Nanophotonics and electronics for bright single-photon sources

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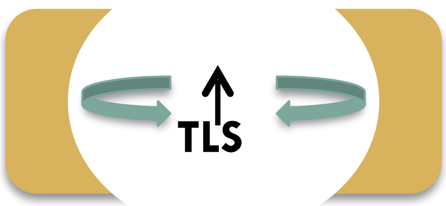
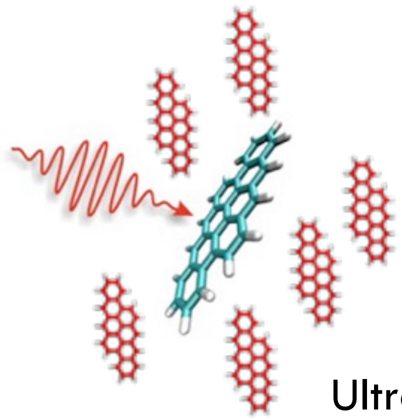
University of Siegen

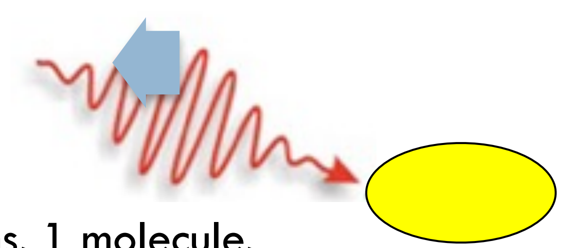
Research goals



Quantum optics:
1 photon, 1 atom, ns to ps time scales



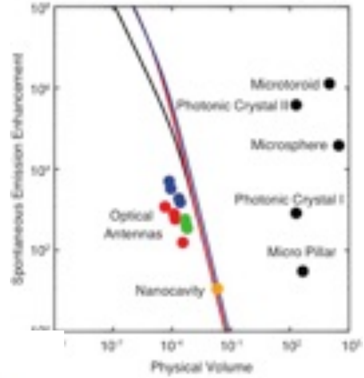
Ultrafast spectroscopy:
many photons, 1 molecule,
fs to as time scales



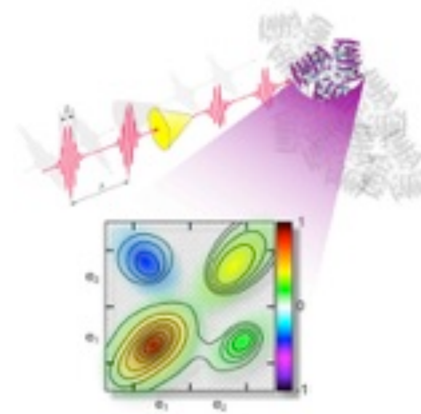
Nano-optics:
many photons, 1 molecule,
nm length scales

Research areas

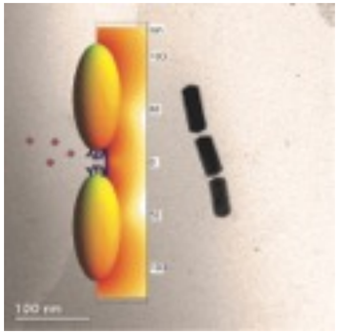
UNIVERSITÄT SIEGEN



- Quantum nano optics

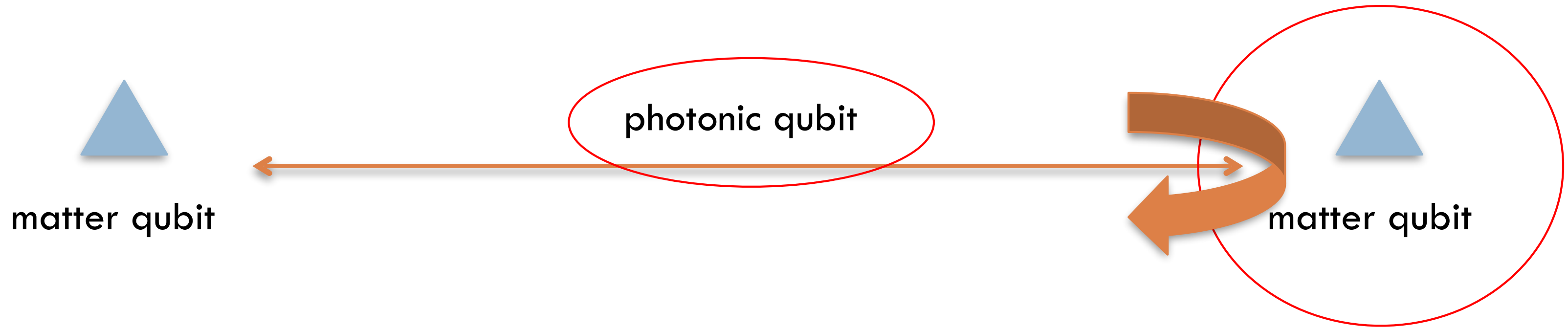


- Nanospectroscopy



- Nanosensing

Quantum photonics – a sketch

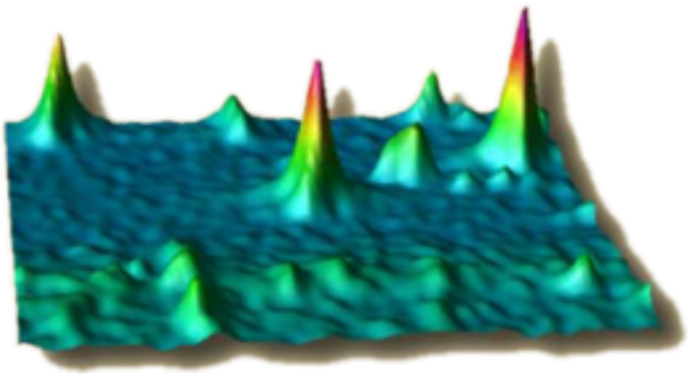


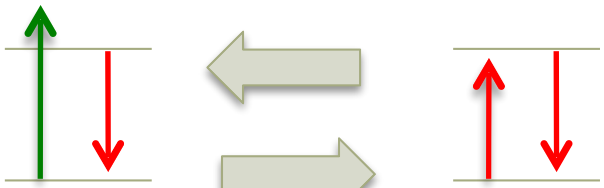
J.L. O'Brien, A. Furusawa, and J. Vučković, Nat. Phot. (2009)
H.J. Kimble, Nature (2008)

Outline

- Part I
  - **Nano quantum optics**
  - Metal nanostructures as optical antennas
  - Fluorescence enhancement
  - Directional emission
  - Quantum optics with optical antennas
- Part II
  - Quantum emitters
  - Intrinsic optical properties of diamond
  - Color centres in diamond
  - Diamond-based nanophotonics
  - Diamond electronics

Quantum emitters, e.g. single molecules





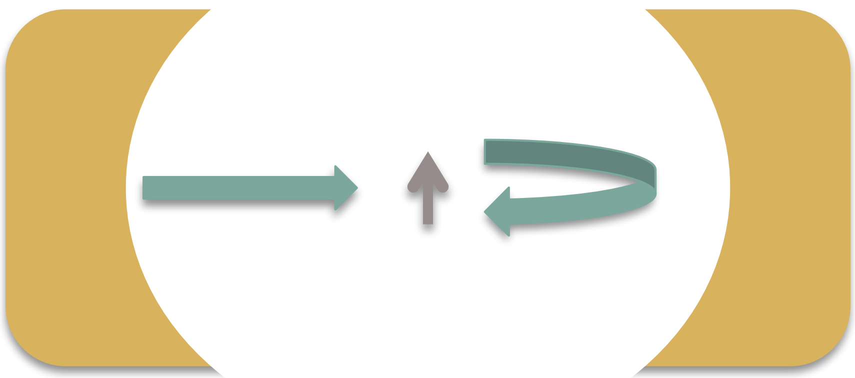
W.E. Moerner, M. Orrit, Phys. Rev. Lett. (1989, 1990)

- Eliminate ensemble average.
- First quantum optical experiments with solid-state emitters.
- Allowed to probe spatial and dynamical heterogeneities at the nanoscale.

- We lack of a robust and flexible light-matter interface.
- Coherence and fast dynamics are difficult to access at the SM level under real-world conditions.

