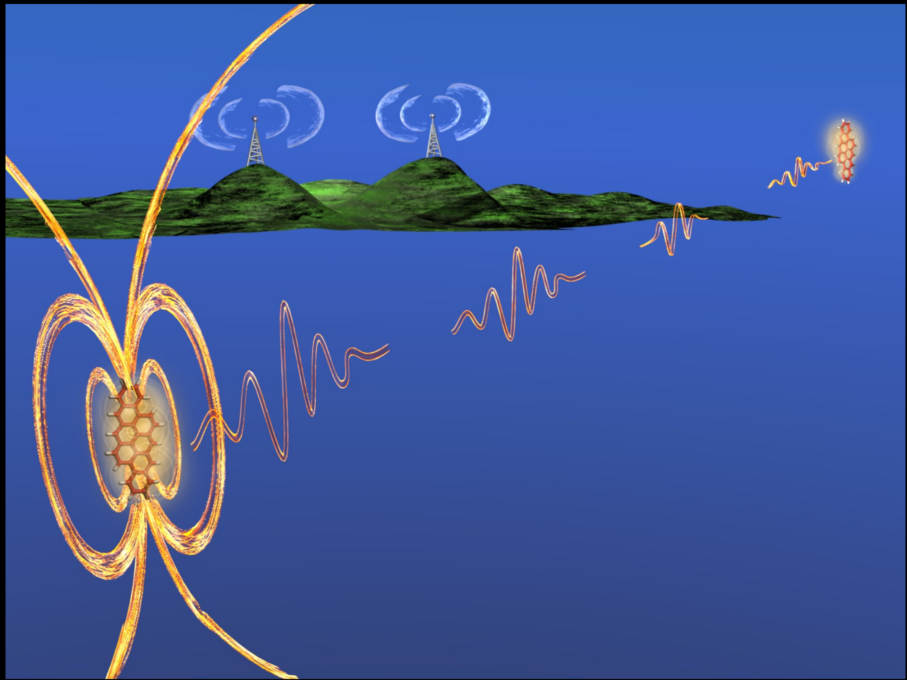
The coherent nature of stimulated emission

Adjust the phase to get a dispersive spectrum

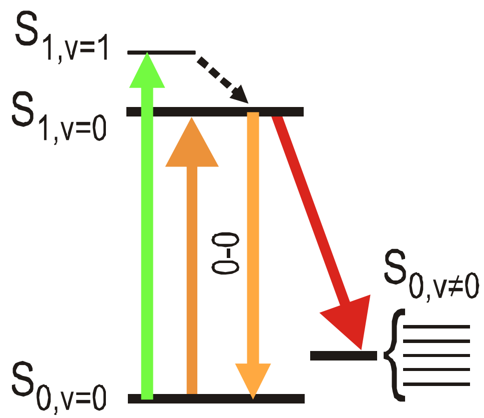


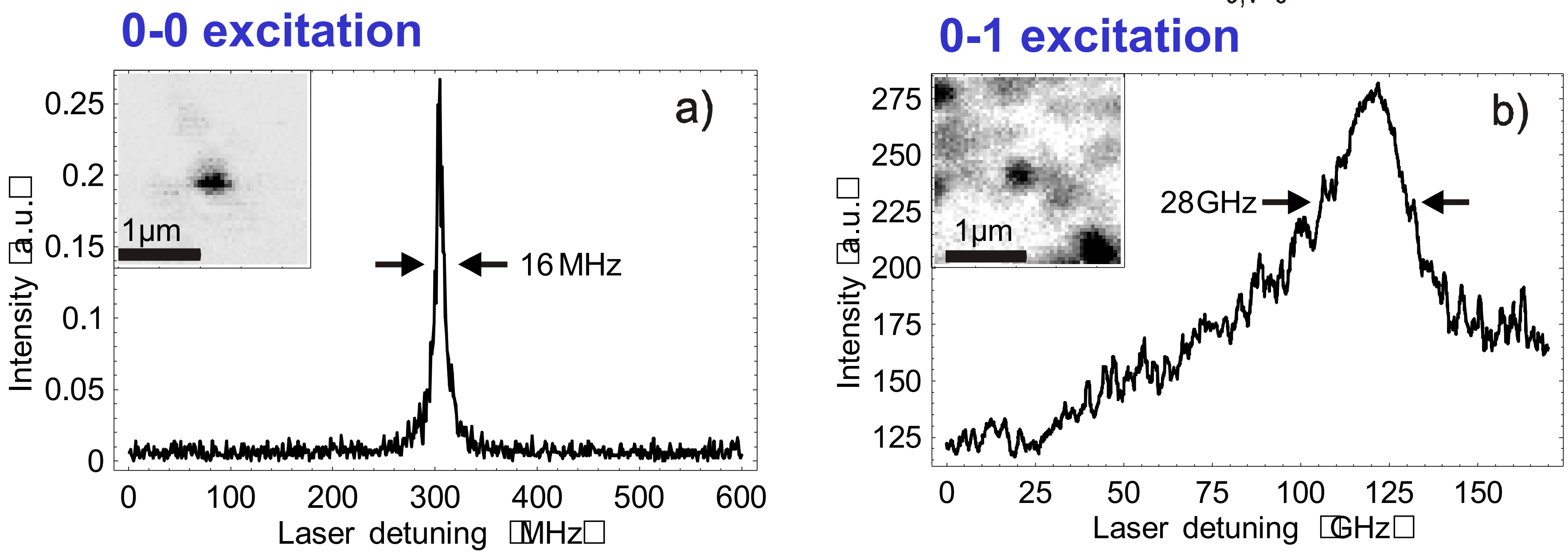
Far-field coupling of two single molecules

Spectroscopy with single photons



Collection of the lifetime-limited fluorescence



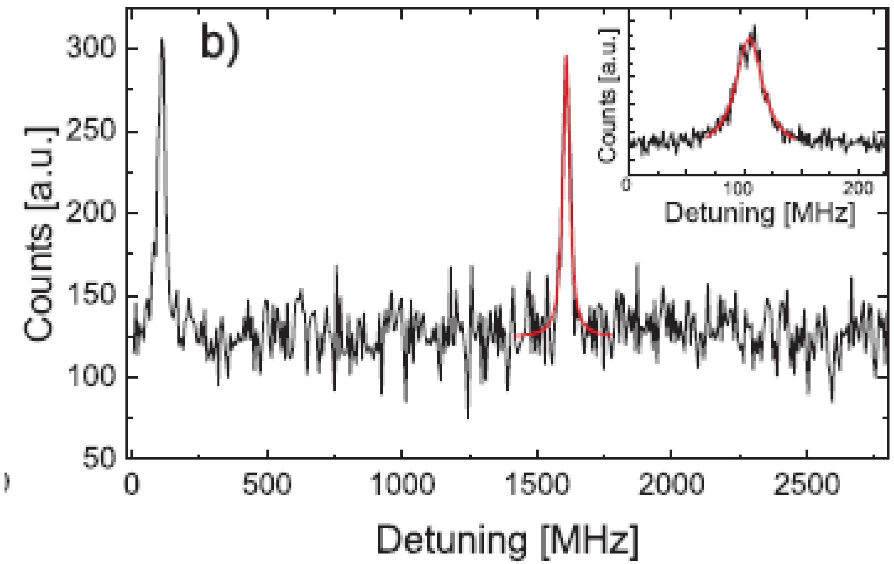


R. Lettow, et al., *Opt. Express* **15**, 15842 (2007).

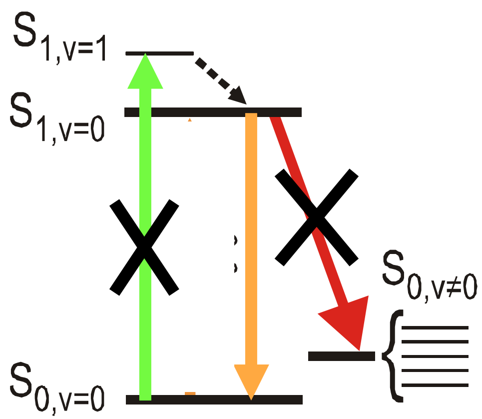
Sinlge molecule as a single-photon source

Narrow-band single photons

Narrow emission

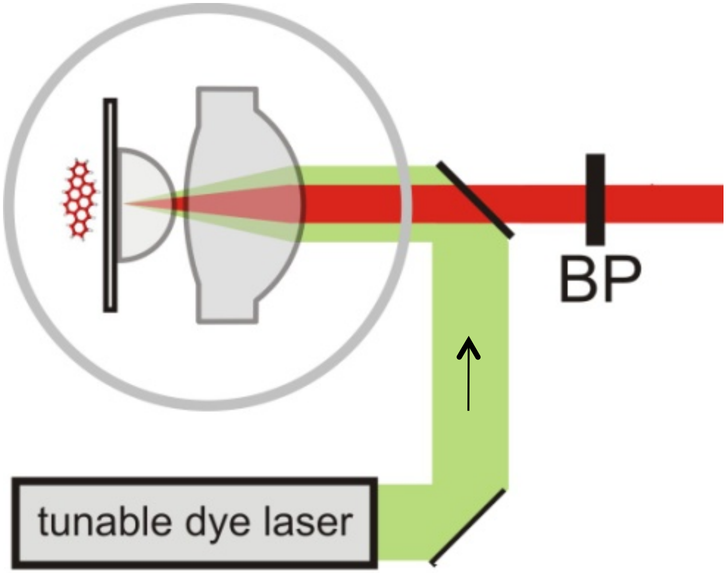


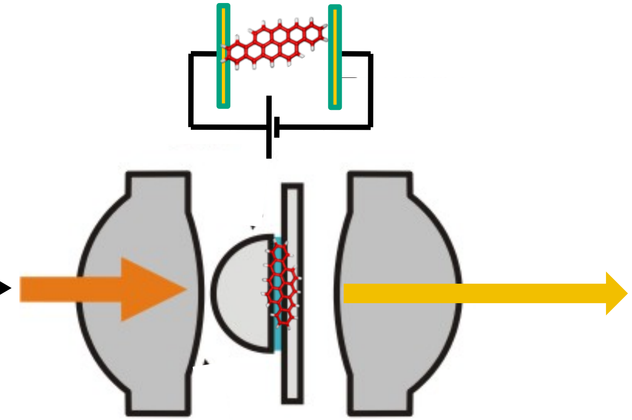
ZPL scanned with Fabry-Perot Cavity



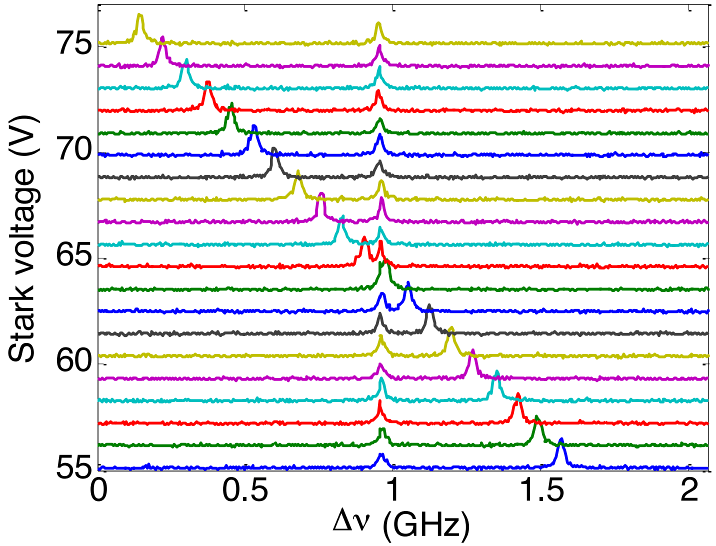
R. Lettow, V. Ahtee, R. Pfab, A. Renn, E. Ikonen, S. Götzinger, V. Sandoghdar
Opt. Express **15**, 15842 (2007).

Exciting a single molecule with a single-photon source





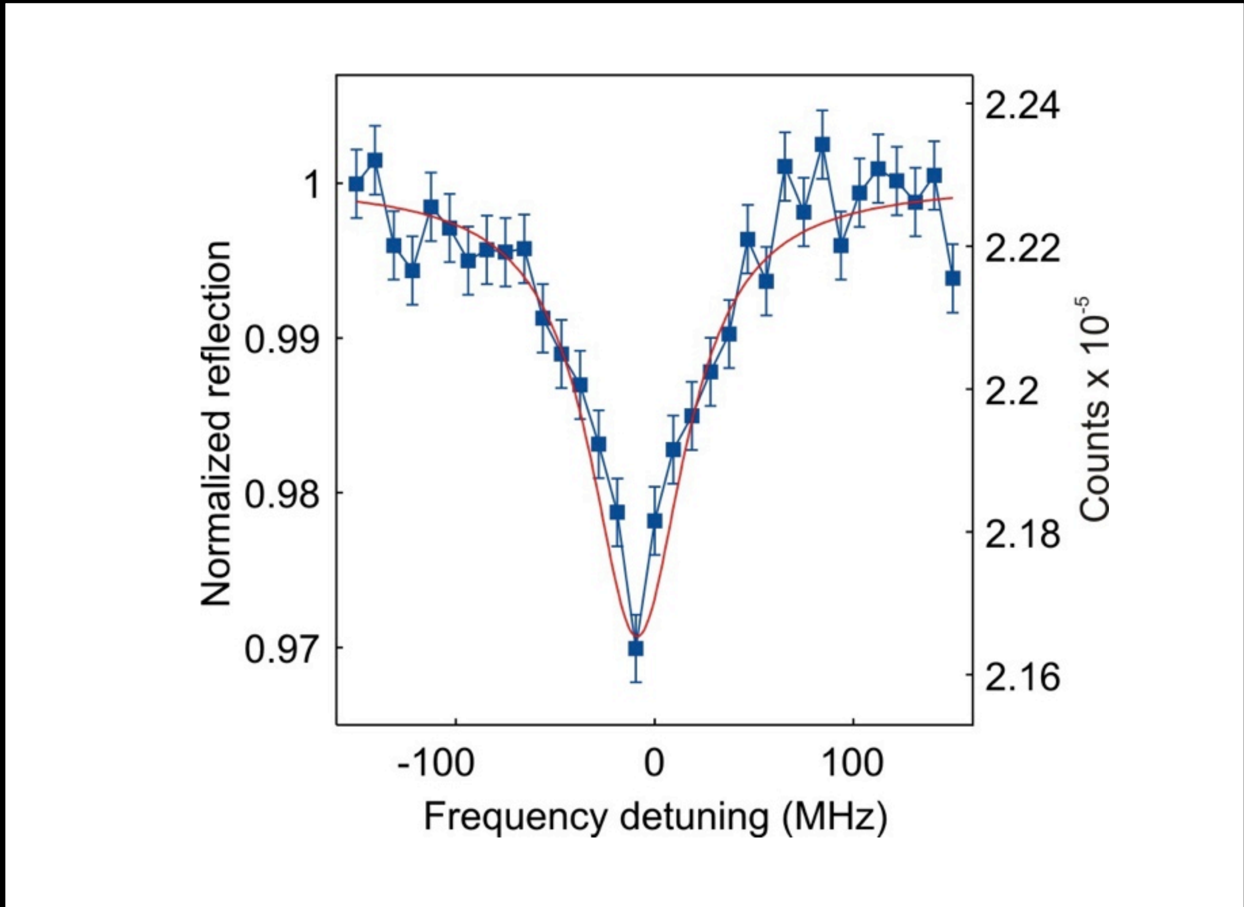
Tunability via Stark shift



- Lifetime-limited photons
 'gas-phase-like in the solid state'
- Cryogenic temperature ⇒ no bleaching
- High brightness, count rates > 1 Mcps

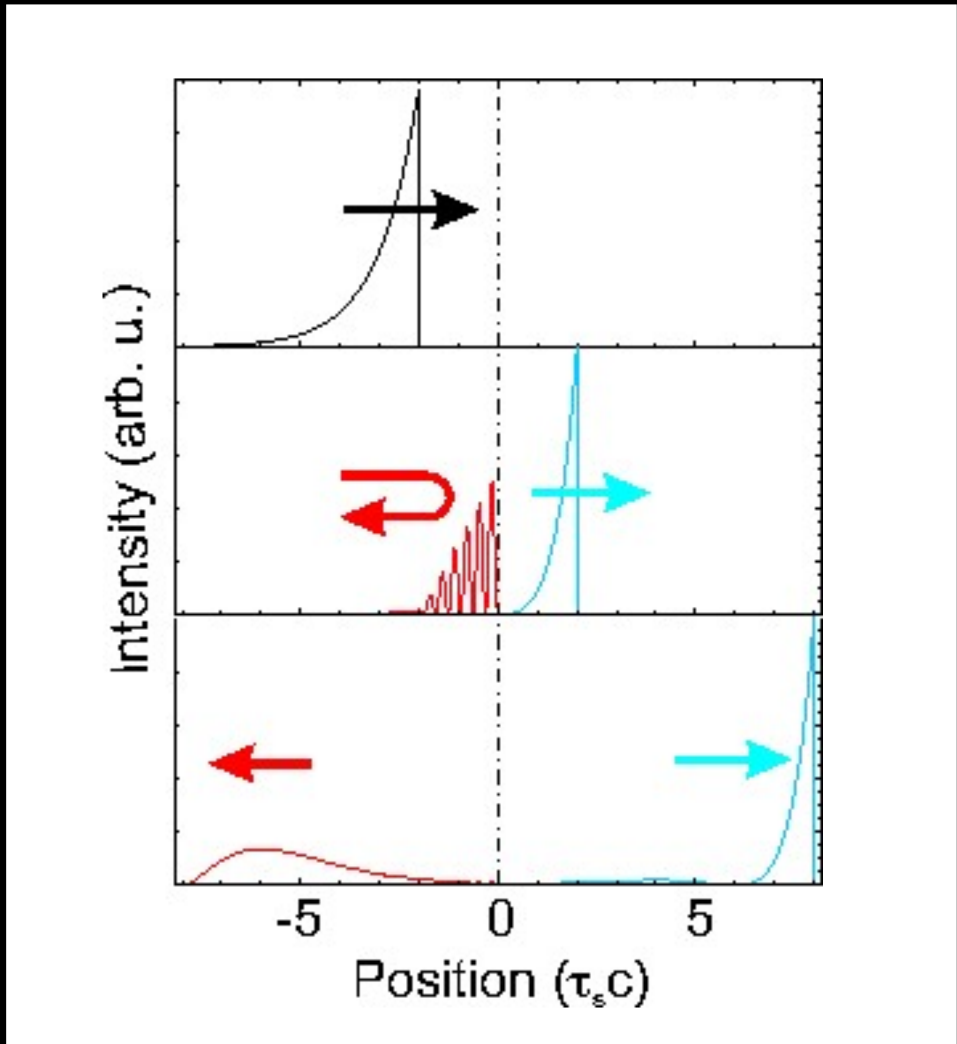
R. Lettow, V. Ahtee, R. Pfab, A. Renn, E. Ikonen, S. Götzinger, V. Sandoghdar, *Opt. Express* **15**, 15842 (2007).

Extinction spectrum of a single molecule using single photons from another molecule



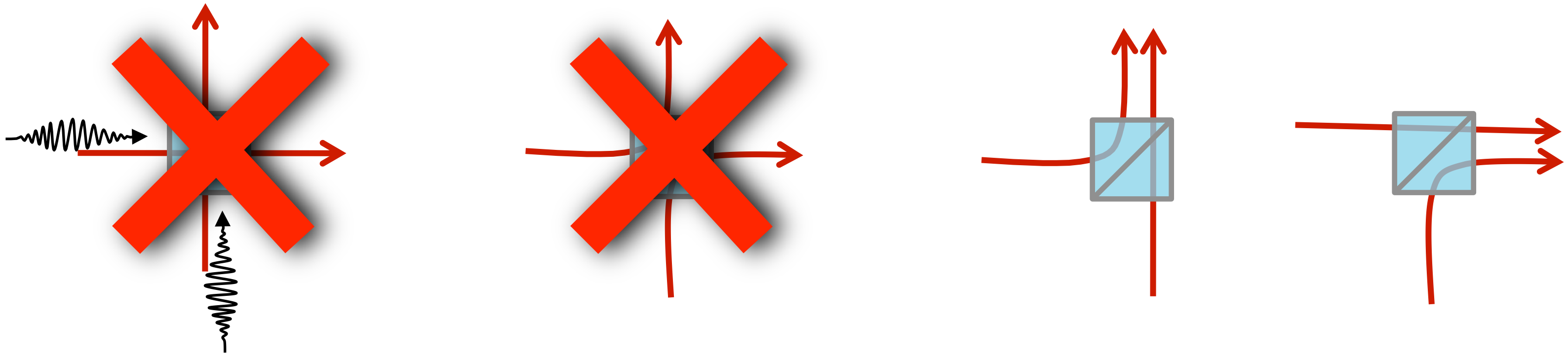
Y. Rezus, S. Walt, R. Lettow, A. Renn, G. Zumofen, S. Götzinger, V. Sandoghdar
Phys. Rev. Lett. **108**, 093601 (2012).

Bouncing of a photon from an atom



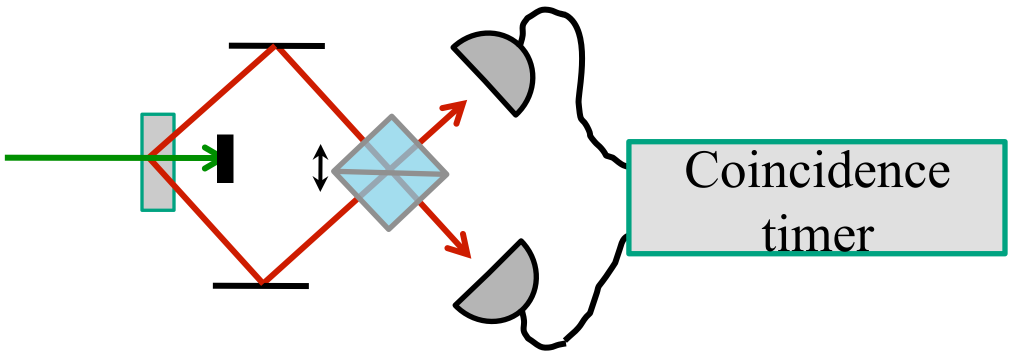
Y. Rezus, S. Walt, R. Lettow, A. Renn, G. Zumofen, S. Götzinger, V. Sandoghdar
Phys. Rev. Lett. **108**, 093601 (2012).

Two-photon interference

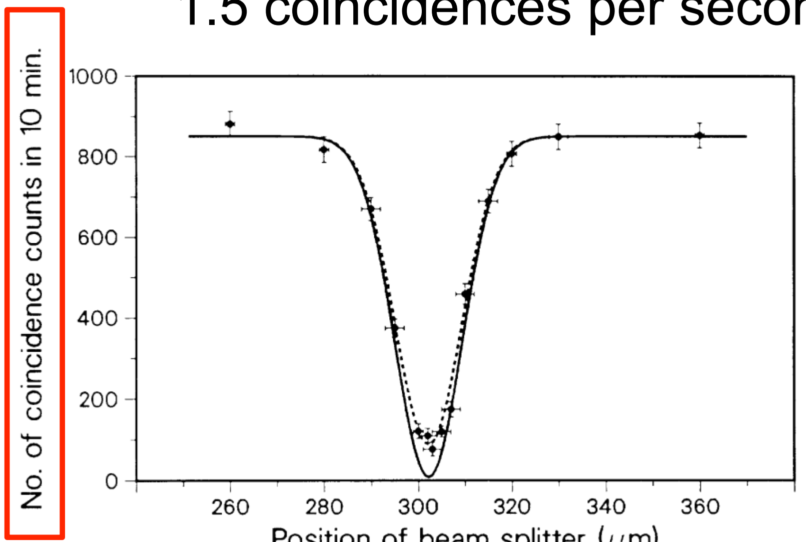


Original HOM experiment

•Originally the two photons were generated by Spontaneous Parametric Down Conversion (SPDC) in a nonlinear crystal



1.5 coincidences per second!



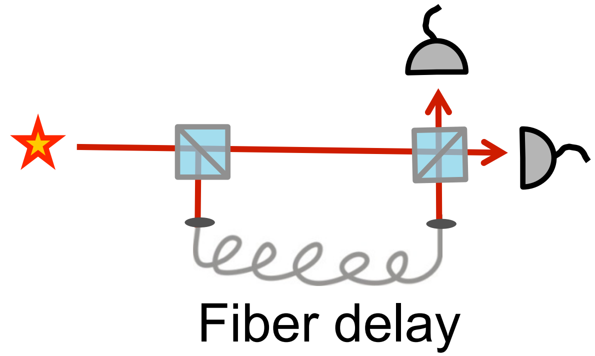
•Indistinguishable photons:

- Frequency
- Polarization
- Coherence length
- Spatial mode

TPI with true single-photon sources

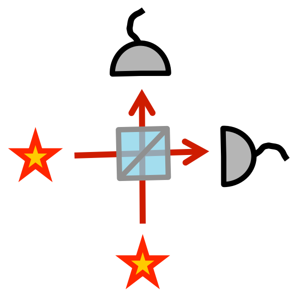
- Two-photon interference with one single-photon source

  Molecules: Zumbusch
  Gas-phase ions: Rempe

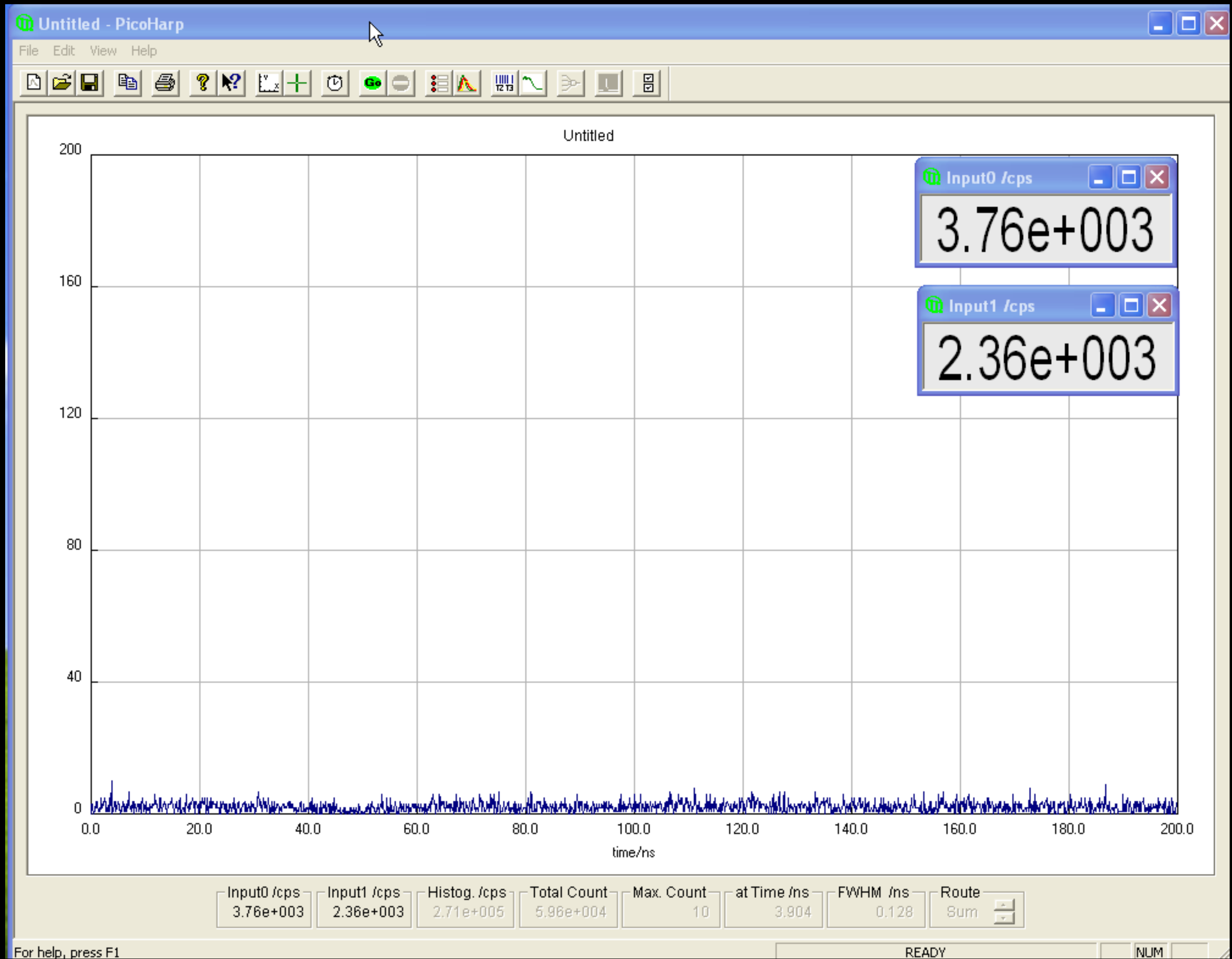


- Two-photon interference with independent single-photon sources

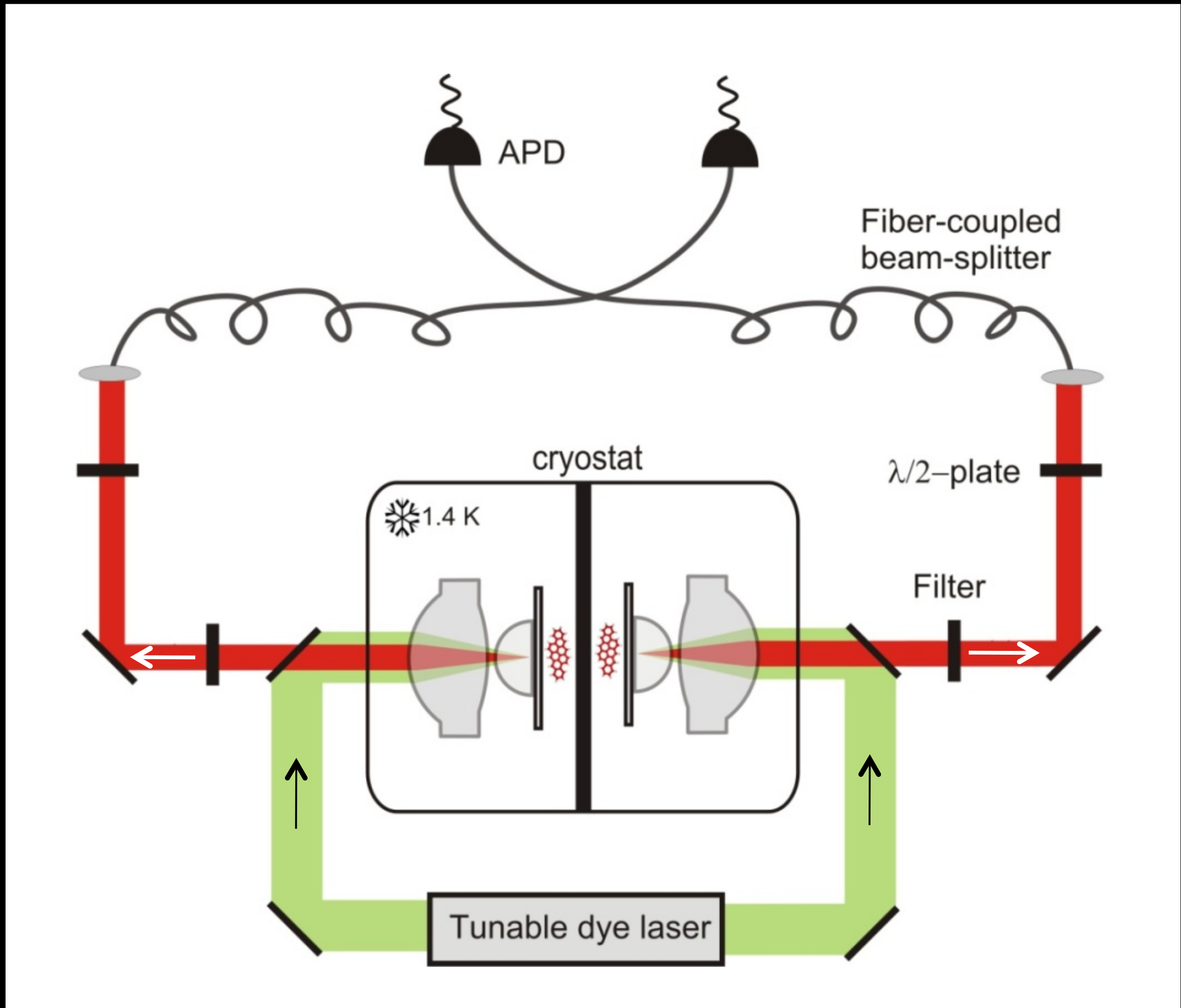
  Gas-phase atoms and ions: Monroe, Grangier



