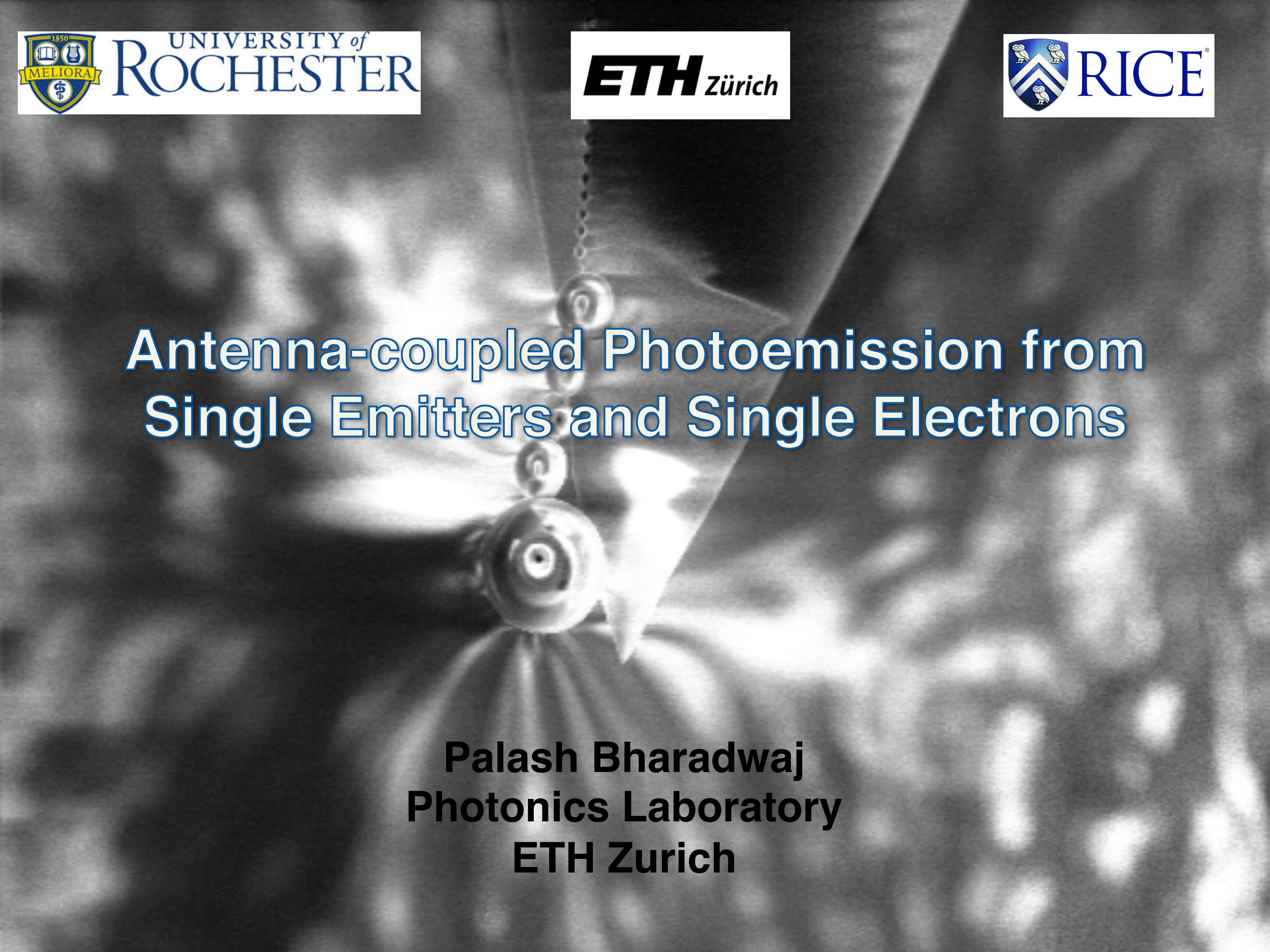


Antenna-coupled Photoemission from Single Emitters and Single Electrons

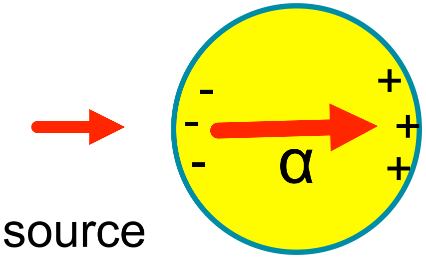
Palash Bharadwaj
Photonics Laboratory
ETH Zurich

Yesterday…

$$\boxed{\rho_{\mathbf{n}}(\boldsymbol{r}_0,\omega)=\frac{6\omega n^2}{\pi c^2}\left\{\boldsymbol{n}_p^{\intercal}\,\mathrm{Im}\left[\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r}_0,\boldsymbol{r}_0;\omega)\right]\boldsymbol{n}_p\right\}}$$

LDOS

$$\boxed{P\propto\left|\boldsymbol{p}_{\mathrm{ind}}\right|^2\propto\frac{|\alpha|^2}{d^6}}$$



Optical Antennas…

- Modulate LDOS on sub-λ length scale
- Can boost decay rates of quantum emitters
- Can direct the emission of quantum emitters

Menu for Today

Plasmonics with Single Emitters and Electrons
Optical antennas
Scanning probe optical microscopy
Single molecules, colloidal quantum dots, Nanotubes

Electrically Active Devices
Tunnel diodes
Transistors

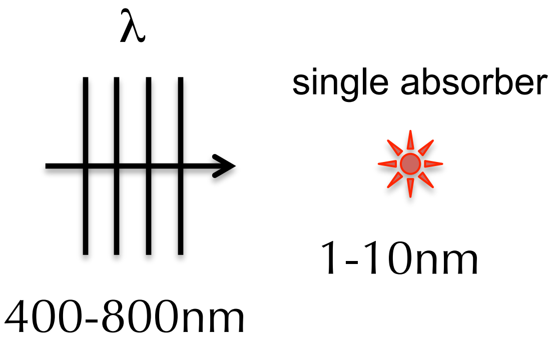
Novel Optical Materials
Layered 2D materials
e. g. MoS_2, h-BN, graphene

1. Optical antennas; fluorescence enhancement & localization
2. 2D semiconductor optoelectronics
3. Electrically excited optical antennas

Part I

Plasmonic Optical Antennas
for
Single Molecule Fluorescence Enhancement, Localization and Imaging

Why Optical Antennas?

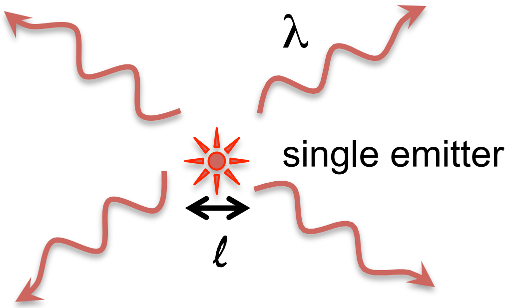


λ

single absorber

1-10nm

400-800nm



λ

single emitter

ℓ

$$\Gamma_{abs} \propto |\mu \cdot \mathbf{E}|^2$$

$$\sigma_{abs} \sim 1\text{nm} \times 1\text{nm}$$

Small Absorption Cross-section

$$\Gamma_{rad} \propto \left(\frac{l}{\lambda}\right)^2$$

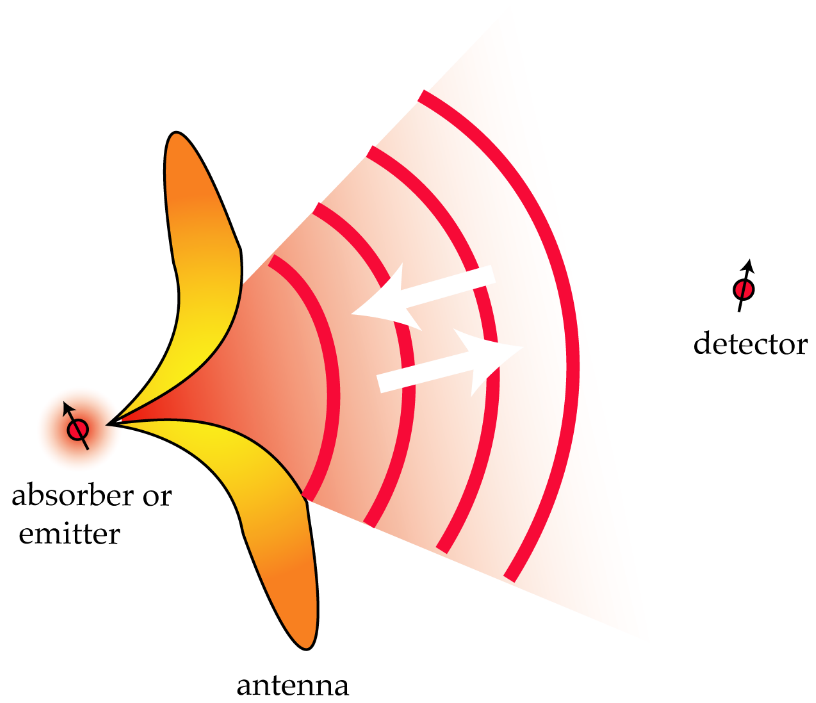
$$\mathrm{R}_{rad} \sim 10^{-2}\ \Omega$$

Small Radiation Resistance

What is an Optical Antenna?

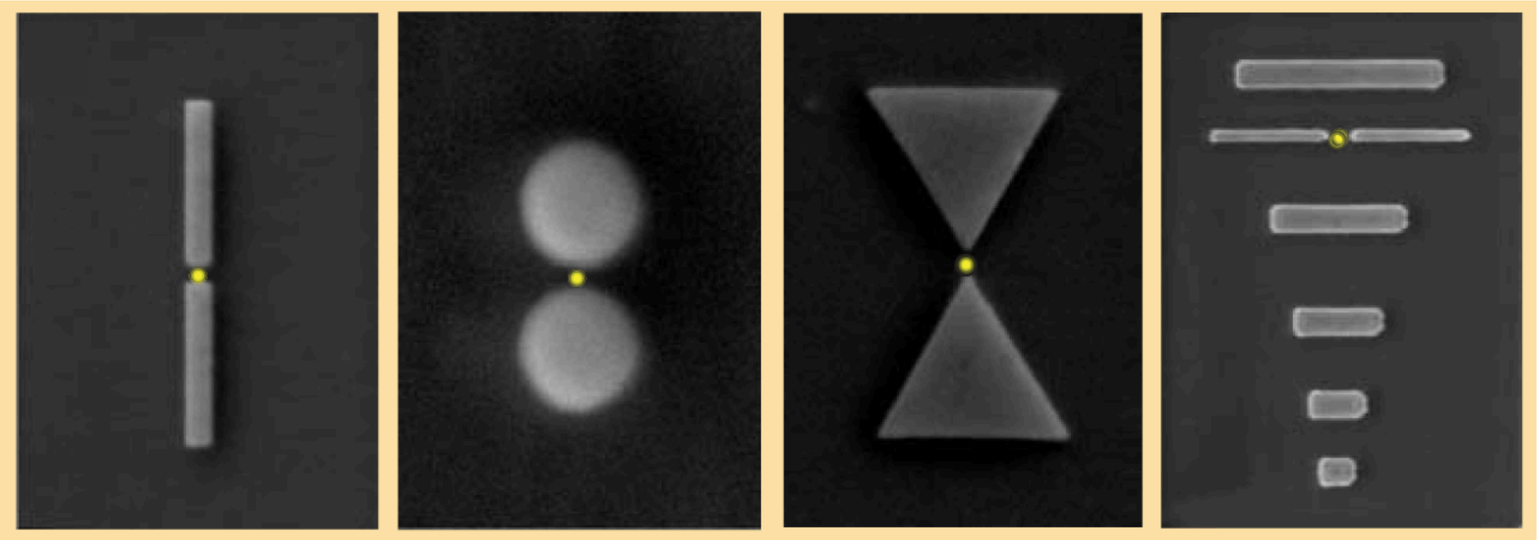
Optical Antenna: "Device that efficiently converts between farfield (propagating) optical radiation and nearfield (localized) energy"

Bharadwaj *et al, Adv. Opt. Photon*., **103**, 186101 (2009).



Antenna **increases** both the **absorption cross-section** and the **radiation resistance**

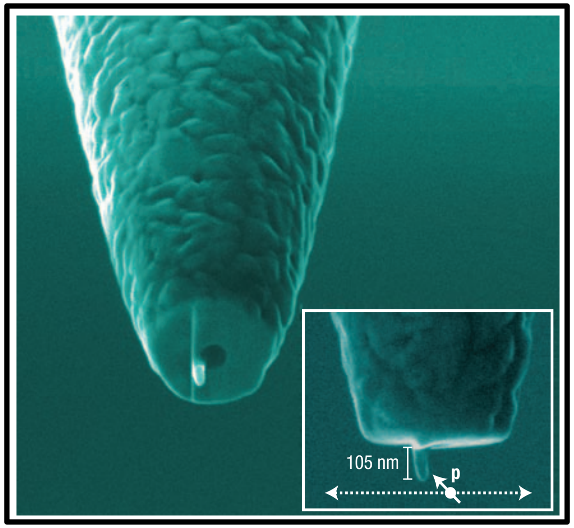
Realizations of Optical Antennas



Dipole antenna Hertzian dimer Bow-tie Log periodic array

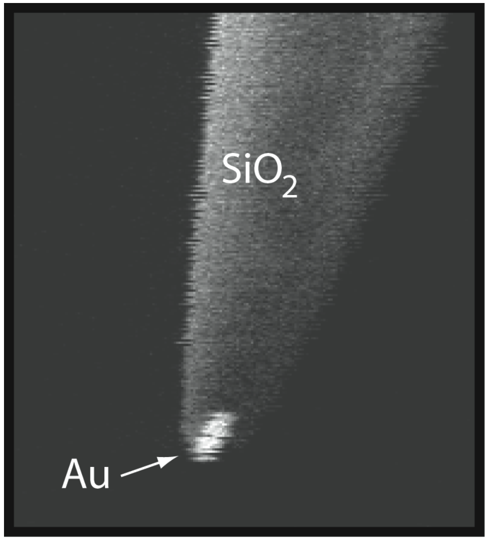
L. Novotny, *Physics Today*, p. 47, July 2011.

Optical Antennas on Scanning Probes



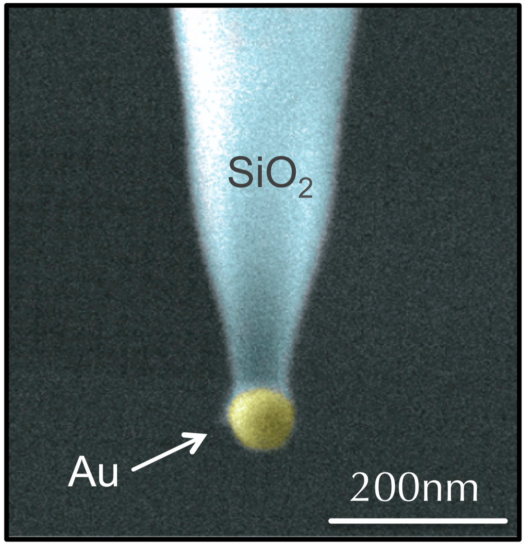
Grounded monopole antenna

Taminiau, Nat. Phot., **329**, 93 (2010)



Au nanorod (20nm x 60nm)