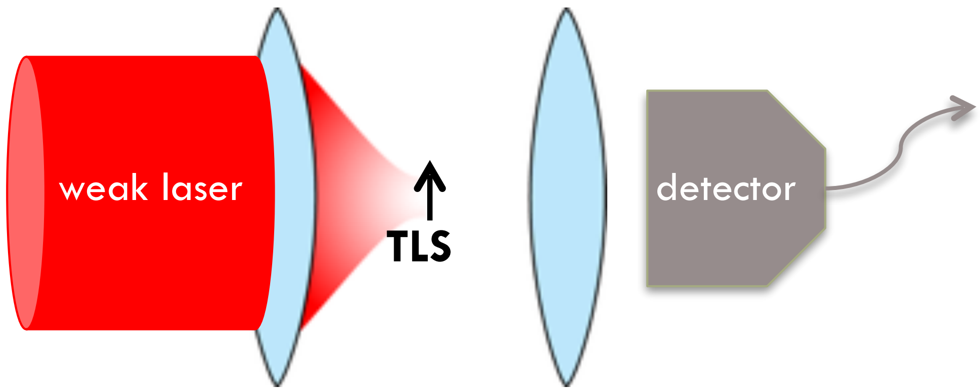
We need to control light matter interaction at a fundamental level

- Enable light-matter interaction and the single-photon-emitter level
- Enable photon-photon interactions at the single-photon level

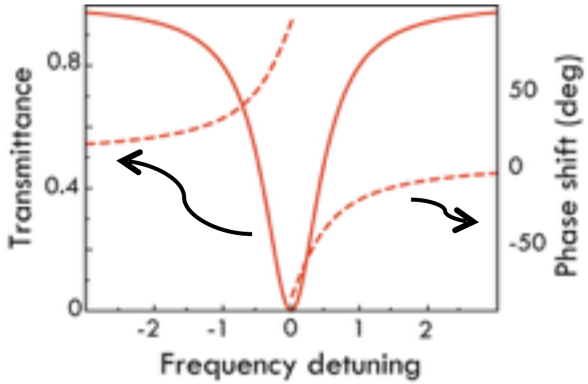


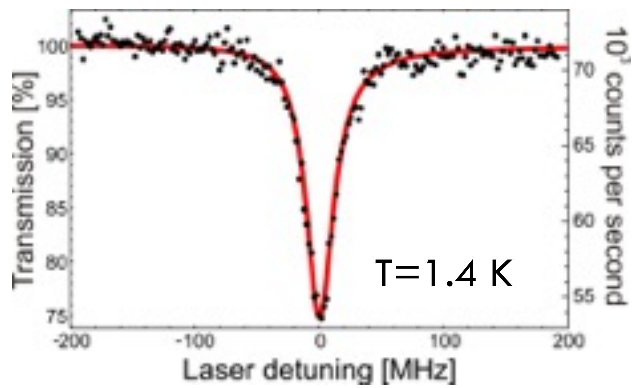
- Optical resonators have played… and will play… a major role in this context
- Bandwidth and footprint limitations

A single emitter is optically thick



$$A_{\text{eff}} = \frac{3\lambda^2}{4\pi} = \frac{\sigma_{\text{TLS}}}{2}$$



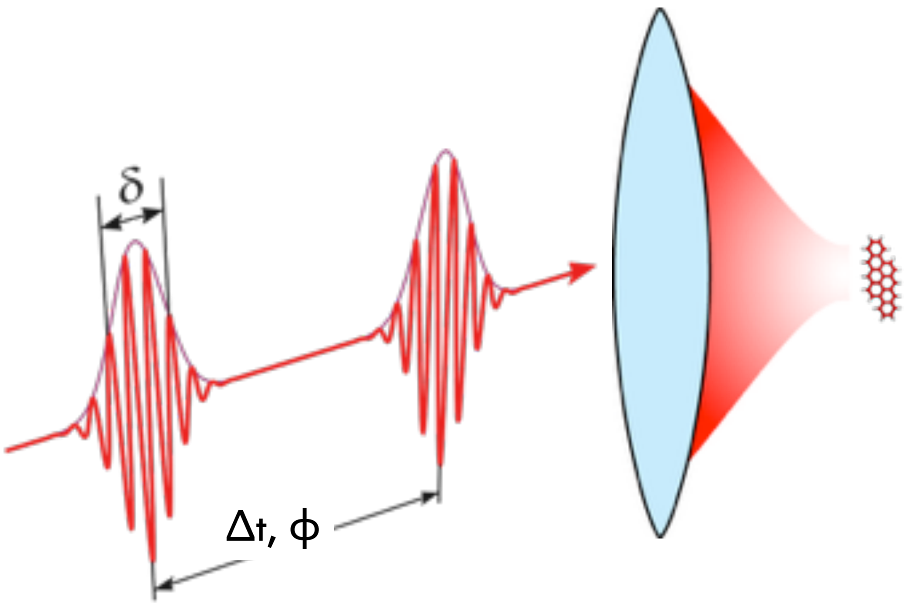


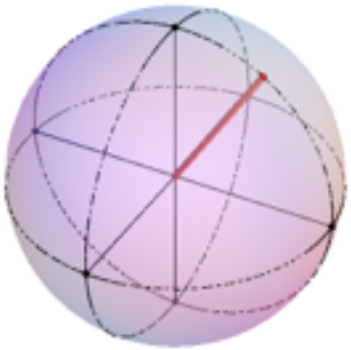
At room temperature a solid-state quantum emitter is barely detectable in extinction (10^{-5})

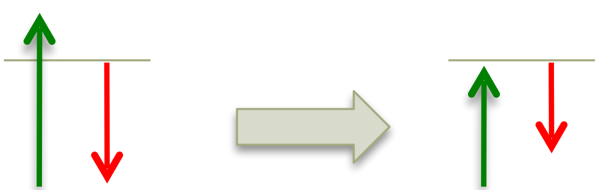
Nano-Optics Group, ETH Zurich (2010)

G. Zumofen, N.M. Mojarad, V. Sandoghdar, M. Agio, Phys. Rev. Lett. (2008)
G. Zumofen, N.M. Mojarad, M. Agio, N. Cimento C (2009)

Ultrafast control of a TLS





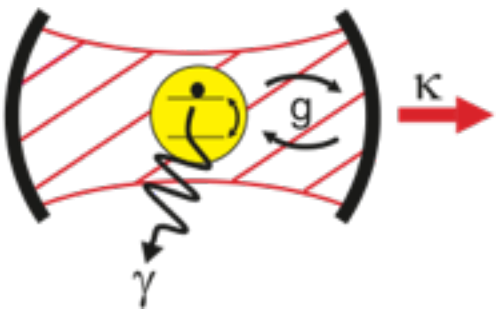


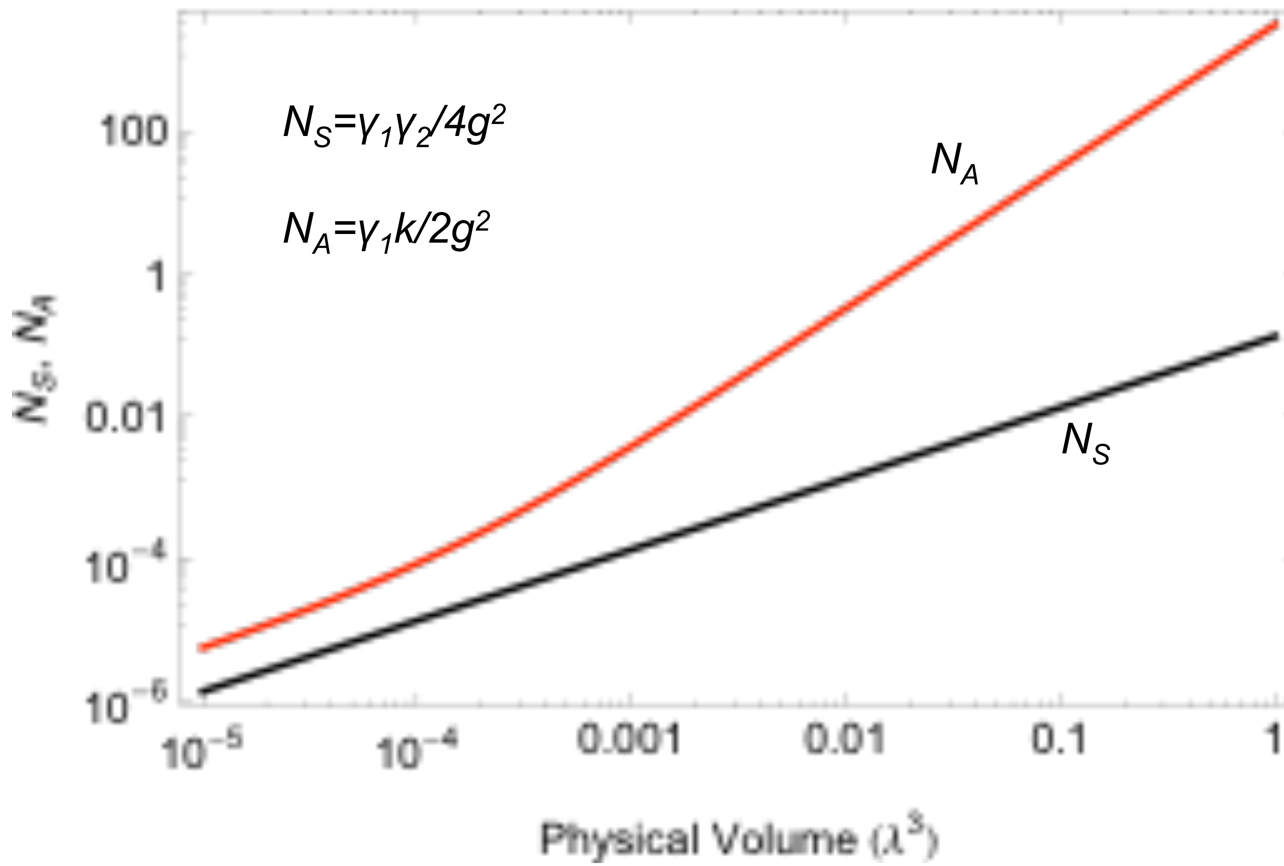
Control coherence of a vibrational wavepacket

Drive coherence between ground and excited state

D. Brinks et al., Nature (2010), A. Curto et al., Nat. Phys. (2011)

Critical parameters





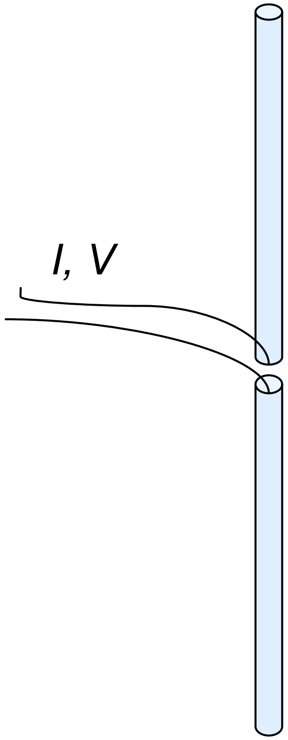
$(g,k,\gamma)=(34,2,1.25)$ MHz

$(g,k,\gamma)=(97,48,-)$ GHz

… ?