Very bright source:
Fast antibunching measurement



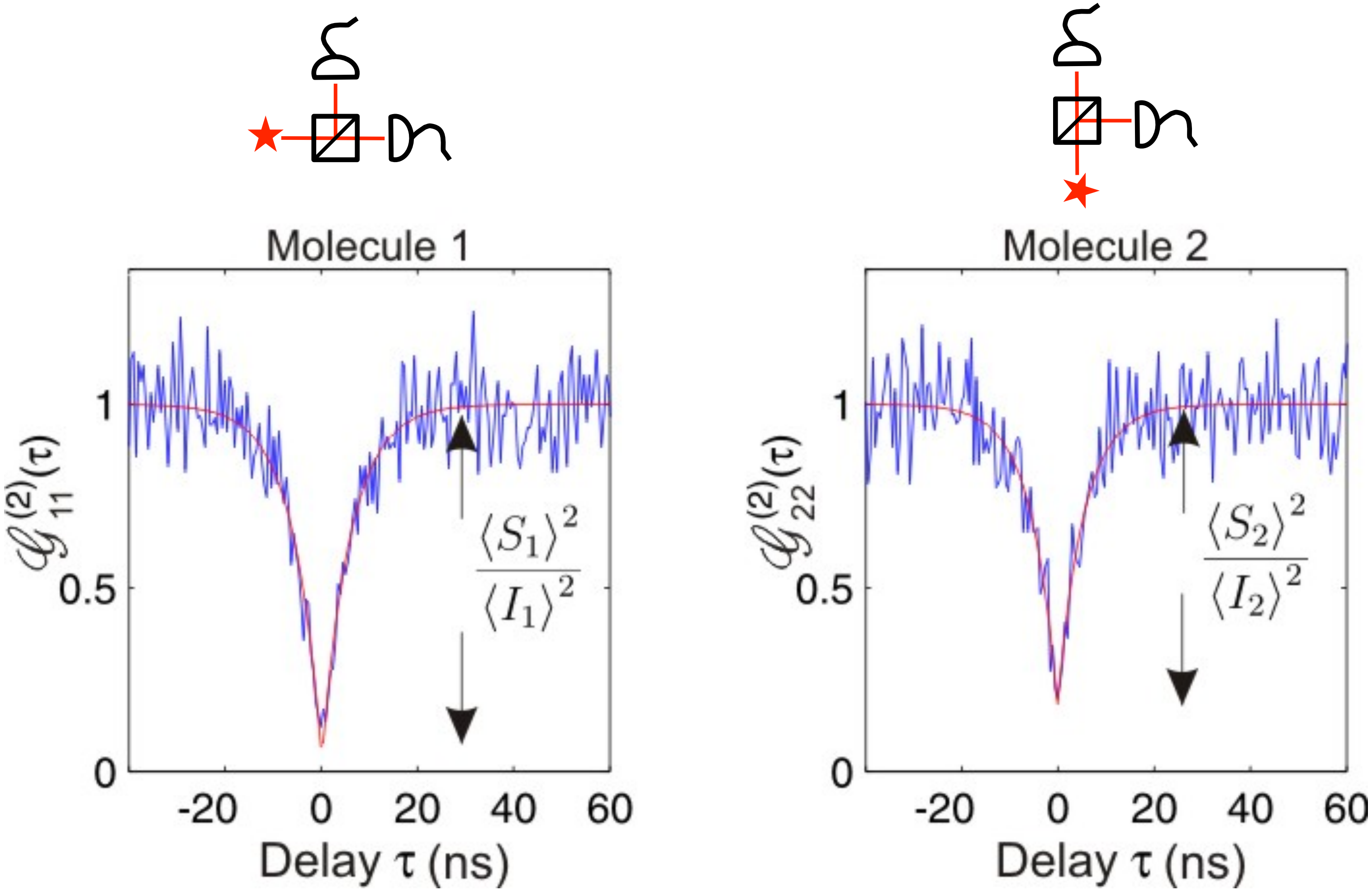
Hong-Ou-Mandel Two-Photon Interference



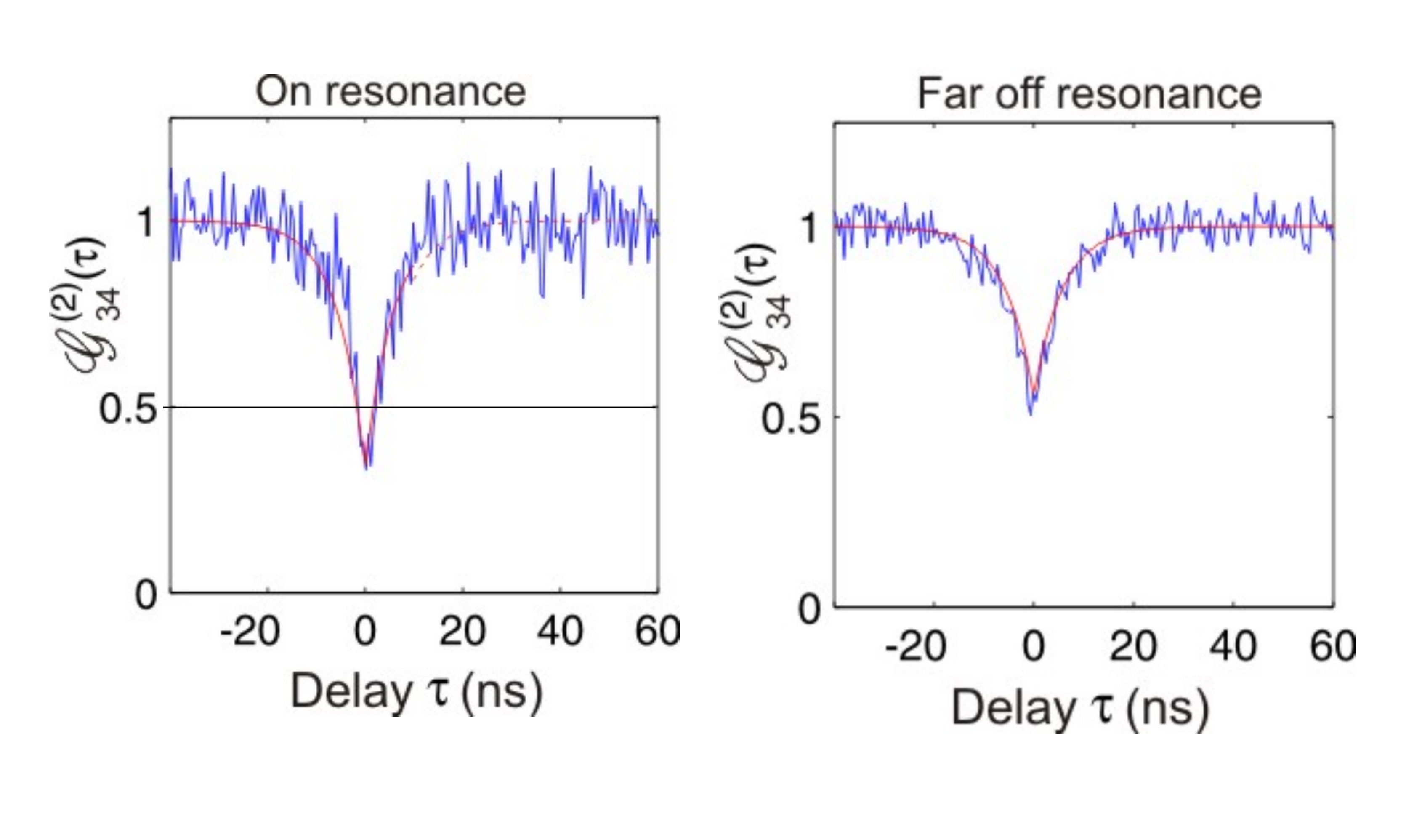
R. Lettow, V. Ahtee, R. Pfab, A. Renn, E. Ikonen, S. Götzinger, V. Sandoghdar
Opt. Express **15**, 15842 (2007).

Checking that each arm is antibunched

Continuous-wave excitation

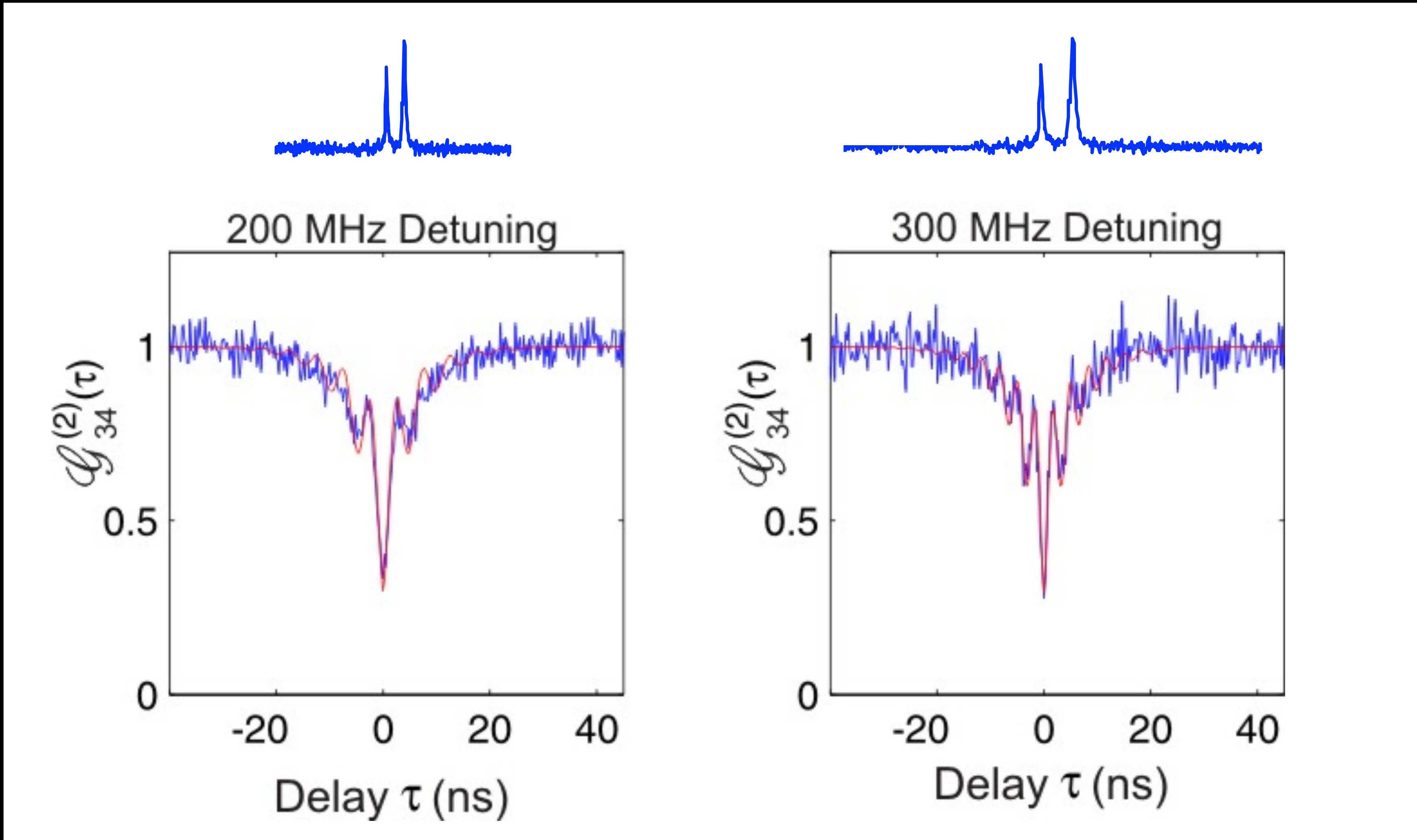


Proof of two-photon interference



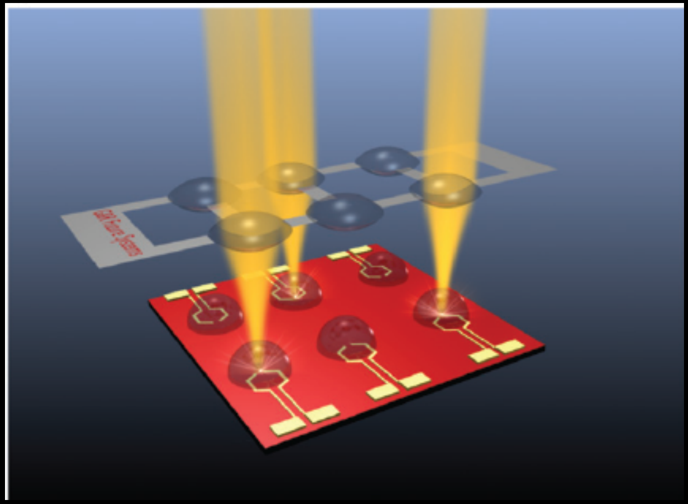
R. Lettow, Y. Rezus, G. Zumofen, A. Renn, E. Ikonen, S. Götzinger, V. Sandoghdar, *Phys. Rev. Lett*. **104**, 123605 (2010).

Controlled tuning of the photons

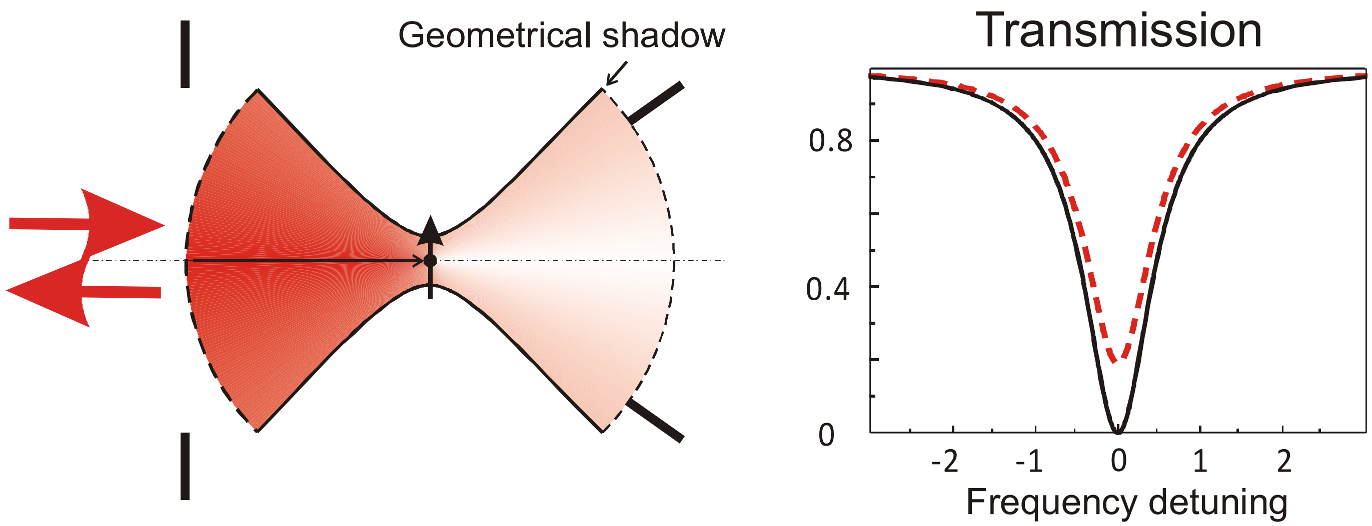


R. Lettow, Y. Rezus, G. Zumofen, A. Renn, E. Ikonen, S. Götzinger, V. Sandoghdar, *Phys. Rev. Lett*. **104**, 123605 (2010).

From two photons to few photons



Perfect reflection of light by a single oscillating dipole

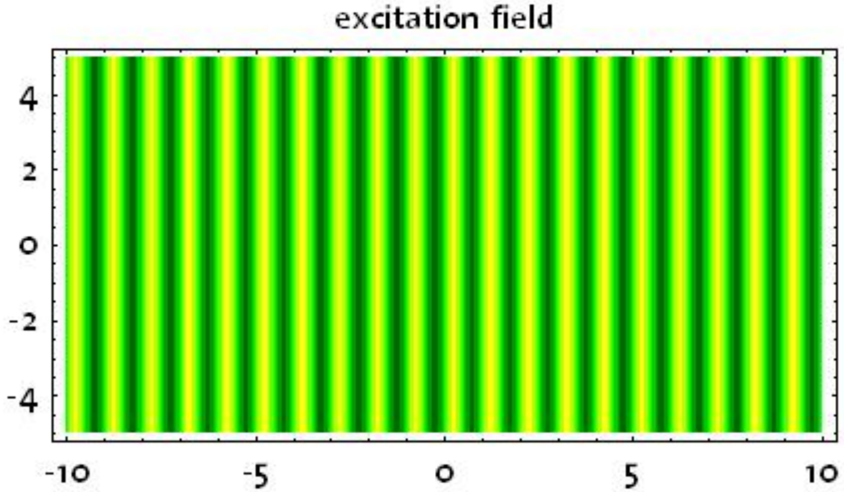


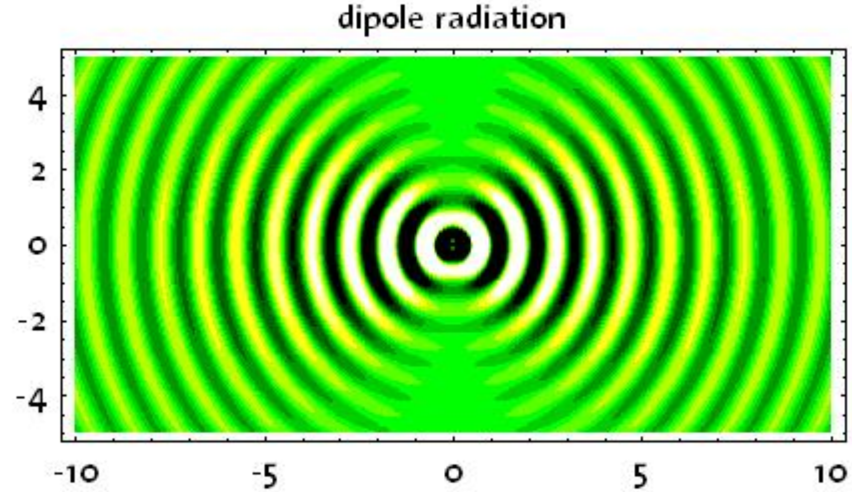
PHYSICAL REVIEW LETTERS **101,** 180404 (2008)

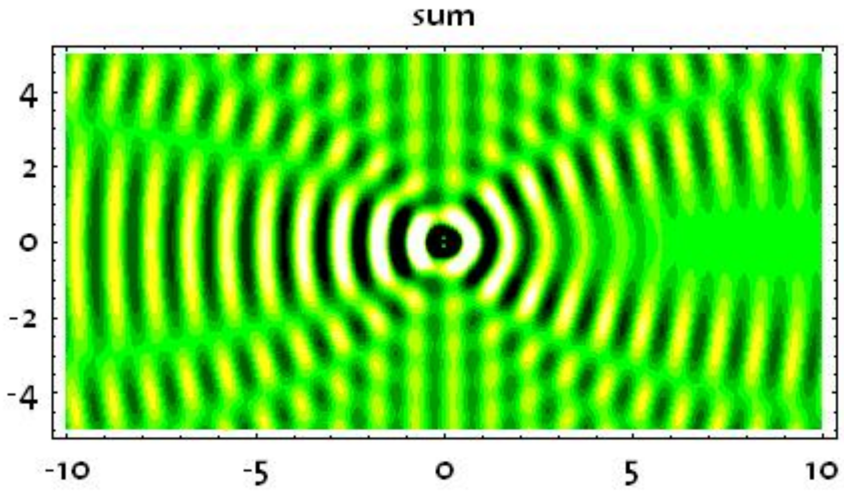
Perfect Reflection of Light by an Oscillating Dipole

G. Zumofen, N. M. Mojarad, V. Sandoghdar, and M. Agio
Nano-Optics Group, Laboratory of Physical Chemistry, ETH Zurich, CH-8093 Zurich, Switzerland
(Received 19 May 2008)

The extinction signal is due to interference







$$I_{total} = |E_{inc}|^2 + |E_{sca}|^2$$

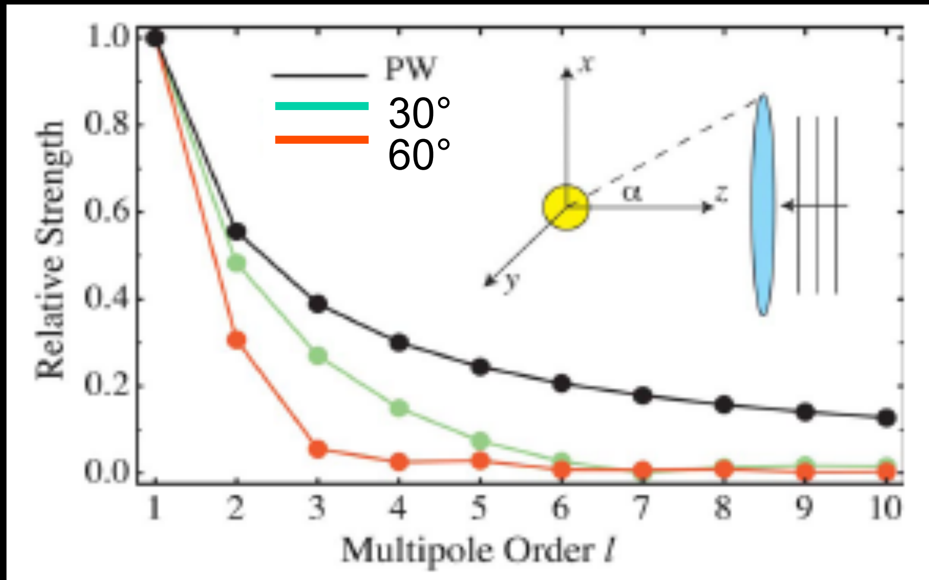
$$+2Re(E_{inc}E^*_{sca})$$

↓

extinction

Complete extinction in the forward direction, hence the definition of the absorption cross section (optical theorem)

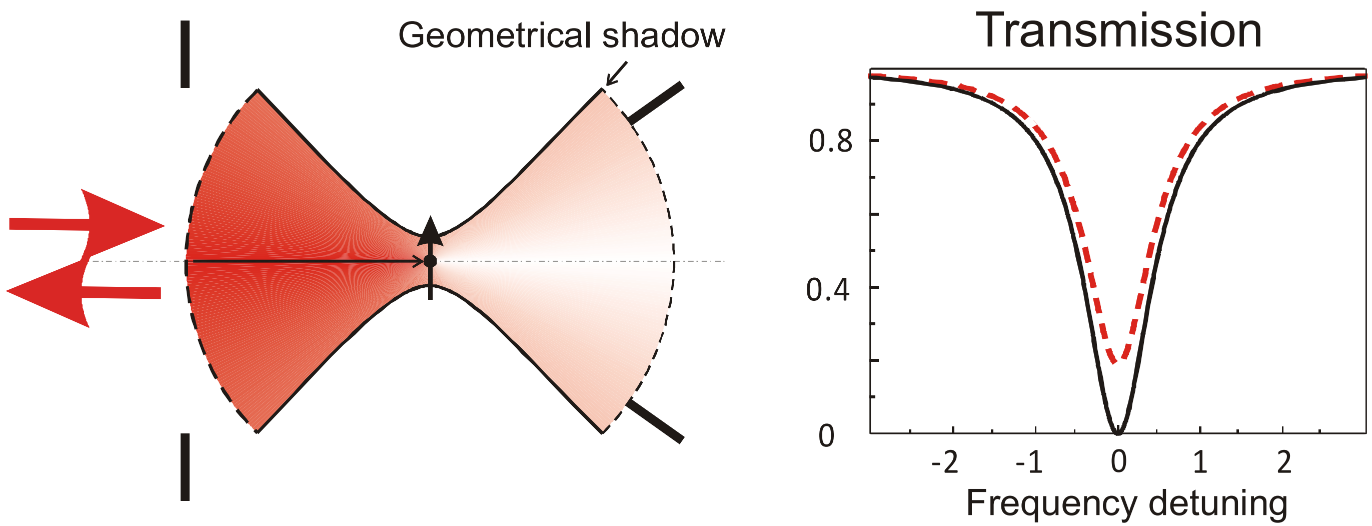
Multipolar content of a focused beam



A tightly focused beam matches the dipolar radiation well

N. M. Mojarad, V. Sandoghdar, M. Agio, *JOSA B* **25**, 651 (2008).

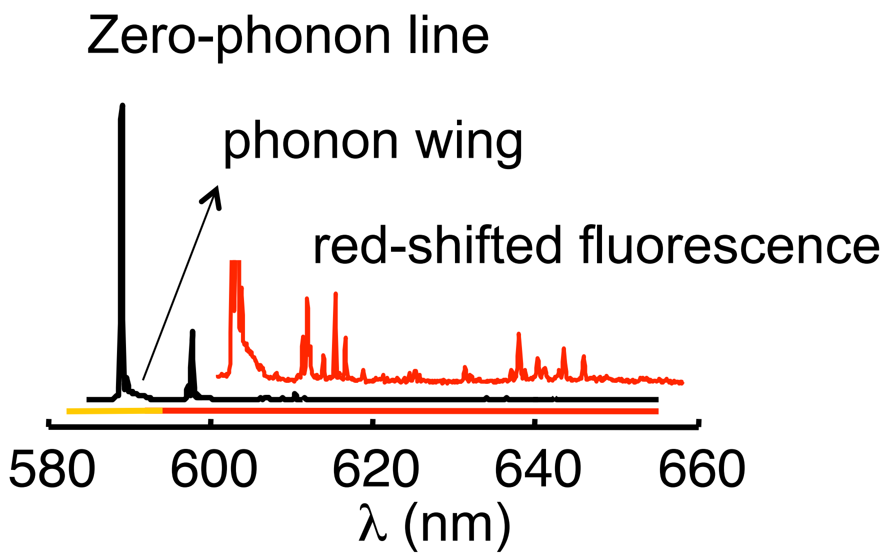
Perfect reflection of light by a single oscillating dipole