Dipole Antenna



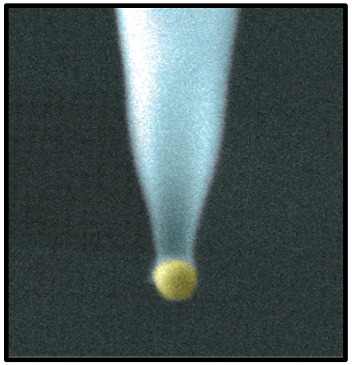
80nm Au nanoparticle

Top-down fabrication

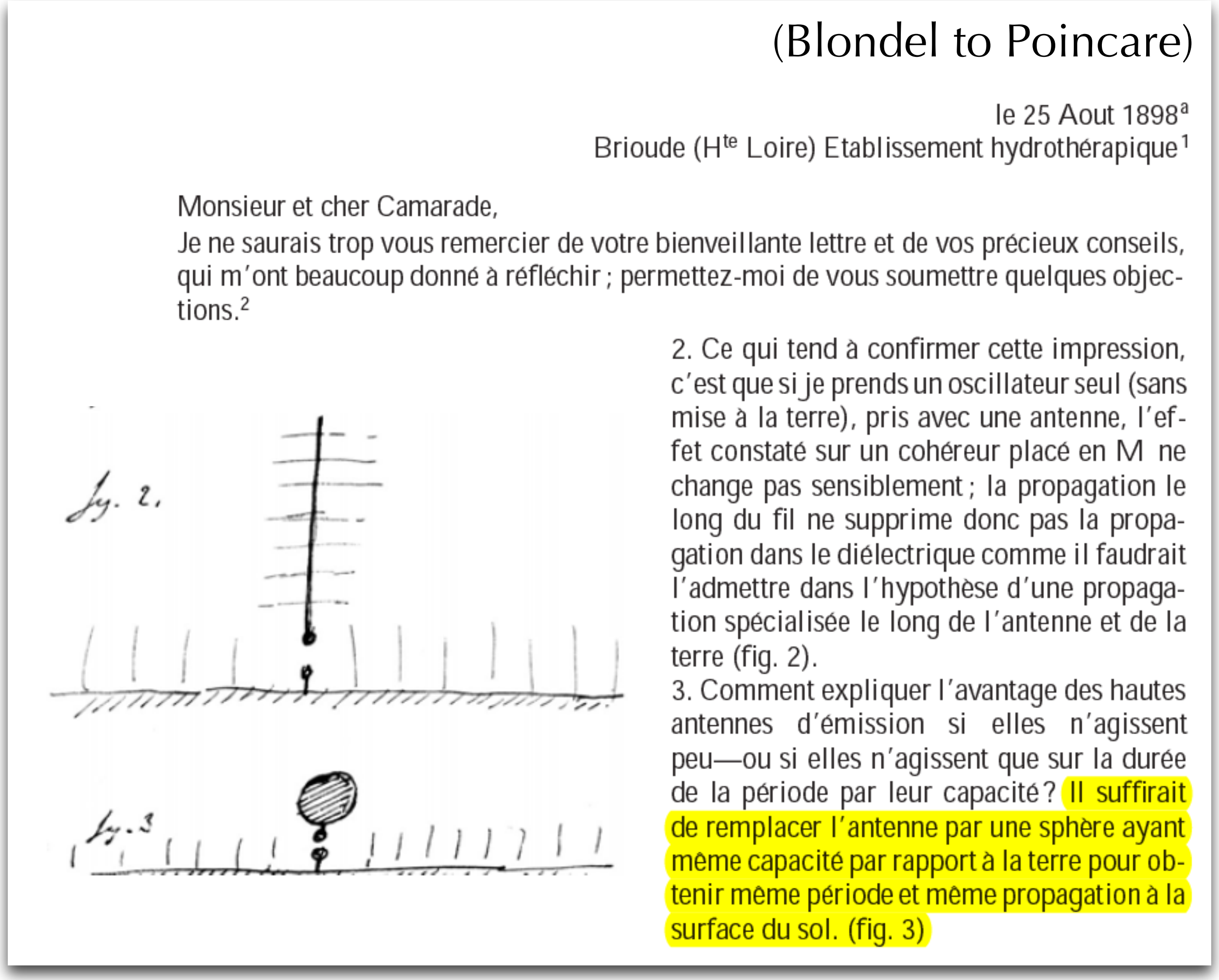
Bottom-up fabrication

Andre Blondel

Andre Blondel



"Blondel Antenna"



(Blondel to Poincare)

le 25 Aout 1898[a]
Brioude (H^{te} Loire) Etablissement hydrothérapique[1]

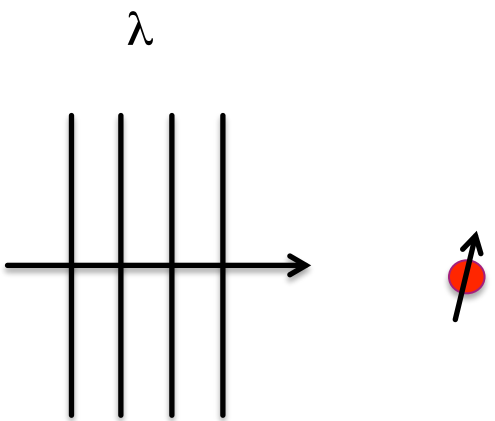
Monsieur et cher Camarade,
Je ne saurais trop vous remercier de votre bienveillante lettre et de vos précieux conseils, qui m'ont beaucoup donné à réfléchir ; permettez-moi de vous soumettre quelques objections.[2]

2. Ce qui tend à confirmer cette impression, c'est que si je prends un oscillateur seul (sans mise à la terre), pris avec une antenne, l'effet constaté sur un cohéreur placé en M ne change pas sensiblement ; la propagation le long du fil ne supprime donc pas la propagation dans le diélectrique comme il faudrait l'admettre dans l'hypothèse d'une propagation spécialisée le long de l'antenne et de la terre (fig. 2).

3. Comment expliquer l'avantage des hautes antennes d'émission si elles n'agissent peu—ou si elles n'agissent que sur la durée de la période par leur capacité ? Il suffirait de remplacer l'antenne par une sphère ayant même capacité par rapport à la terre pour obtenir même période et même propagation à la surface du sol. (fig. 3)

La Correspondance d'Henri Poincaré, Volume 2, p. 32, Basel: Birkhäuser, (2007)

Antenna-emitter Interaction



$$\Gamma^o_{exc} \propto |\mu \cdot \mathbf{E}|^2$$

$$\Gamma^o_{fl} = \Gamma^o_{exc}\, Q^o$$

where $Q^o = \frac{\Gamma^o_r}{\Gamma^o_r + \Gamma^o_{nr}}$

$$\frac{\Gamma_{fl}}{\Gamma^o_{fl}} = \frac{\Gamma_{exc}}{\Gamma^o_{exc}}\,\frac{Q}{Q^o}$$

$$= \frac{\Gamma_{exc}}{\Gamma^o_{exc}}\,\frac{\Gamma_r}{\Gamma_r + \Gamma^o_{nr} + \Gamma_{abs}}\,\frac{\Gamma^o_r + \Gamma^o_{nr}}{\Gamma^o_r}$$

$$= \left(\frac{\Gamma_{exc}}{\Gamma^o_{exc}}\right)\left(\frac{\Gamma_r}{\Gamma^o_r}\right)\left(\frac{\Gamma^o_r + \Gamma^o_{nr}}{\Gamma_r + \Gamma^o_{nr} + \Gamma_{abs}}\right)$$

Antenna-emitter Interaction

$$\frac{\Gamma_{fl}}{\Gamma_{fl}^{o}} = \left(\frac{\Gamma_{exc}}{\Gamma_{exc}^{o}}\right)\left(\frac{\Gamma_{r}}{\Gamma_{r}^{o}}\right)\left(\frac{\Gamma_{r}^{o}+\Gamma_{nr}^{o}}{\Gamma_{r}+\Gamma_{nr}^{o}+\Gamma_{abs}}\right)$$

“good” emitter

$$Q^{o} \approx 1 \equiv \Gamma_{nr}^{o} \ll \Gamma_{r}^{o}, \text{ and } \Gamma_{abs} \approx 0$$

$$\frac{\Gamma_{fl}}{\Gamma_{fl}^{o}} = \frac{\Gamma_{exc}}{\Gamma_{exc}^{o}}$$

excitation enhancement
(abs cross-section ↑)

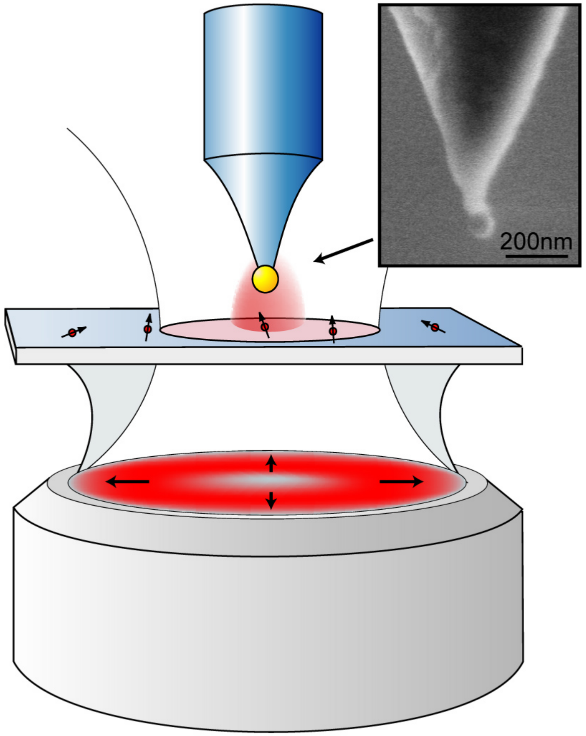
“poor” emitter

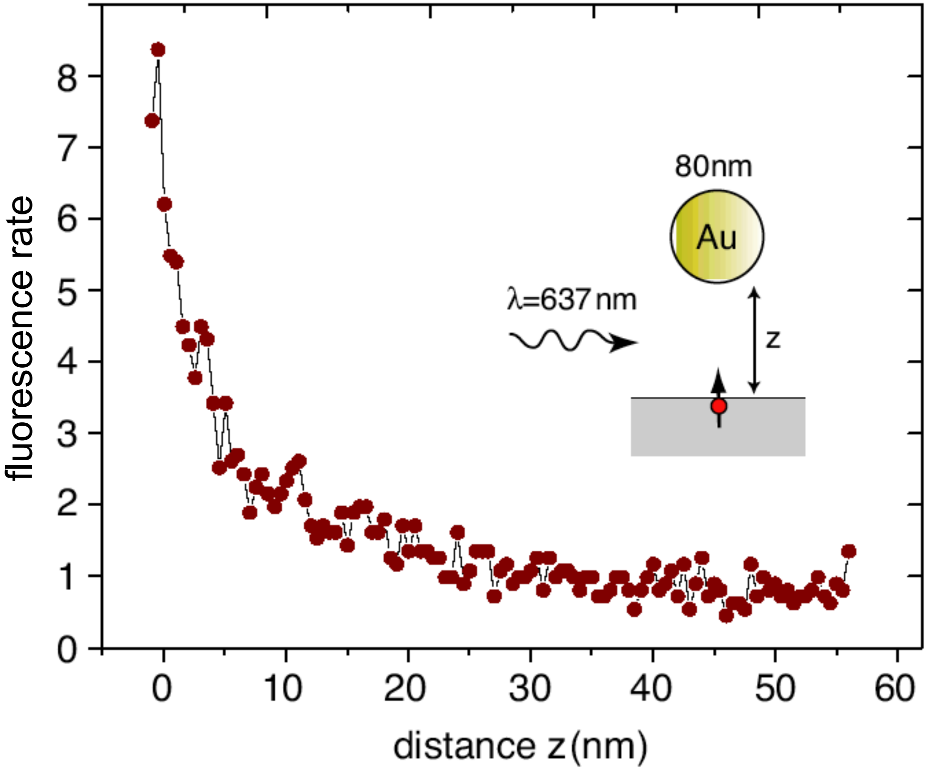
$$Q^{o} \approx 0 \equiv \Gamma_{nr}^{o} \gg \Gamma_{r}^{o}, \text{ and } \Gamma_{abs} \approx 0$$

$$\frac{\Gamma_{fl}}{\Gamma_{fl}^{o}} = \left(\frac{\Gamma_{exc}}{\Gamma_{exc}^{o}}\right)\left(\frac{\Gamma_{r}}{\Gamma_{r}^{o}}\right)$$

excitation + emission enhancement
(abs cross-section ↑ ***AND*** R_{rad} ↑)

High Q° molecule: Excitation Enhancement





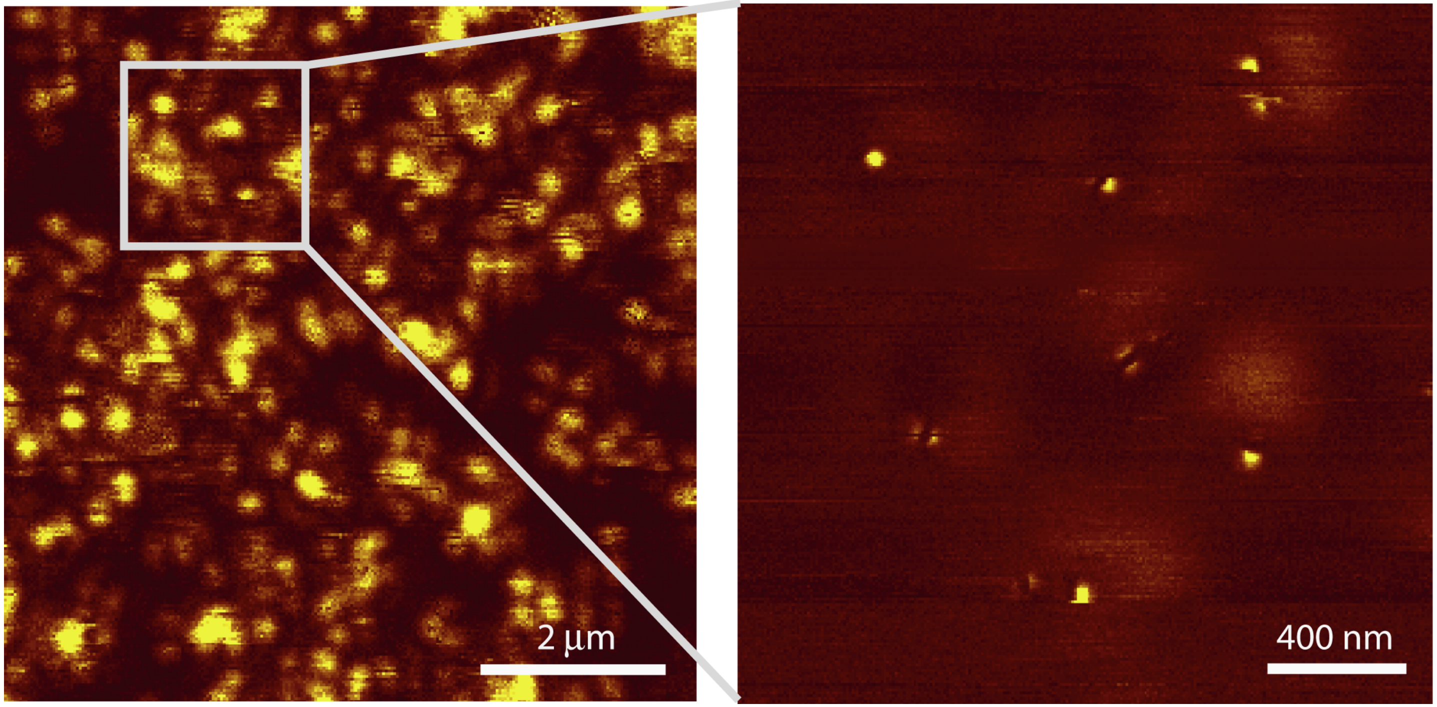
Nile blue molecules excited at 635nm

Phys. Rev. Lett. **96**, 113002 (2006)

Enhancement → Improved Imaging Resolution

without antenna

with Au antenna



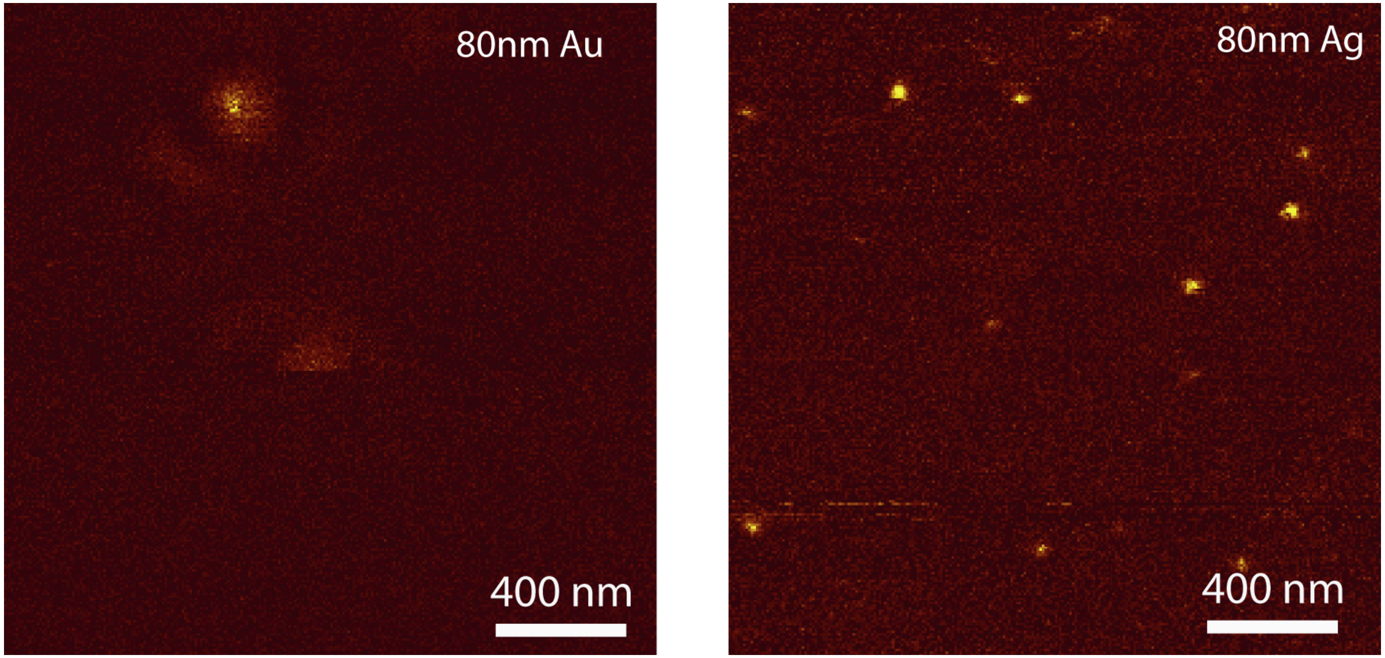
Fluorescence image of Nile blue molecules