Critical photon number (N_S) (black curve) and critical atomic number (N_A) (red curve) as a function of the cavity volume. The calculation was performed assuming dephasing times T_2=100 fs and T_1 =2.7 ns at λ=740 nm.

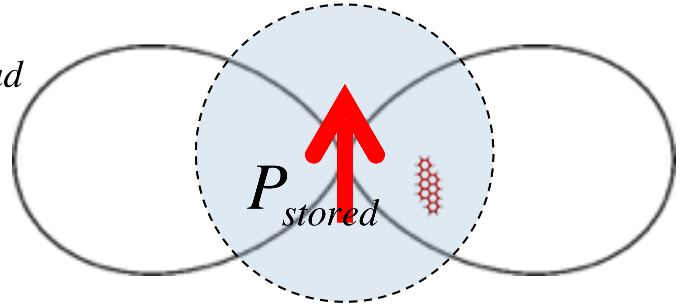
A dipole antenna



$$E \approx \frac{1}{r^3} + \frac{k}{r^2} + \frac{k^2}{r}$$

$$H \approx \frac{k}{r^2} + \frac{k^2}{r}$$

$$r_B = \frac{1}{k} \approx 100 \text{ nm}$$



$$P = P_{rad} + P_{stored}$$

$$P_{stored} \approx \frac{1}{(kr)^3}$$

M. Agio, A. Alù, eds. *Optical Antennas* (Cambridge University Press, 2013)

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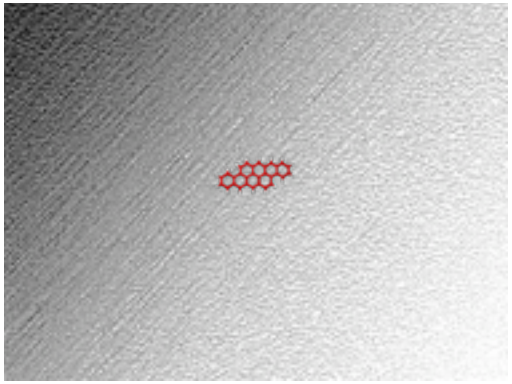
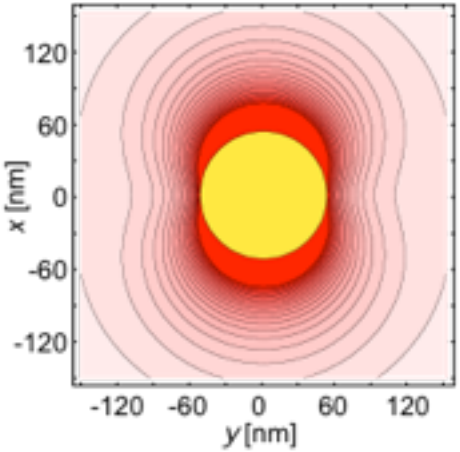
From field-enhanced spectroscopy to optical antennas

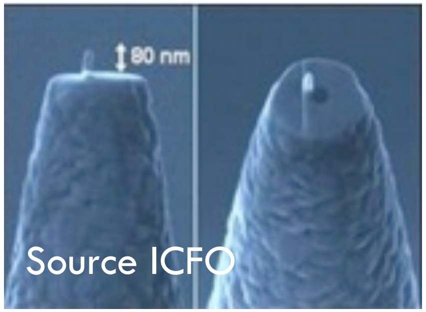
scattering ratio: $K_s = \frac{\sigma}{A}$

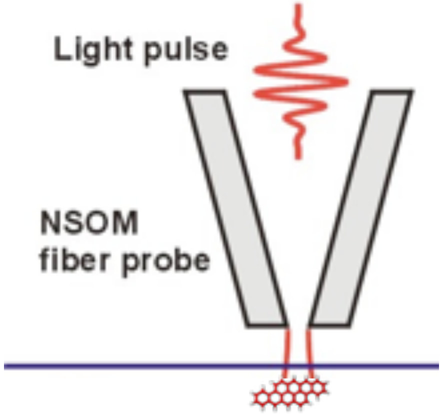
$A \geq \lambda^2$ and $\sigma << \frac{3\lambda^2}{2\pi}$

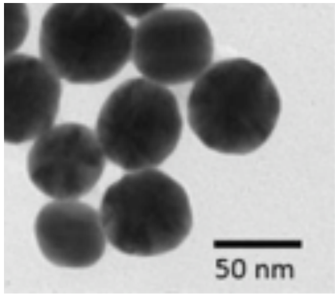




RAMAN spectroscopy:
$\sigma = 10^{-30}$ cm^2 – $A = 10^{-10}$cm^2: $K_s = 10^{-20}$



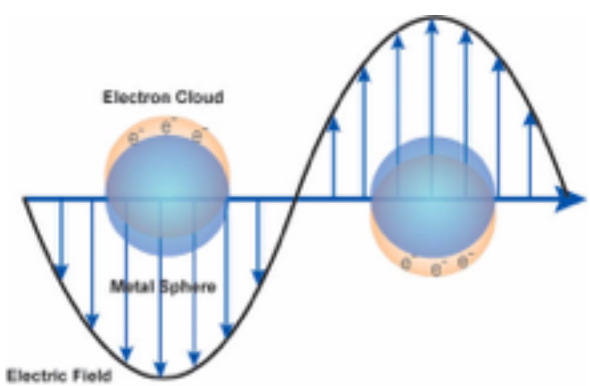


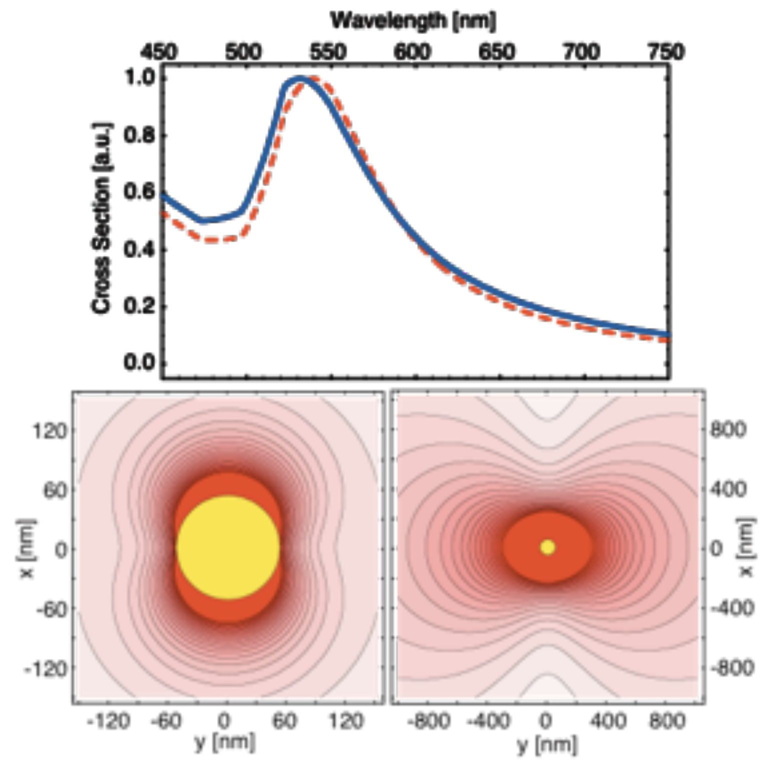


M. Moskovits, Surface-enhanced spectroscopy, Rev. Mod. Phys. (1985)
D. Pohl, Near-field optics as an antenna problem, World Sci. Publ. (2000)

Surface plasmon-polariton resonances

Lycurgus cup, Roman period, IV b.C.