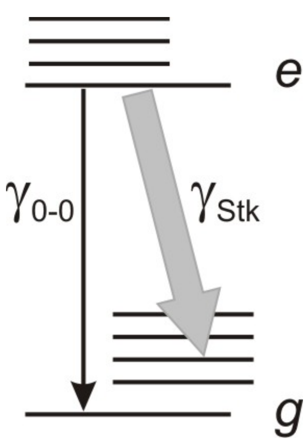


PHYSICAL REVIEW LETTERS **101,** 180404 (2008)

Perfect Reflection of Light by an Oscillating Dipole

G. Zumofen, N. M. Mojarad, V. Sandoghdar, and M. Agio
Nano-Optics Group, Laboratory of Physical Chemistry, ETH Zurich, CH-8093 Zurich, Switzerland
(Received 19 May 2008)





$$\sigma_{ext} = \frac{3\lambda^2}{2\pi}\frac{\gamma_0}{\gamma_{hom}}\alpha$$

Mode matching:

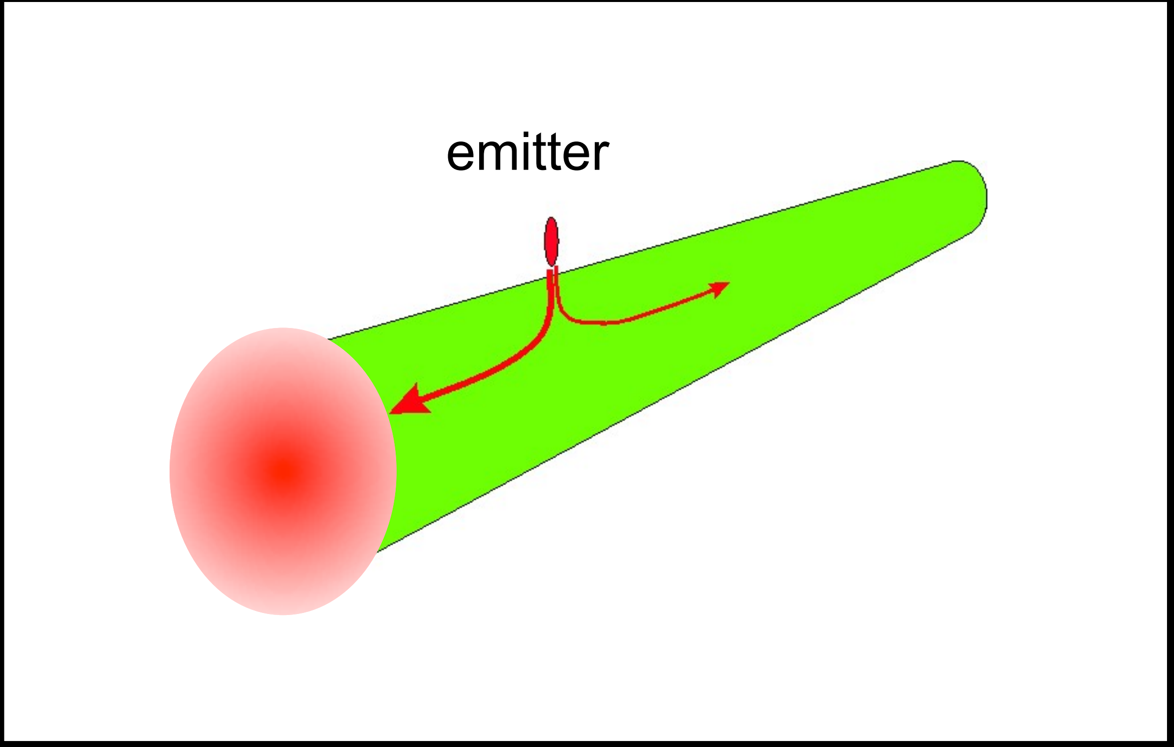
Match the light to the emitter

Match the emitter to the light

Coupling to nanofibers

Mode conversion: dipolar radiation to something like a TEM mode

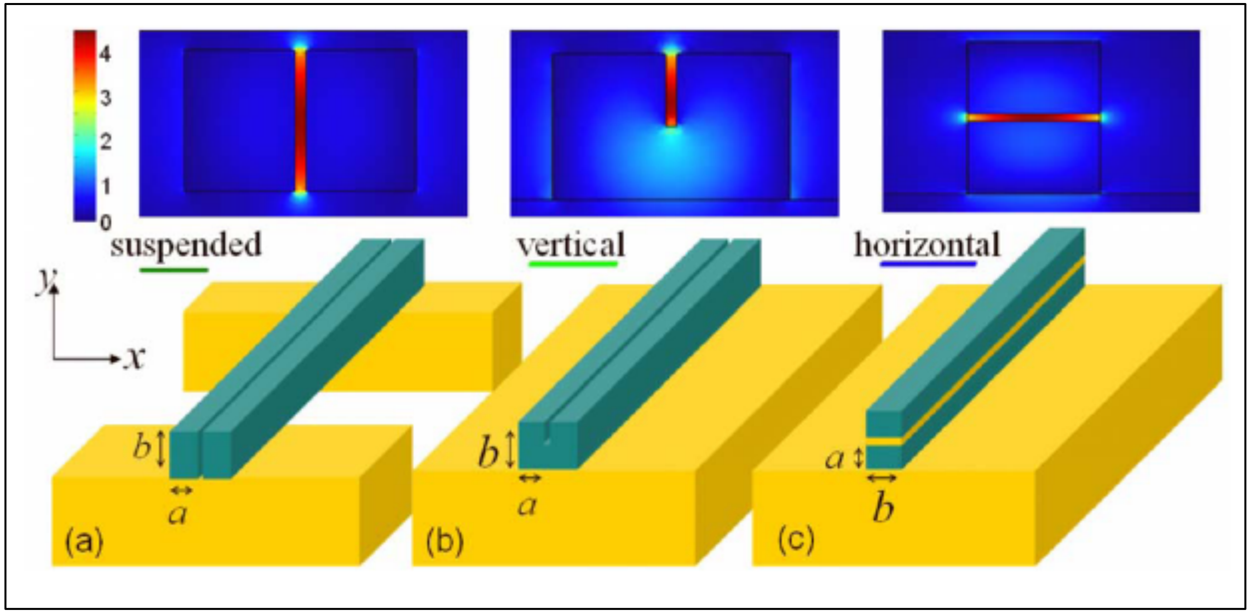
The higher the refractive index, the larger the coupling



PHYSICAL REVIEW A **80**, 011810(R) (2009)

Broadband waveguide QED system on a chip

Qimin Quan, Irfan Bulu, and Marko Lončar



Broadband enhancement of light emission in silicon slot waveguides

Young Chul Jun[1], Ryan M. Briggs[2], Harry A. Atwater[2], and Mark L. Brongersma[1*]

27 April 2009 / Vol. 17, No. 9 / OPTICS EXPRESS 7479

nature Vol 450|15 November 2007|

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

A. V. Akimov[1,4]*, A. Mukherjee[1]*, C. L. Yu[2]*, D. E. Chang[1], A. S. Zibrov[1,4], P. R. Hemmer[3], H. Park[1,2] & M. D. Lukin[1]

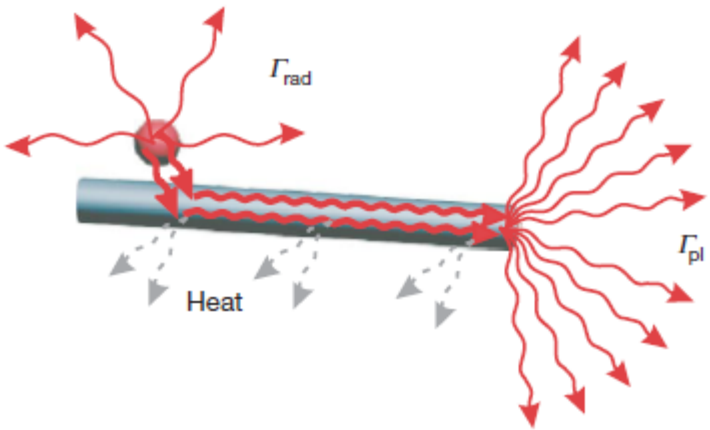


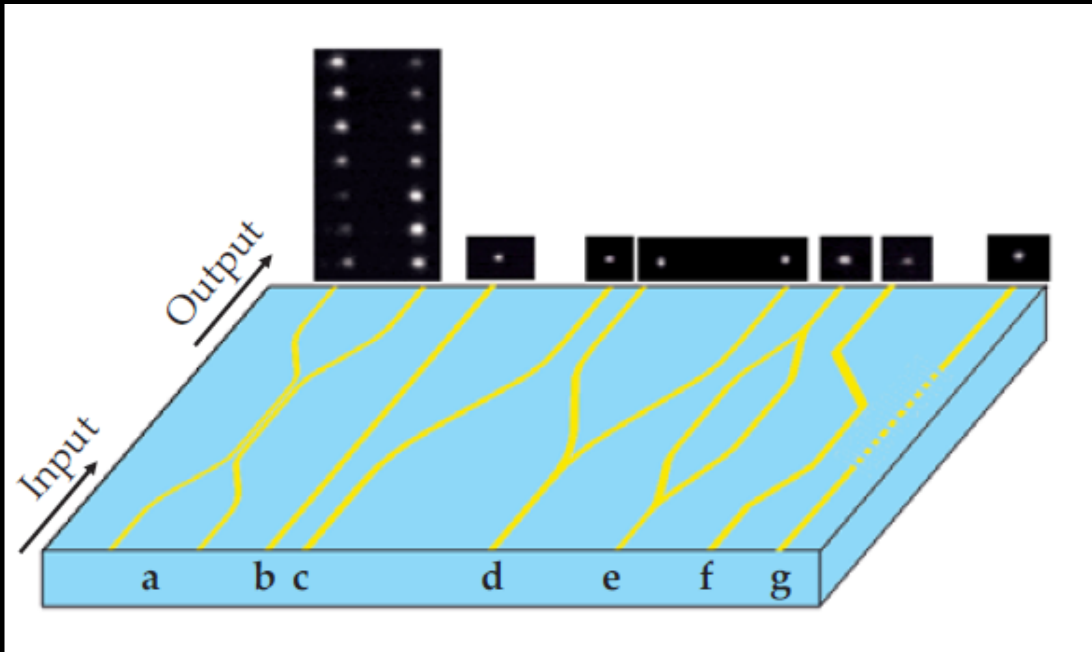
Figure 3 | Demonstration of single surface plasmon generation.

Surface-plasmon circuitry

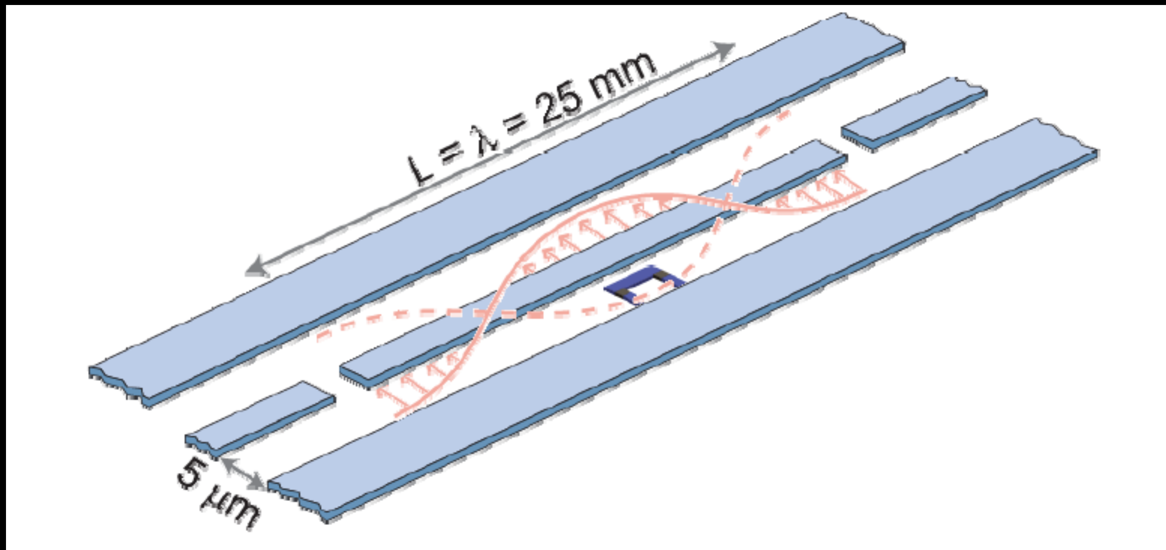
Thomas W. Ebbesen, Cyriaque Genet, and Sergey I. Bozhevolnyi

www.physicstoday.org
physics today
May 2008

feature article

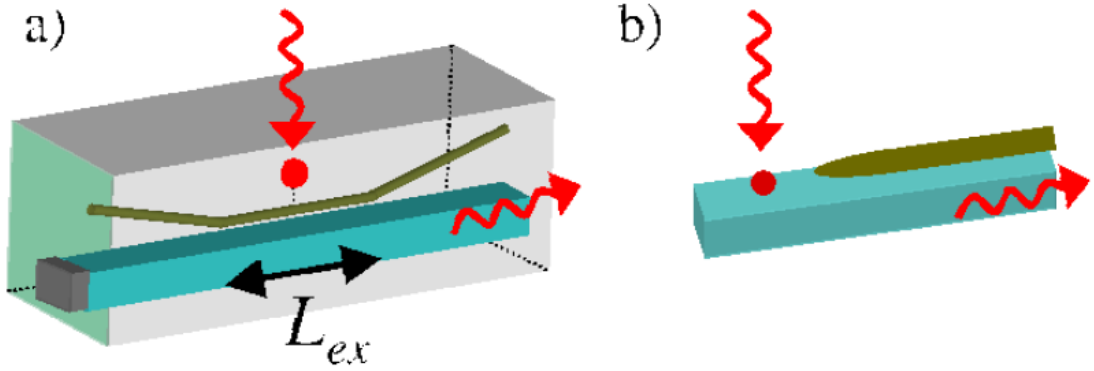


Similarities to circuit QED



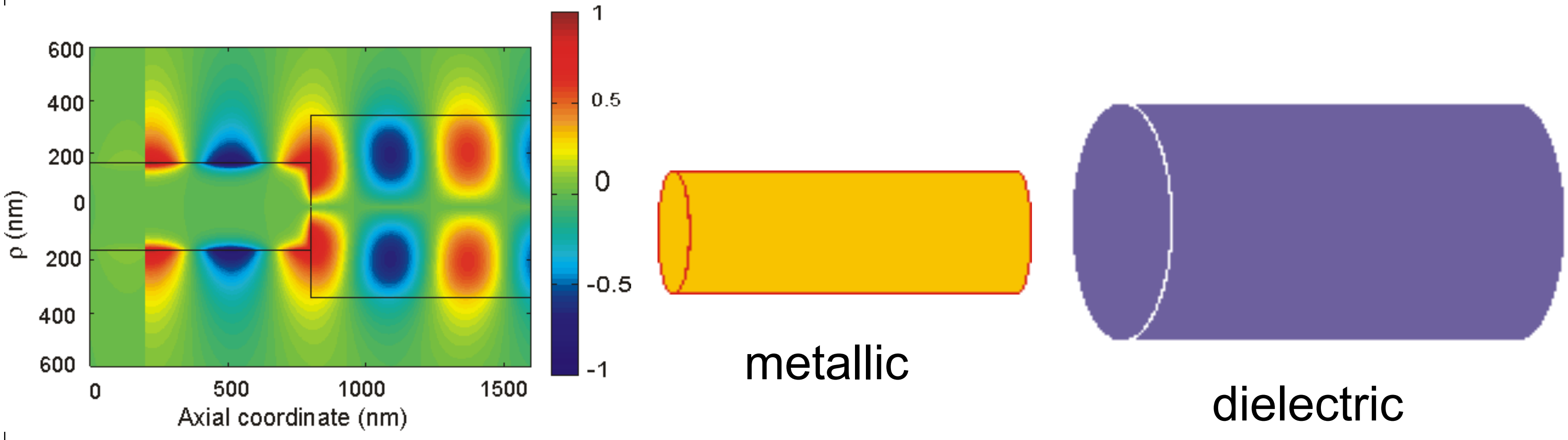
Quantum Optics with Surface Plasmons

D. E. Chang,[1] A. S. Sørensen,[2] P. R. Hemmer,[1,3] and M. D. Lukin[1]



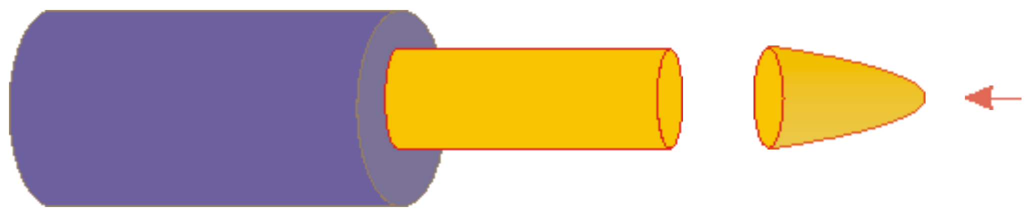
>95% coupling of plasmonic and dielectric channels

broadband

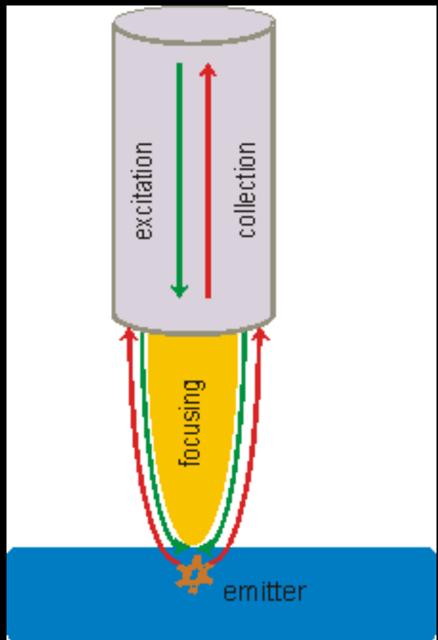


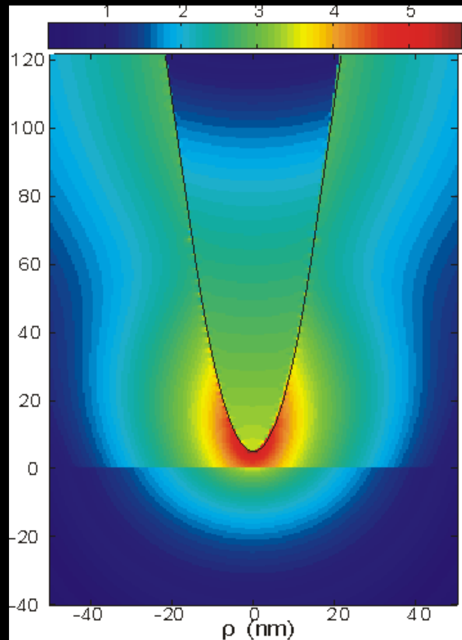
X. Chen, V. Sandoghdar, M. Agio, *Nano Lett.*, **9**, 3756 (2009).

… and super efficient couplig to a single emitter



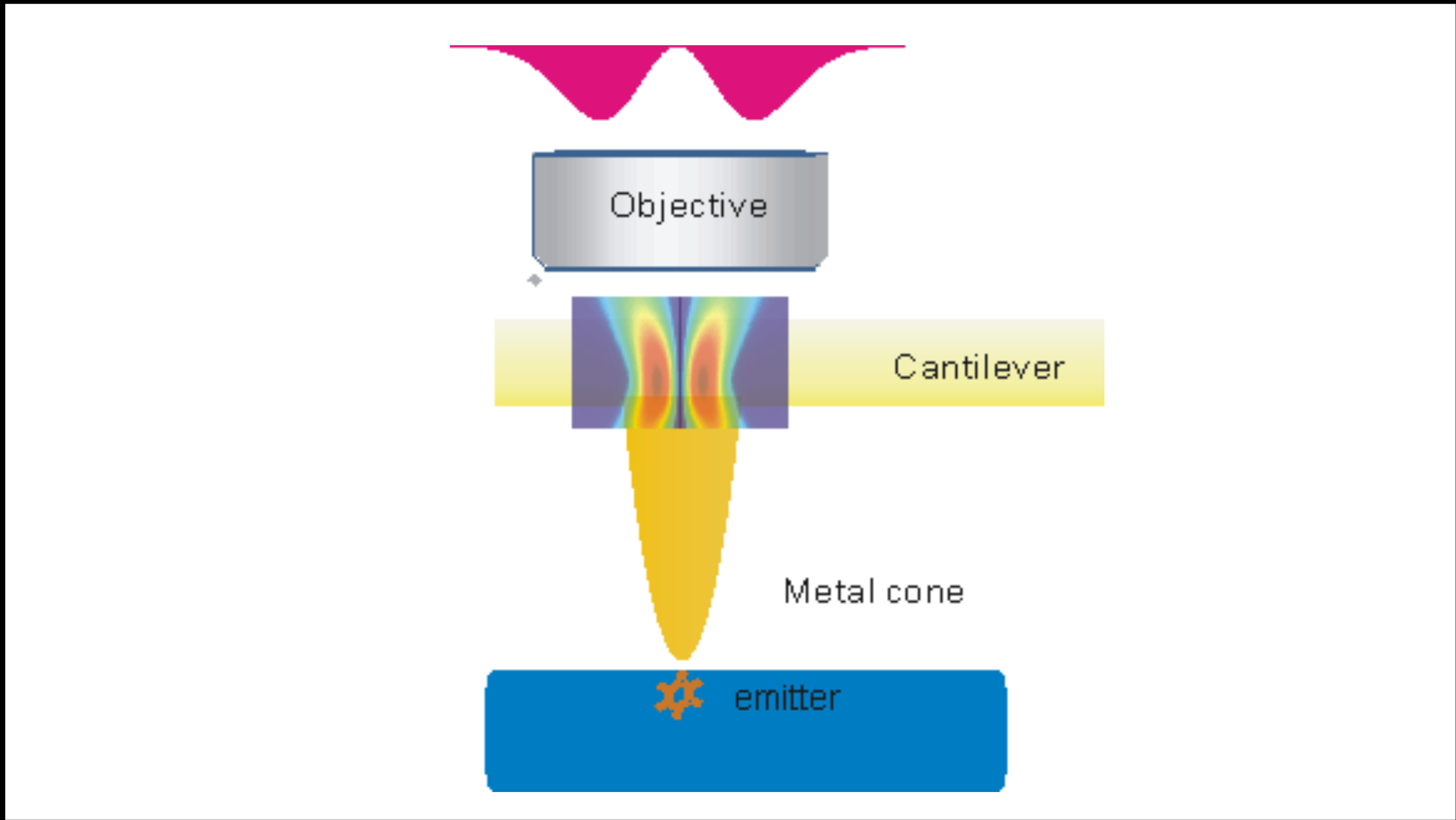
X.-W. Chen, V. Sandoghdar, M. Agio, Opt. Express, Focus issue: Unconventional Polarization States of Light, 18, 10878 (2010)





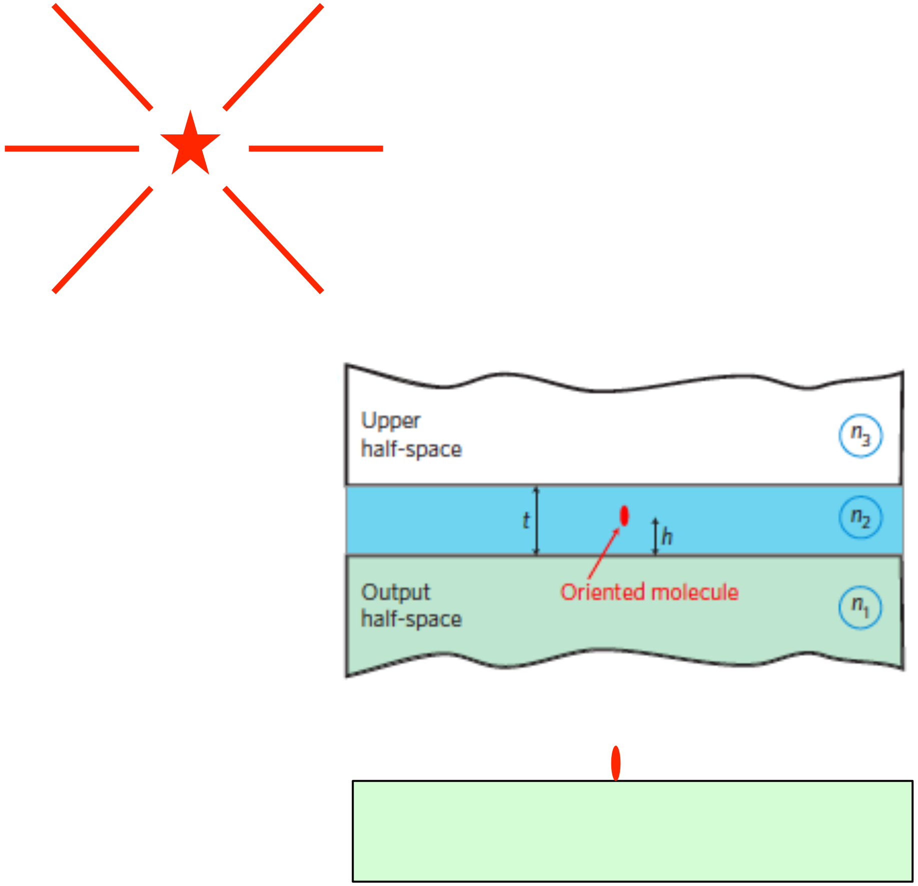
M.I. Stockman, Phys. Rev. Lett. 93, 137404 (2004), *and many others*

High-throughput high resolution SNOM



X.-W. Chen, V. Sandoghdar, M. Agio, *Opt. Express*, **18**, 10878 (2010);
Focus issue: Unconventional Polarization States of Light.