Enhancement **~8x**
Resolution **~65nm** FWHM

Phys. Rev. Lett. **96**, 113002 (2006)

Different Antennas for Different Colors

Alexa488 molecules excited at 488nm

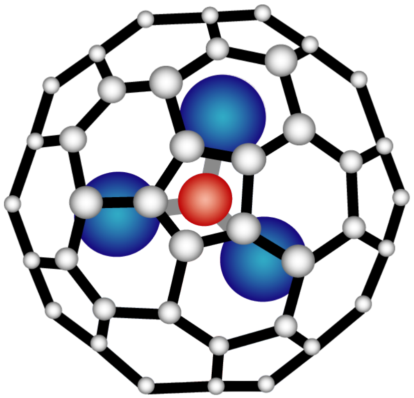


Enhancement ~**1x**

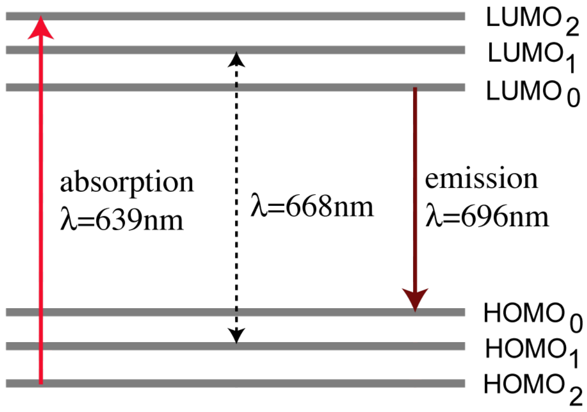
Enhancement **~15x**

Opt. Express **15**, 14266 (2007)

“Poor” Emitter (Low Q°): Excitation *and* Emission Enhancement



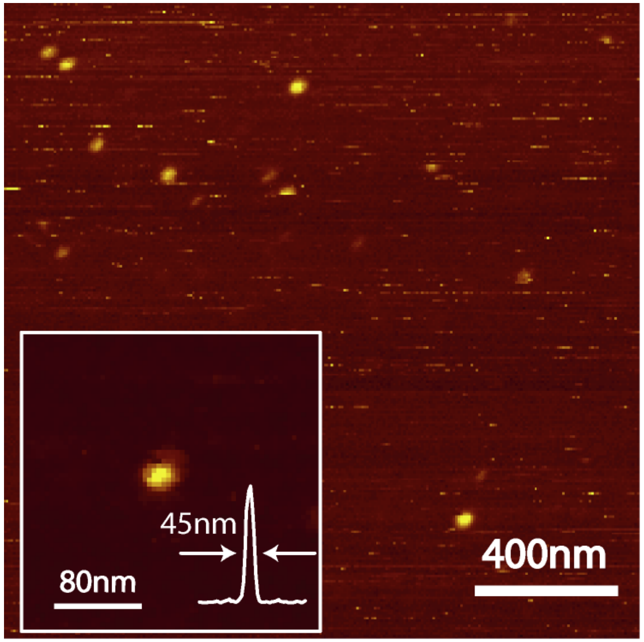
$Y_3N@C_{80}$
(Y-Trimetaspheres^{TM})



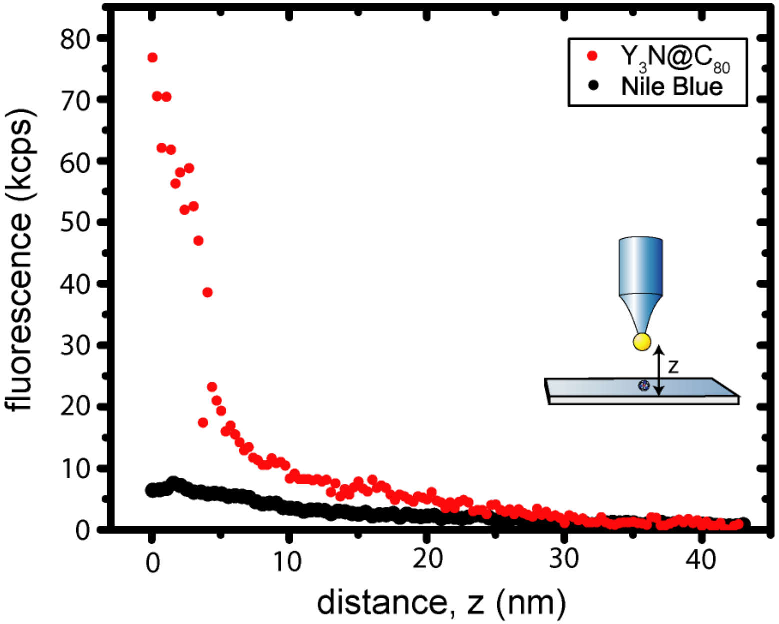
Quantum yield ~1%

Antenna Enhances Absorption AND Emission

Fluorescence map of $Y_3N@C_{80}$
with 80nm Au antenna



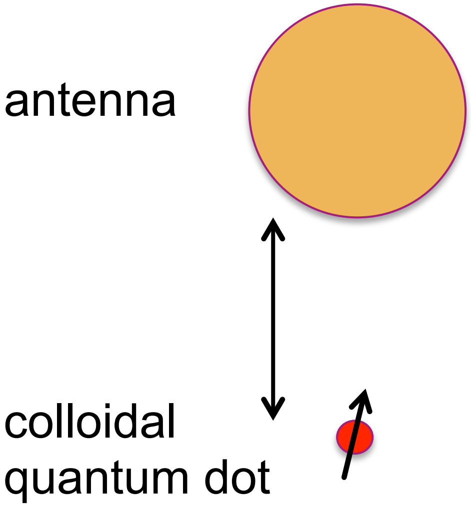
Huge enhancements of **>100x**

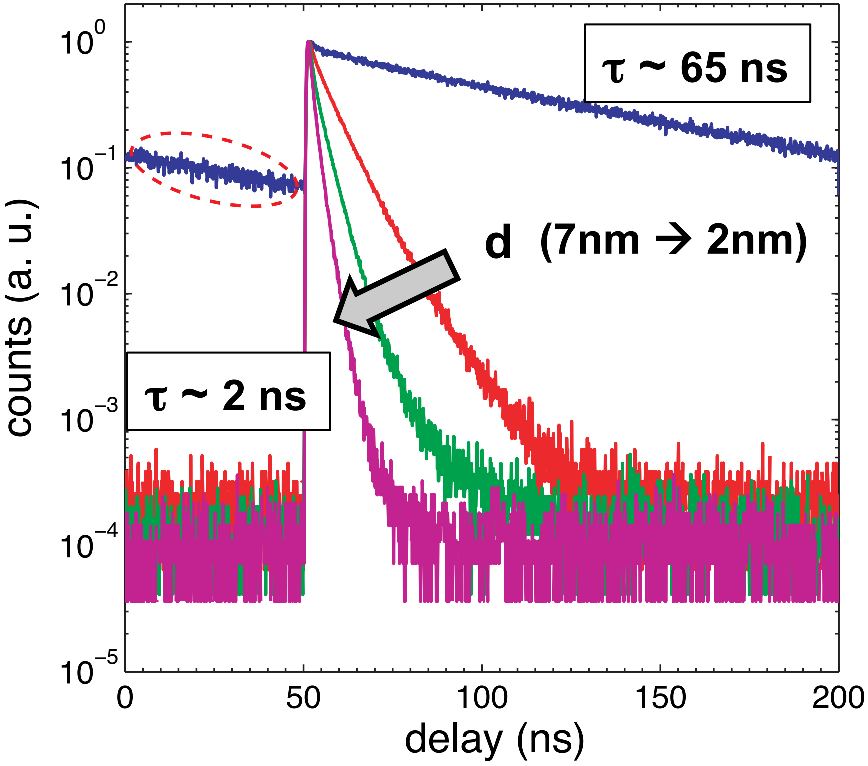


Both absorption (Γ_{exc}) ***and*** emission (Q) are enhanced!

J. Phys. Chem. C, ***114****,* 7444 (2010)

Antenna Shortens Lifetime → Faster Devices



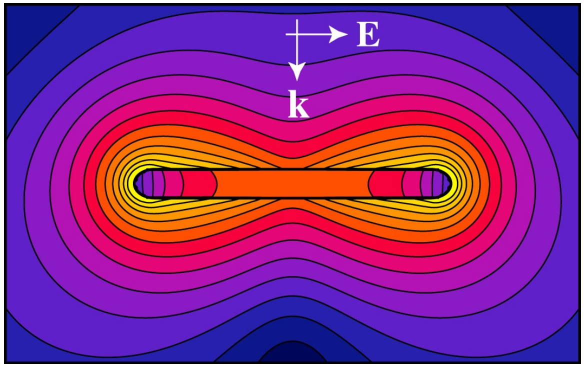


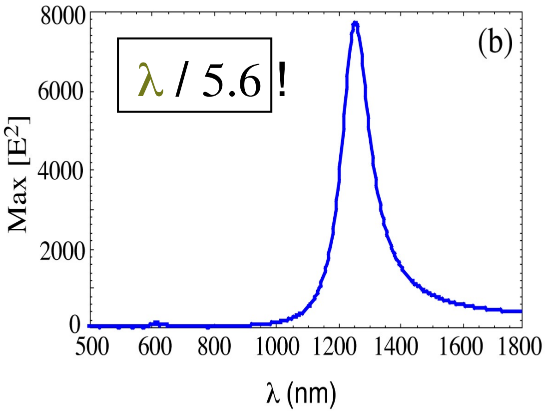
Nano Lett., **11**, 2137 (2011)

Non-spherical Antennas

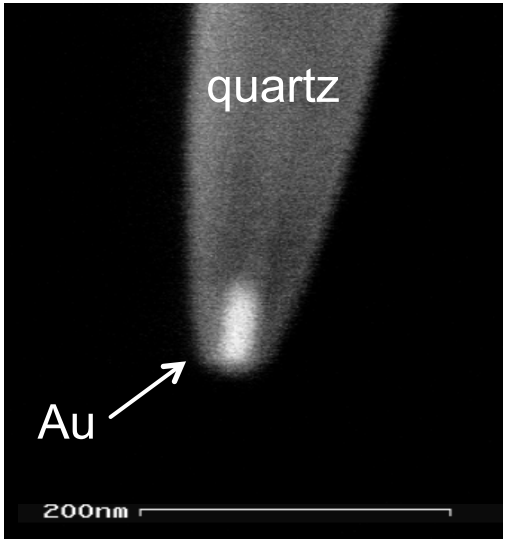
- Higher enhancements
- Better localization

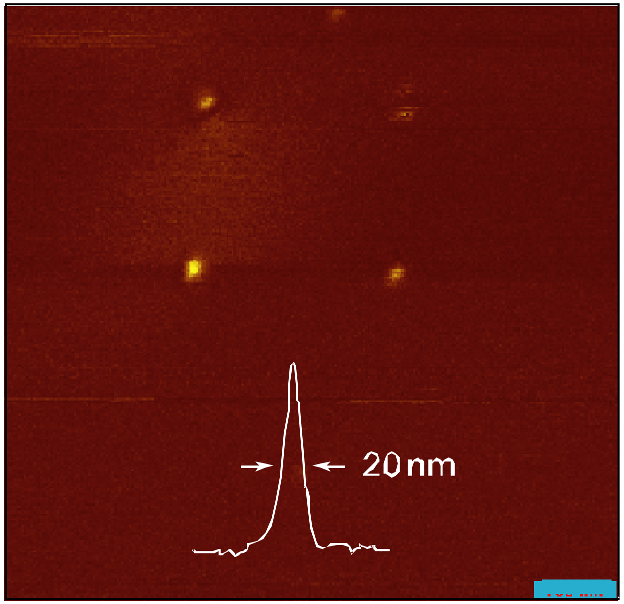
Nanorod Dipole Antennas





L. Novotny, *PRL*, **98**, 266802 (2007)





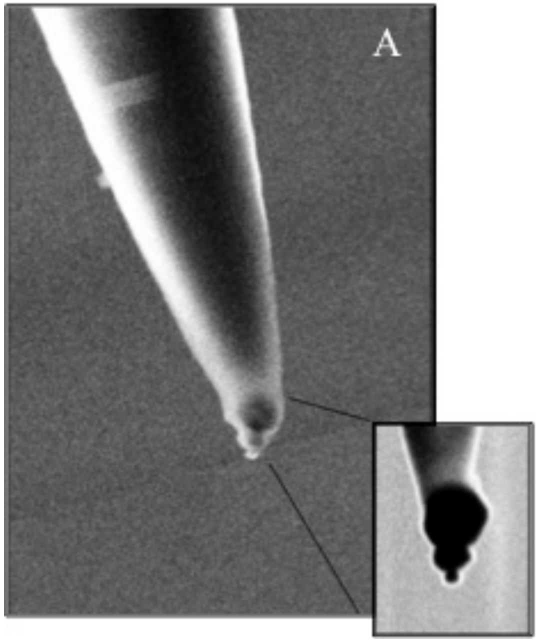
Fluorescence image of single molecules

20nm resolution, enhancement **15x**

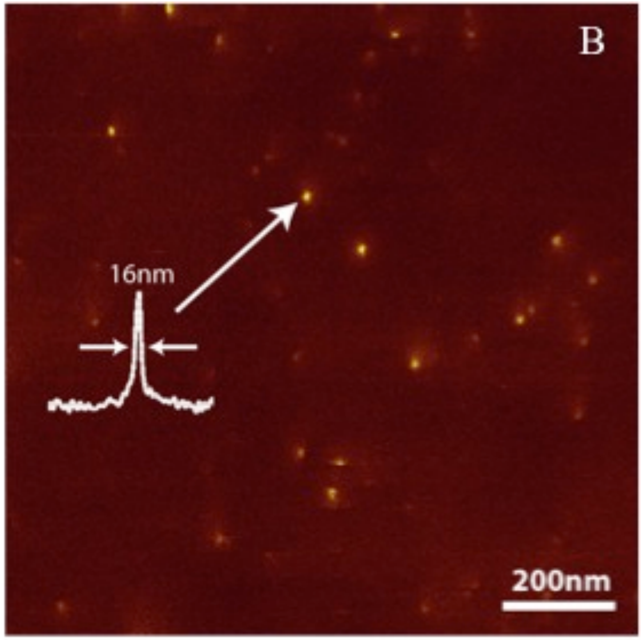
Chem. Sci. **2**, 136 (2011)

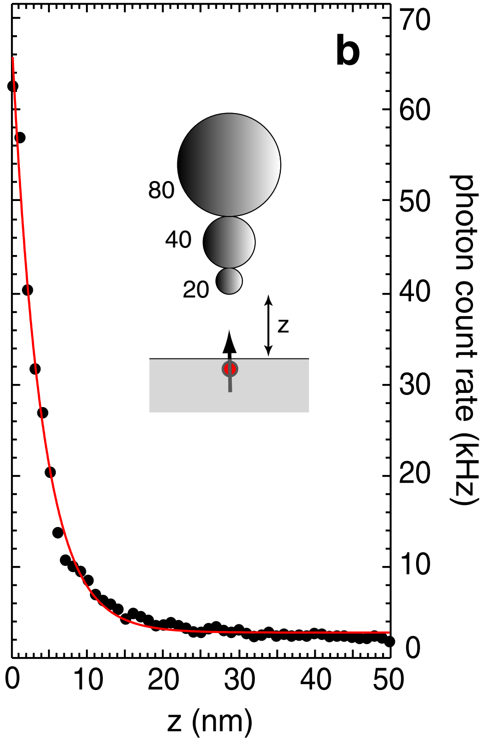
Multiparticle Antennas: 80/40/20nm Au Trimer

High Resolution + High Enhancement



Self-similar trimer antenna





~15nm resolution (~λ/40)
Fluorescence enhancement 40x

Phys. Rev. Lett. **109**, 017402 (2012)