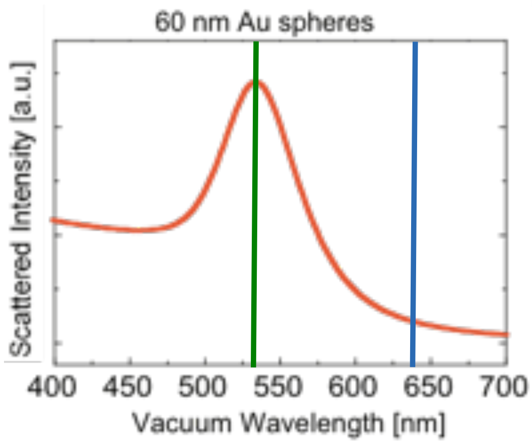


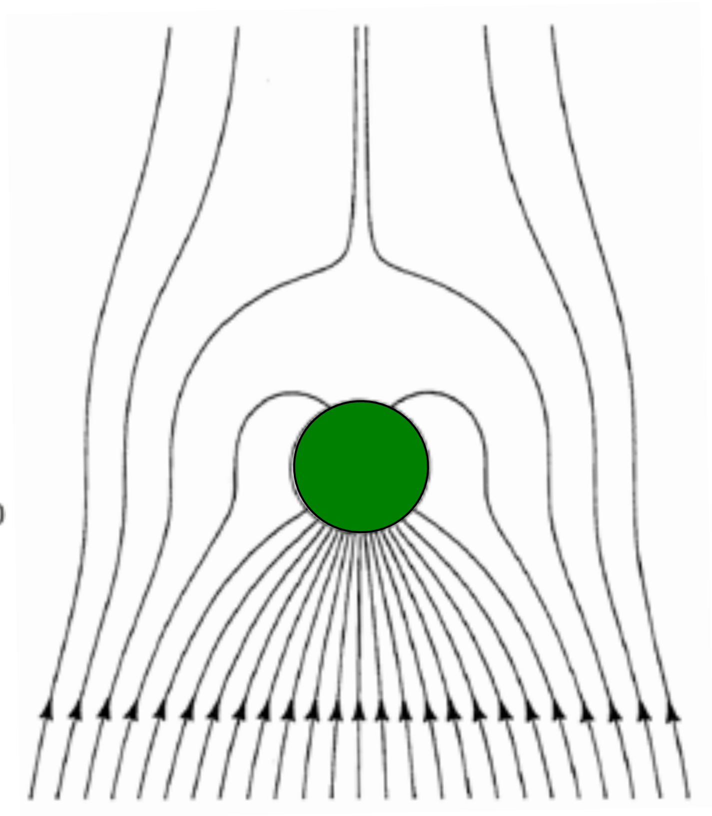


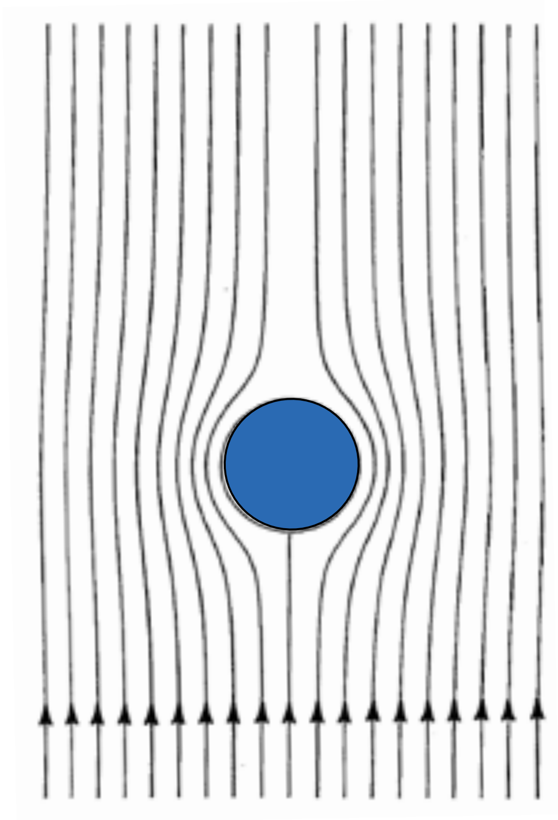
Dipolar polarizability: $\alpha(\omega) = 4\pi a^3 \dfrac{\varepsilon(\omega) - \varepsilon_b}{\varepsilon(\omega) + 2\varepsilon_b}$

C.F. Bohren, and D.R. Huffman, Absorption and scattering of light by small particles (Wiley, 1983)