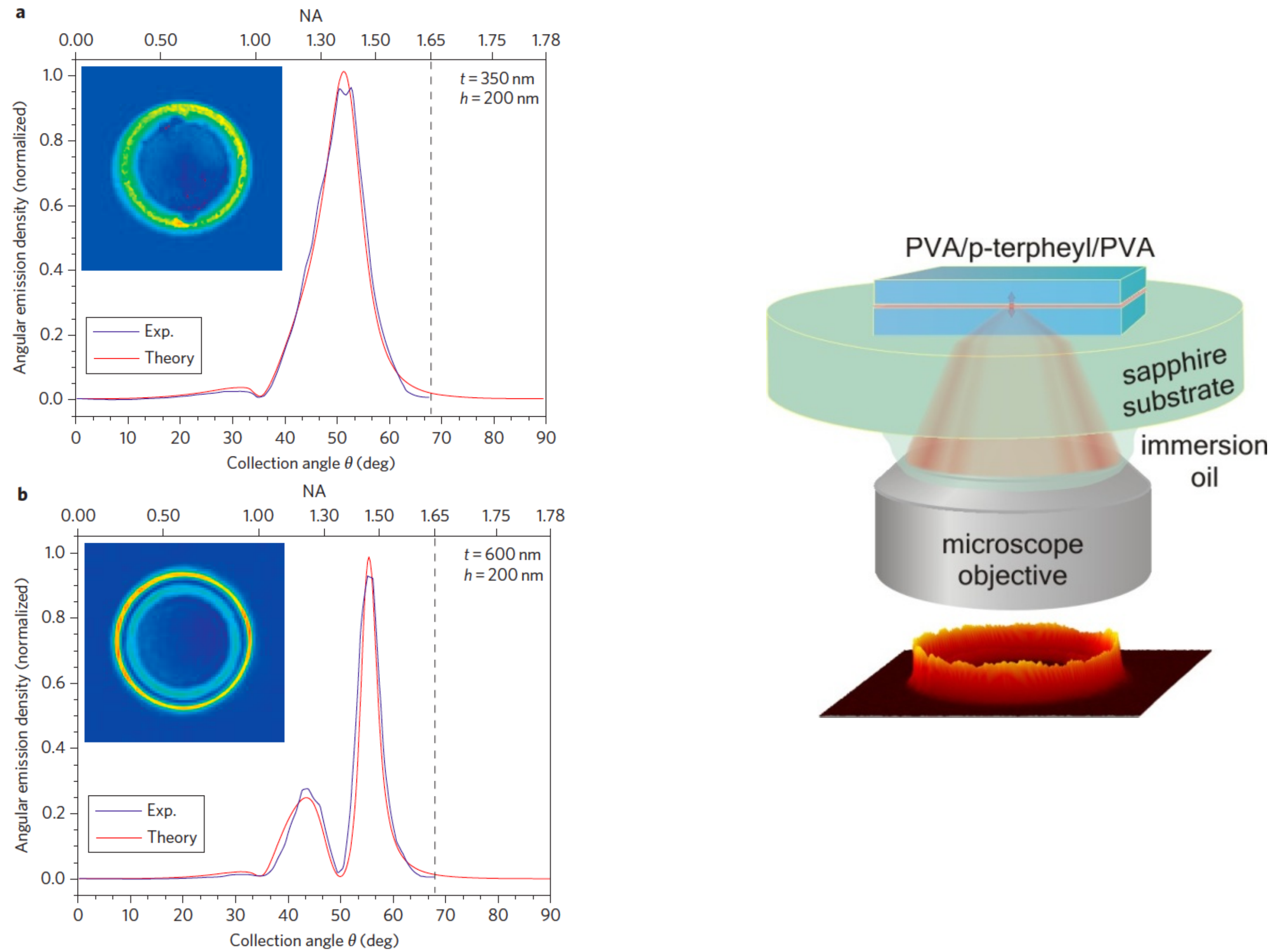
A planar dielectric antenna: 96% collection efficiency of single photons



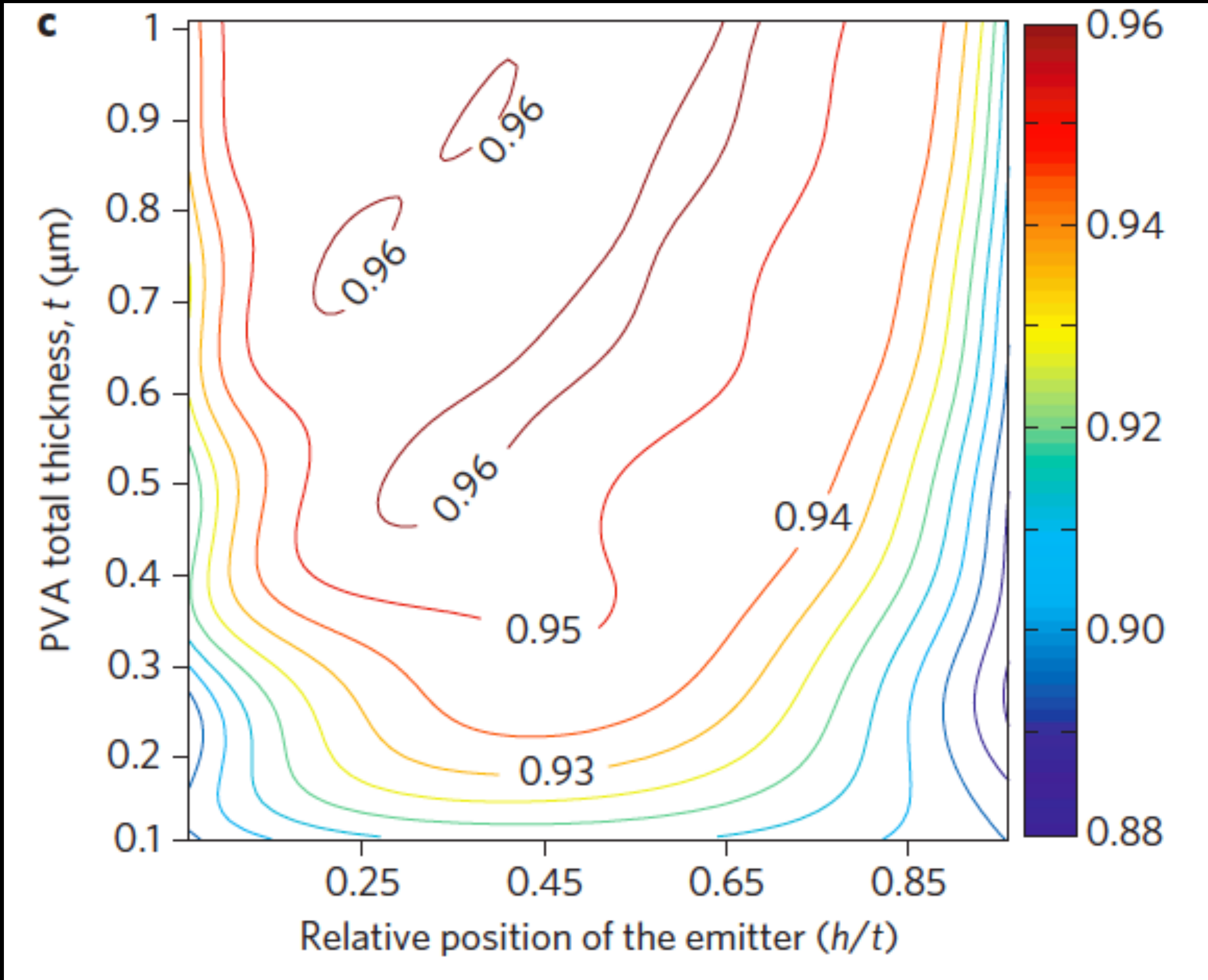
K-G. Lee, X-W. Chen, H. Eghlidi, P. Kukura. R. Lettow, A. Renn, V. Sandoghdar, S. Götzinger, *Nature Photonics,* **5**, 166 (2011).

96% collection efficiency of single photons

experimental results

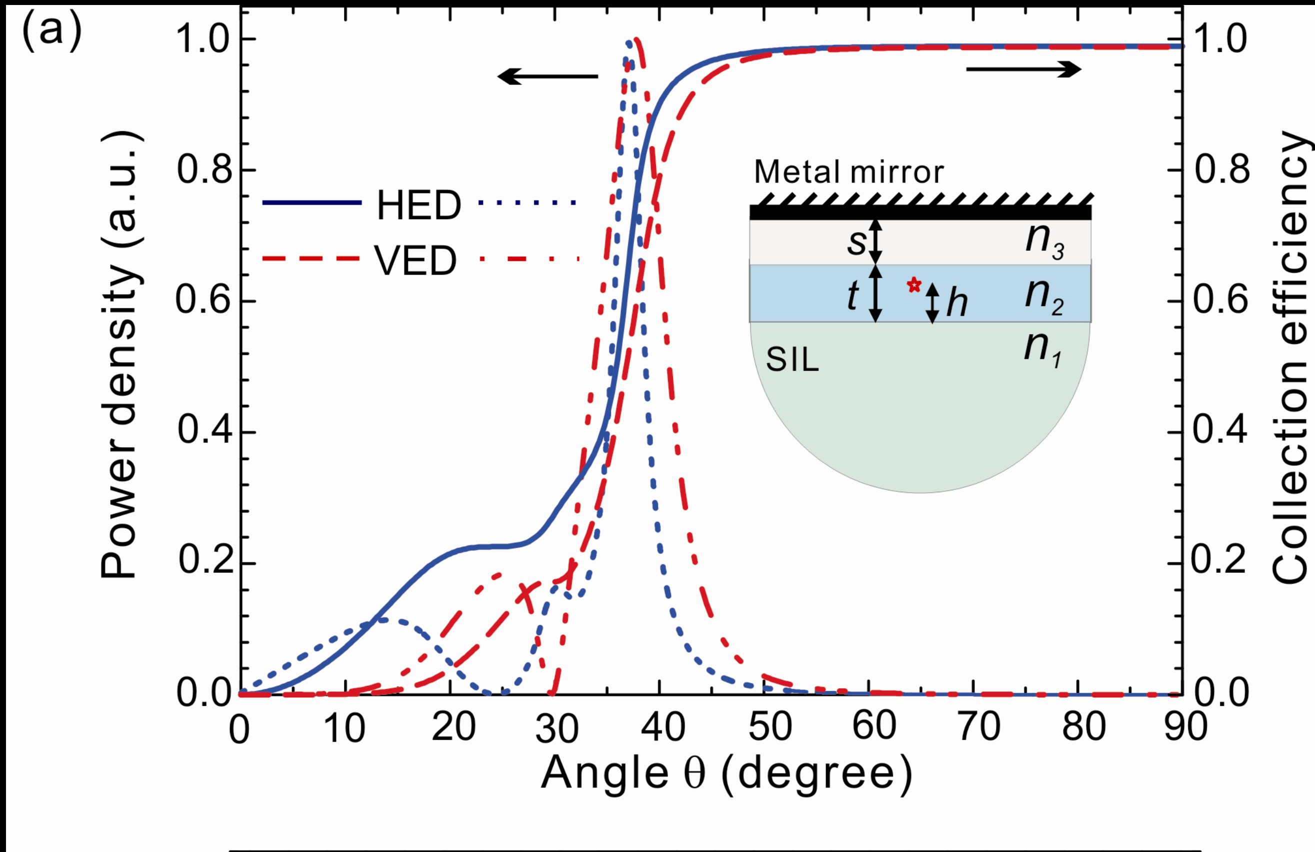


K-G. Lee, X-W. Chen, H. Eghlidi, P. Kukura. R. Lettow, A. Renn, V. Sandoghdar, S. Götzinger, *Nature Photonics*, **5**, 166 (2011).



c
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
PVA total thickness, t (μm)
0.25
0.45
0.65
0.85
Relative position of the emitter (h/t)
0.96
0.96
0.96
0.95
0.94
0.93
0.96
0.94
0.92
0.90
0.88

A metallo-dielectric antenna: 99% collection efficiency



X-W. Chen, S. Götzinger, V. Sandoghdar, *Opt. Lett.* **36**, 3545 (2011).

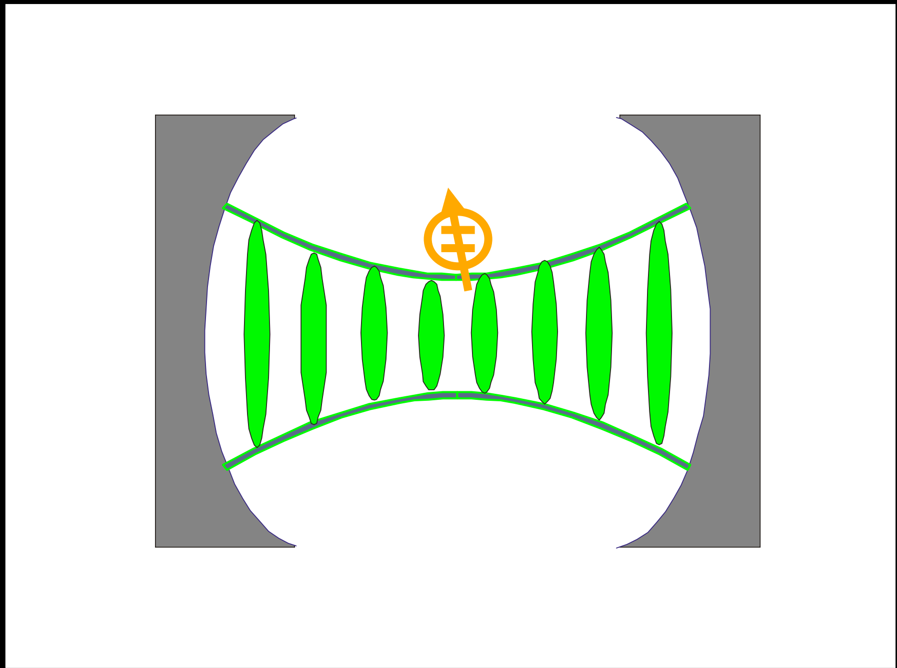
Connection to the efforts in atomic physics, where collective effects are used

Spatial cooperative effect:

antenna theory

Connection to cavity QED

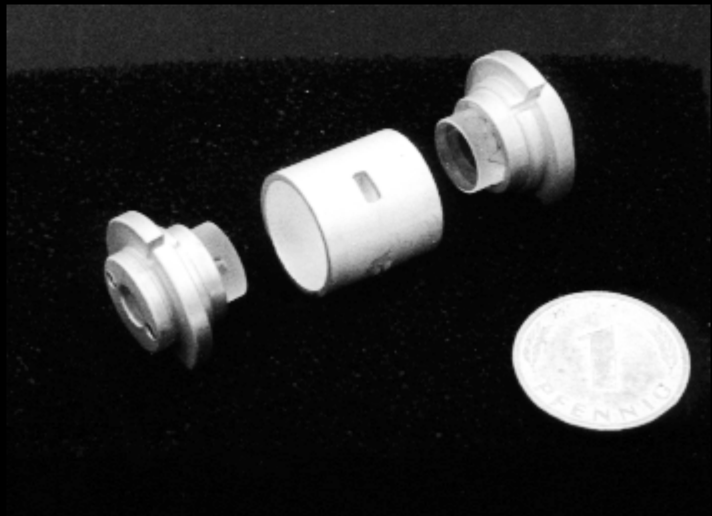
Confining light in a microcavity

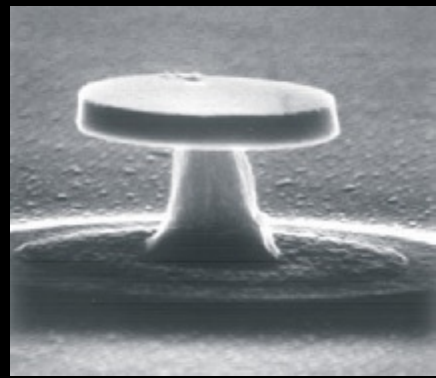


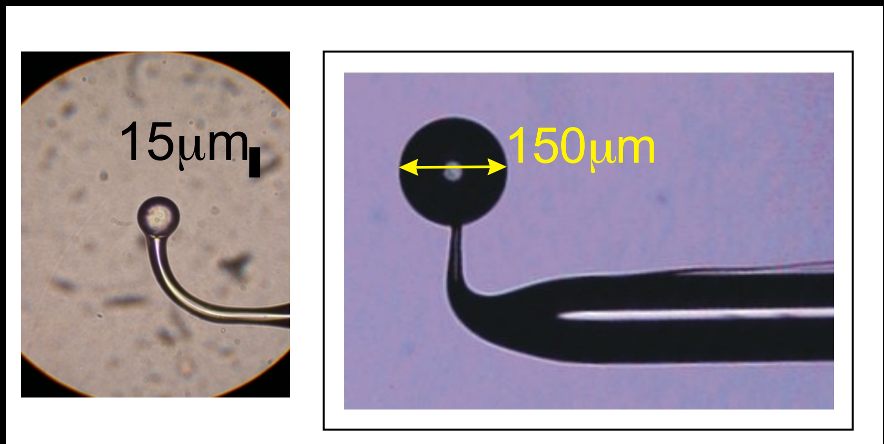
Multiple reflection → interference → resonance

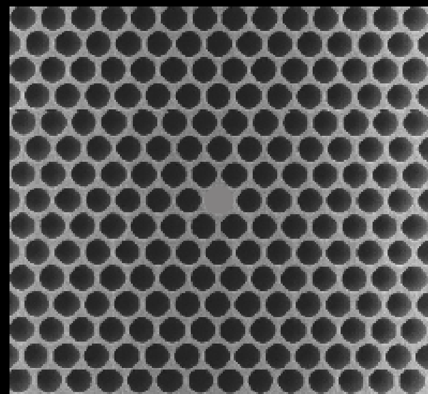
Long but finite storage time, Q

Cavity finesse: mode volume, density of states

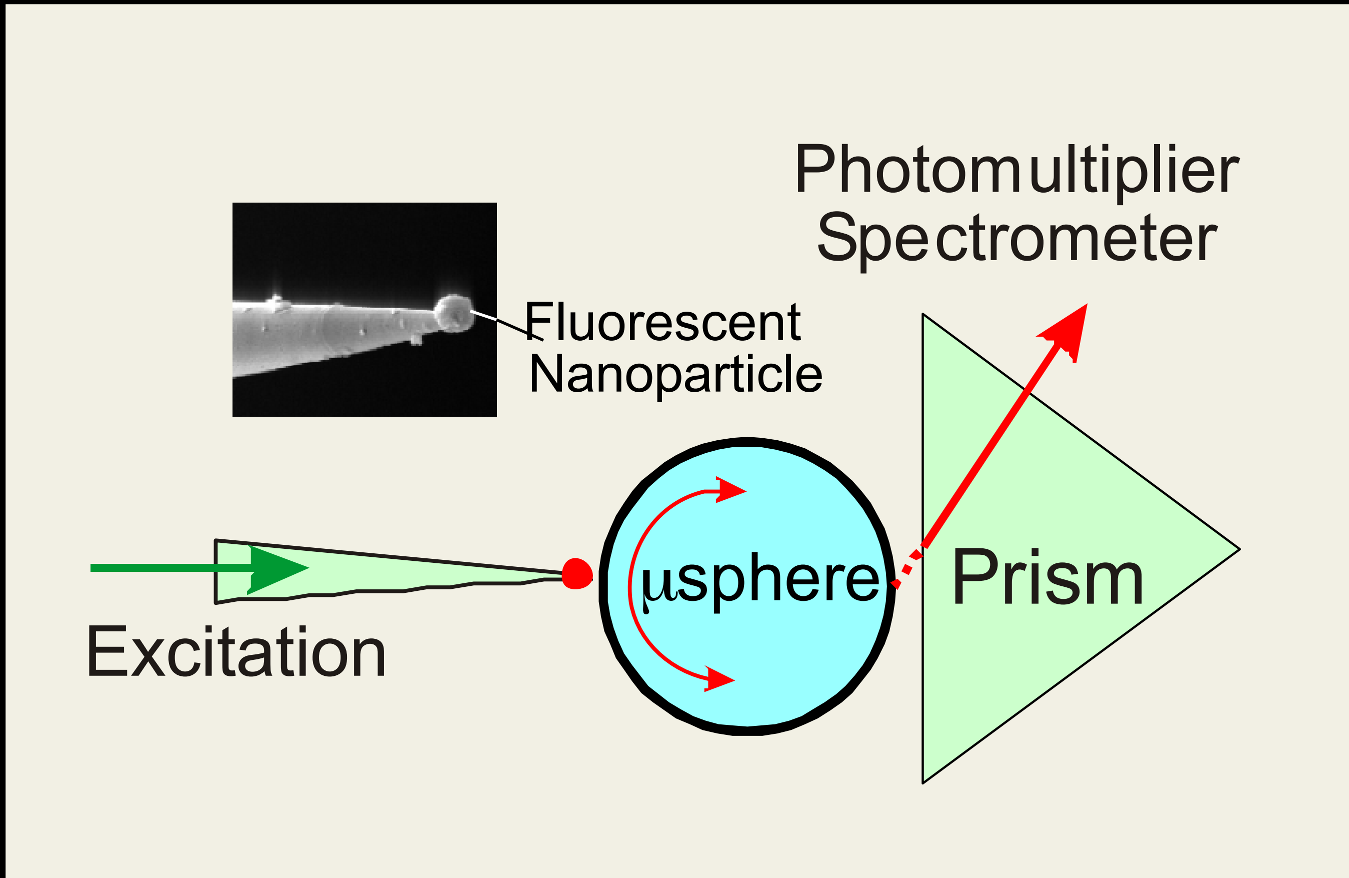





15μm
150μm



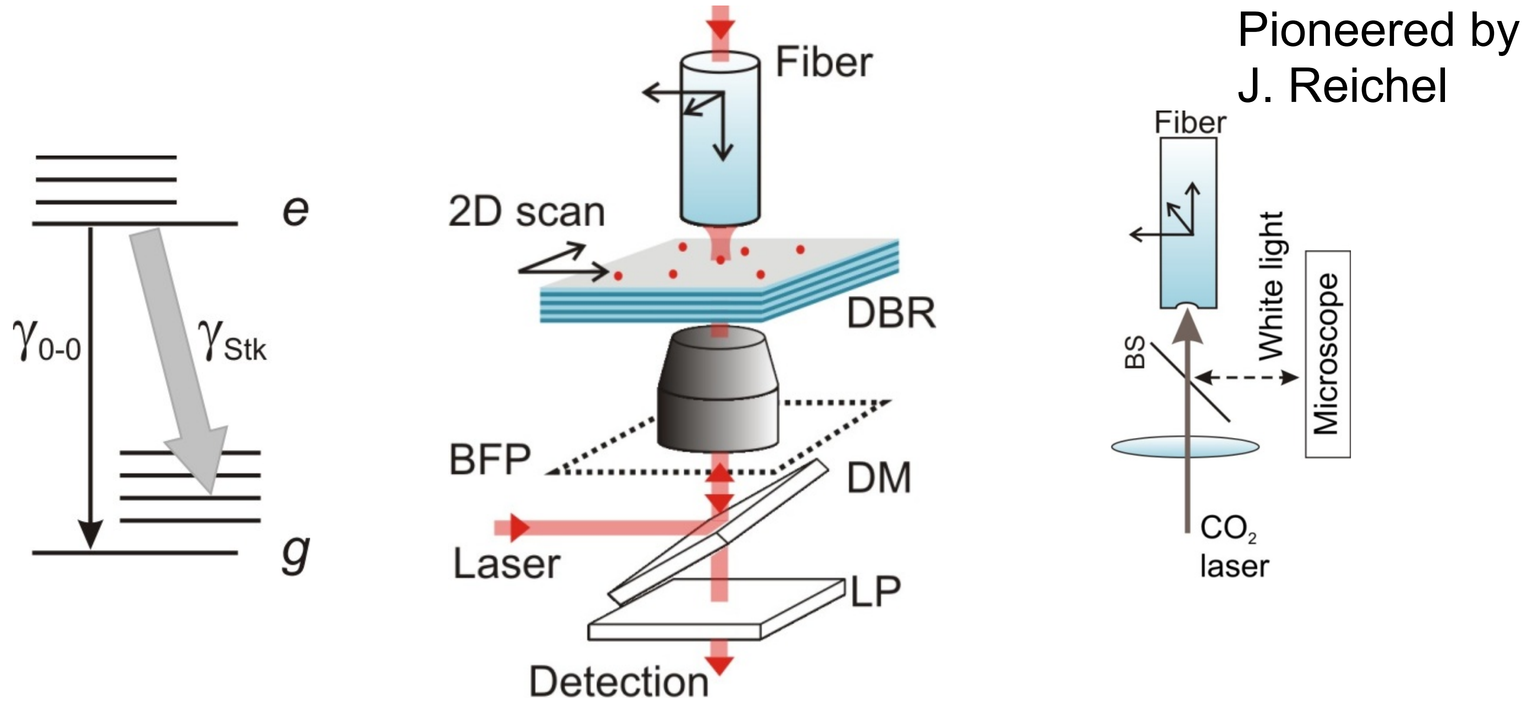
Coupling a nanoscopic emitter to the microsphere



S. Götzinger, L. de S. Menezes, A. Mazzei, S. Kühn, V. Sandoghdar & O. Benson, *Nano Letters* **6**, 1151 (2006).

S. Götzinger, L. de S. Menezes, O. Benson, D. V. Talapin, N. Gaponik, H. Weller, A. L. Rogach, V. Sandoghdar, *J. Opt. B* **6**, 154 (2004).

Enhancing the narrow-band emission
Eliminating the broad Stokes-shifted photons



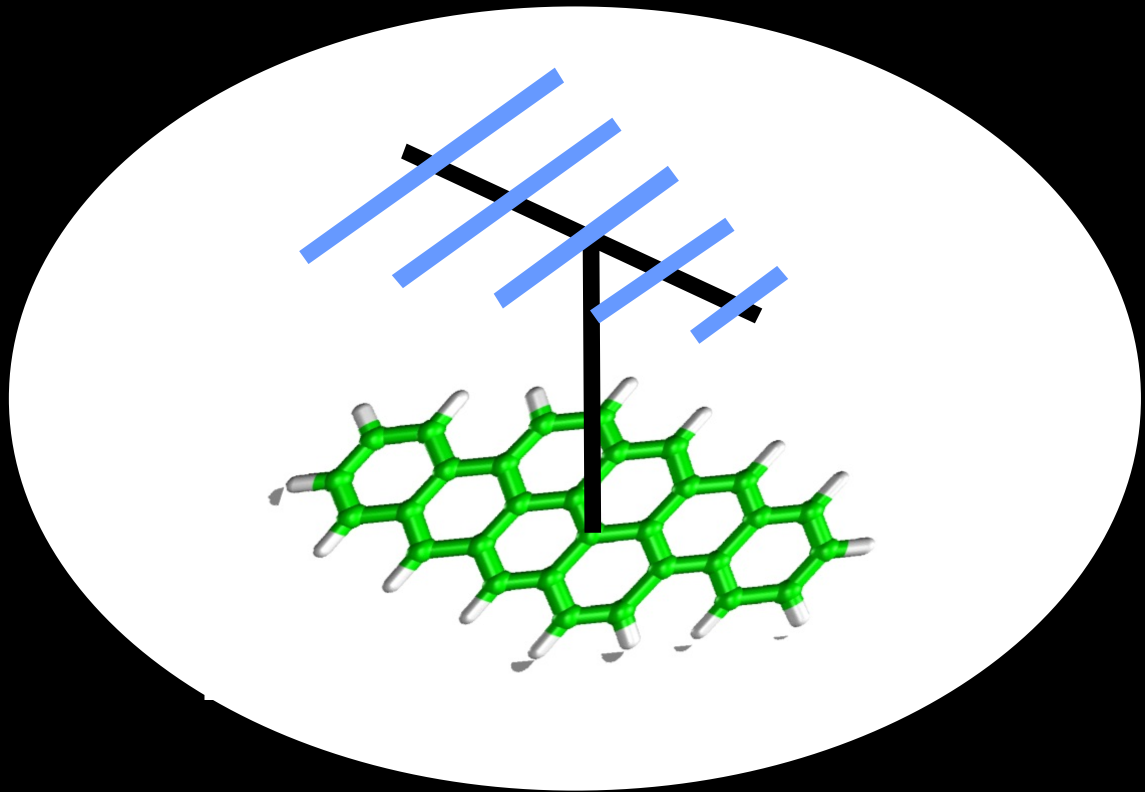
A scannable microcavity

C. Toninelli, Y. Delley, T. Stöferle, A. Renn, S. Götzinger, V. Sandoghdar, *Appl. Phys. Lett.* **97**, 021107 (2010).

Engineering the excitation and emission of an emitter by using *nano-antennas*

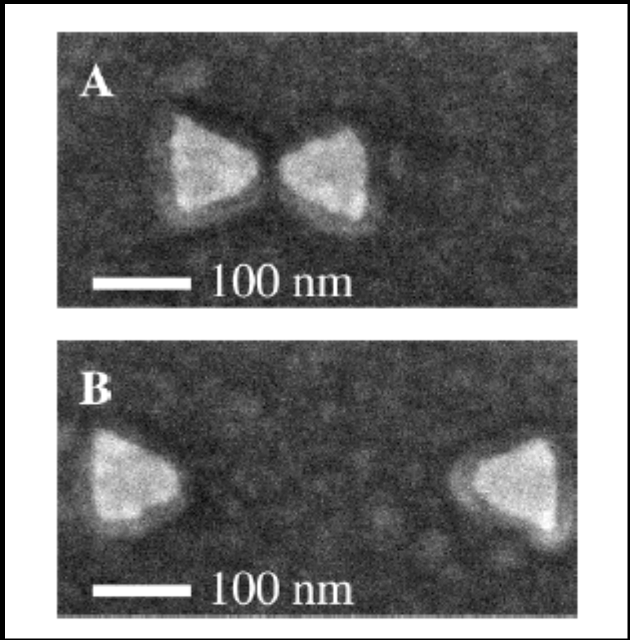
Cavity QED concepts have been already implemented to modify spontaneous emission and to reach strong coupling between an atom and a photon

Goal: achieve the same in the near field (without any quality factor!)