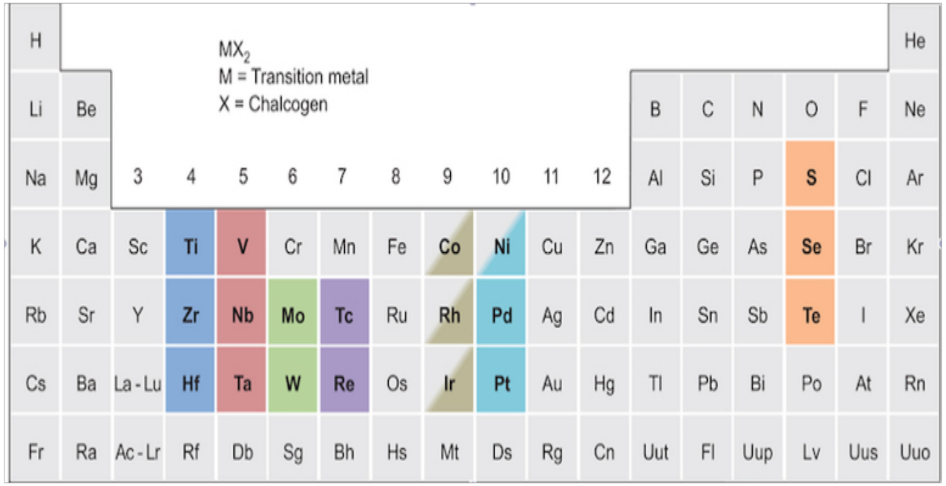
Part II

2D Semiconductor Optoelectronics

Transition Metal Dichalcogenides (TMDCs)

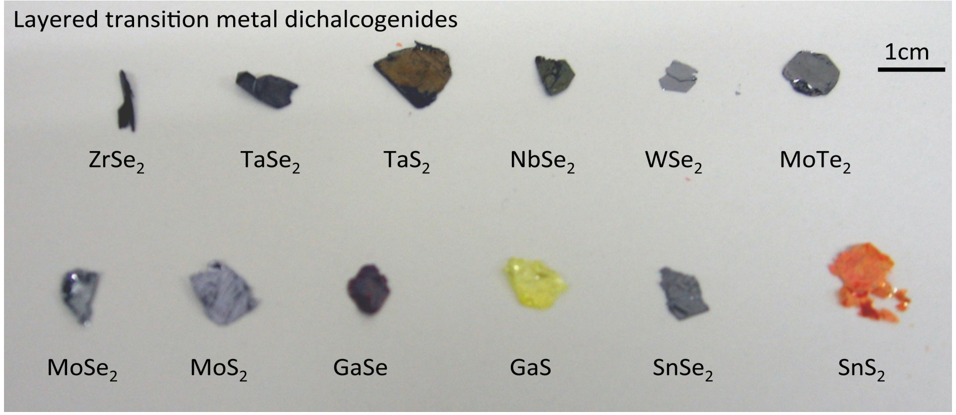
Formula: MX_2

M = Transition Metal
X = Chalcogen



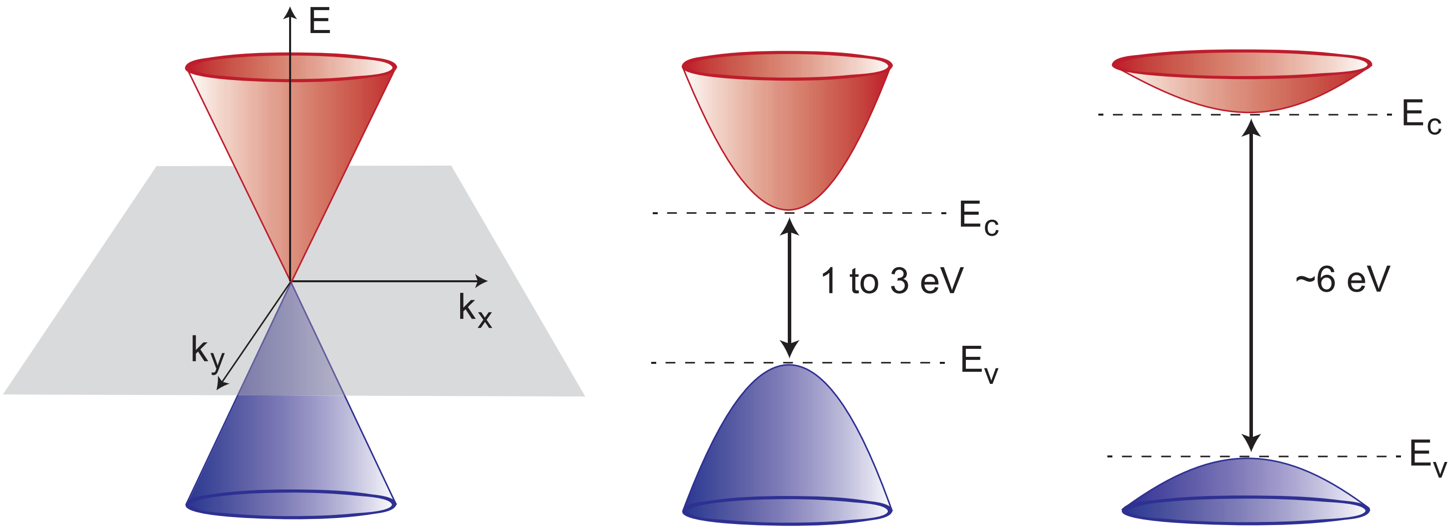
MX_2
M = Transition metal
X = Chalcogen

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg	3	4	5	6	7	8	9	10	11	12	Al	Si	P	**S**	Cl	Ar
K	Ca	Sc	**Ti**	**V**	Cr	Mn	Fe	**Co**	**Ni**	Cu	Zn	Ga	Ge	As	**Se**	Br	Kr
Rb	Sr	Y	**Zr**	**Nb**	**Mo**	**Tc**	Ru	**Rh**	**Pd**	Ag	Cd	In	Sn	Sb	**Te**	I	Xe
Cs	Ba	La - Lu	**Hf**	**Ta**	**W**	**Re**	Os	**Ir**	**Pt**	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac - Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fl	Uup	Lv	Uus	Uuo



M. Chhowalla et al. *Nature Chemistry* **5**, 263 (2013).

The 2-D Landscape



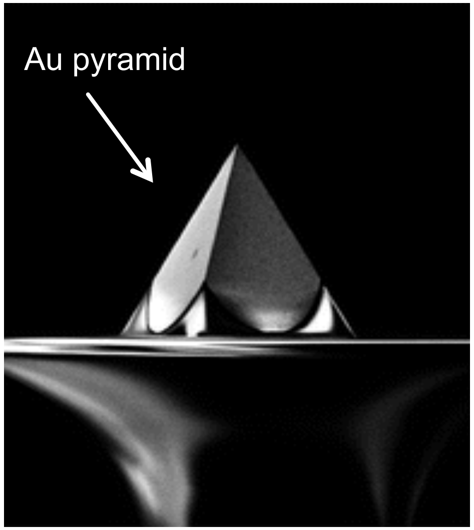
Graphene
(metallic)

TMDC monolayers
(semiconducting)

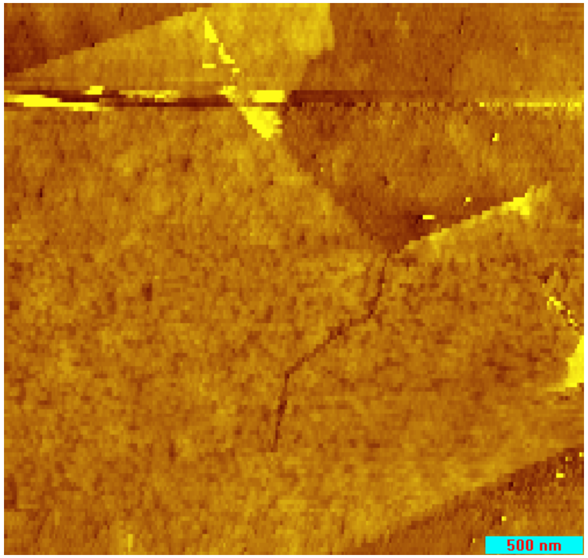
Boron Nitride
(insulating)

Optics and Photonics News, July/August 2015

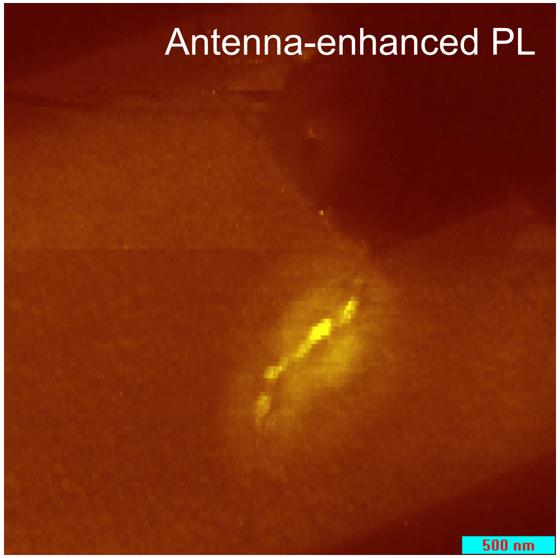
Nanoscopy of 2D Materials



Template-stripped
Au pyramid antennas,
Courtesy **S.-H. Oh,**
University of Minnesota



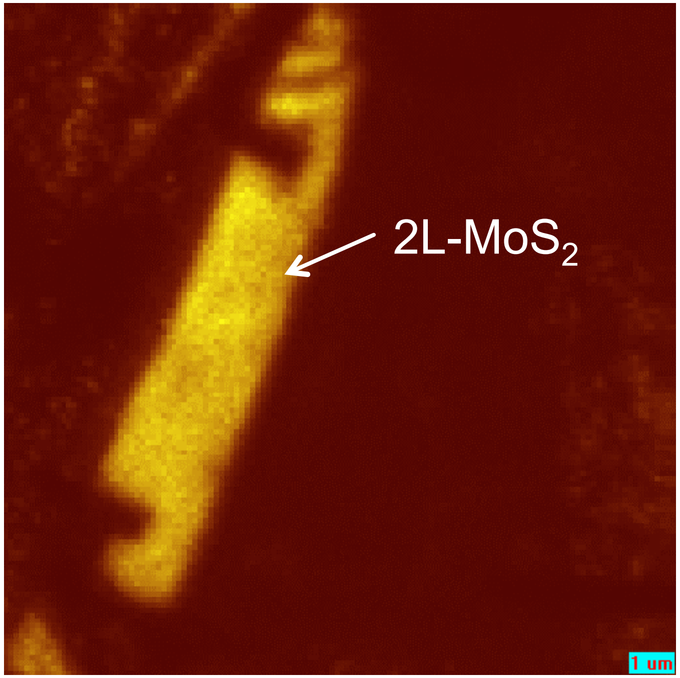
Topography of MoS_2 flake



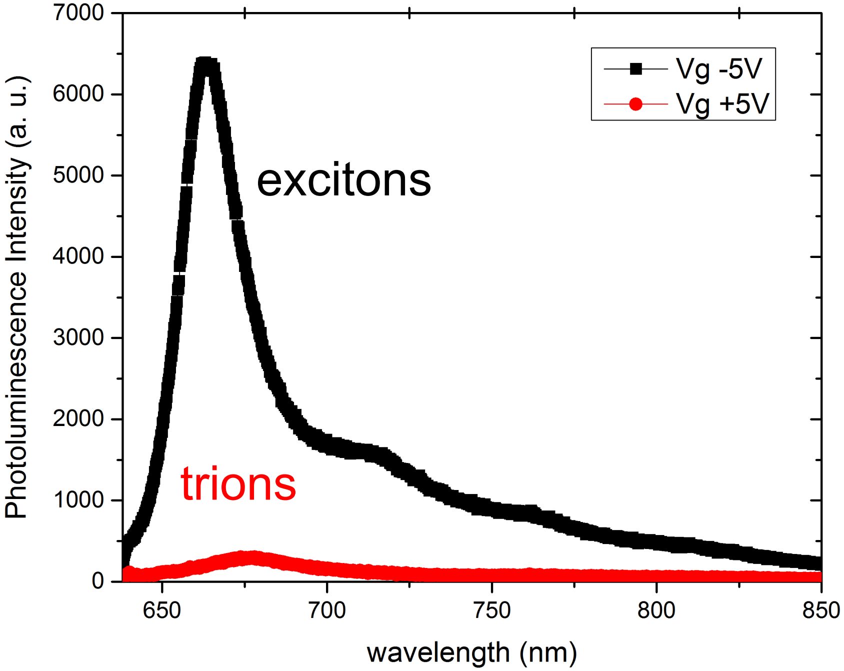
2.5x2.5 μm photoluminescence
633nm excitation

(Bharadwaj, unpublished)

Gate modulates Light Emission in an MoS_2 FET



15x15 μm confocal PL
MoS_2 FET with graphene-hBN backgate



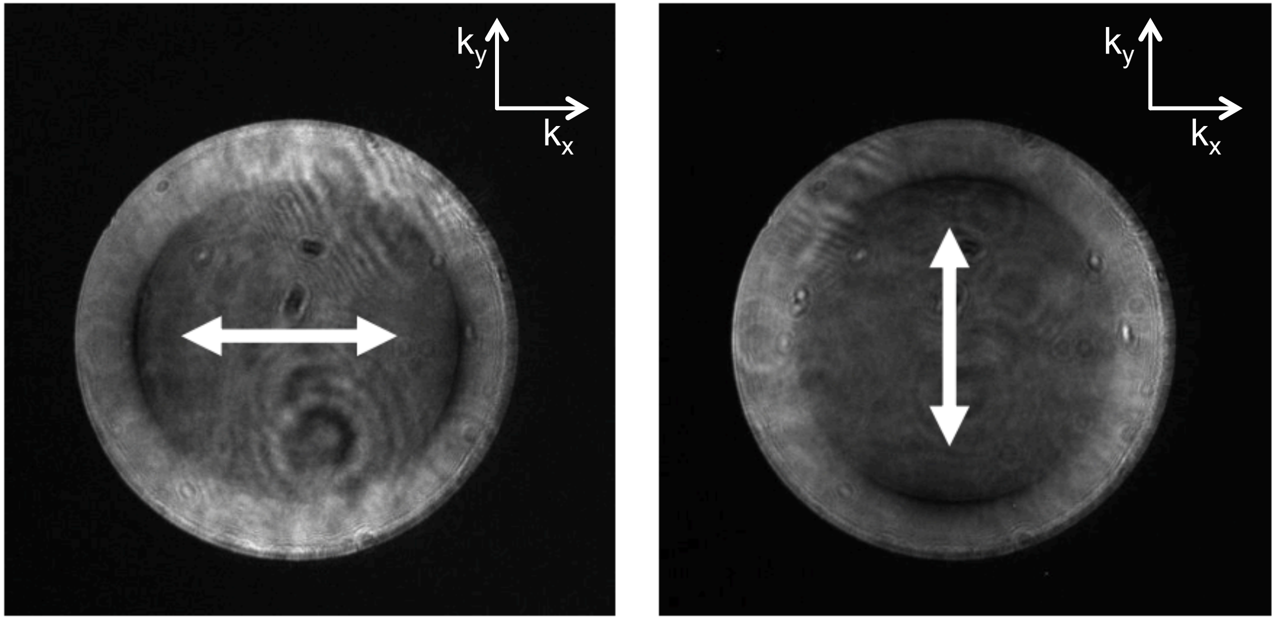
Gate modulation of PL

(Bharadwaj, unpublished)

Where are the Photons Going?