Metal nanoparticle as optical antennas

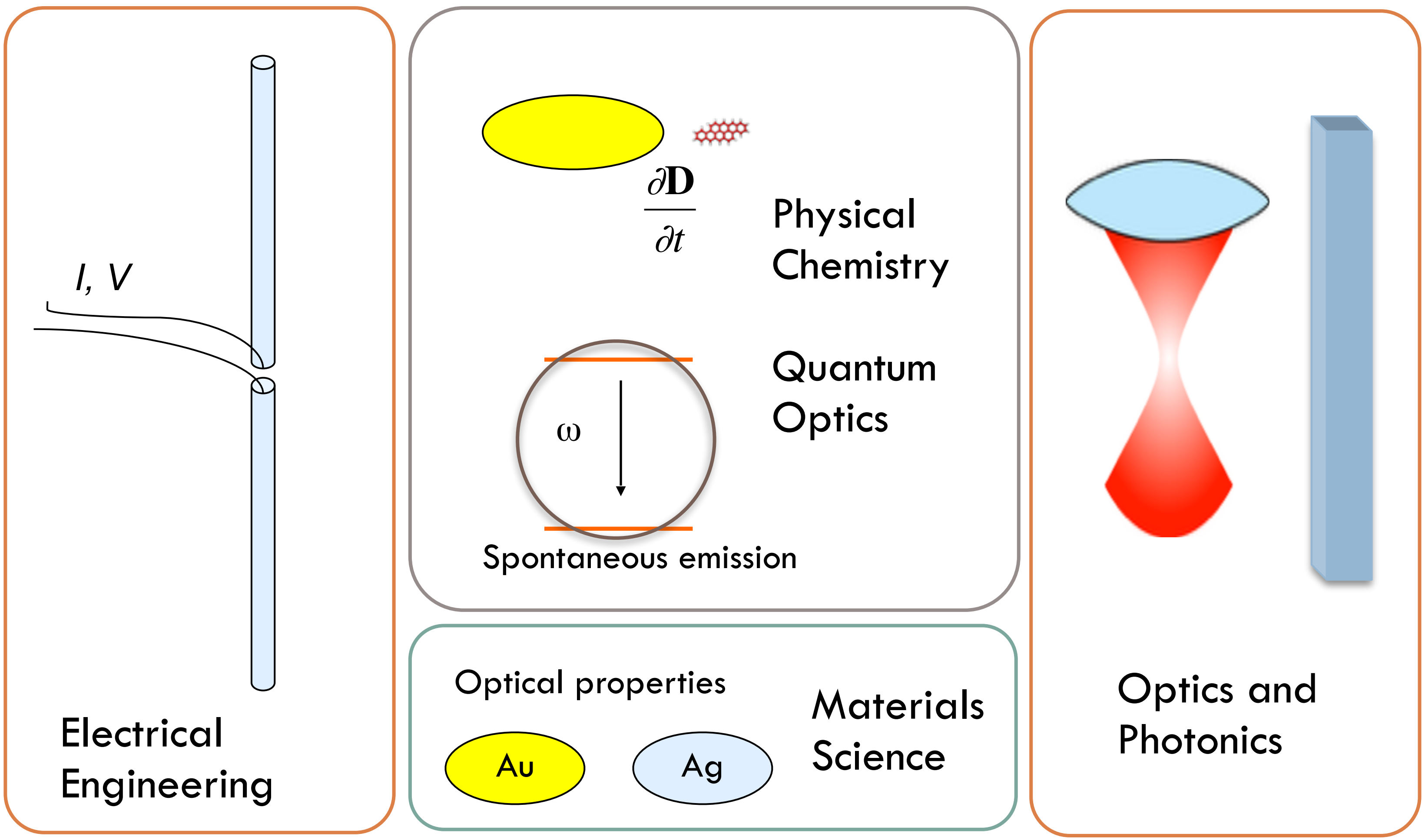






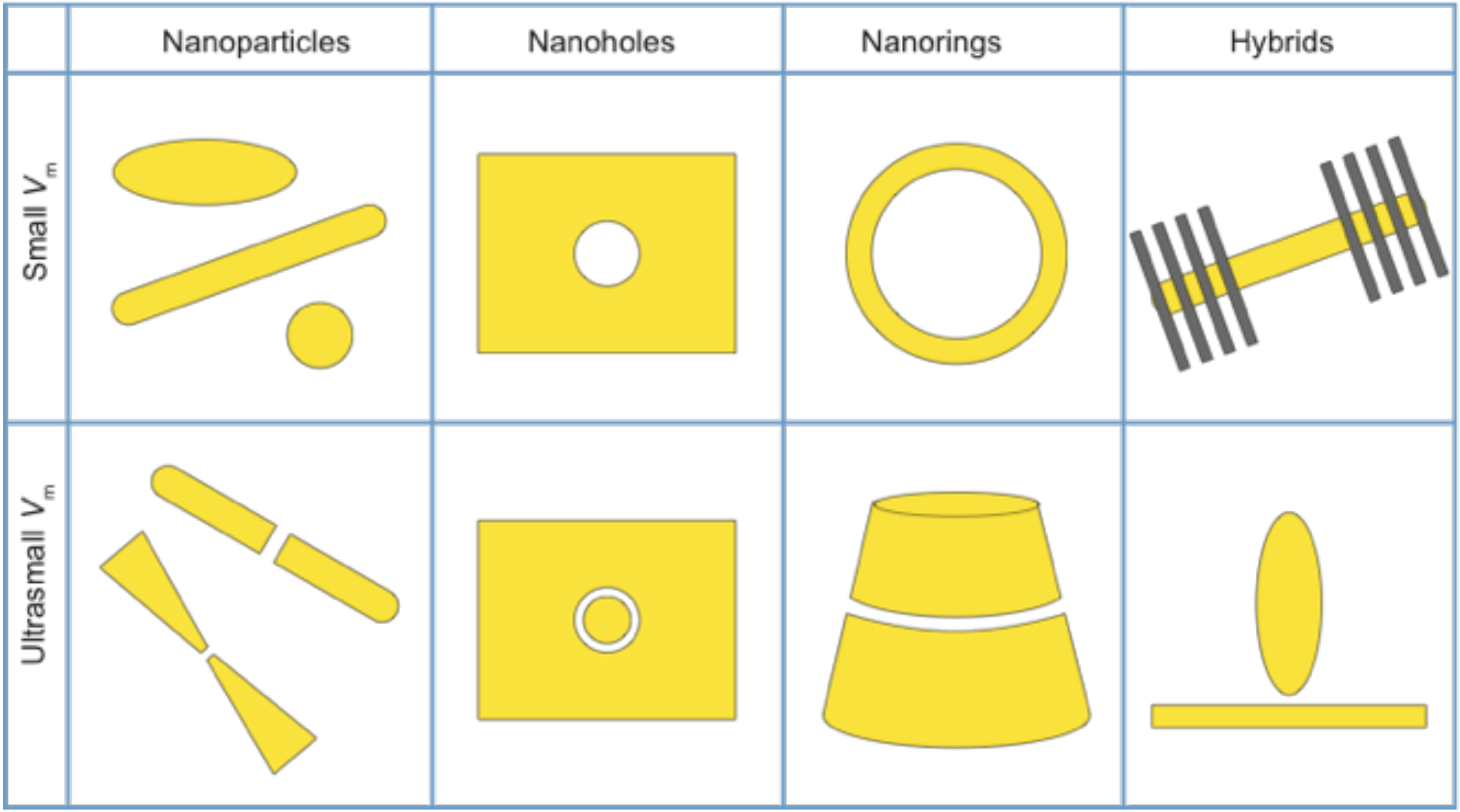
B. J. Messinger, et al., Phys. Rev. B (1981)
C. Bohren, Am. J. Phys. (1982)

Optical antennas



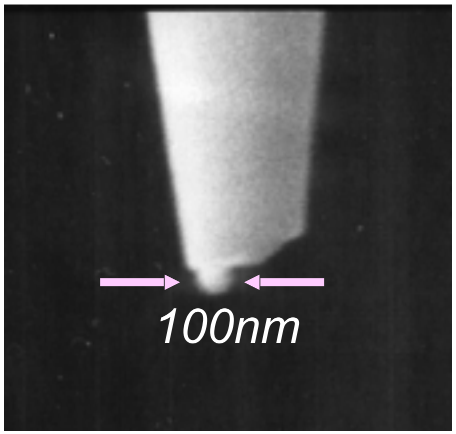
M. Agio, Habilitation Thesis (ETH Zurich 2011) http://dx.doi.org/10.3929/ethz-a-007049705

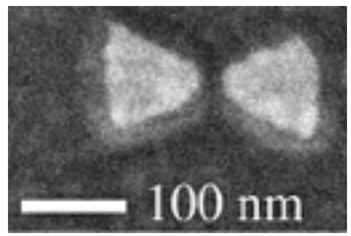
Optical nanocavities/antennas

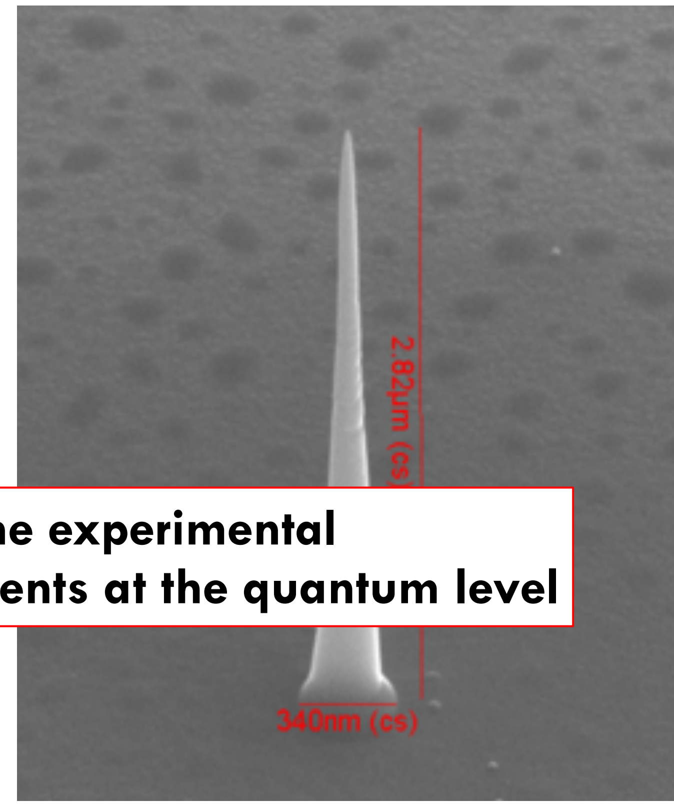


M. Agio, Nanoscale (2012)

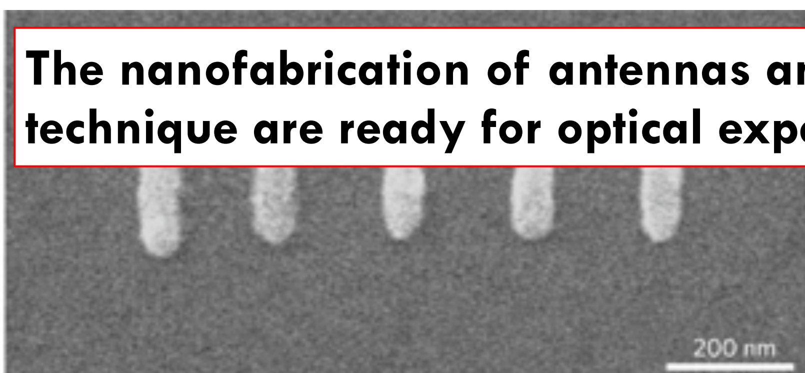
Progress in optical antennas







The nanofabrication of antennas and the experimental technique are ready for optical experiments at the quantum level



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Single-molecule fluorescence

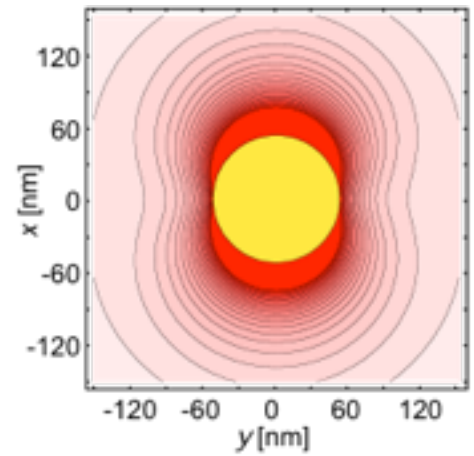
field enhancement

$$S_o \propto \eta_o |\mathbf{E}_o|^2, \quad \eta_o = \frac{\gamma_r^o}{\gamma_t^o}, \quad \gamma_t^o = \gamma_r^o + \gamma_{nr}^o$$

quantum efficiency
(radiation efficiency)

$$S \propto \eta |\mathbf{E}|^2$$

100 nm gold NP



decay rates

antenna efficiency

$$\eta = \frac{\gamma_r}{\gamma_t} = \frac{\eta_o}{(1-\eta_o)\dfrac{\gamma_r^o}{\gamma_r} + \dfrac{\eta_o}{\eta_a}}, \quad \eta_a = \frac{\gamma_r}{\gamma_r + \gamma_{nr}}$$

$$\gamma_t = \gamma_r + \gamma_{nr}^o + \gamma_{nr}$$

Fluorescence spectroscopy:
$\sigma = 10^{-15}$ cm^2 – $A = 10^{-10}$ cm^2: $K_s = 10^{-5}$

$$\frac{|\mathbf{E}|^2}{|\mathbf{E}_o|^2} \approx \frac{\gamma_r}{\gamma^o}$$

The spontaneous emission rate

$$P = -\frac{1}{2}\int_V \mathrm{Re}\left\{\mathbf{j}^*(\mathbf{r},\omega)\cdot\mathbf{E}(\mathbf{r},\omega)\right\}\mathrm{d}V$$

$$\mathbf{E}(\mathbf{r}) = \frac{1}{\varepsilon_0}\frac{\omega^2}{c^2}\overleftrightarrow{G}(\mathbf{r},\mathbf{r}_0,\omega)\cdot\mathbf{p}$$