Optical nanoantennas

Inspired by radio antennae

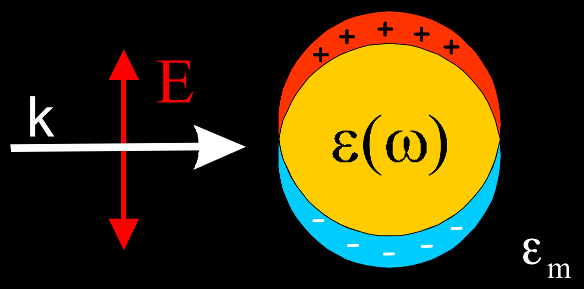


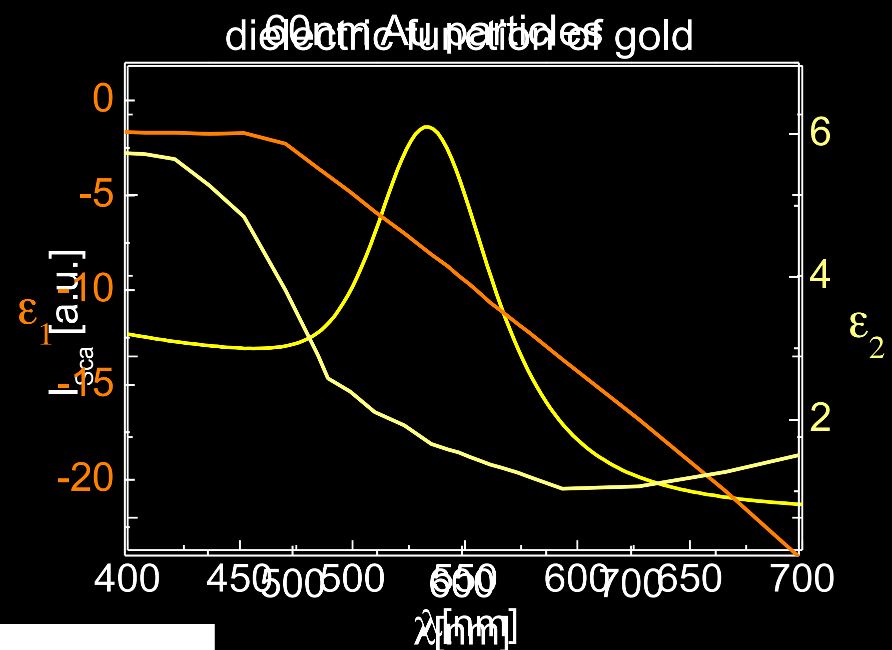
Fromm, et al, NanoLett (2004)

Mühlschlegel, et al, Science (2005)

Antennas in fantasy

Optical Properties of Small Metal Particles





$$s(\lambda) = \eta\alpha(\lambda) = \eta\epsilon_{\mathrm{med}}(\lambda)\frac{\pi D^3}{2}\frac{\epsilon_{\mathrm{part}}(\lambda) - \epsilon_{\mathrm{med}}(\lambda)}{\epsilon_{\mathrm{part}}(\lambda) + 2\epsilon_{\mathrm{med}}(\lambda)}.$$

for $\varepsilon(\omega) = -2\varepsilon_m$

➡ Resonance

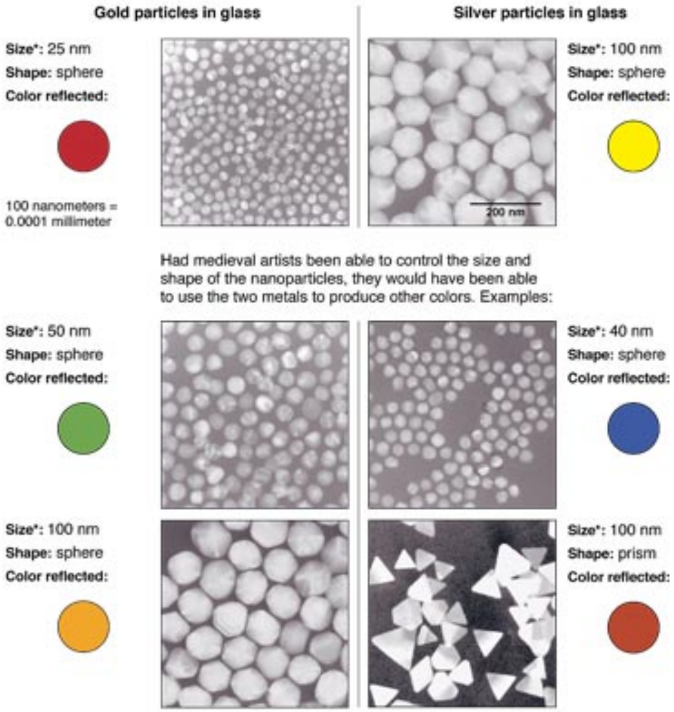
full electrodynamic calculation: Mie theory

G. Mie, *Ann. Phys.* **25** (1908)

The color of the particle depends on its material, size and shape

The First Nanotechnologists

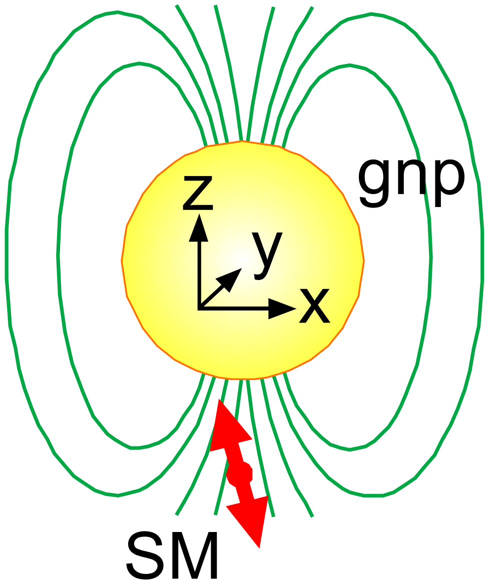
Ancient stained-glass makers knew that by putting varying, tiny amounts of gold and silver in the glass, they could produce the red and yellow found in stained-glass windows. Similarly, today's scientists and engineers have found that it takes only small amounts of a nanoparticle, precisely placed, to change a material's physical properties.

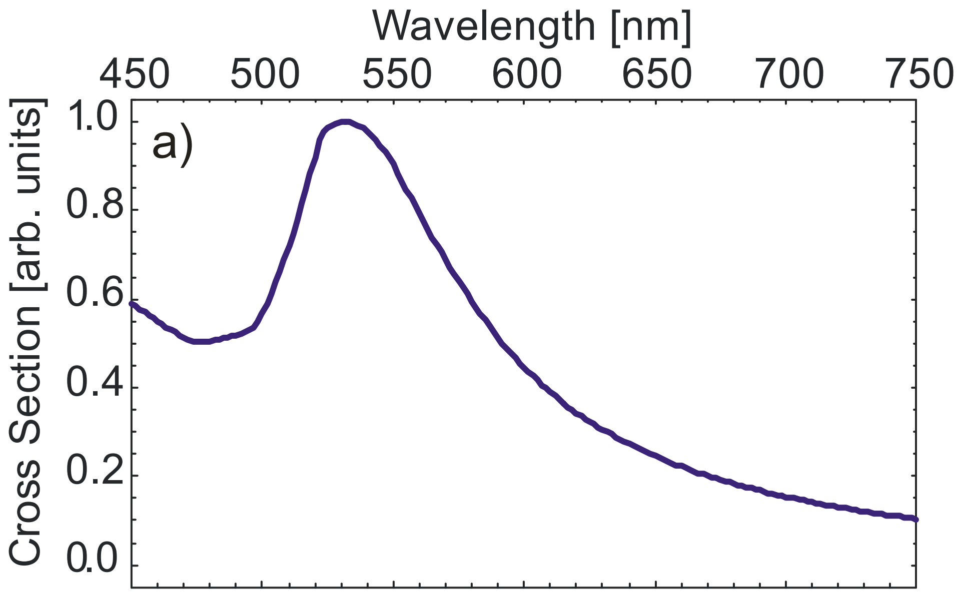


Source: Dr. Chad A. Mirkin, Institute of Nanotechnology, Northwestern University

*Approximate

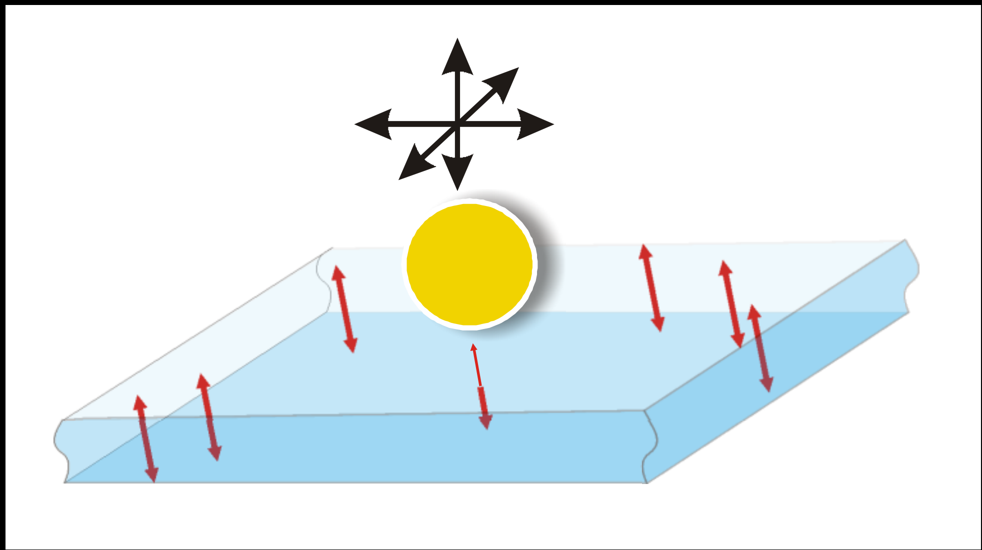
A single gold nanoparticle as a Nano-Antenna

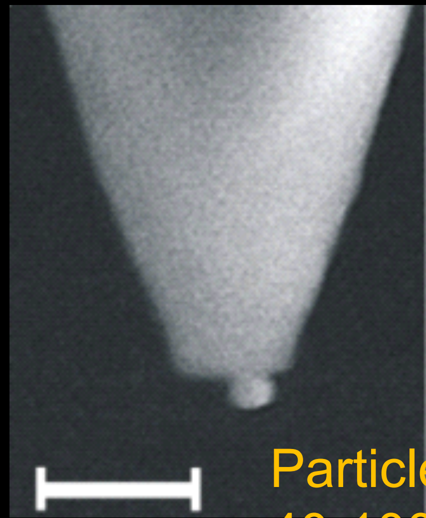




The subwavelength gold particle acts as the extension/magnification of the molecular dipole moment
→ Faster emission

Controlled positioning of the nanoantenna





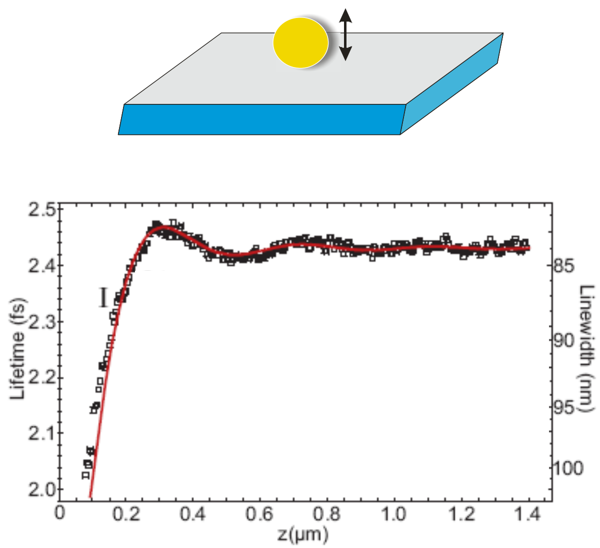
200 nm

Particle diameter
40-100 nm

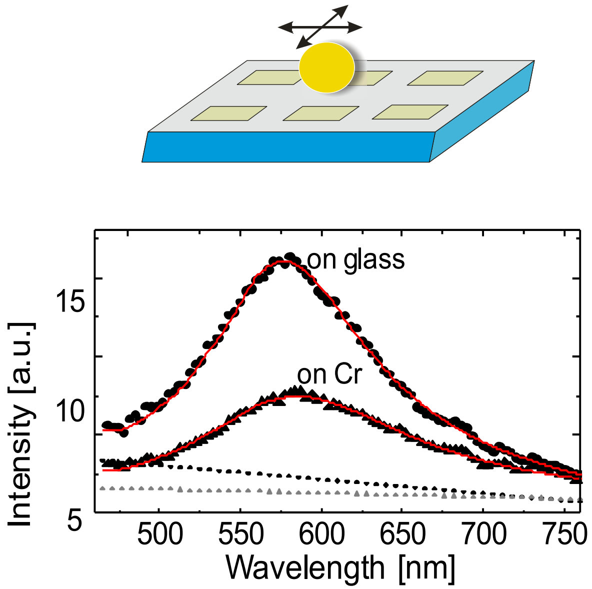
T. Kalkbrenner, M. Ramstein, J. Mlynek, V. Sandoghdar
J. Microscopy **202,** 72 (2001).

T. Kalkbrenner, U. Hakanson, & V. Sandoghdar,
Nano Lett. **4**, 2309 (2004).

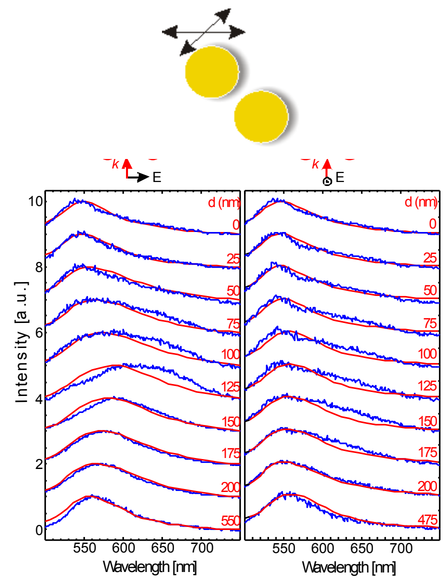
Local modification/control of antenna resonances



B. C. Buchler, et al, *Phys. Rev. Lett.* **95**, 063003 (2005).



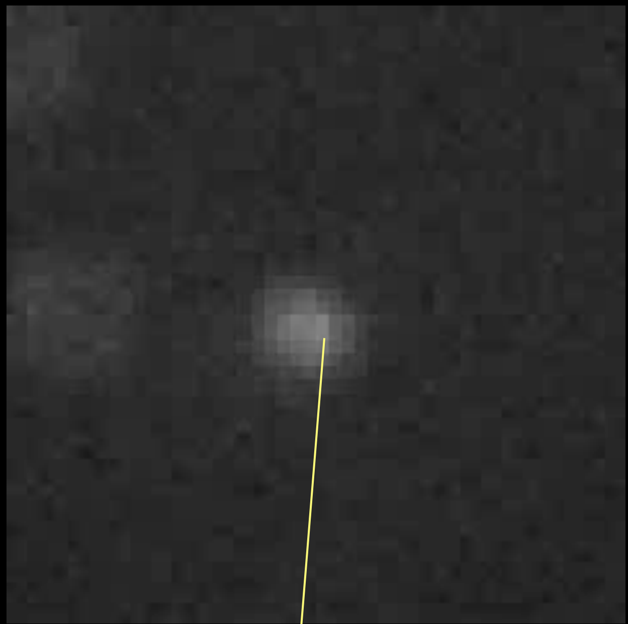
T. Kalkbrenner, et al, *Phys. Rev. Lett.* **95,** 200801 (2005).



U. Hakanson, et al, *Phys. Rev. B* **77**, 155408 (2008).

Controlled Interaction of a Single Gold Nanoparticle with a Single Molecule

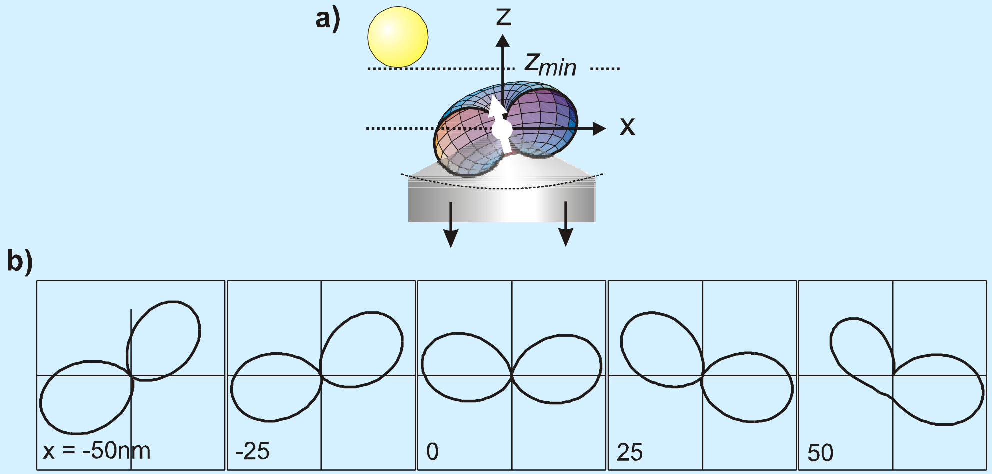
Scanning the molecule

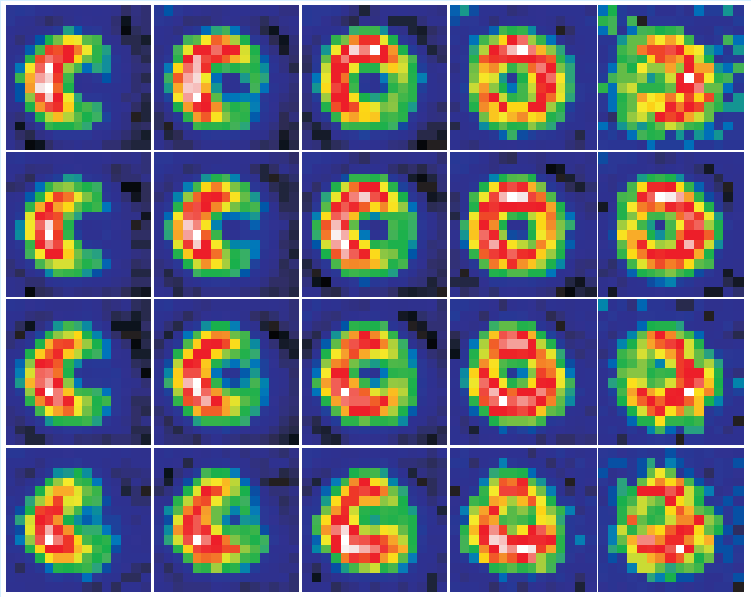


gold
nanoparticle

Scanning the particle

Modification of the molecular emission pattern





S. Kühn, et al, *PRL.* **97**, 017402 (2006).

S. Kühn, et al., *Mol. Phys*. **106**, 893 (2008).

enhancement of the excitation field
modification of the radiative lifetime
modification of the nonradiative lifetime

| Field enhancement | ²

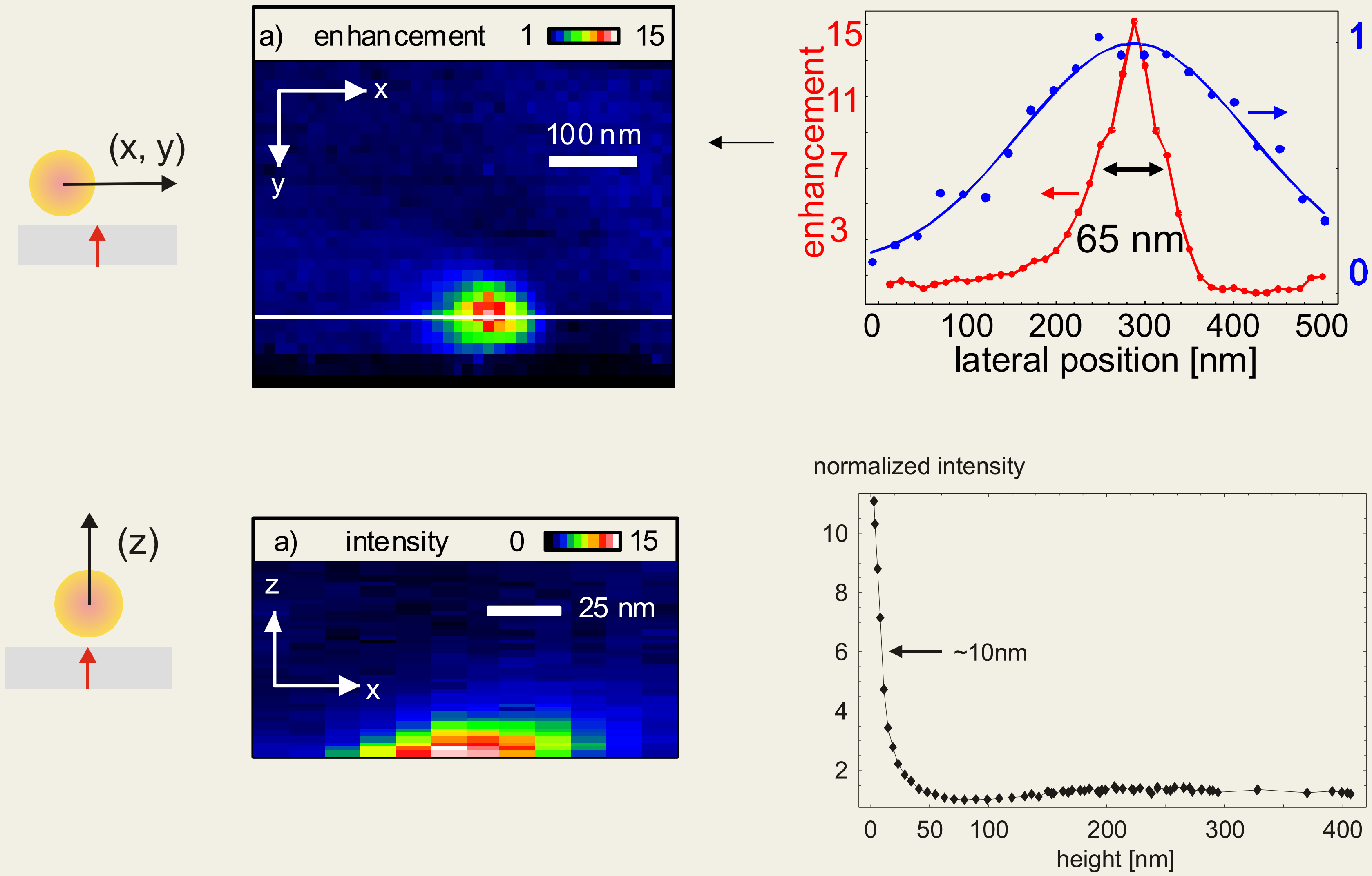
Overall collection efficiency

$$S_{fluor,enhanced} = \xi_{collection} \cdot K_{exciatation} \cdot \eta_{system} \cdot S_{fluor,0}$$

Quantum efficiency
(of complete system)

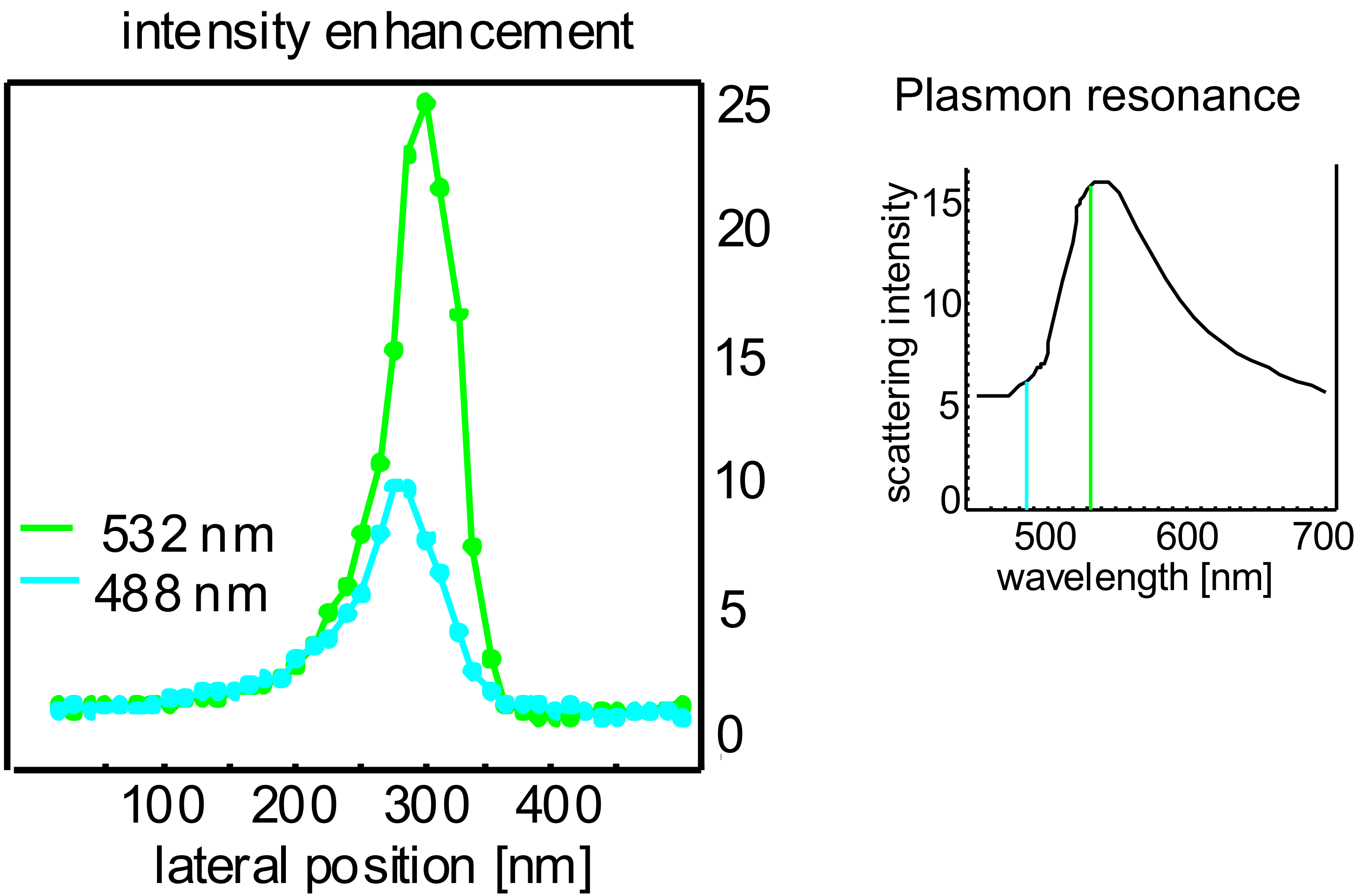
$$\eta = \frac{\Gamma_{rad}}{\Gamma_{rad} + \Gamma_{nr}}$$