Orientational Imaging of MoS_2 Excitons

Angular distribution of PL (Fourier Plane Imaging)



Polarizer along x

Polarizer along y

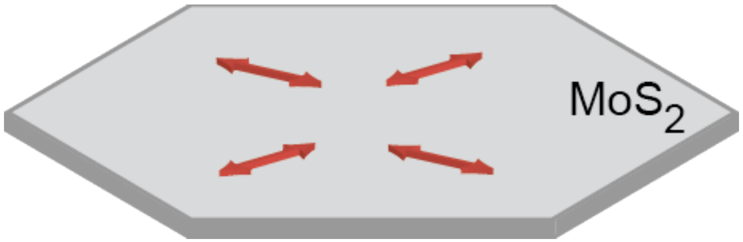
MoS_2 excitons are randomly oriented in-plane

Similar behavior for Vg +5V (trions)

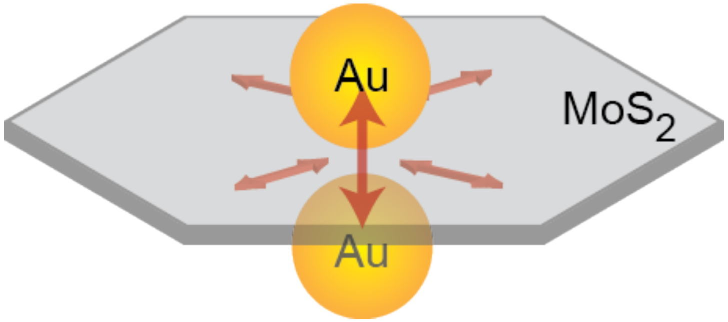
(Bharadwaj, unpublished)

Coupling Antennas to MoS_2

MoS_2 Electrons and Holes Live In-plane

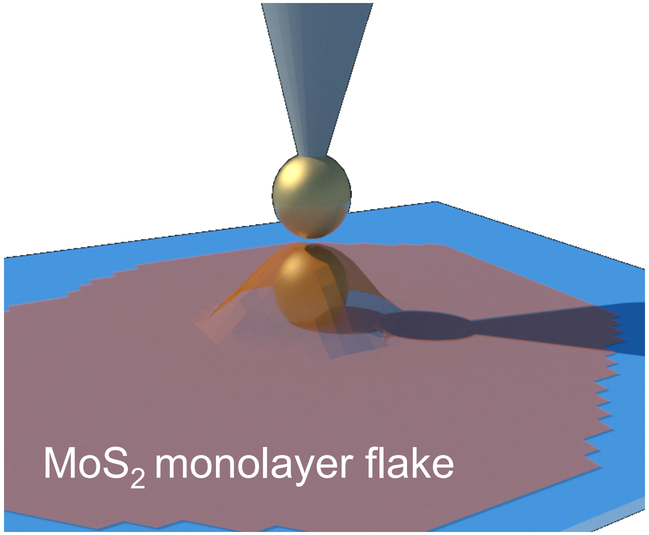


In-plane excitons radiate out-of-plane

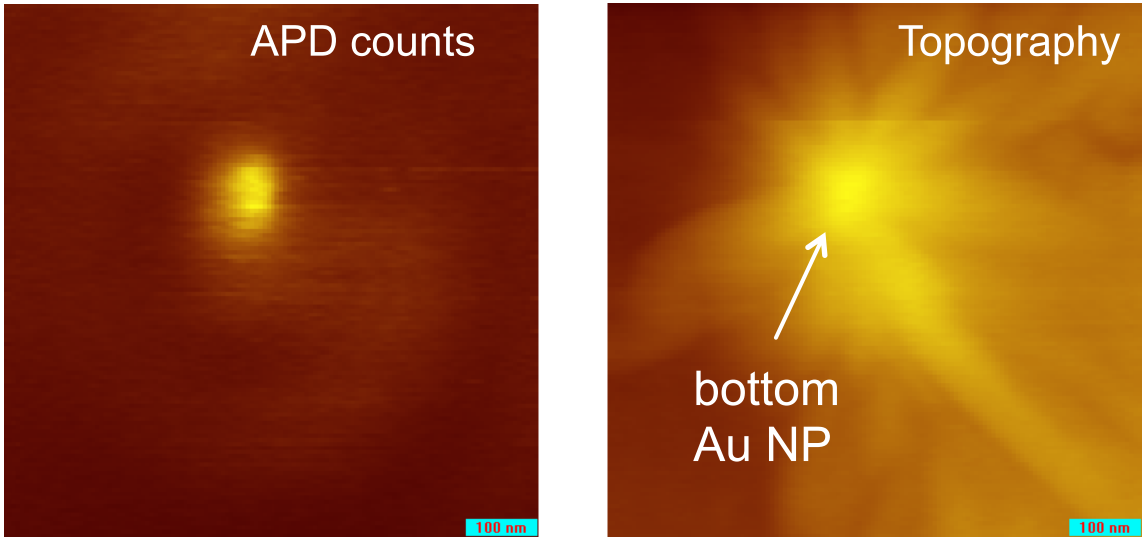


Coupling excitons to a dimer antenna induces an out-of-plane dipole, which radiates in-plane

MoS_2 sandwiched between a gold dimer

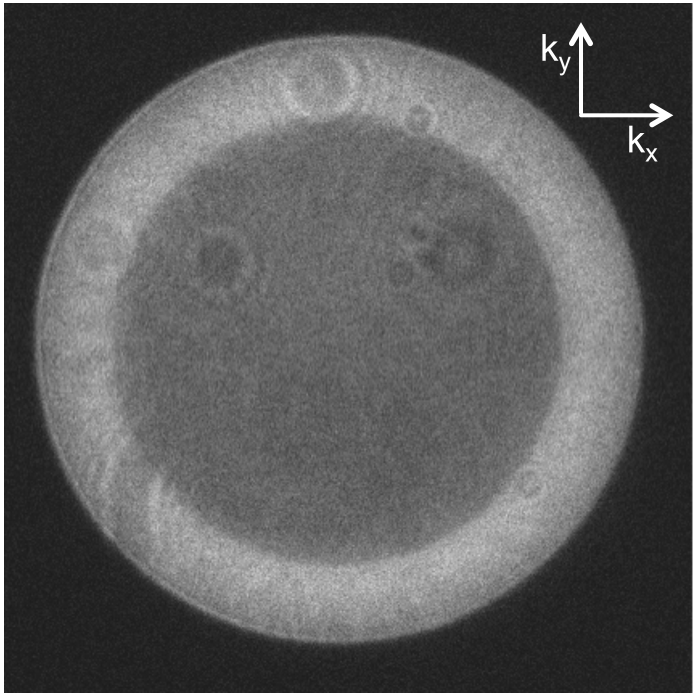


Schematic of experiment

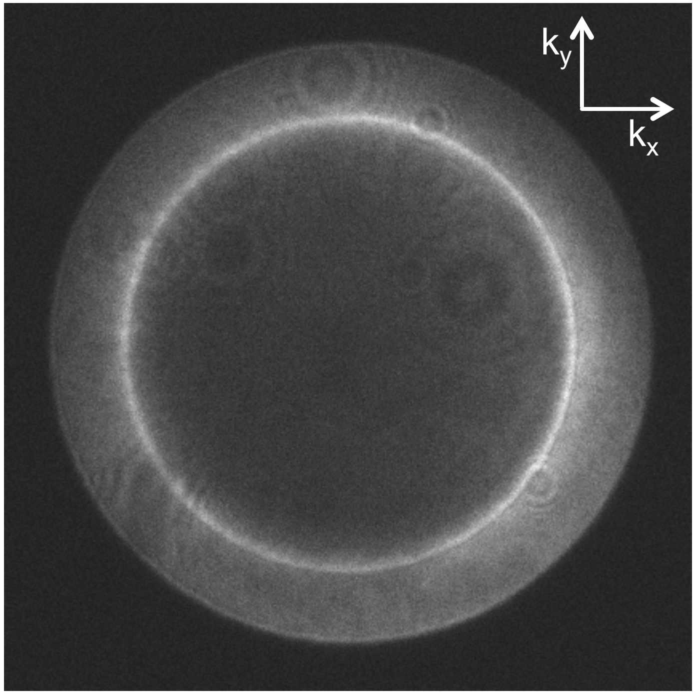


Photoluminescence is enhanced 5x by the dimer antenna

Where is the light going?



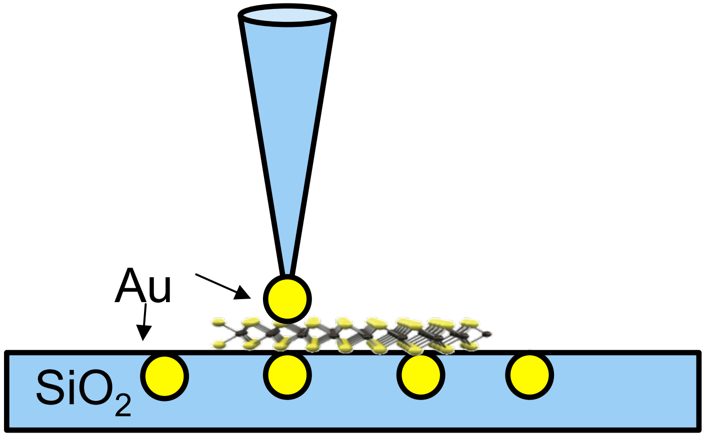
Not coupled to antenna --
Ensemble of in-plane dipoles

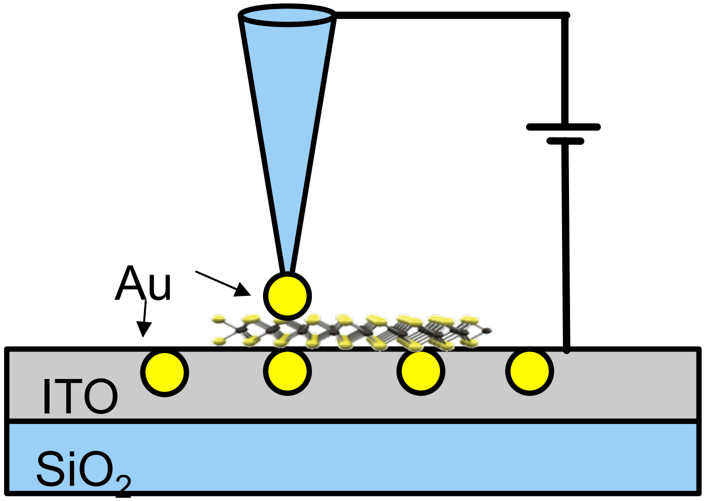


Coupled to dimer antenna --
Out-of-plane dipolar emission

(Bharadwaj, unpublished)

Can Excitons be Reoriented?

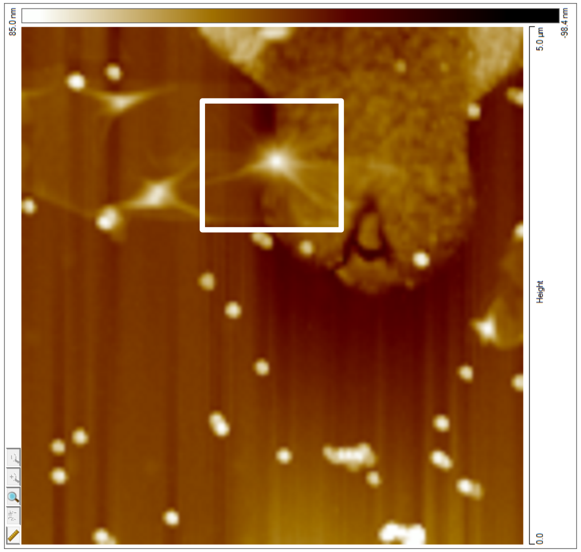




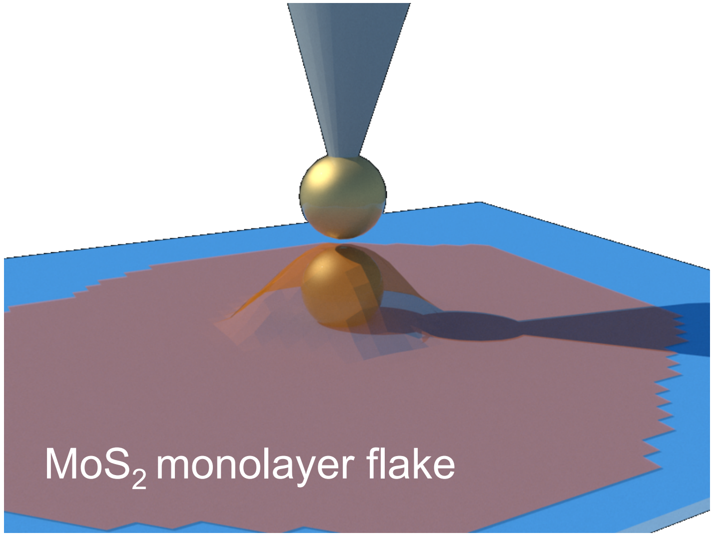
Absorption still in-plane
But *z*-dipole like emission pattern?

Excitons in strong dc fields
Stark modulation of PL?

MoS_2 over Au Nanoparticles

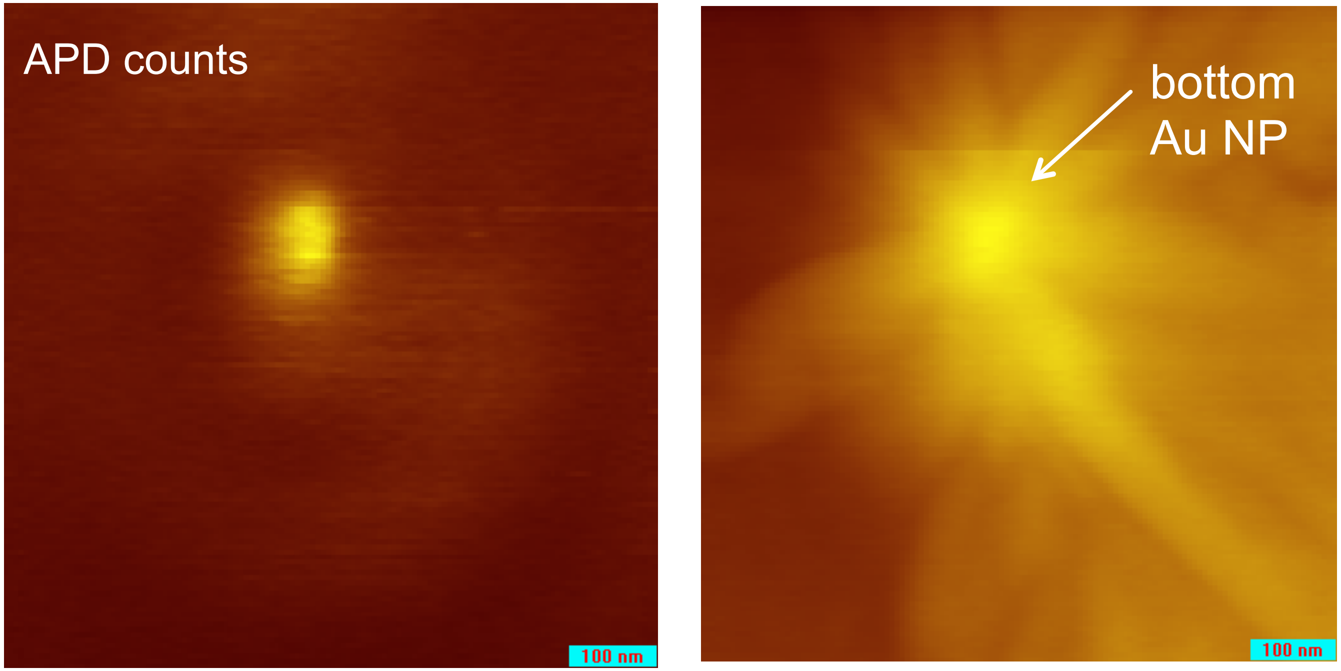


AFM topography



Record PL counts and angular distribution of photons as a function of the position of scanning top particle

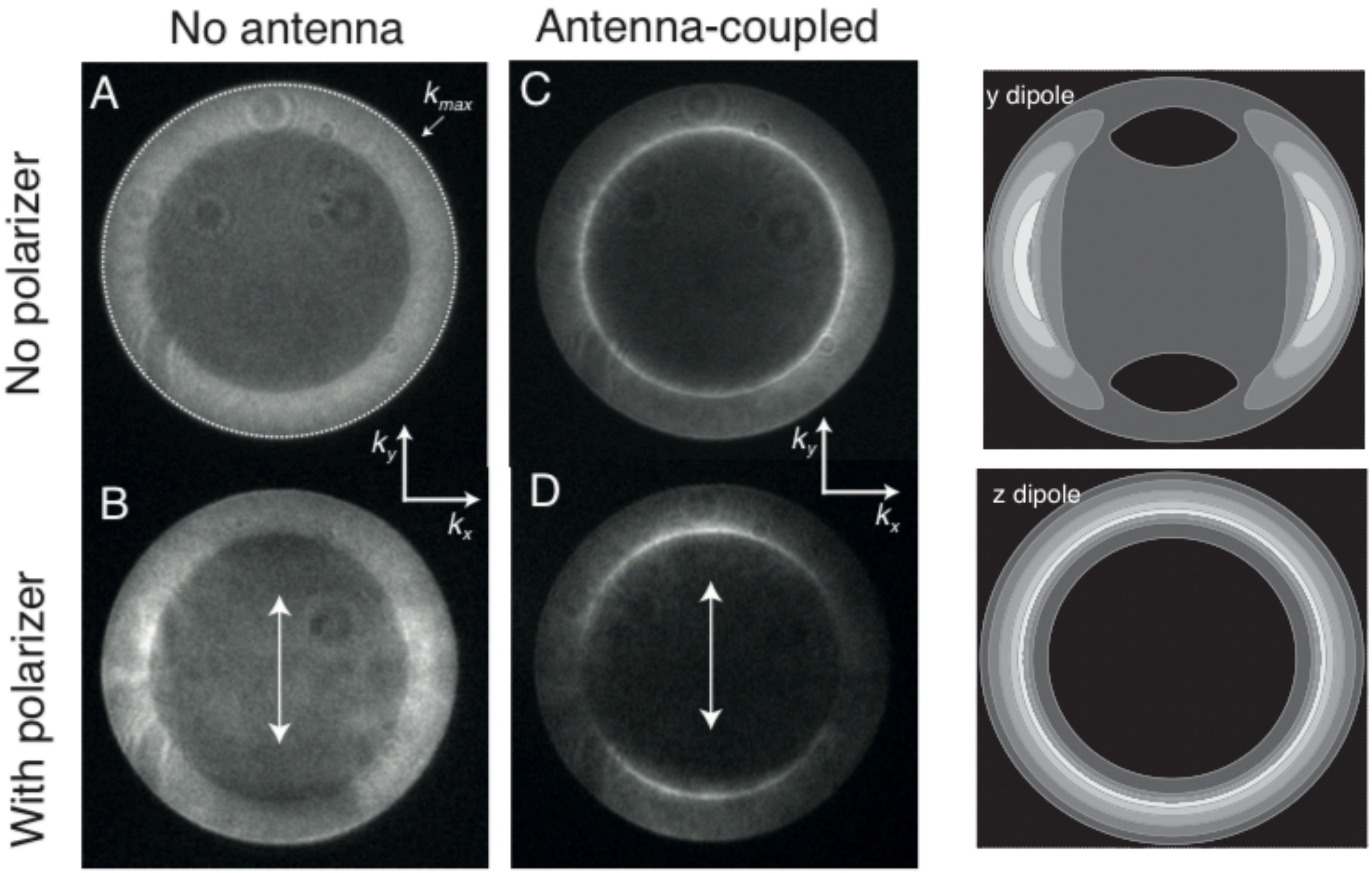
Dimer-coupled MoS_2 Photoluminescence



1x1 μm PL with 80nm Au NP Topography

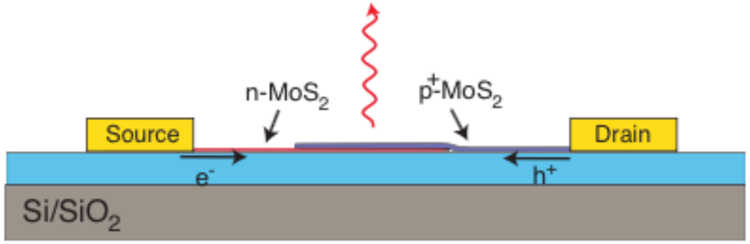
Bharadwaj *et al*, manuscript in preparation