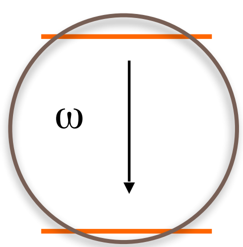
$$\mathbf{j}(\mathbf{r},\omega) = -i\omega\mathbf{p}\delta(\mathbf{r}-\mathbf{r}_0)$$

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

$$P = \frac{\omega}{2}\mathrm{Im}\left\{\mathbf{p}^*\cdot\mathbf{E}(\mathbf{r}_0)\right\}$$

$$P = \frac{\pi\omega^2}{12\varepsilon_0}|\mathbf{p}|^2\rho(\mathbf{r}_0,\omega)$$

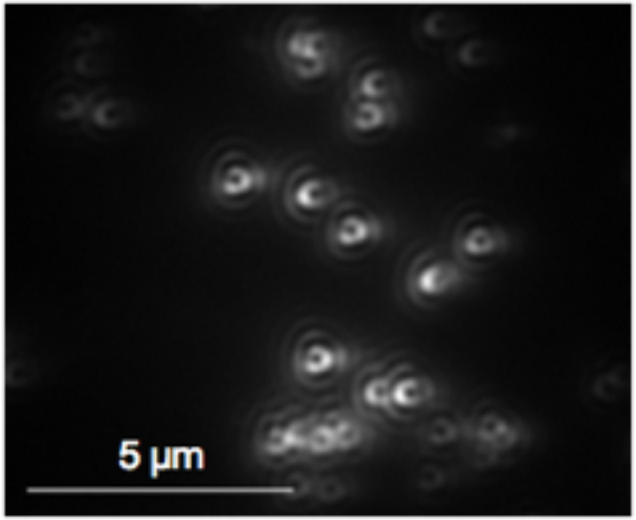
$$\frac{\rho(\mathbf{r}_0,\omega)}{\rho_0(\mathbf{r}_0,\omega)} = \frac{P}{P_0}$$

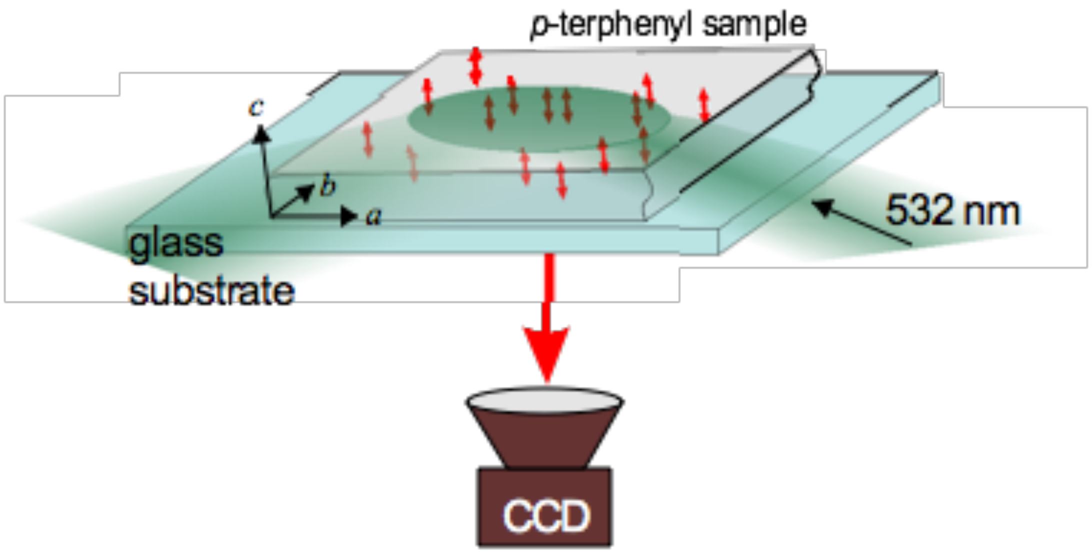


ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Photostable molecules in a thin film

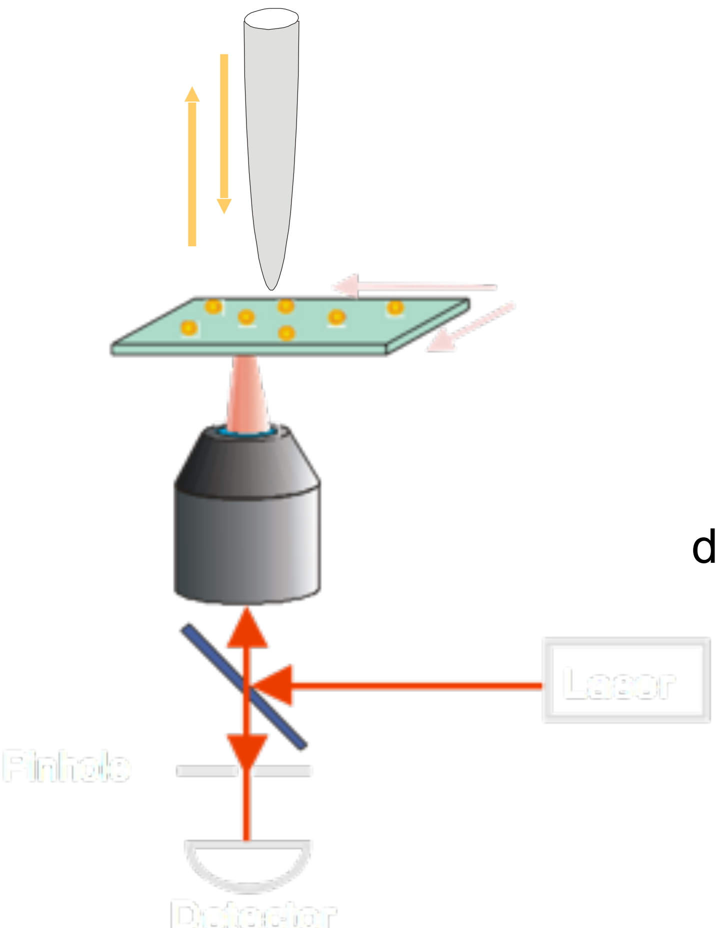
The p-terphenyl film embedding aligned terrylene molecules is about 20 nm thick