Influence on the Fluorescence Intensity

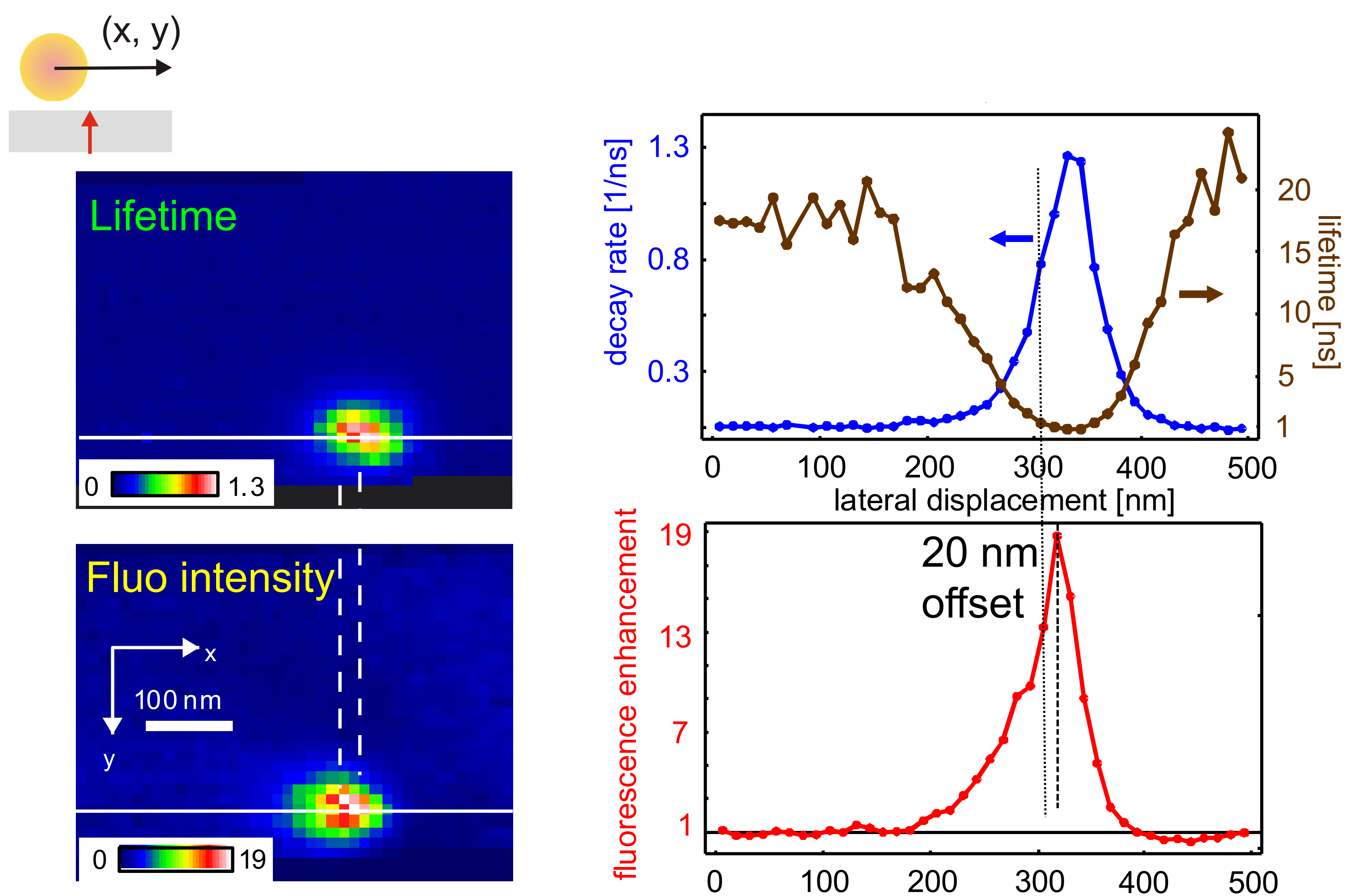


S. Kühn, U. Hakanson, L. Rogobete & V. Sandoghdar,
Phys. Rev. Lett. **97**, 017402 (2006) .

Direct demonstration of the antenna resonance effect

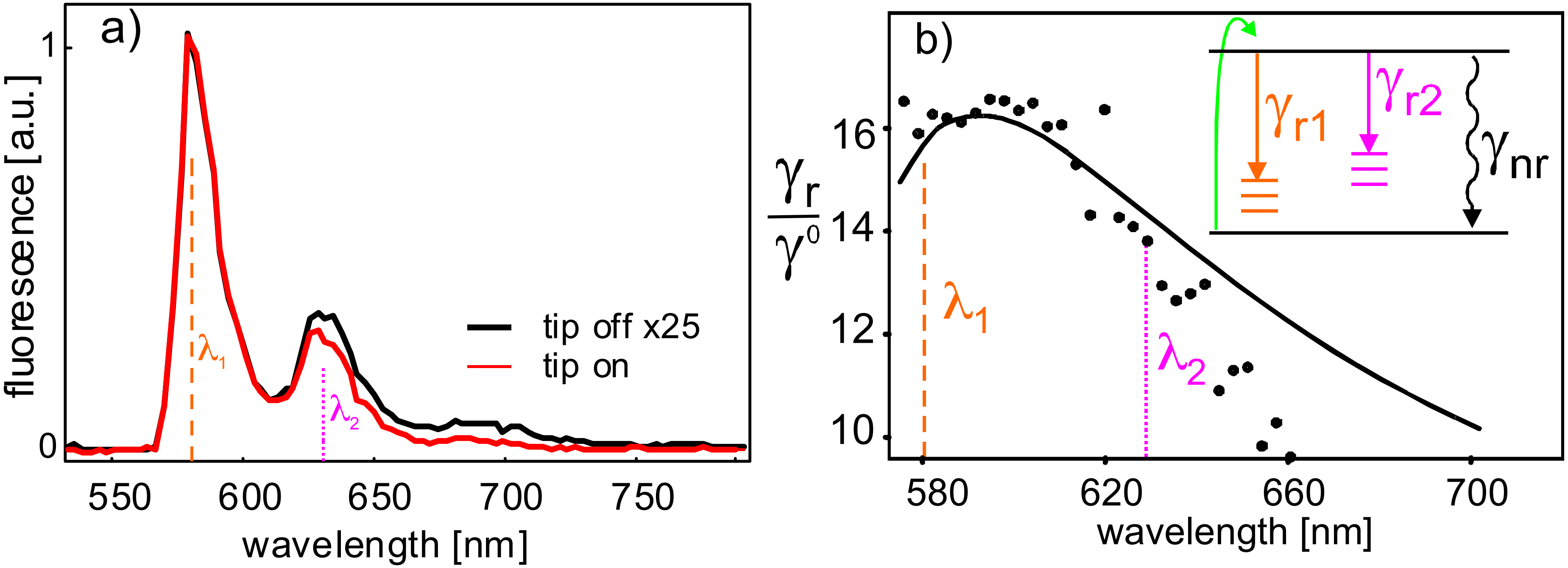


Influence on the Emission Lifetime



S. Kühn, U. Hakanson, L. Rogobete & V. Sandoghdar,
Phys. Rev. Lett. **97**, 017402 (2006) . AND the **Supplementary**

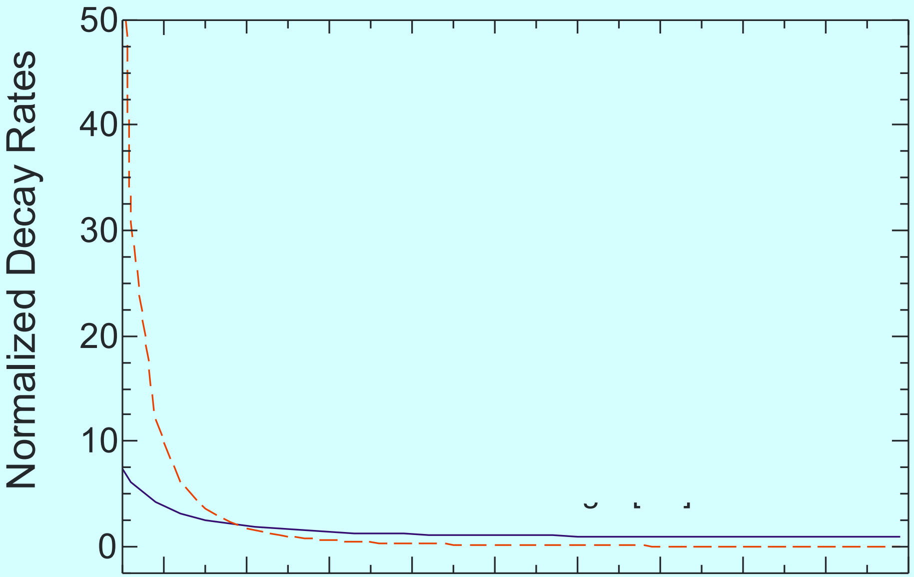
Modification of the emission spectrum

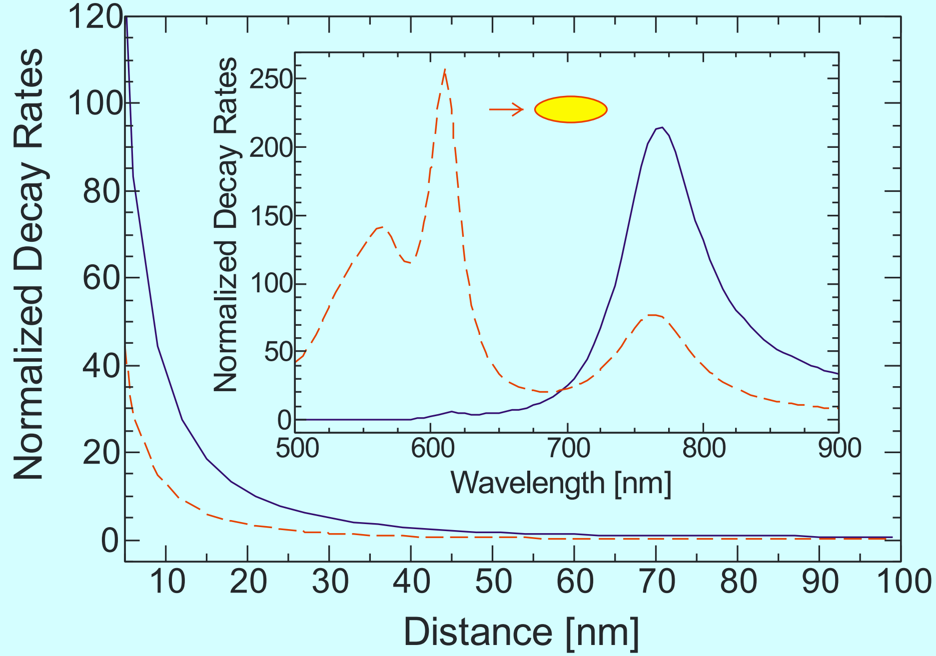


S. Kühn, U. Hakanson, L. Rogobete & V. Sandoghdar,
Phys. Rev. Lett. **97**, 017402 (2006) ; **Supplementary Material**

S. Kühn, et al., *Mol. Phys.*, **106**, 893 (2008).

Engineering the quantum efficiency

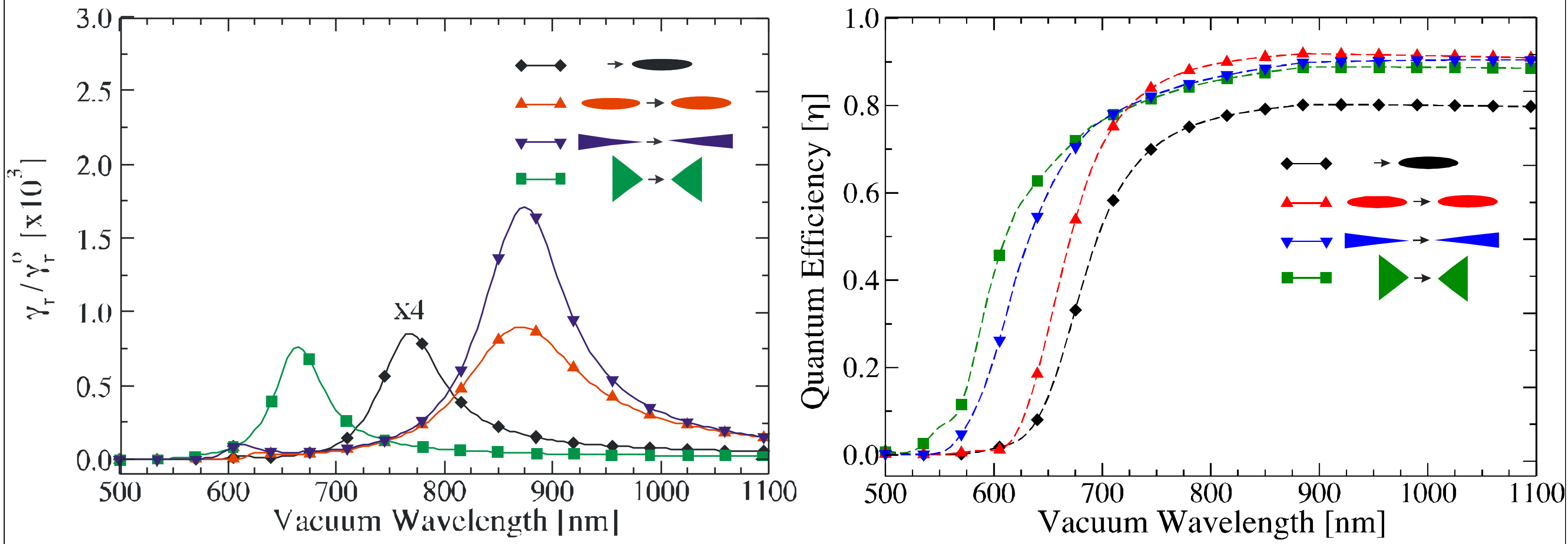




L. Rogobete, F. Kaminski, M. Agio, V. Sandoghdar, *Opt. Lett.* **32**, 1623 (200

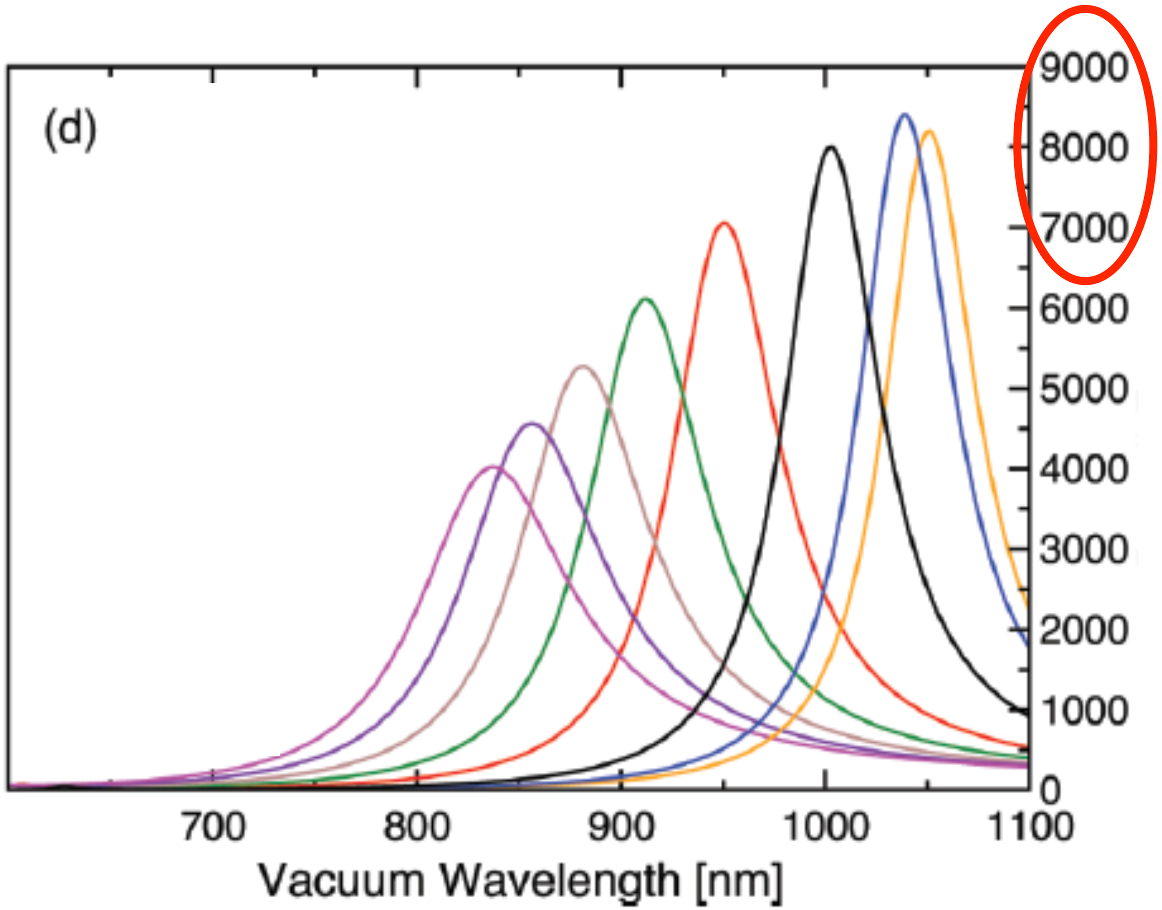
Optimizing the radiative emission rate

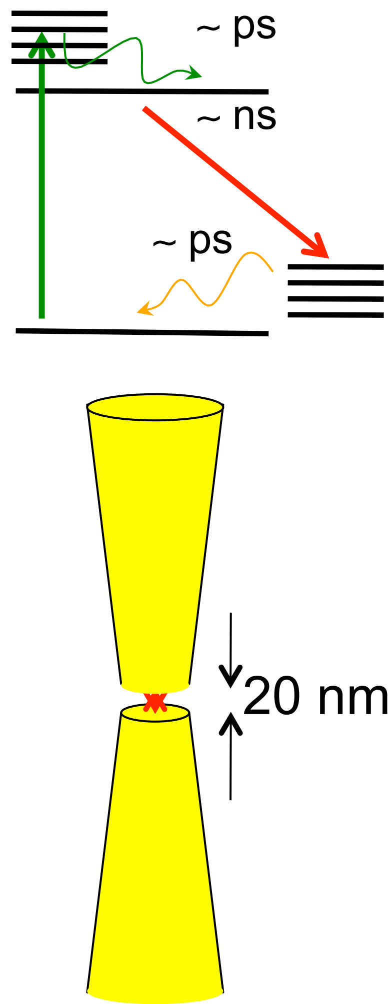
Key: mode matching



L. Rogobete, F. Kaminski, M. Agio, V. Sandoghdar,
Opt. Lett. **32**, 1623 (2007).

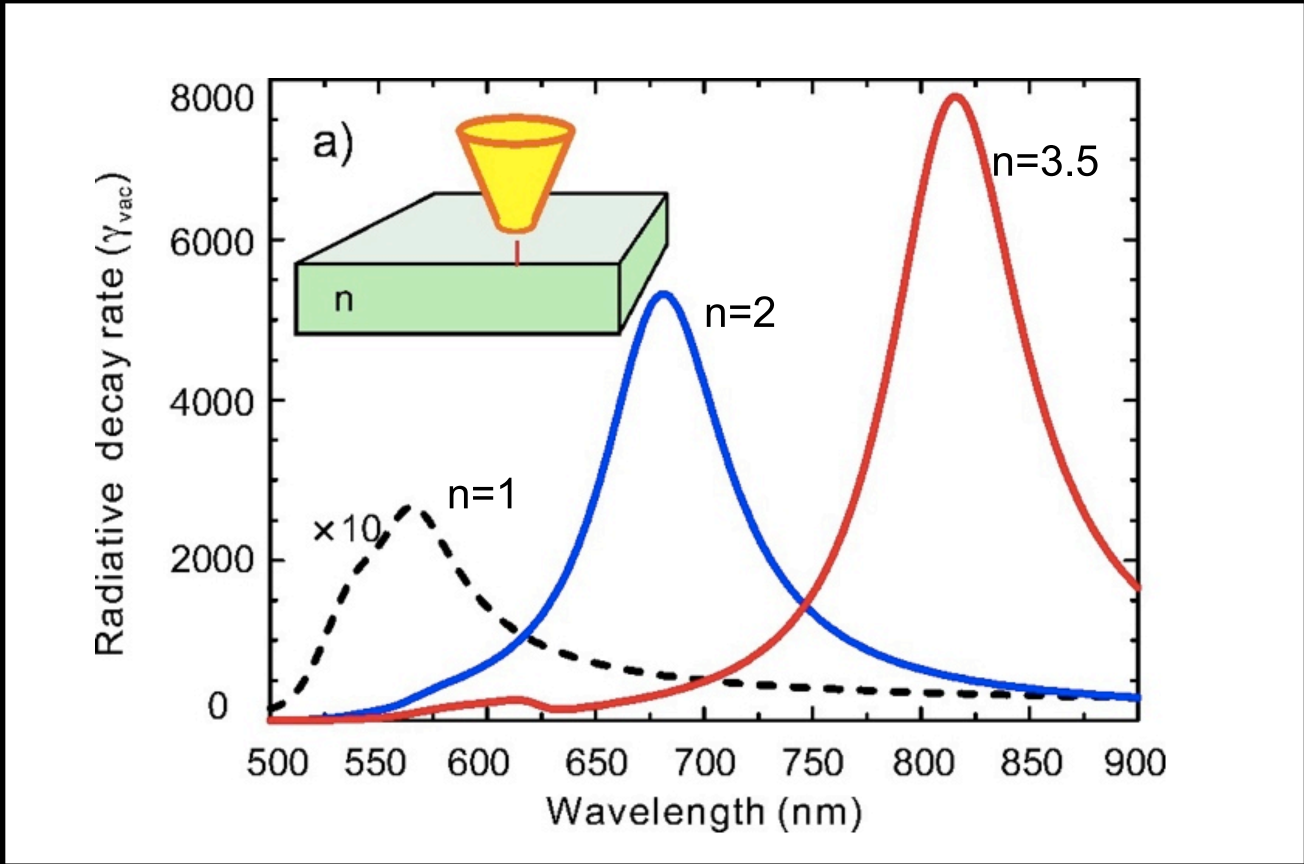
Antenna enhancement of spontaneous emission rate





A. Mohammadi, F. Kaminski, V. Sandoghdar, M. Agio,
J. Phys. Chem. C 114, 7372 (2010).

Ultrastrong enhancement of spontaneous emission



X-W. Chen, M. Agio, V. Sandoghdar, *Phys. Rev. Lett.* to appear (2012).

Room temperature experiments

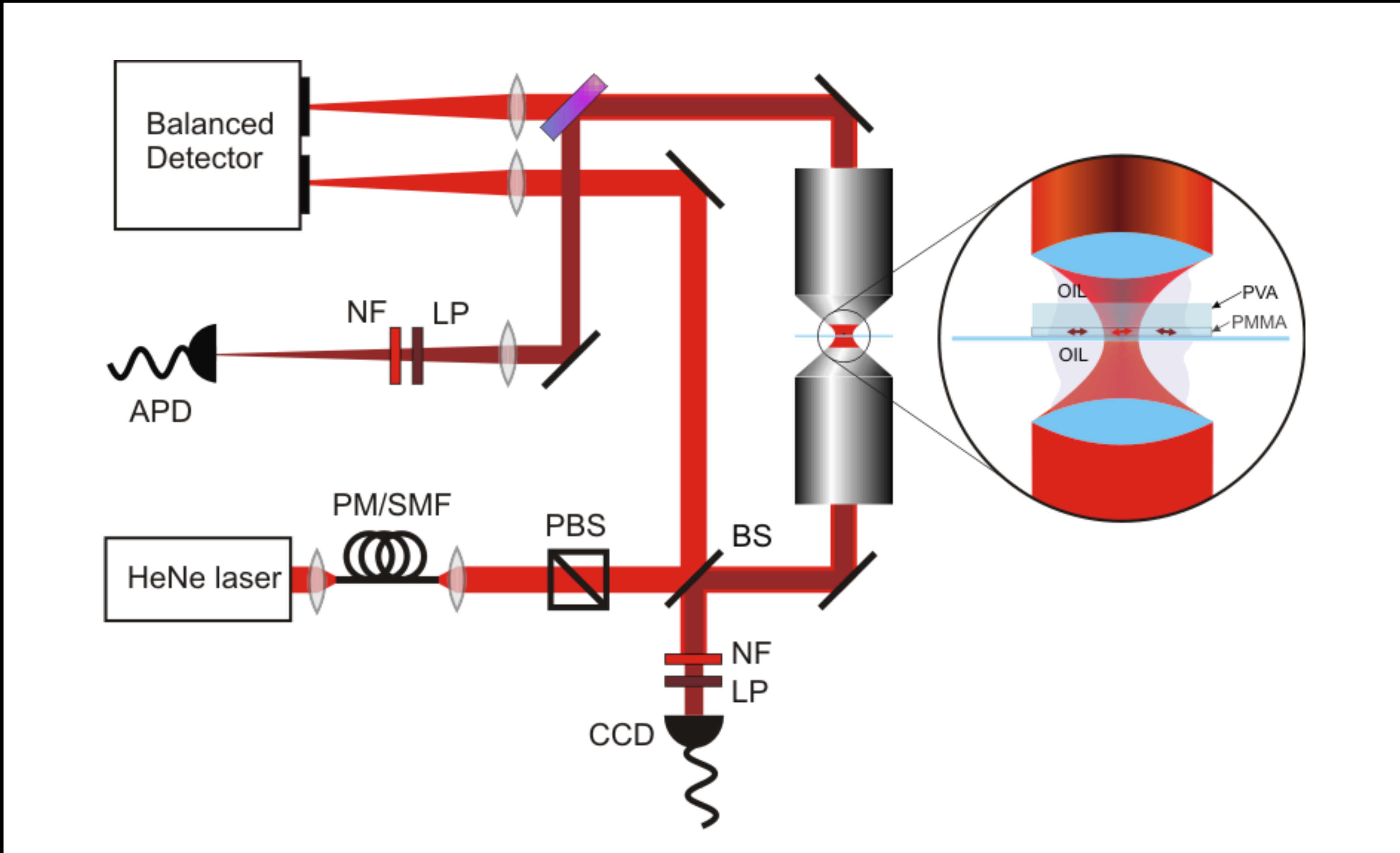
$$\sigma_{abs} = \frac{3\lambda^2}{2\pi}\frac{\gamma_0}{\gamma} \qquad 10^{-6}$$

Limit of A ~ (200 nm) ^{2}

Room-temperature in the solid state $\sigma \sim 10^{-15}$ cm ^{2} ~ 0.1 (nm) ^{2}

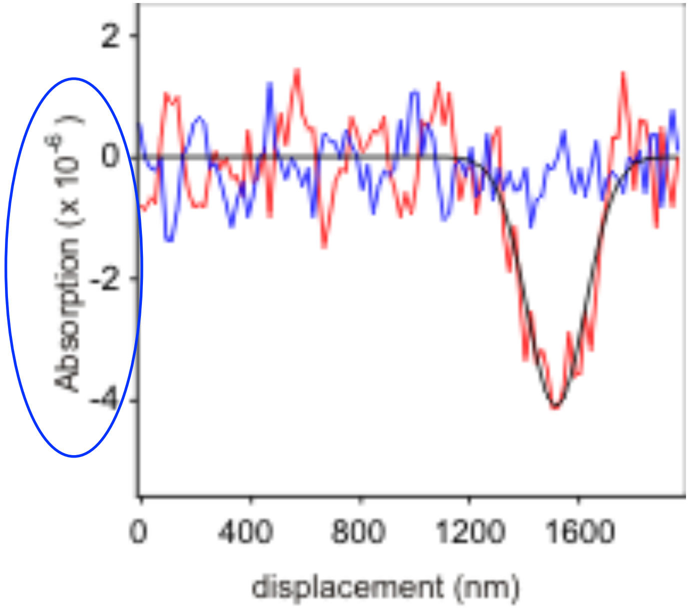
Need a signal-to-noise ratio of 10^{-6} → laser noise suppresion

Transmission measurement of *single* ***molecules***



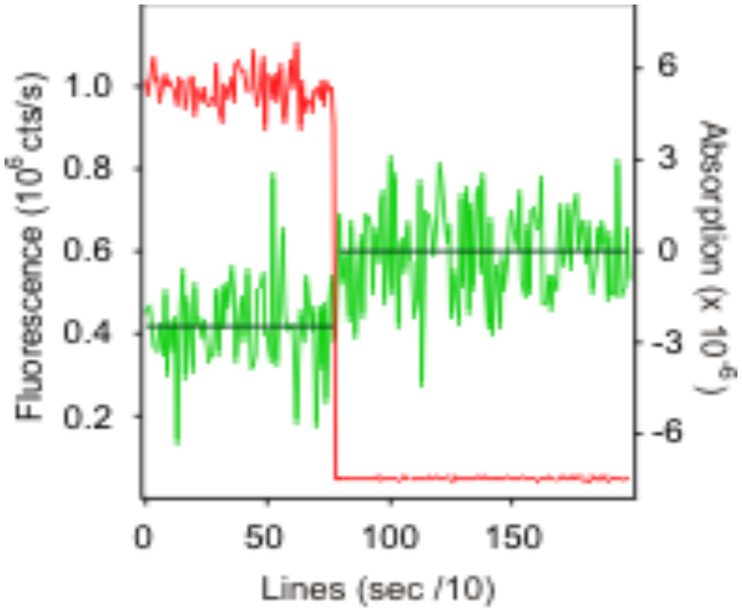
P. Kukura, M. Celebrano, A. Renn, V. Sandoghdar,
J. Phys. Chem. Lett. **1**, 3323 (2010).

The first demonstration of single-molecule sensitivity in absorption
@ room temperature



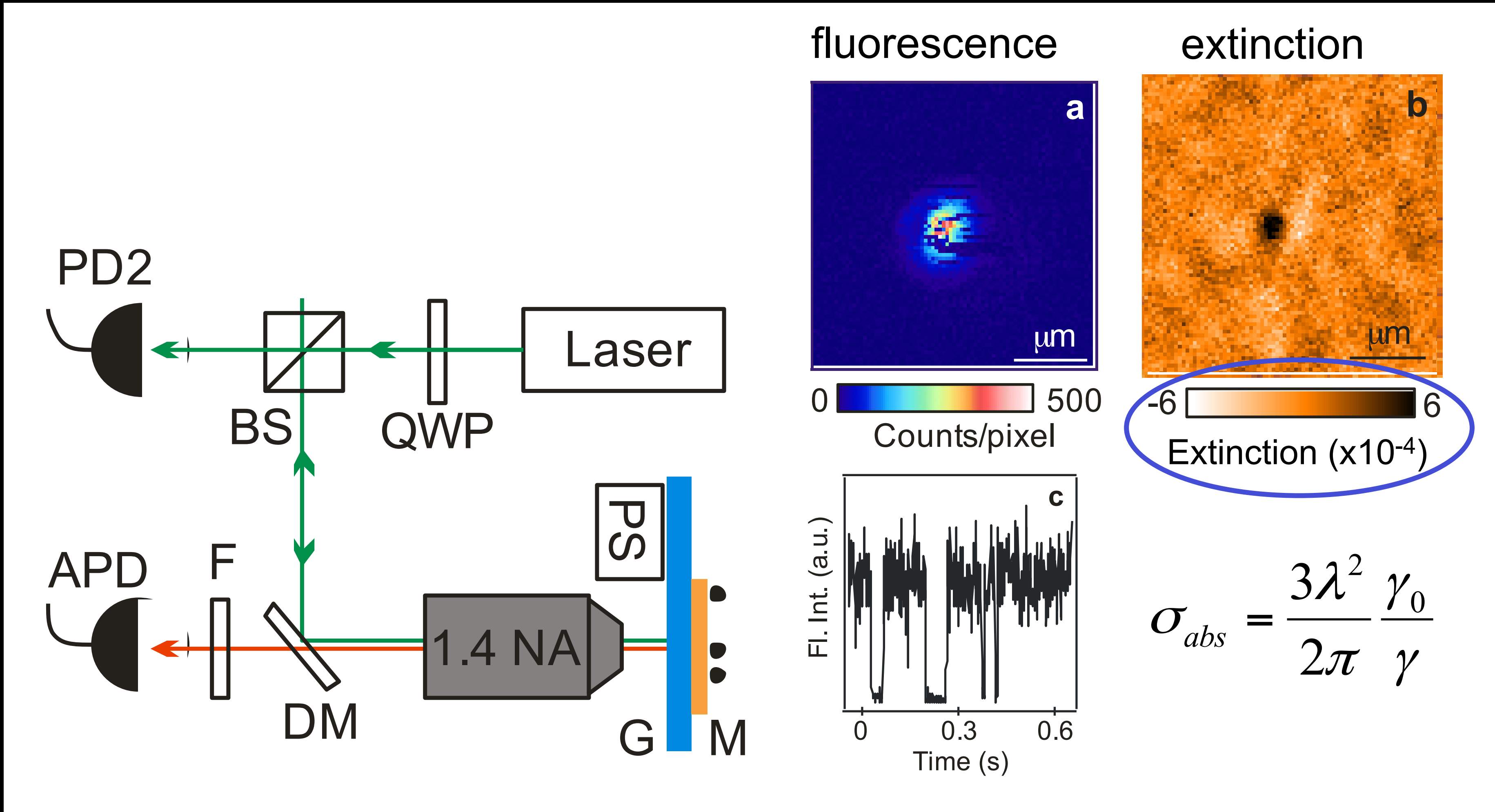
after bleaching

before bleaching



P. Kukura, M. Celebrano, A. Renn, V. Sandoghdar, *J. Phys. Chem. Lett.* **1**, 3323 (2010).

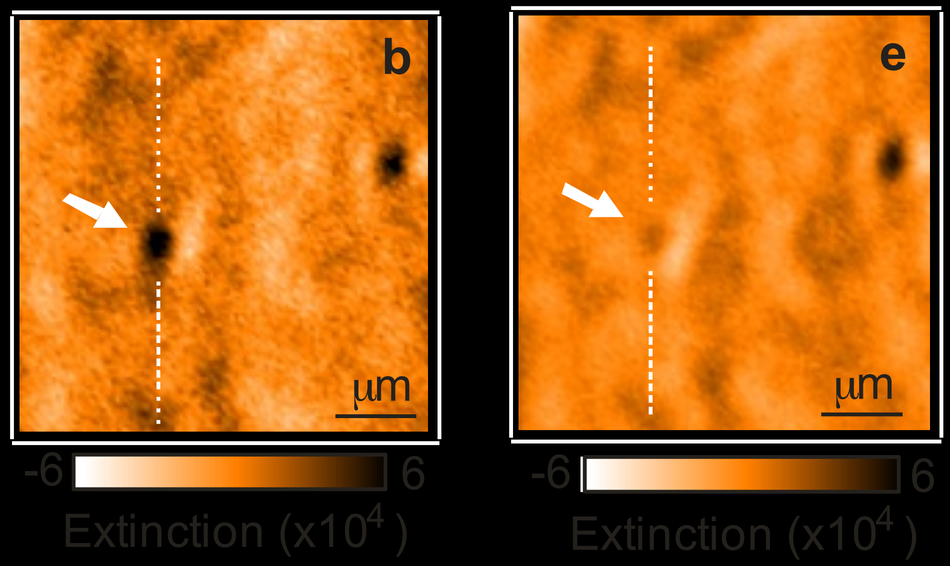
Room-Temperature Imaging of a Single (nonfluorescent) Quantum Dot