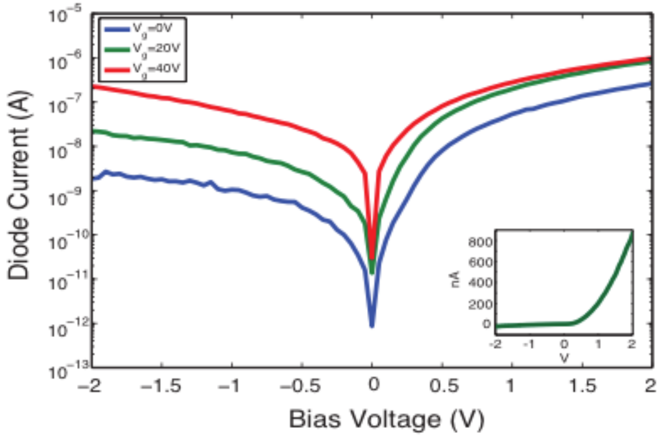
Dimer Antenna Strongly Redirects the PL!

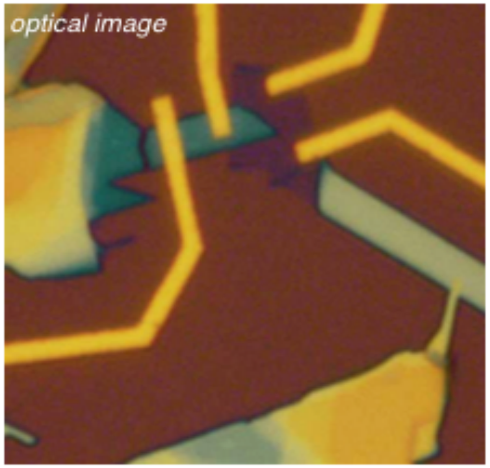


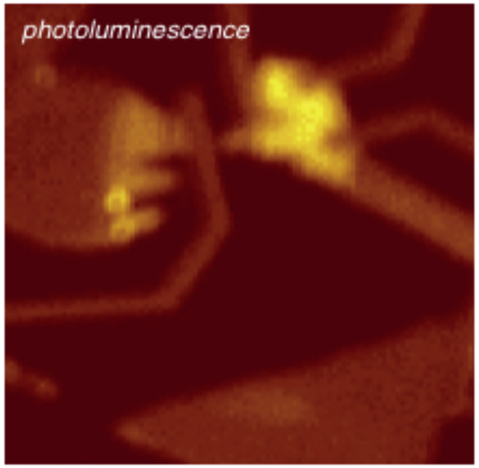
Bharadwaj *et al*, manuscript in preparation

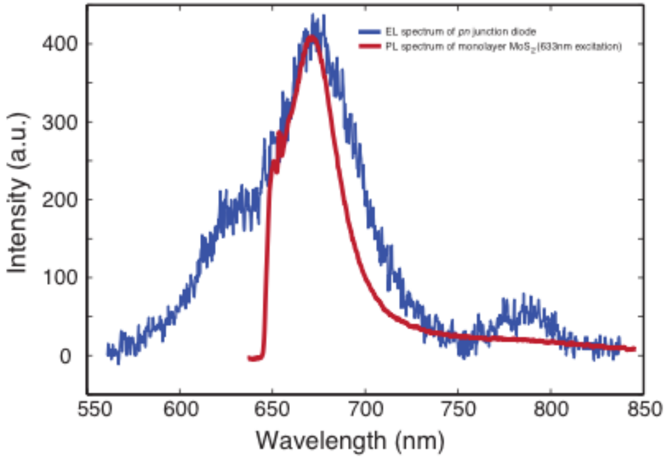
First all-MoS_2 LED







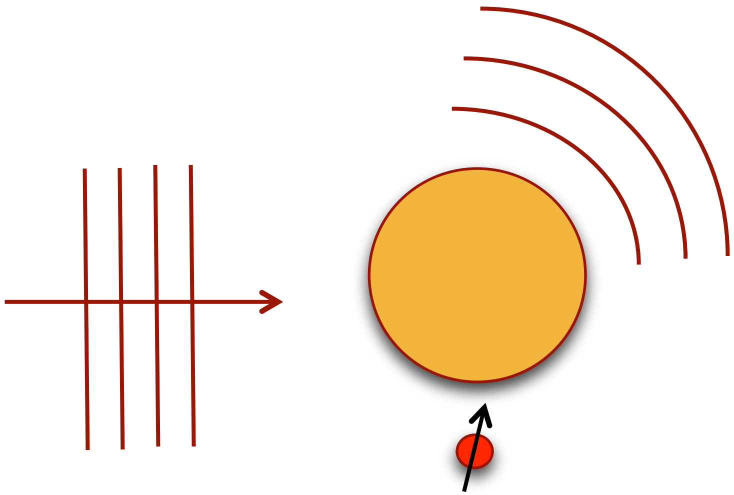




A. Jain, Bharadwaj *et al*, manuscript in preparation

Part III

Electrical Excitation of Optical Antennas

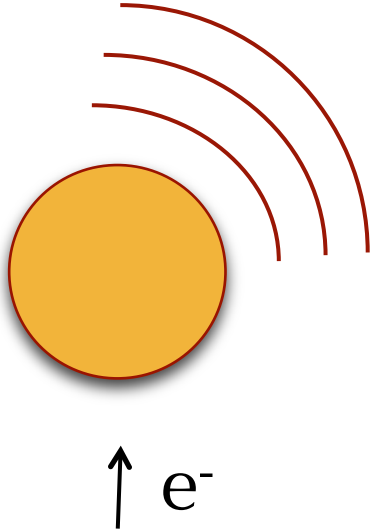


Part III

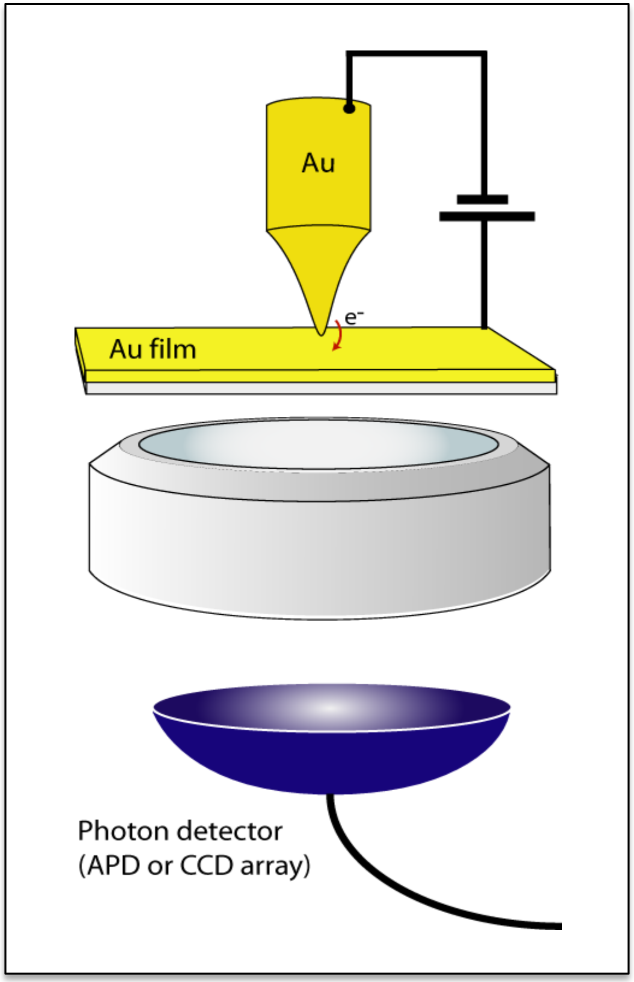
Electrical Excitation of Optical Antennas

Can **optical** antennas be excited **electrically**?

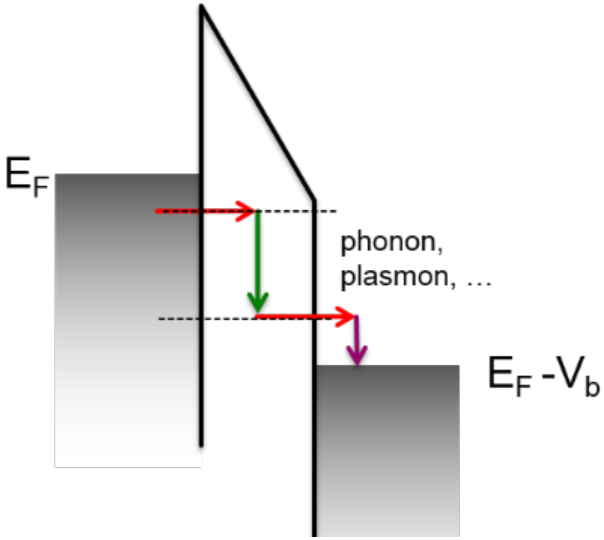
Can electrons excite localized / propagating plasmons?



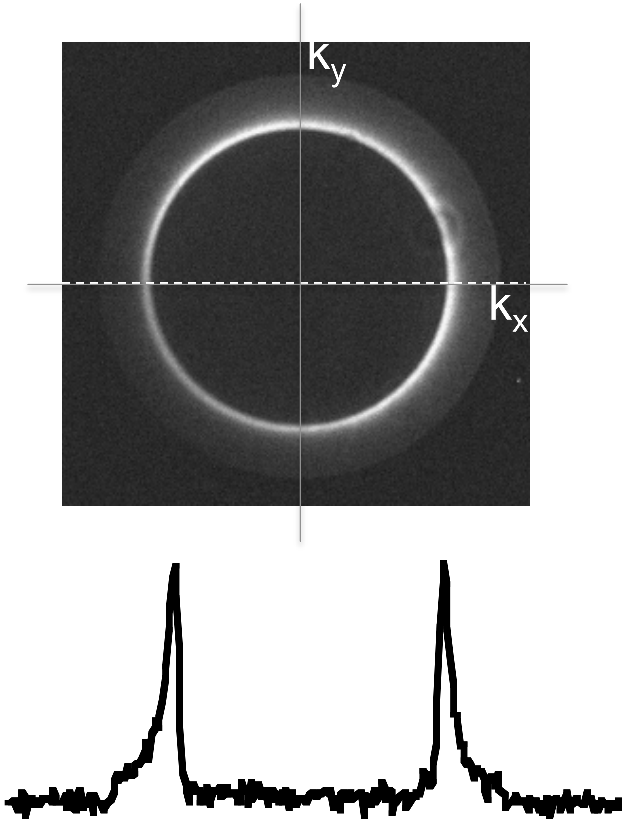
Electrically Excited Plasmons on a Metal Film

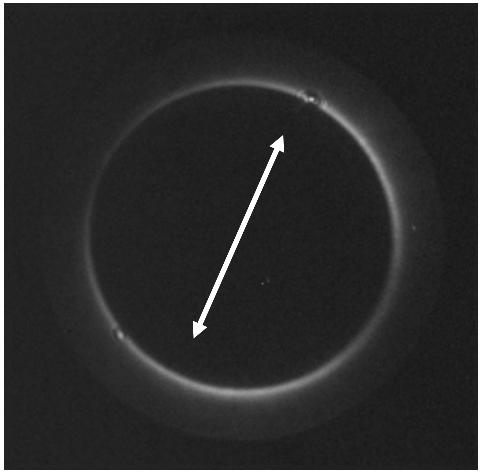


Can propagating surface plasmon polaritons (SPPs) be excited via local electron tunneling?



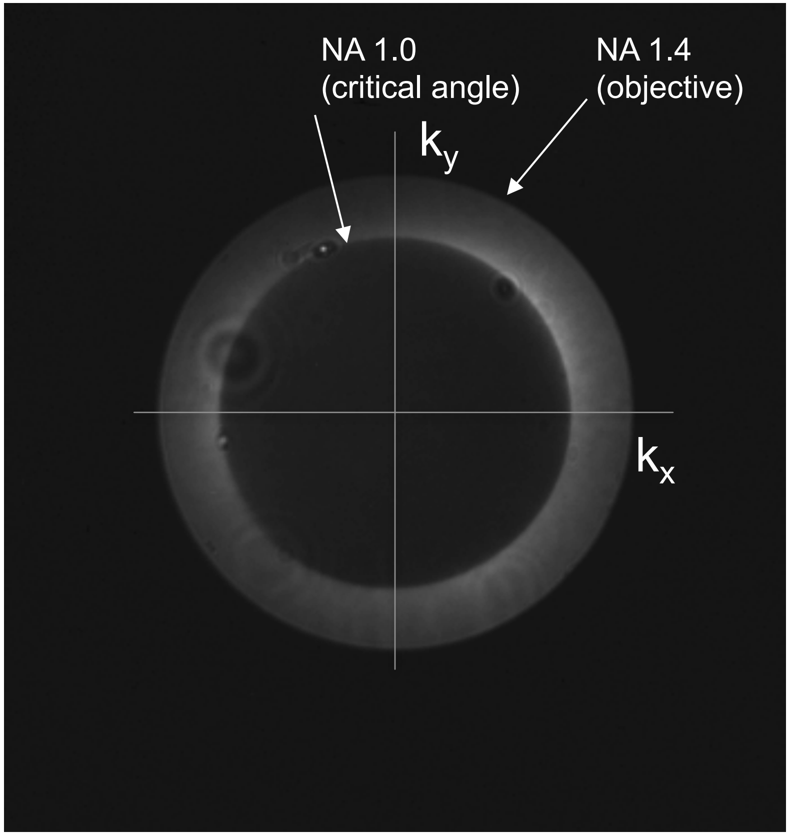
Plasmon Decay at Kretchmann Angle





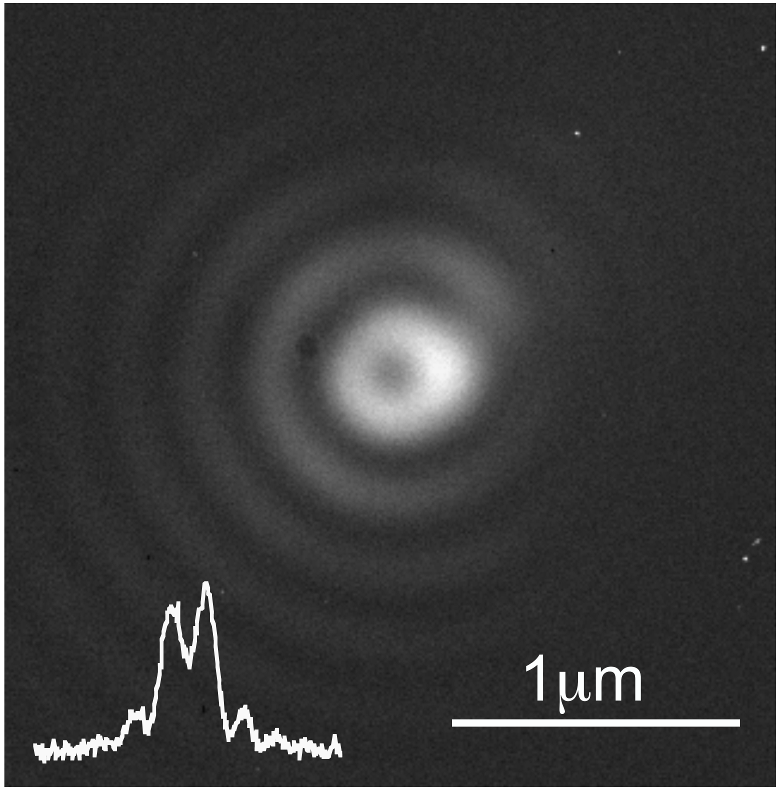
analyzer in front of the CCD (oriented along the arrow)