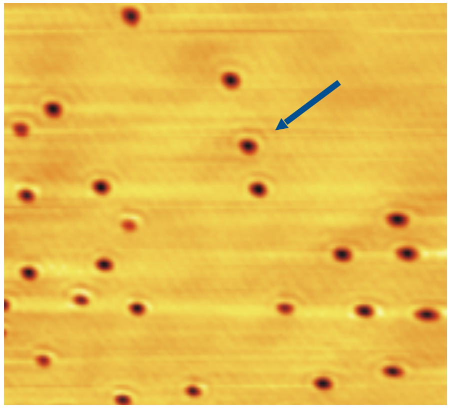


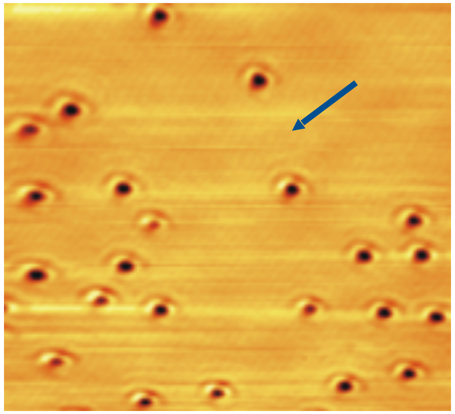


R. Pfab, J. Zimmermann, C. Hettich, I. Gerhardt, A. Renn, and V. Sandoghdar, Chem. Phys. Lett. 387, 490 (2004).

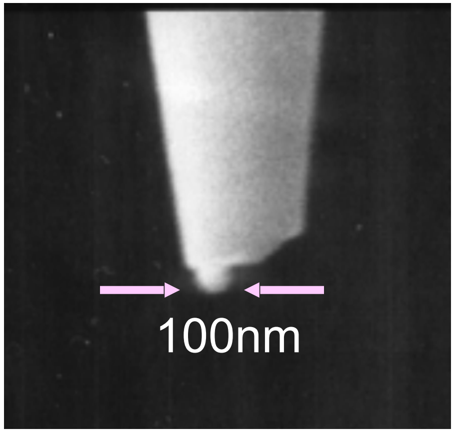
Manipulating a single GNP





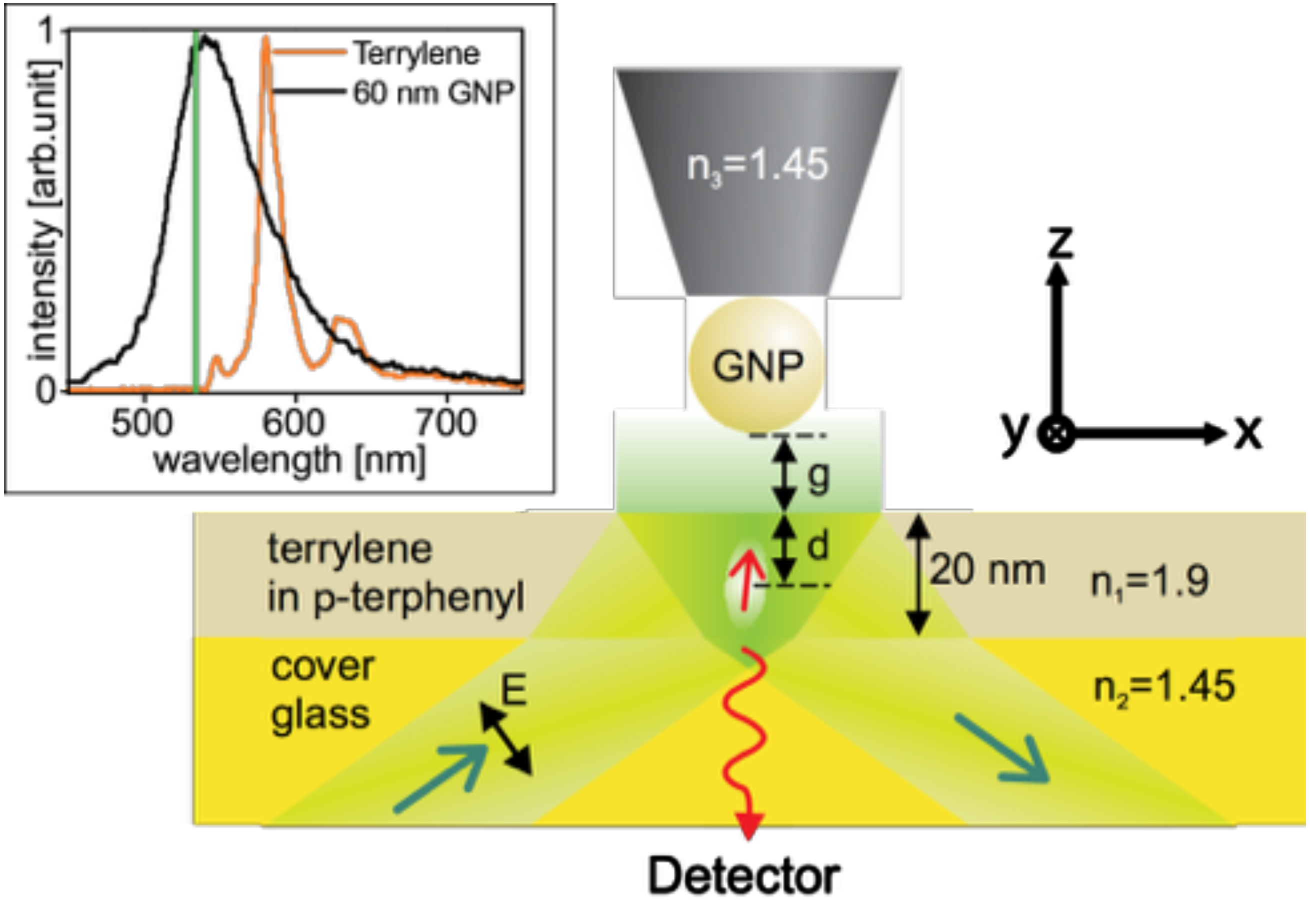


diameter: 30-100nm



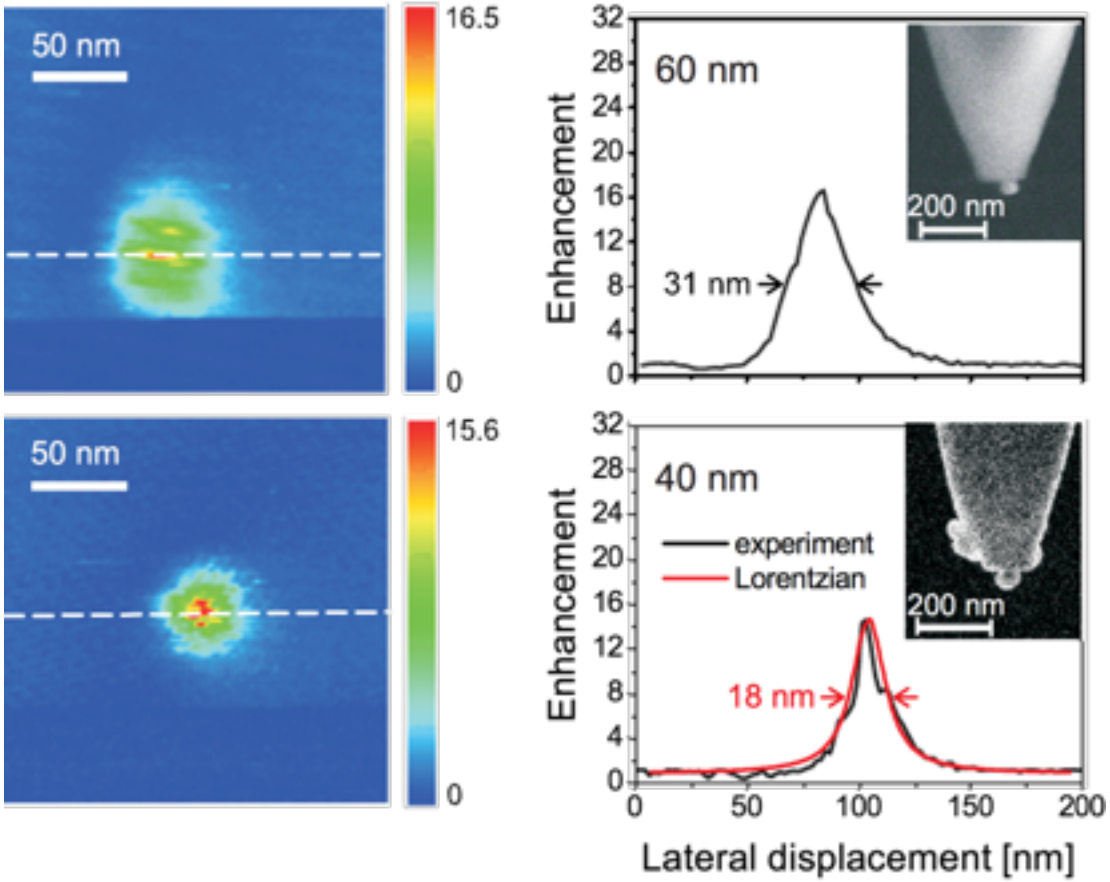
T. Kalkbrenner, et al., J. Microscopy 202, 72 (2001); Nano Lett. 4, 2309 (2004)

Interaction between a single molecule and a single GNP



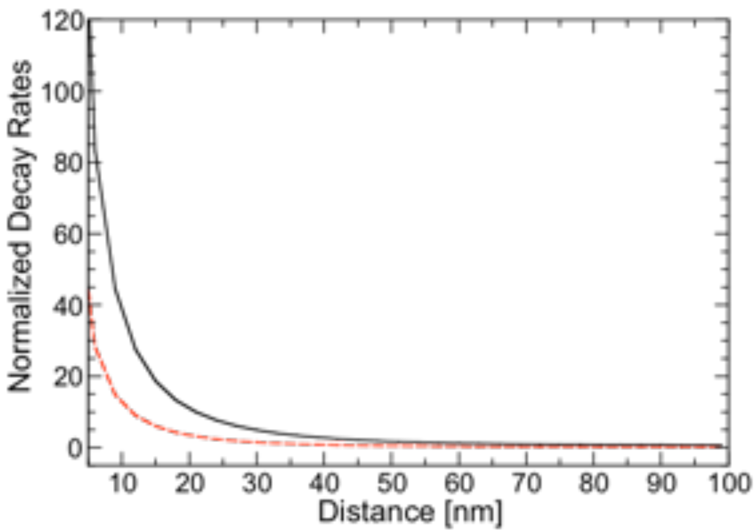
S. Kühn, U. Håkanson, L. Rogobete, and V. Sandoghdar, Phys. Rev. Lett. 97, 017402 (2006)

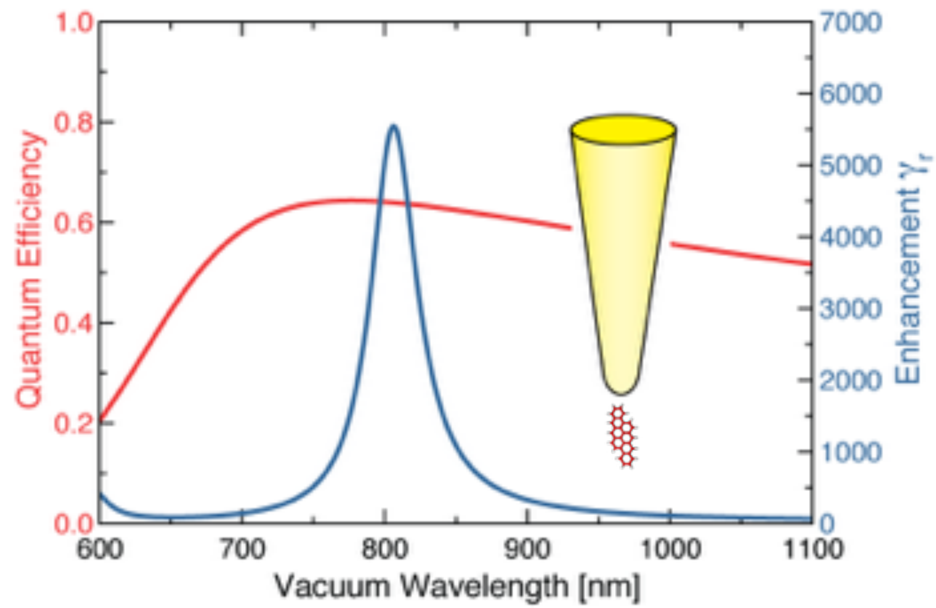
Ultrahigh resolution with fluorescence enhancement