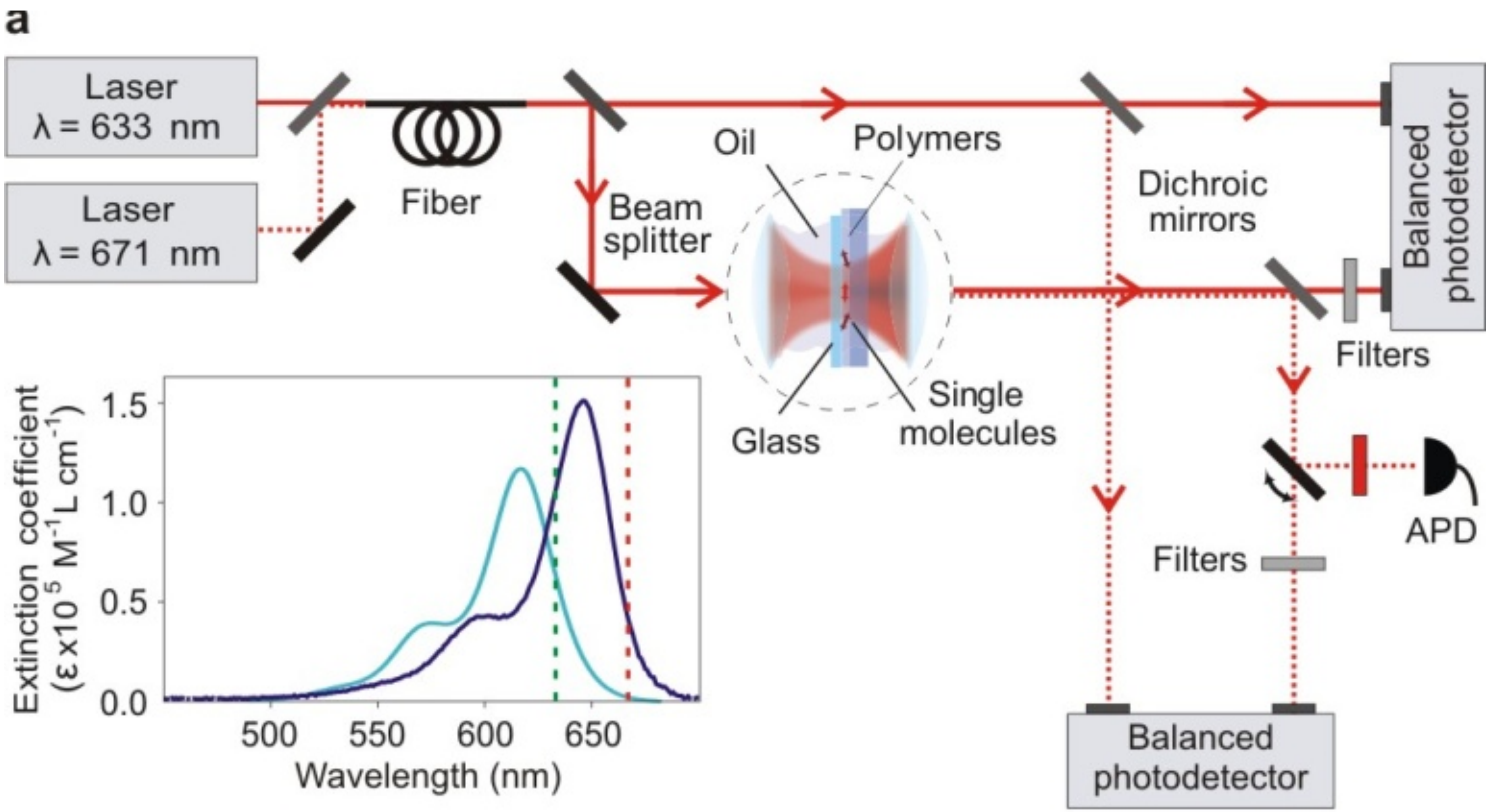
$$\sigma_{abs} = \frac{3\lambda^2}{2\pi}\frac{\gamma_0}{\gamma}$$

P. Kukura, M. Celebrano, A. Renn, V. Sandoghdar, *Nano Lett.* **9**, 926 (2009).

Main limitation: background scattering

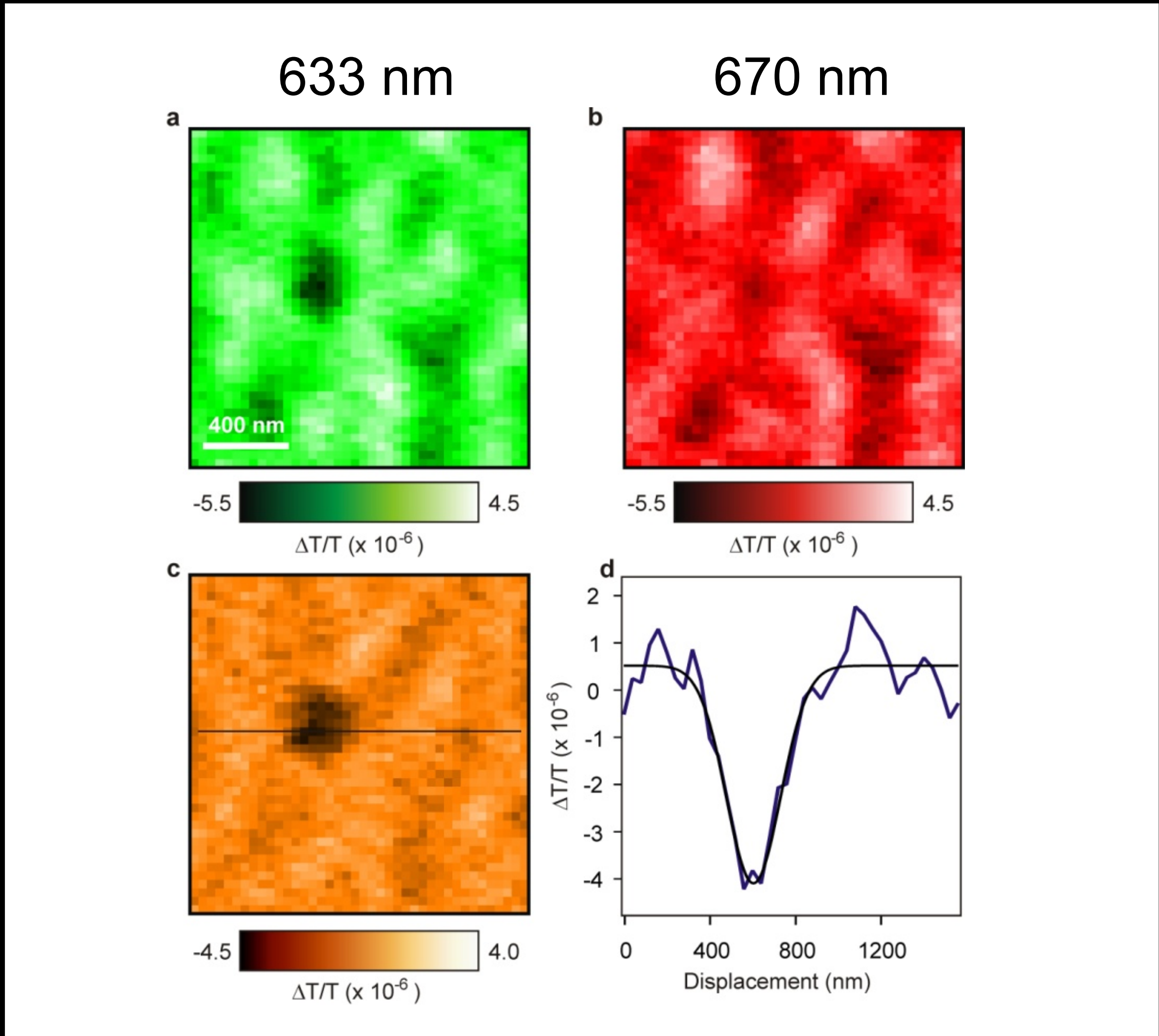


Single-molecule absorption *imaging* and *spectroscopy*



M. Celebrano, P. Kukura, A. Renn, V. Sandoghdar
Nature Photonics **5**, 95 (2011).

Imaging a single molecule in absorption



M. Celebrano, P. Kukura, A. Renn, V. Sandoghdar
Nature Photonics **5**, 95 (2011).

Extinction spectroscopy/microscopy lets us see strongly quenched systems

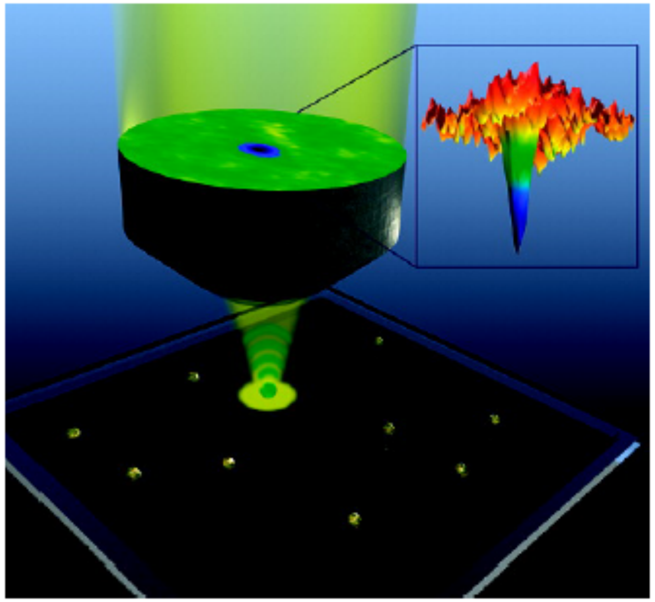
NANO LETTERS

Letter

Imaging a Single Quantum Dot When It Is Dark

P. Kukura, M. Celebrano, A. Renn, and V. Sandoghdar

Nano Lett., **2009**, 9 (3), 926-929• DOI: 10.1021/nl801735y • Publication Date (Web): 01 August 2008



Fluorescence

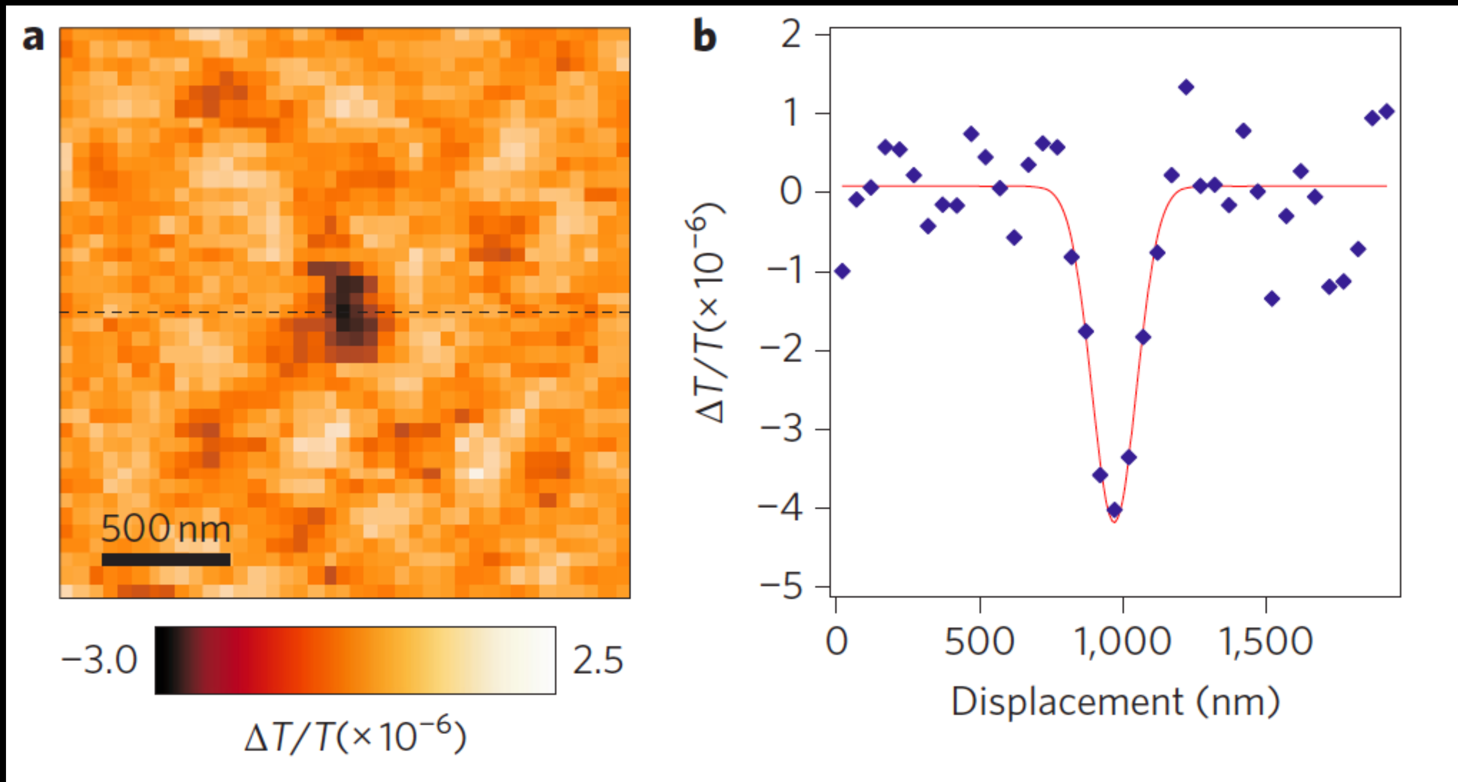
$$\eta \propto \frac{\gamma_r}{\gamma_r + \gamma_{nr}}$$

Extinction

$$\sigma \propto \frac{\gamma_r}{\gamma_r + \gamma_{nr} + \gamma_{deph}}$$

$$\gamma_{deph} \approx 10^{4-5}\gamma_r$$

Imaging a *quenched* molecule



During the same summer !

Room-Temperature Detection of a Single Molecule's Absorption by Photothermal Contrast

A. Gaiduk, M. Yorulmaz, P. V. Ruijgrok, M. Orrit*

SCIENCE VOL 330 15 OCTOBER 2010

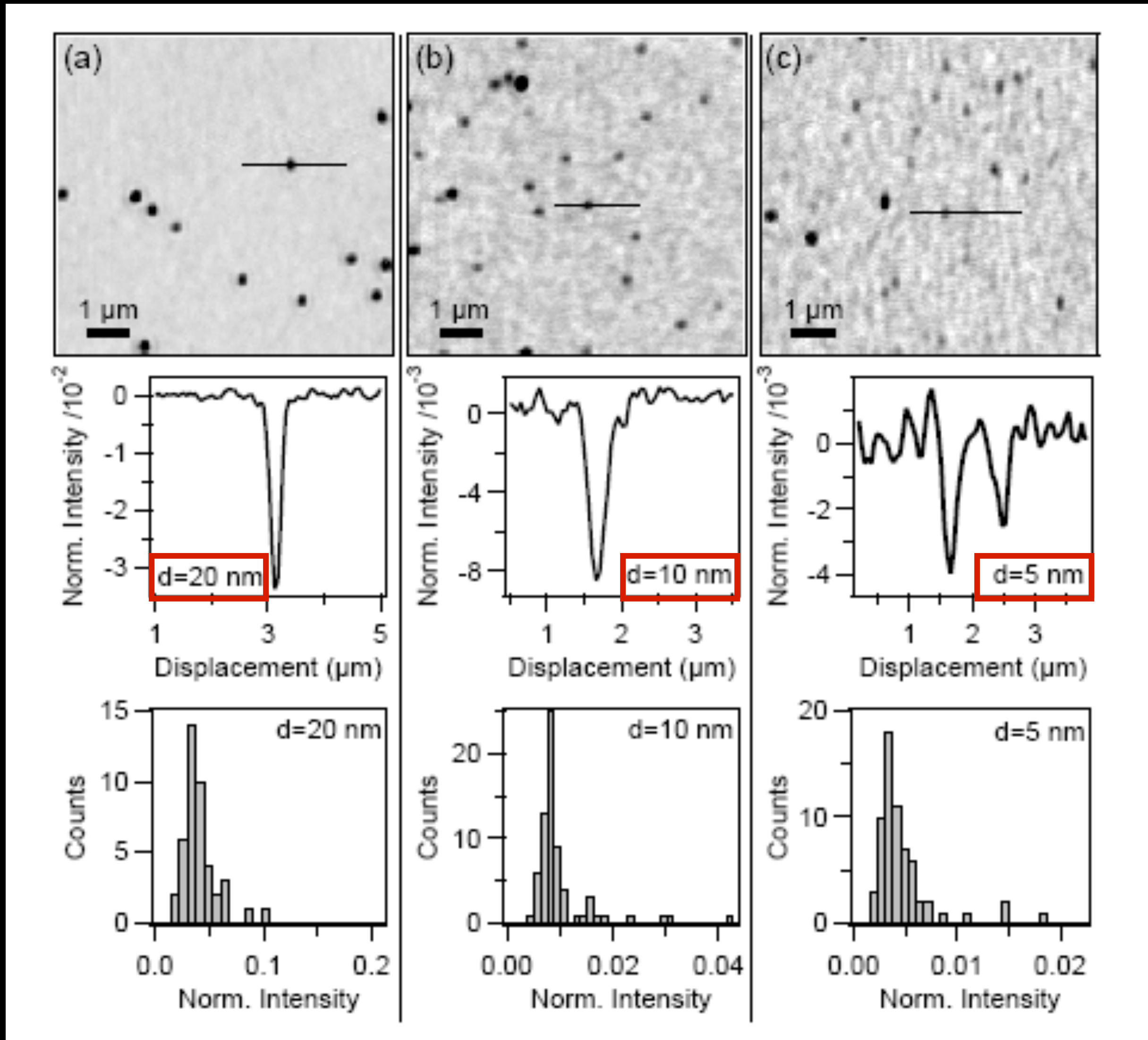
Ground-State Depletion Microscopy: Detection Sensitivity of Single-Molecule Optical Absorption at Room Temperature

Shasha Chong,[‡] Wei Min,[†,‡] and X. Sunney Xie*

J. Phys. Chem. Lett. 2010, *1*, 3316–3322

Scattering without absorption

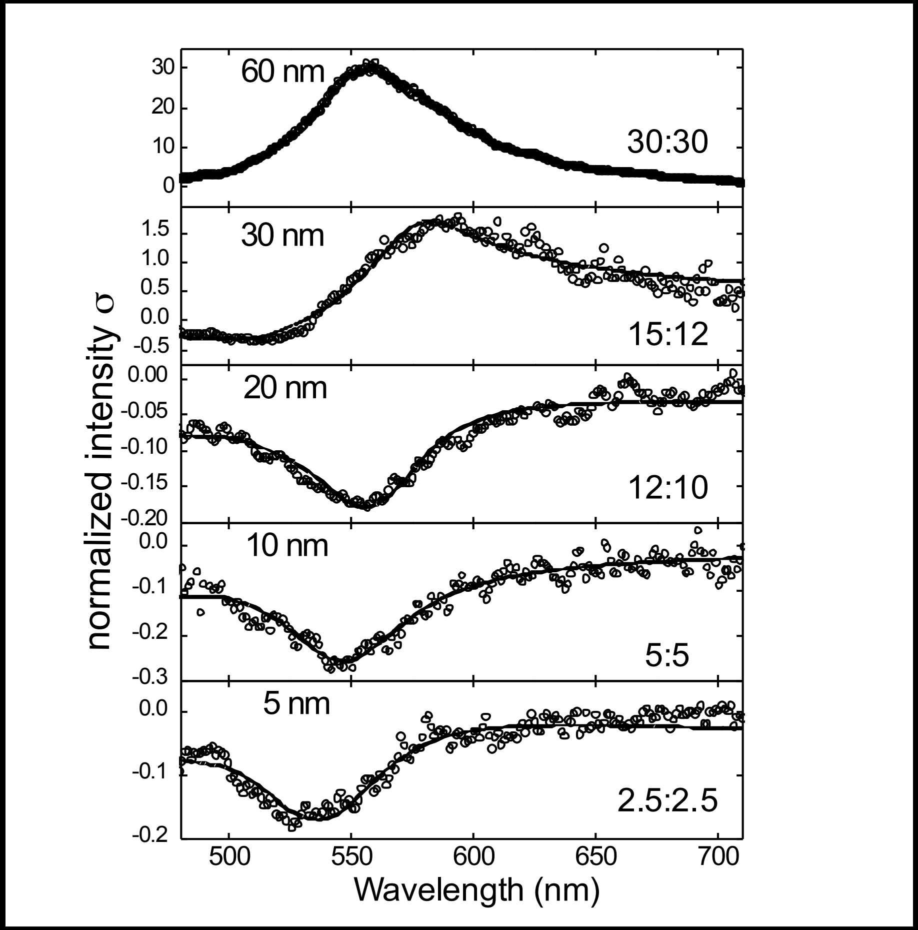
Detection of gold nanoparticles as small as 5 nm



V. Jacobsen, P. Stoller, C. Brunner, V. Vogel, V. Sandoghdar, *Opt. Exp.* **14**, 405 (2006).

Plasmon Spectra of Single Gold Nanoparticles

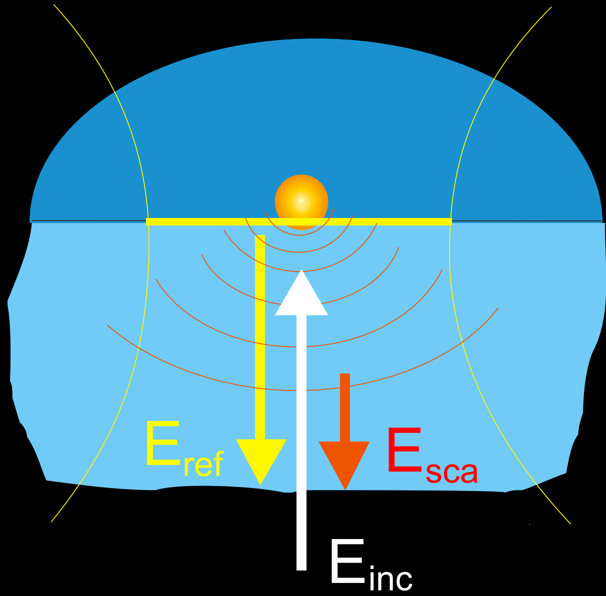
Excitation by quasi-white light from photonic crystal fibers



K. Lindfors, T. Kalkbrenner, P. Stoller, & V. Sandoghdar, *PRL* **93**, 037401 (2004).

P. Stoller, V. Jacobsen, V. Sandoghdar, *Opt. Lett*. **31**, 2474 (2006).

Interferometric detection of *nonfluorescent* nanoparticles



$$E_{ref} \sim r e^{-i\pi/2} E_{inc}$$

polarizability

$$E_{sca} = sE_{inc} \sim D^3 \frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m} \frac{e^{ikR}}{R} E_{inc}$$

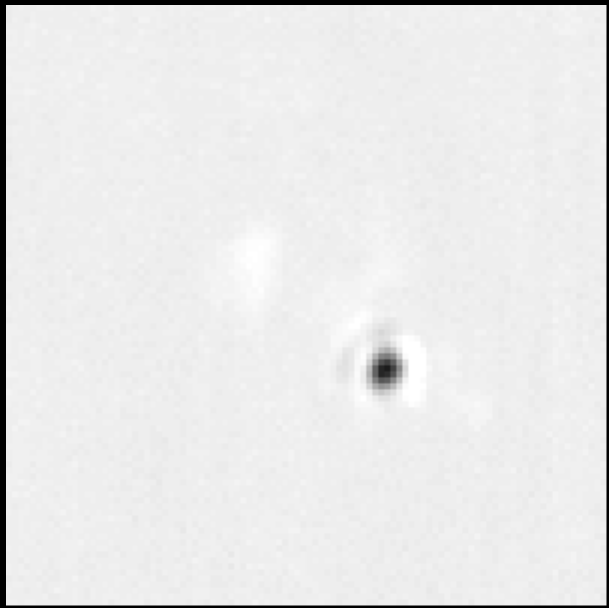
φ: scattering phase: $s = |s| e^{i\varphi}$

scales as D^3

$$I_{det} = |E_{ref} + E_{sca}|^2 = |E_{inc}|^2 \left\{ r^2 + |s|^2 - 2r|s|\sin\varphi \right\}$$

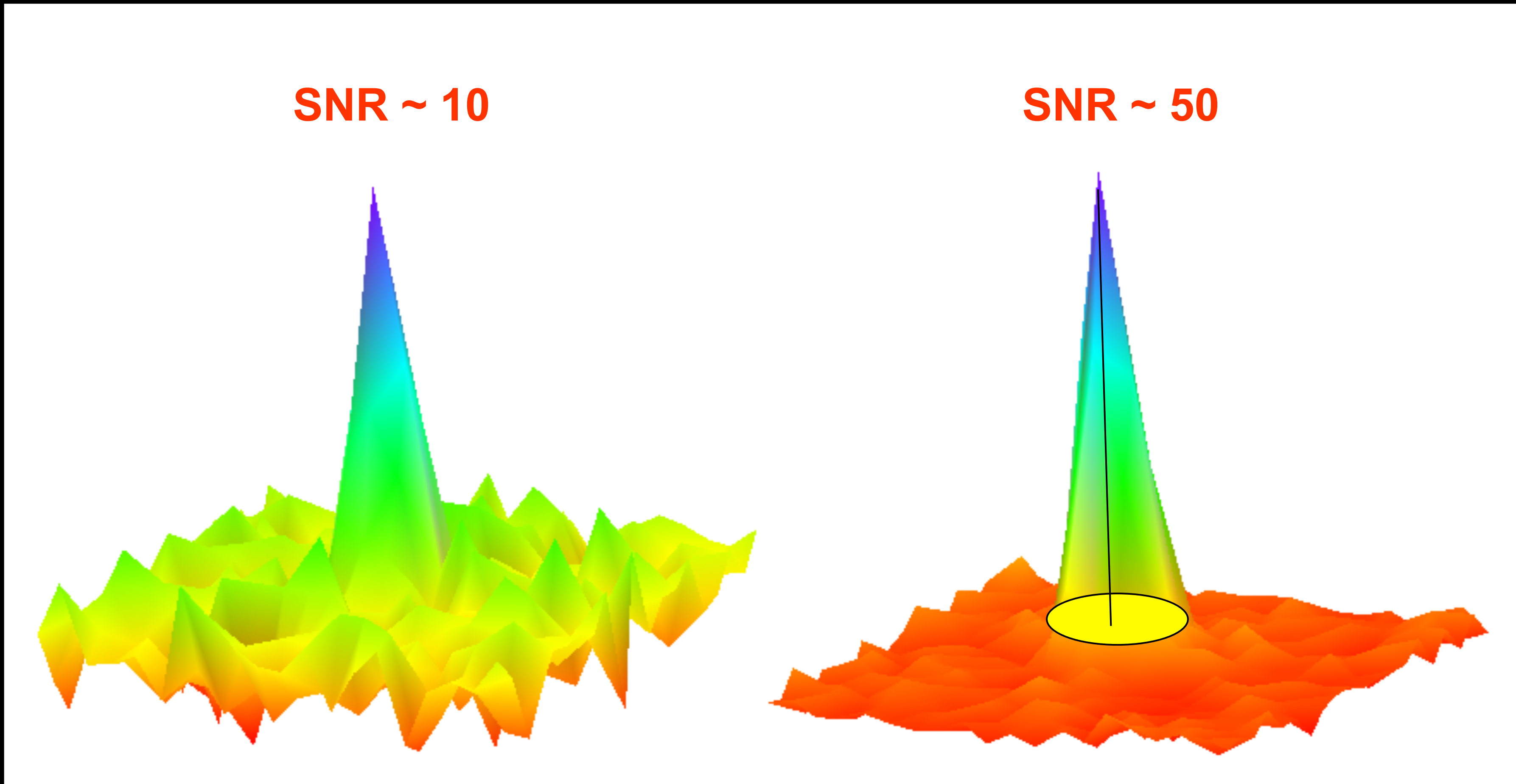
K. Lindfors, T. Kalkbrenner, P. Stoller, & V. Sandoghdar, *PRL* **93**, 037401 (2004).