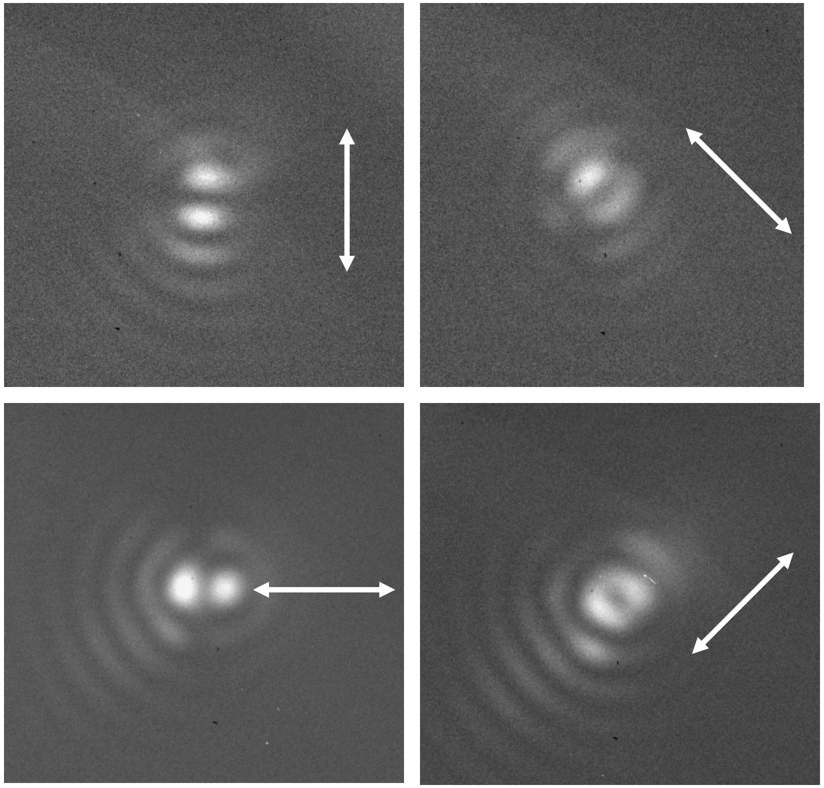
Fourier Plane Imaging of 5nm Au on glass



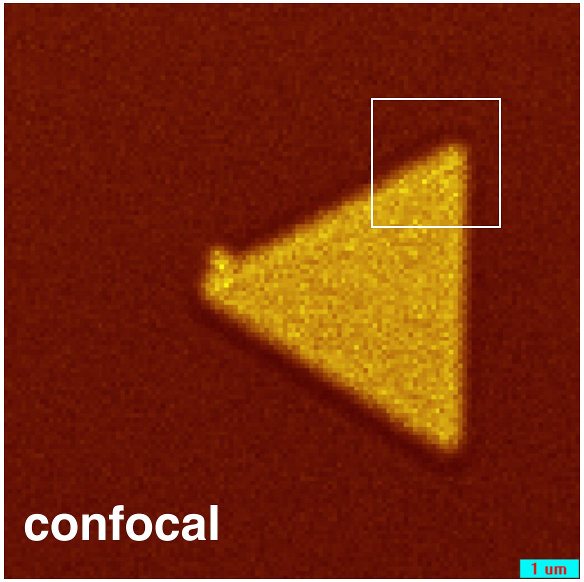
Au tip; 2V bias;
2nA tunnel current

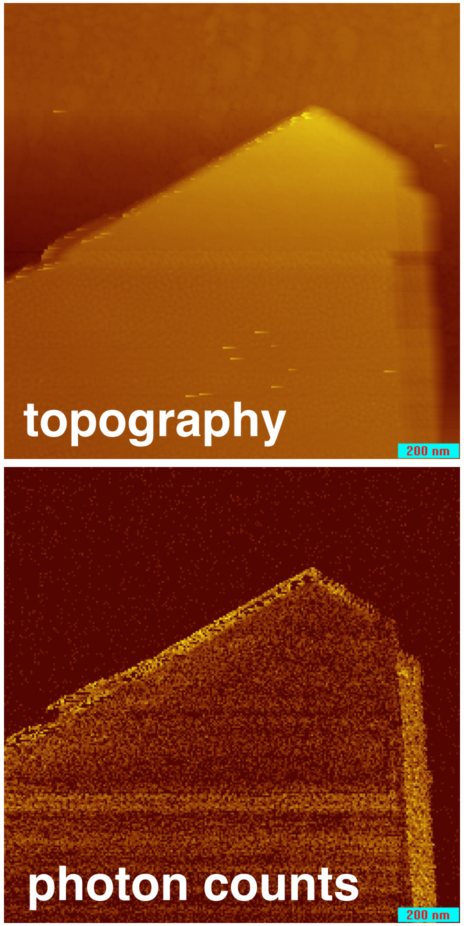
Real Space Imaging of 20nm Au on Glass

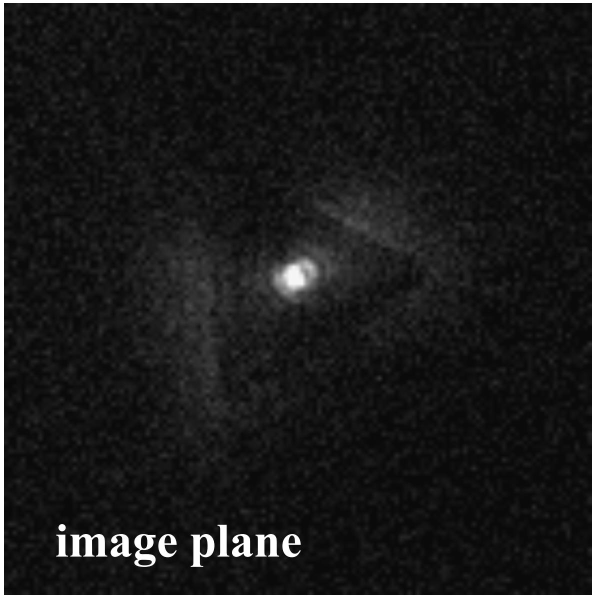




Single crystal Au flake on ITO (Au tip)

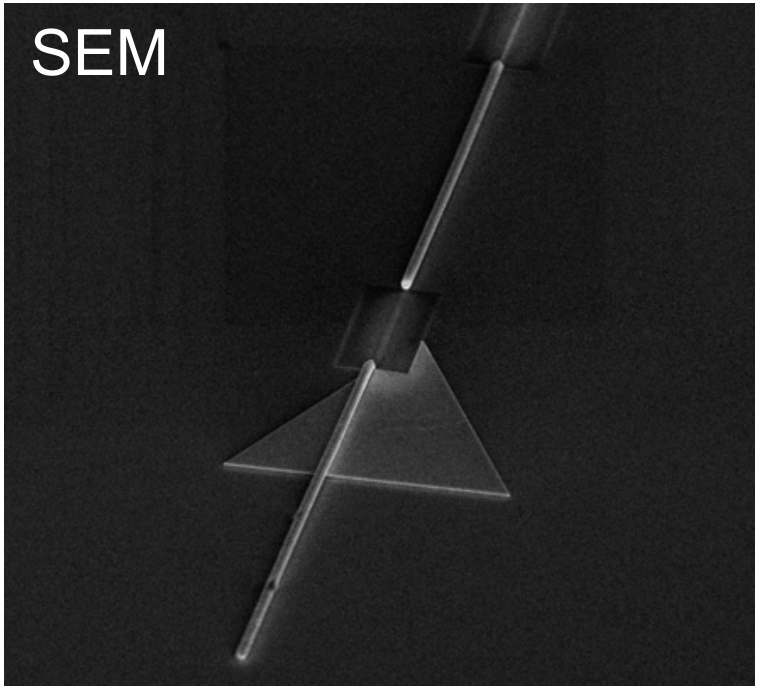


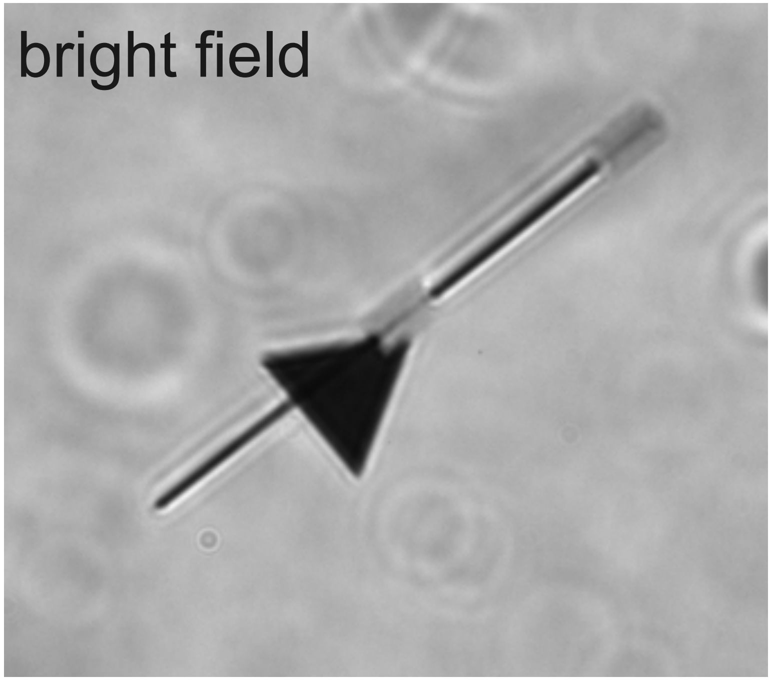




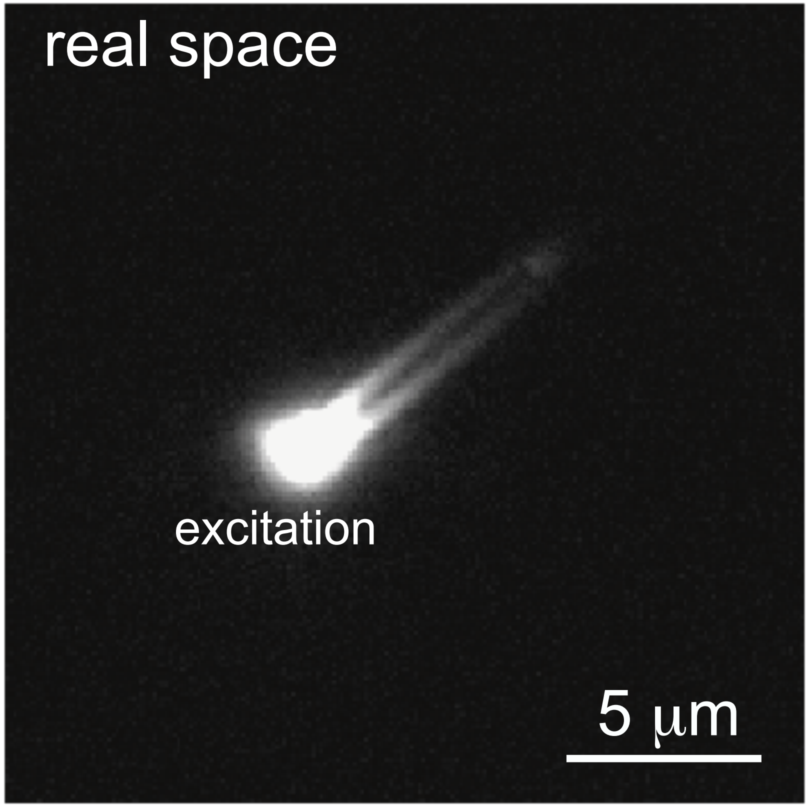
Au tip; 2V bias,
1.5nA tunnel current

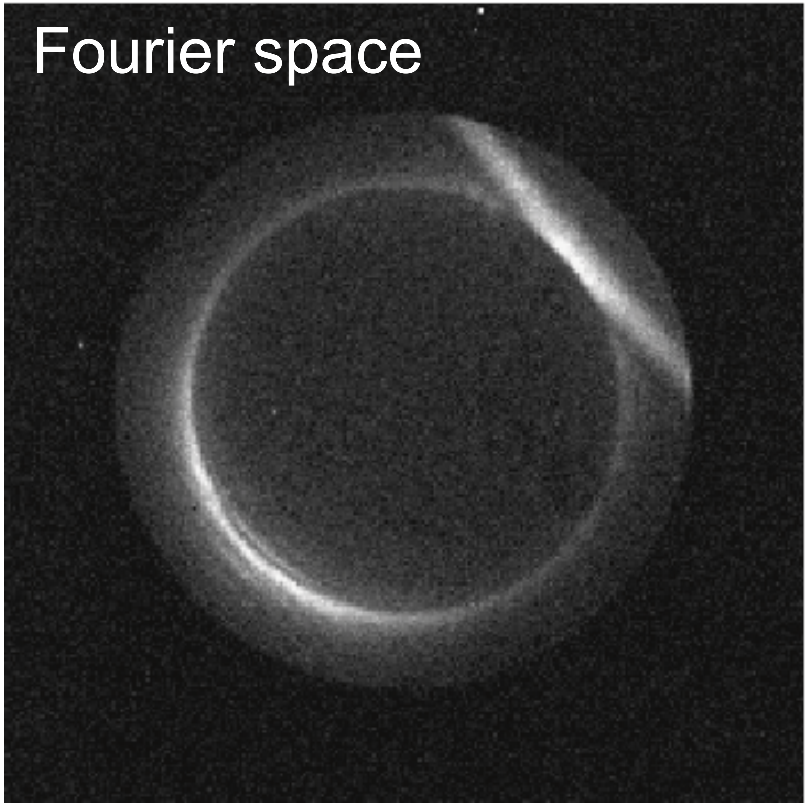
Single Crystal Au Nanowire on ITO



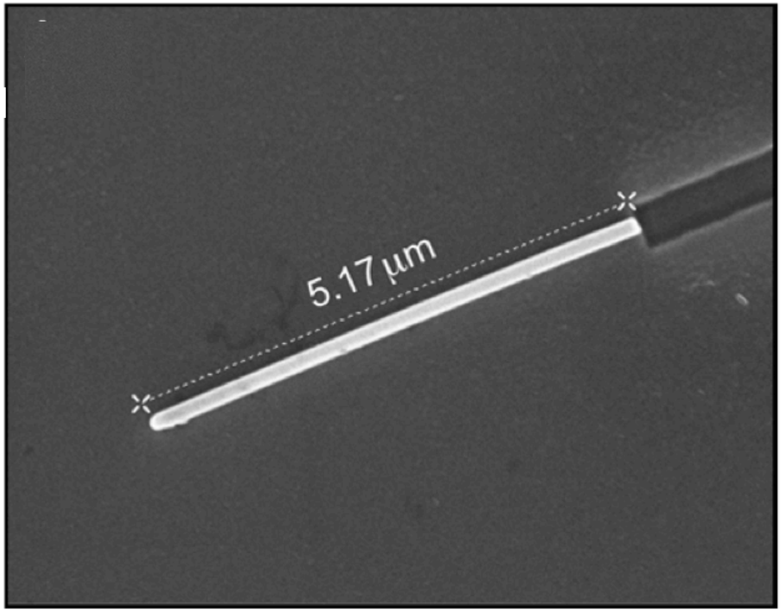


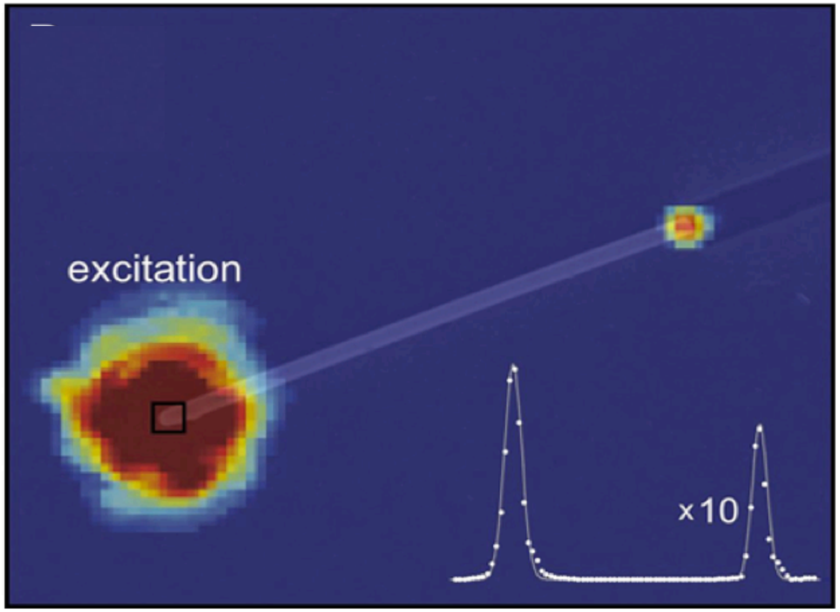
Electrically Excited Plasmons on a Nanowire