Free diffusion of gold nanoparticles on supported membranes



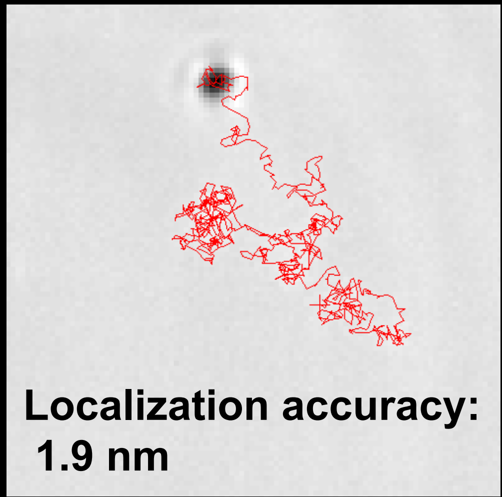
40nm AuNPs diffusing on a DOPC supported membrane (0.8% GM1)
Labeling scheme: AuNP – CTxB – GM1 lipid molecule
Frame rate: 200 Hz (video is 10x slow)
Field of view: 6.3 x 6.3 μm^2

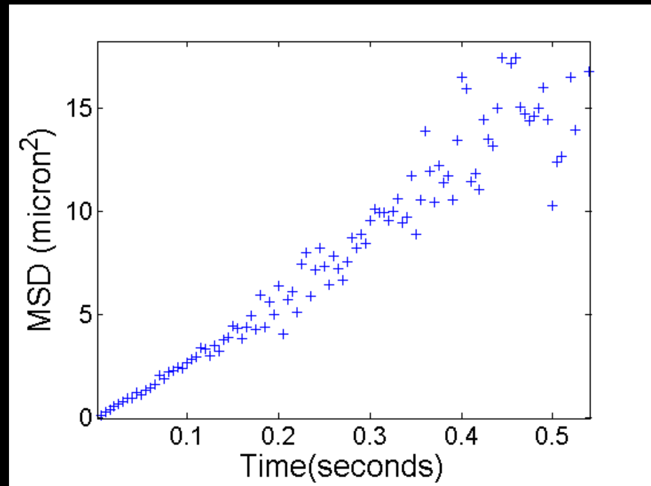
Accuracy in localizing a single nano-object



Localization accuracy: 10 nm

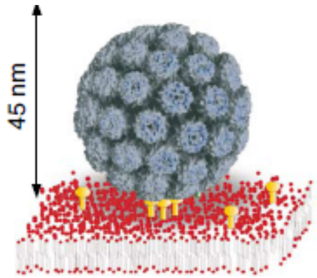
Localization accuracy: 2 nm

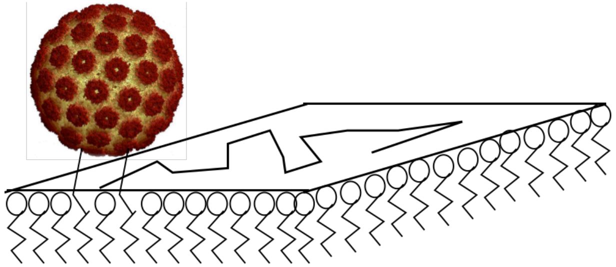


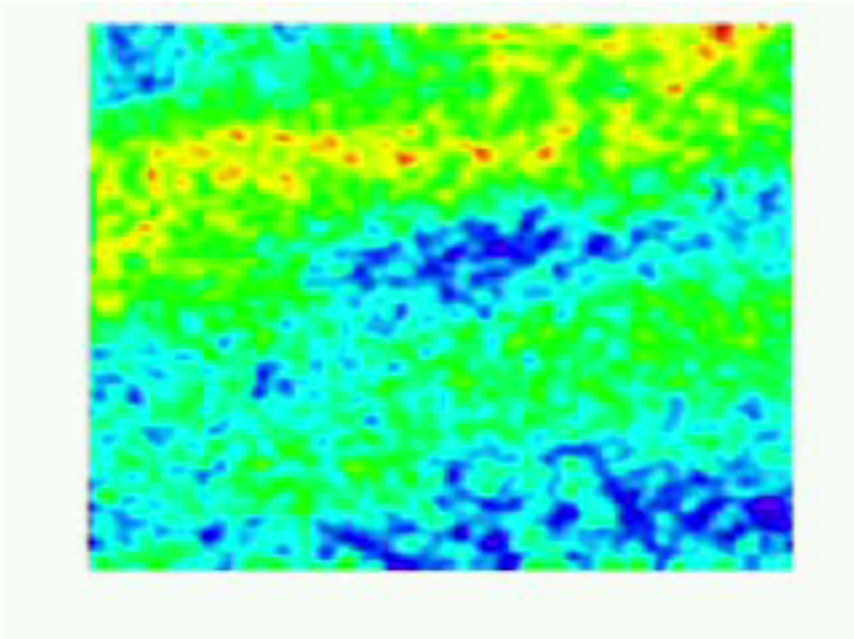


Diffusion coefficient: 8.32 μm^2/sec

Interferometric Tracking of a single *naked* virion on a supported bilayer membrane containing GM1 receptors





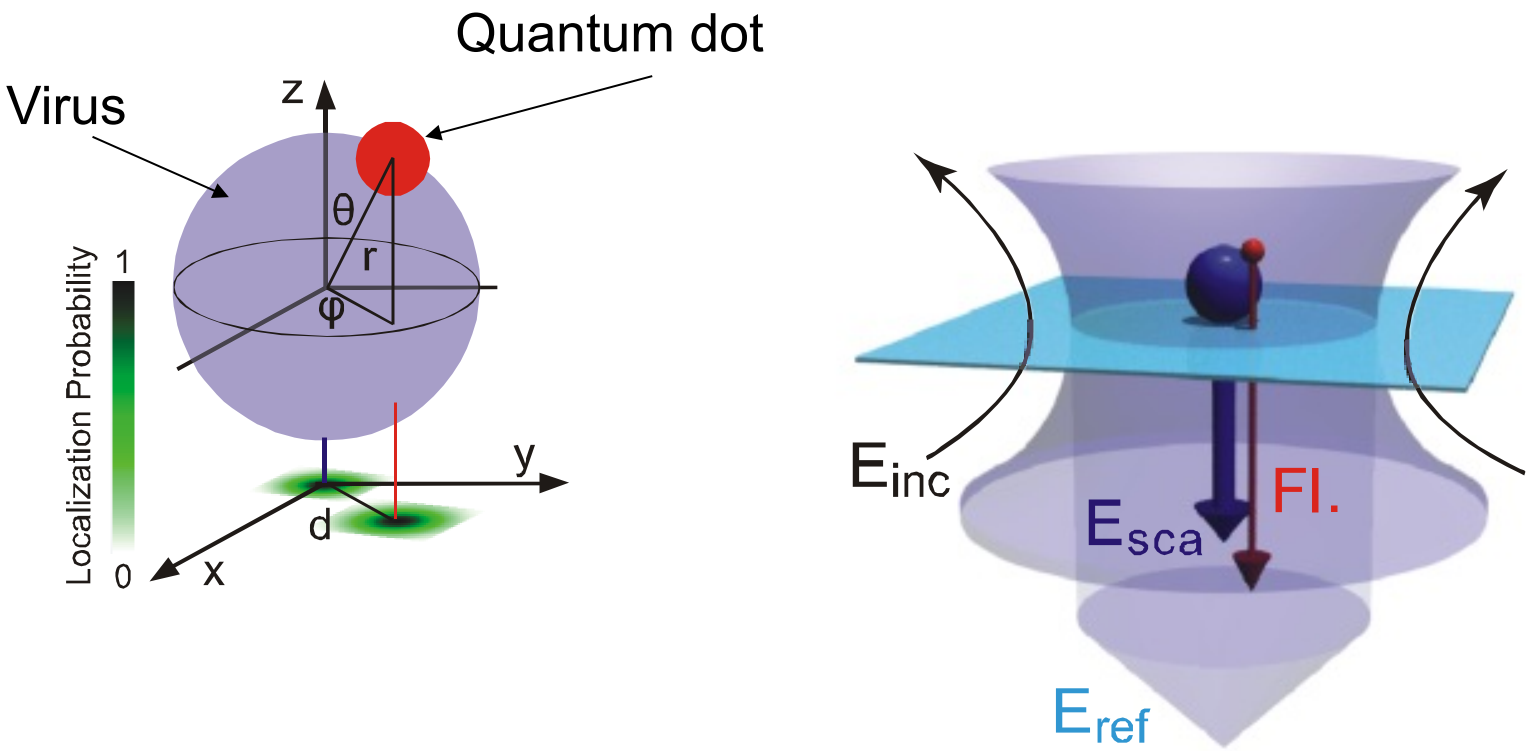


H. Ewers, V. Jacobsen, E. Klotzsch, A. Smith, A. Helenius, V. Sandoghdar, *Nano Lett.* **7**, 2263 (2007).

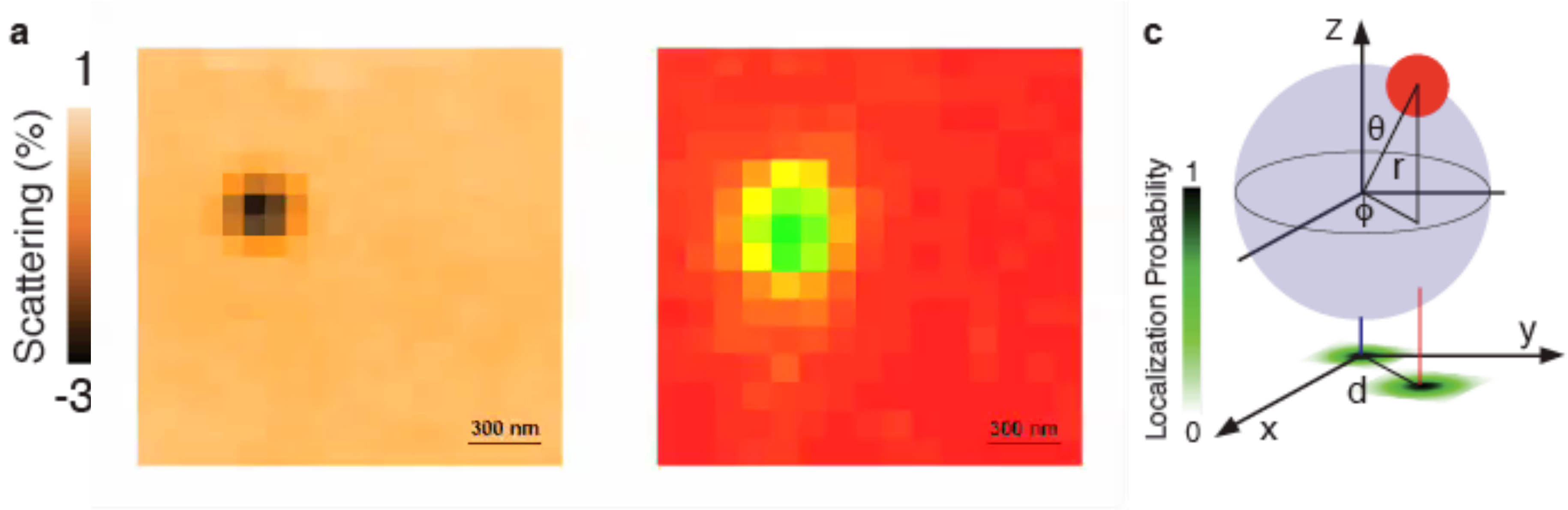
P. Kukura, H. Ewers, C. Müller, A. Renn, A. Helenius, V. Sandoghdar *Nature Methods*, **6**, 923 (2009);

How does a virus move on the membrane: rolls or slides?

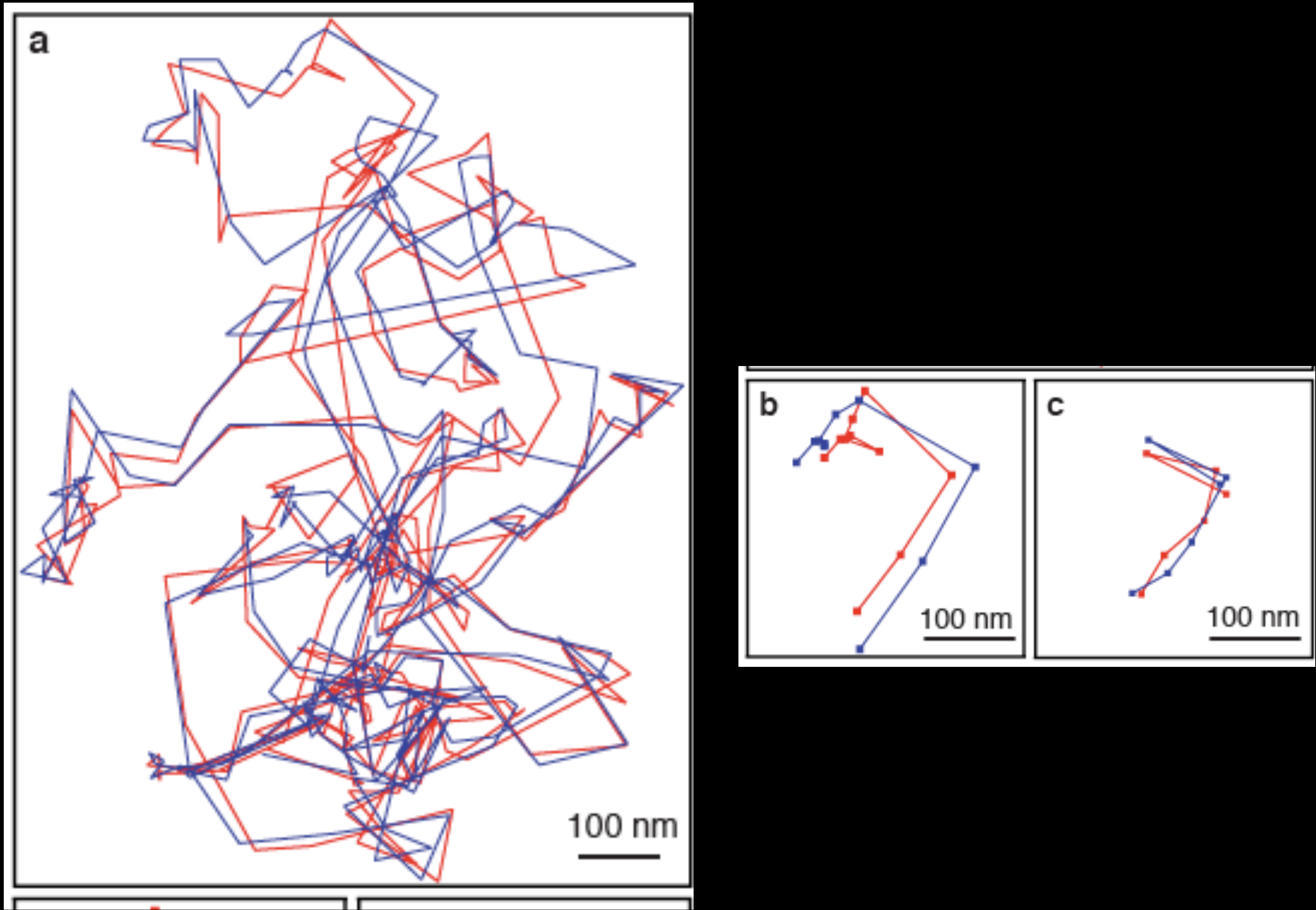
Translational & orientational nano-motion of a virus



Colocalization of scattering and fluorescence



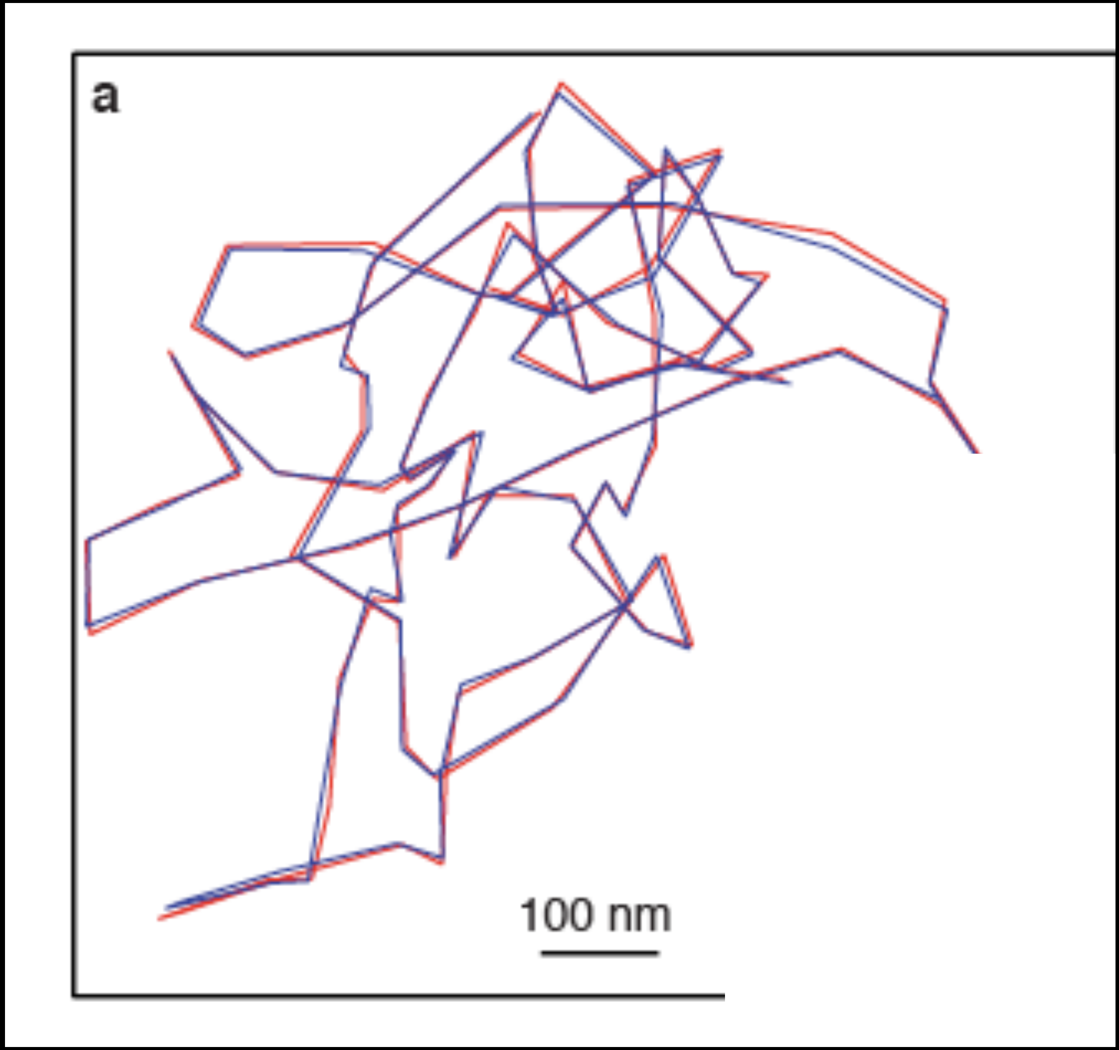
Simultaneous trajectories of the virus center of mass and of the quantum dot: *0.05% receptor concentration*



The virus dance

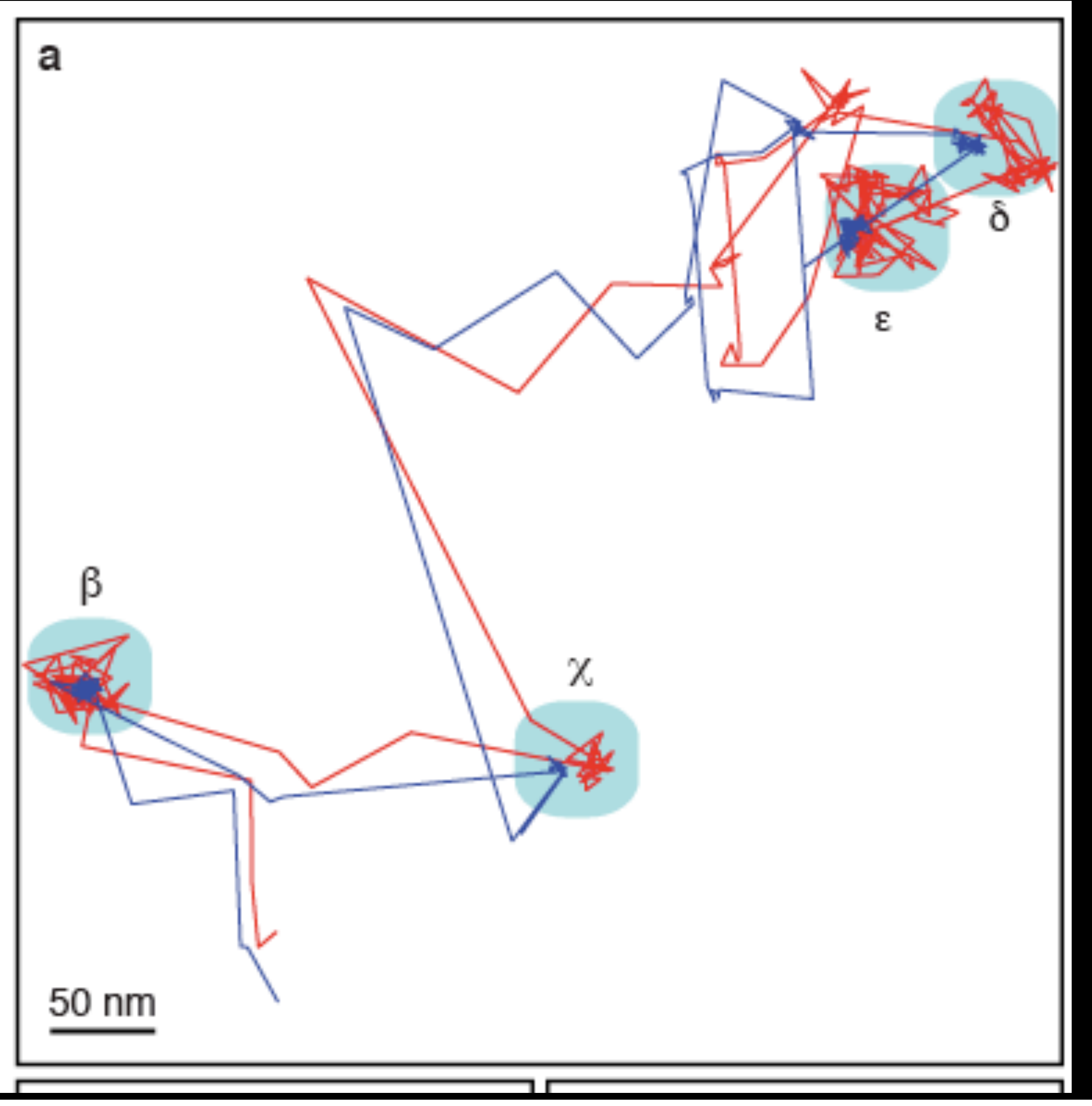
Checking

Fluo and scattering trajectories of a homogeneously doped bead



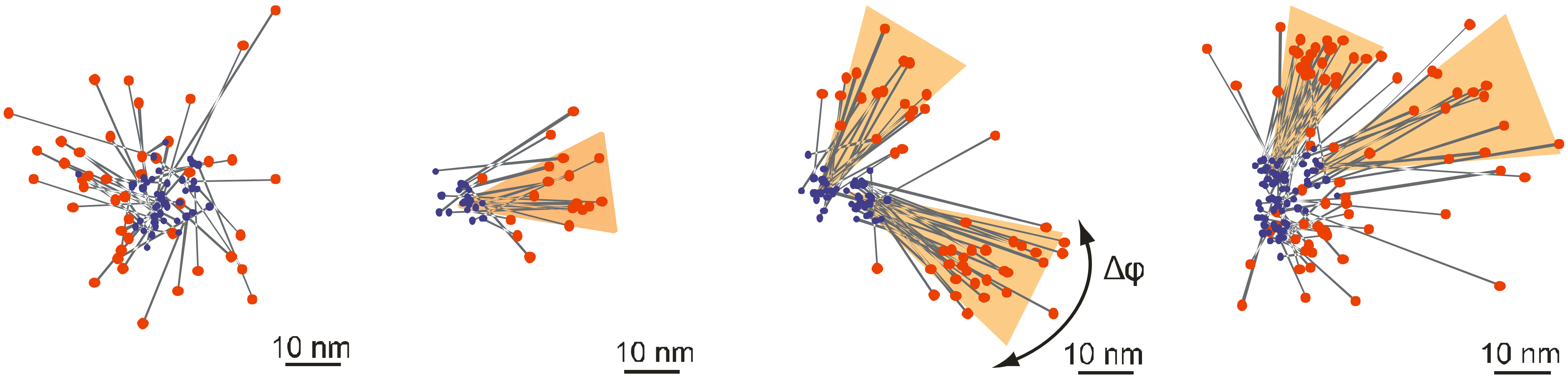
Fluo and scattering „trajectories“ of an immobilized Qdots-labeled virus

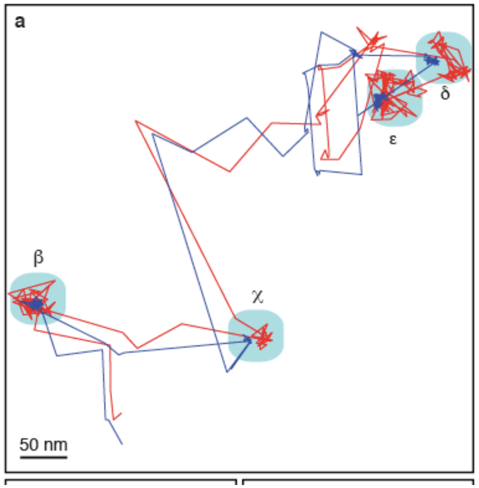
Simultaneous trajectories of the virus center of mass and of the quantum dot: *1% receptor concentration*

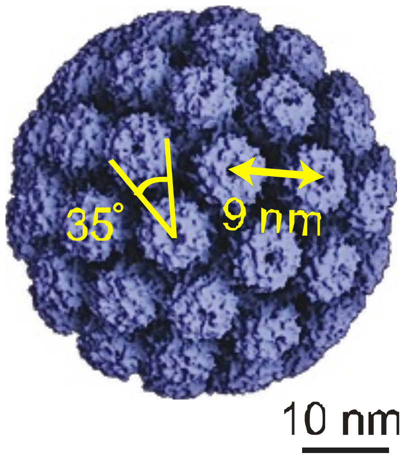


Resolving the nanomotion of the virus

Sliding, tumbling, rocking of a single virus

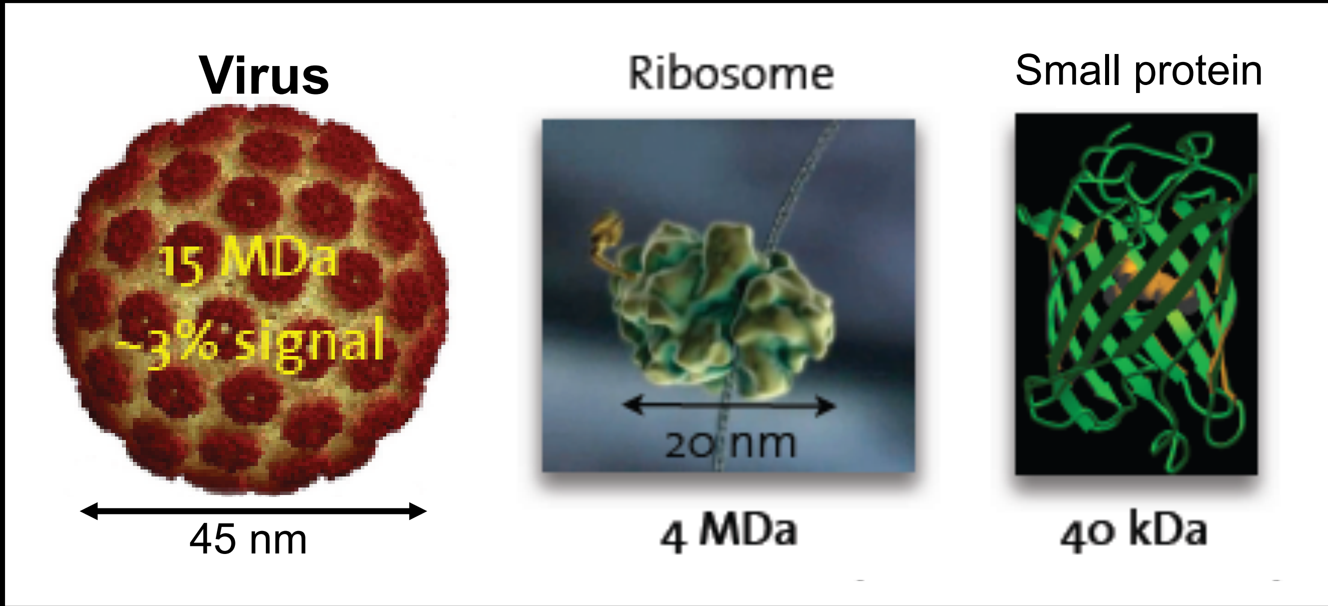






P. Kukura, H. Ewers, C. Müller, A. Renn, A. Helenius, V. Sandoghdar
Nature Methods, **6**, 923 (2009);

Direct detection of smaller biological nano-objects: *no absorption, no fluorescence*



We are close to the detection of a single protein.

Goal: *Detect and image single nonfluorescent atoms on a surface*

Conclusions

Direct detection of single nanoparticles and molecules using LIGHT and *without fluorescence* is now possible

Focused light is one of the most sensitive sensors of matter