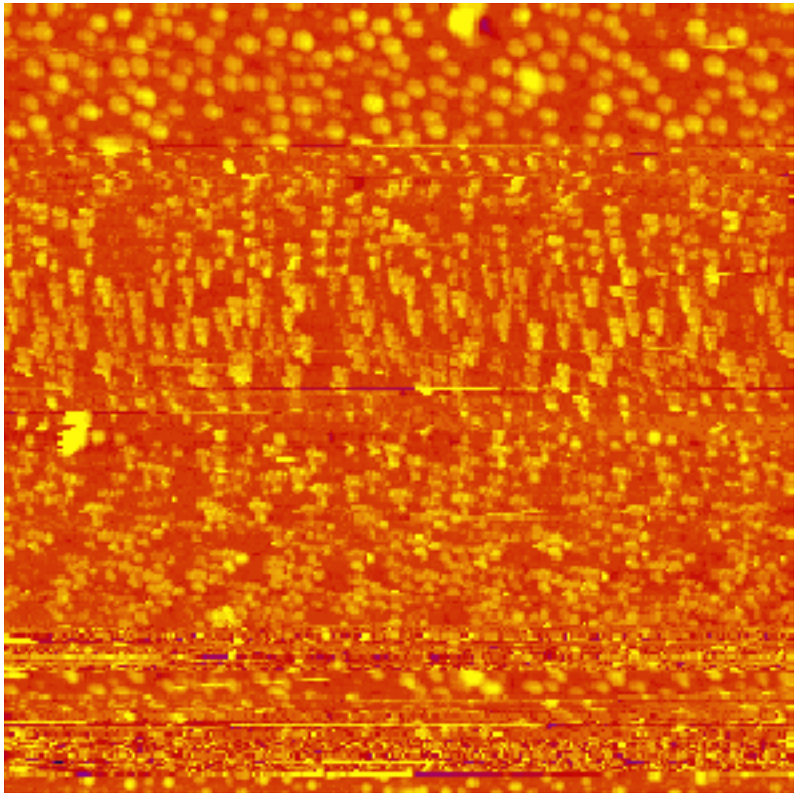
Electrically Excited Plasmons on a Nanowire



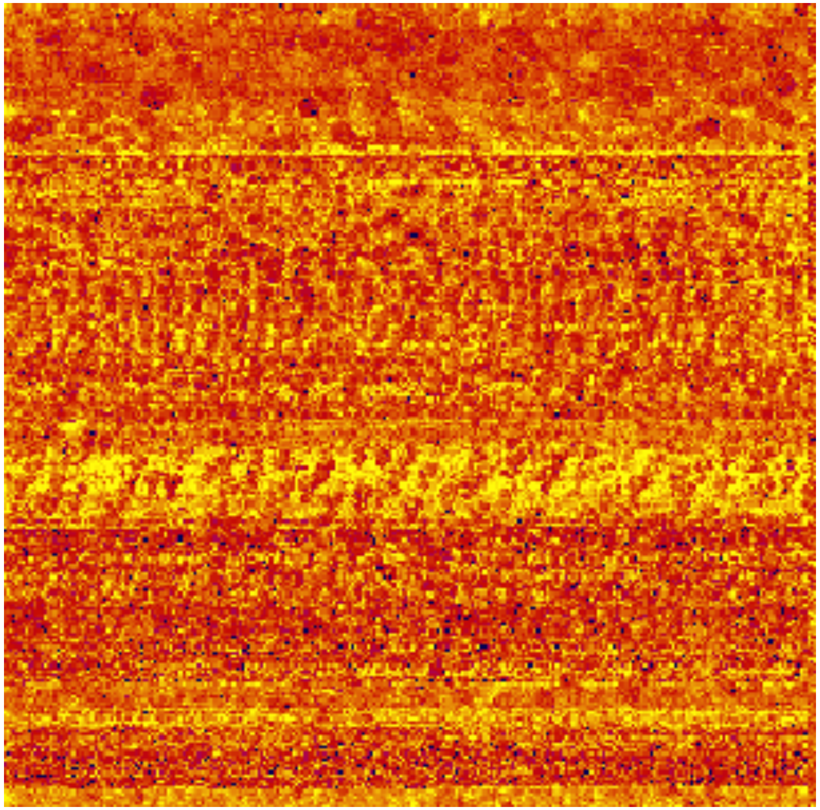


Phys. Rev. Lett., **106**, 226802 (2011)

Ambient STM is not stable!

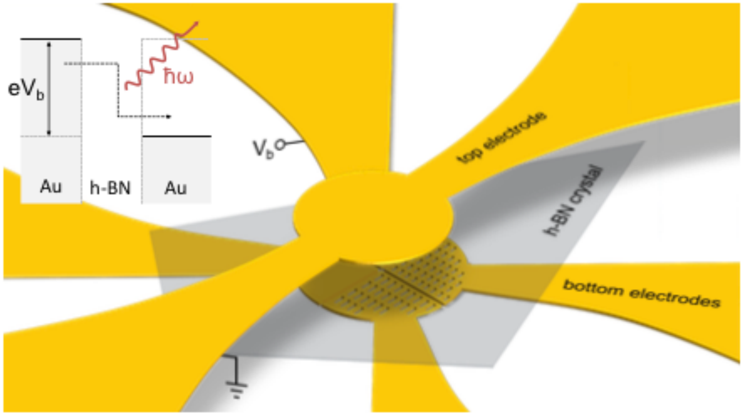


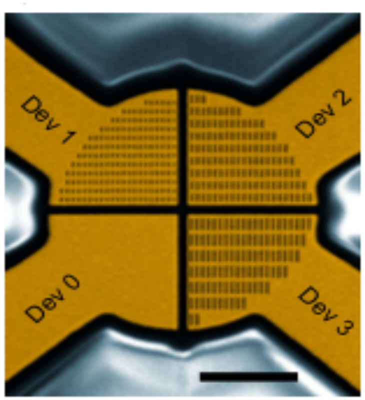
Topography
3x3 μm

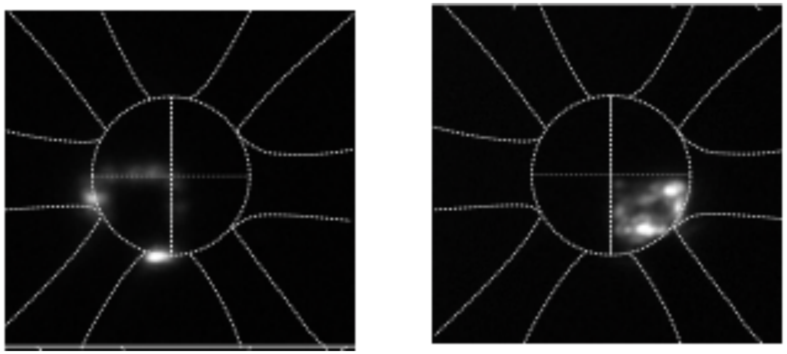


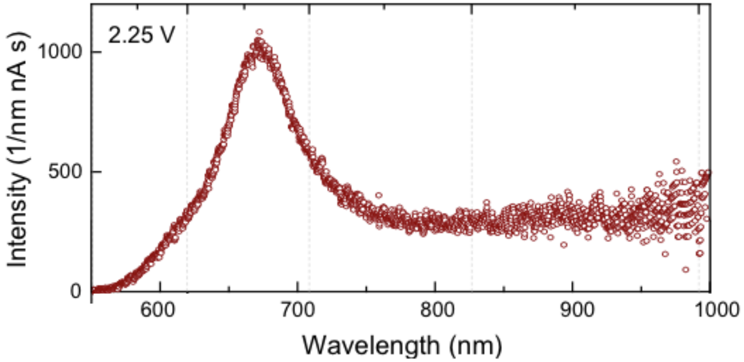
APD Counts 3-30 kHz
3x3 μm

Integrated Light Emitting Antennas with hBN





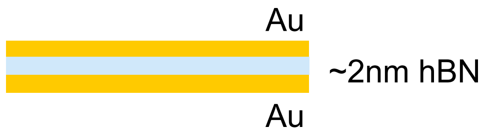




Light emission on CCD

M. Parzefall, P. Bharadwaj *et al*
Nature Nano. **(accepted)**

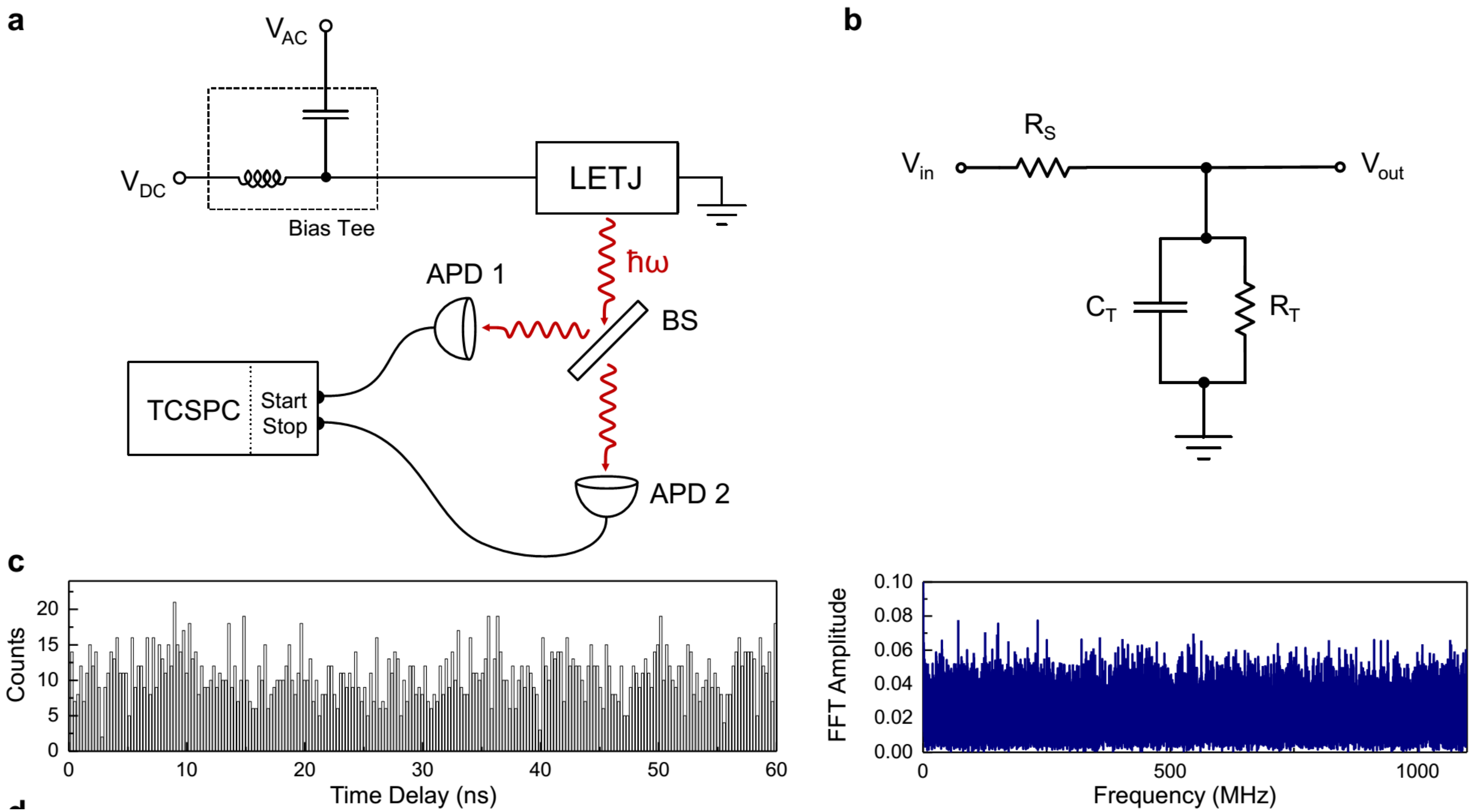
How fast can one modulate the light emission?



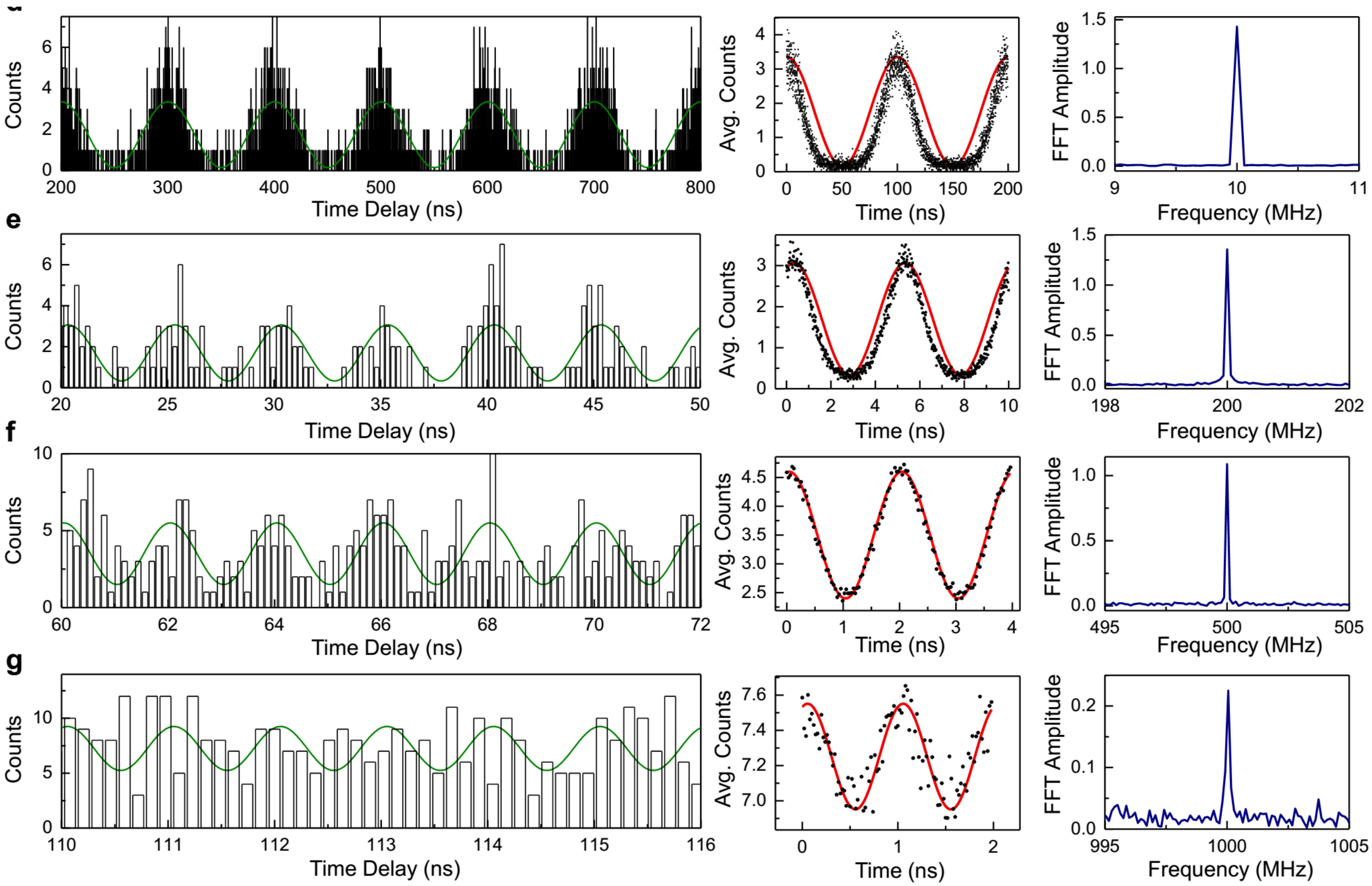
Capacitance of junction ~0.1 pF
Series Resist. of junction ~100 Ω

RC ~ 10 ps

f_{max} ~ 100 GHz



Time-modulation of Light Emission



Concluding Remarks

Plasmonics with Single Emitters and Electrons

- Optical antennas enhance absorption and emission of light by single emitters
- Scanning antennas enable high resolution imaging
- Antennas can be electrically excited using tunneling electrons

Active Devices using Novel Optical Materials

- Electrical modulation of photoluminescence from an MoS_2 FET
- All-MoS_2 *pn* light-emitting diode
- Antennas strongly redirect light emission from excitons in MoS2

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- U.S. DoE and NSF, and Swiss SNF for funding

U.S. DEPARTMENT OF
ENERGY

NSF
National Science Foundation
WHERE DISCOVERIES BEGIN

FNSNF
FONDS NATIONAL SUISSE
SCHWEIZERISCHER NATIONALFONDS
FONDO NAZIONALE SVIZZERO
SWISS NATIONAL SCIENCE FOUNDATION

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