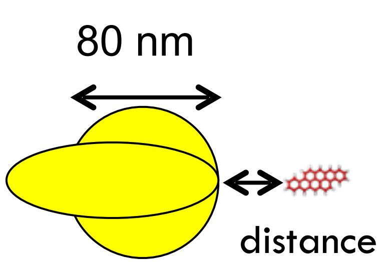


H. Eghlidi, K. G. Lee, X.-W. Chen, S. Götzinger, and V. Sandoghdar, Nano Lett. (2009).

Fighting against quenching



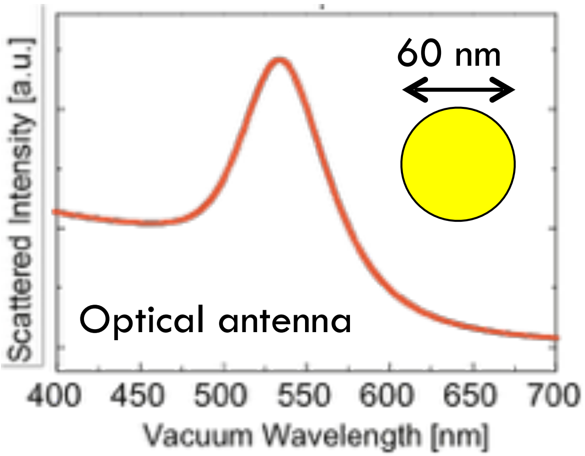




Improve the quantum yield of molecules
Photophysics under unusual regimes

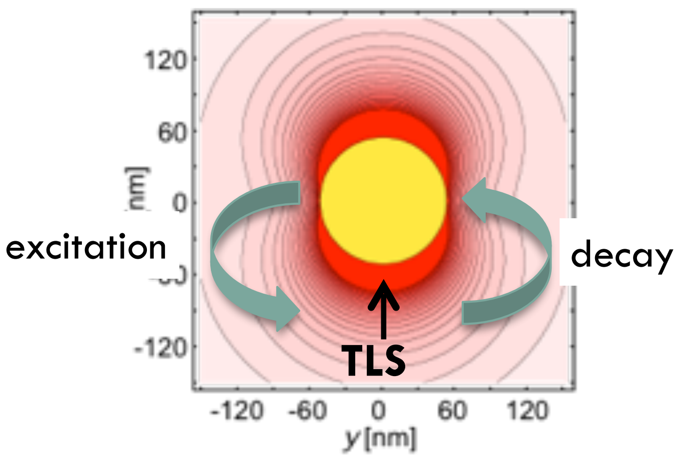
L. Rogobete, F. Kaminski, M. Agio, V. Sandoghdar, Opt. Lett. (2007)
M. Agio, Nanoscale (2012)
X.-W. Chen, M. Agio, V. Sandoghdar, Phys. Rev. Lett. (2012)

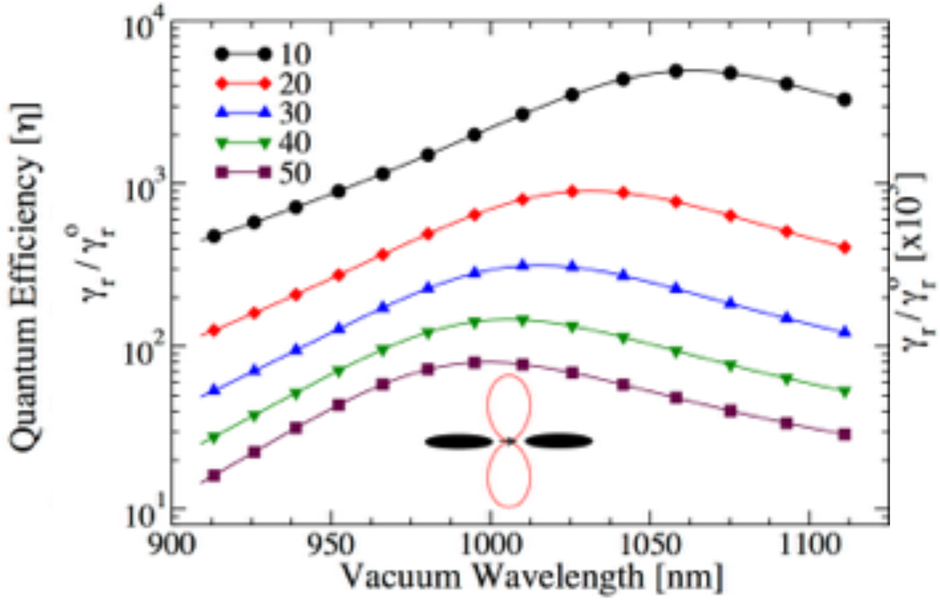
Enhancing quantum emitters



Strong enhancement with a high antenna efficiency

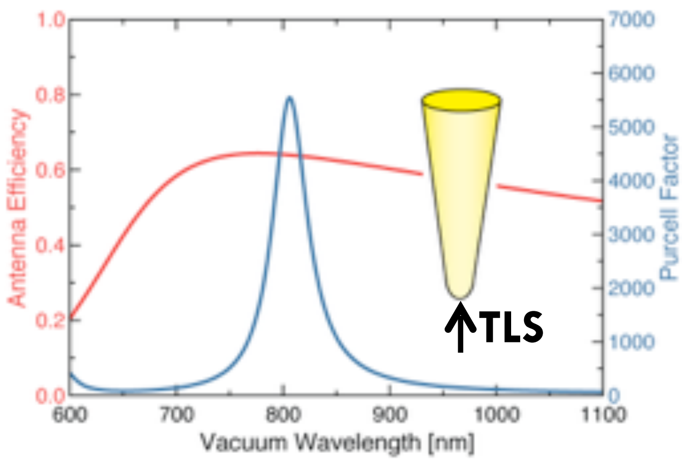
$$\eta_a = \frac{P_{rad}}{P_{rad} + P_{abs}} \approx 1$$

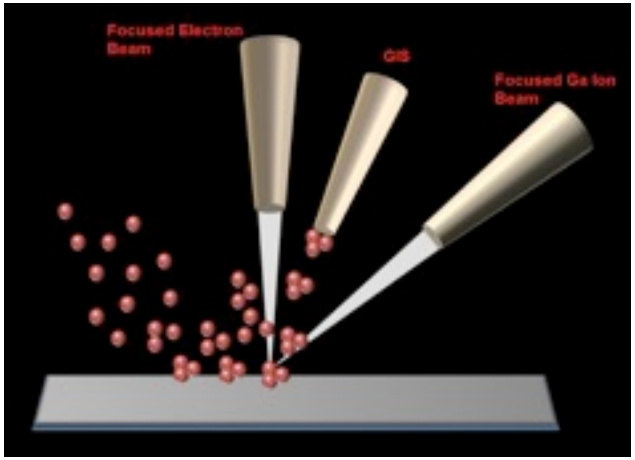




L. Rogobete, F. Kaminski, M. Agio, V. Sandoghdar, Opt. Lett. (2007), M. Agio, Nanoscale (2012)

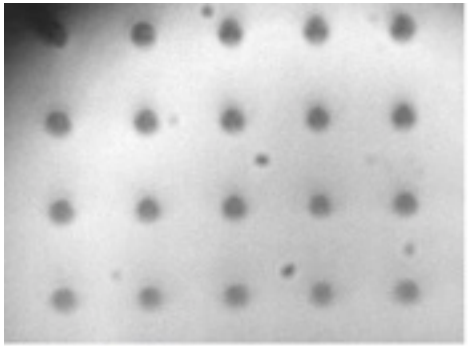
Resonant nanocones



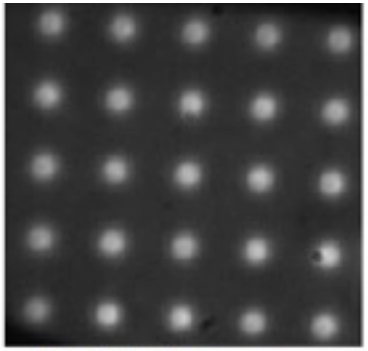


A. Mohammadi, F. Kaminski, V. Sandoghdar, M. Agio, J. Phys. Chem. C (2010)
A. Flatae in collaboration with the Nanostructures Department at IIT, Genoa, Italy

Nanocones - Nanofabrication

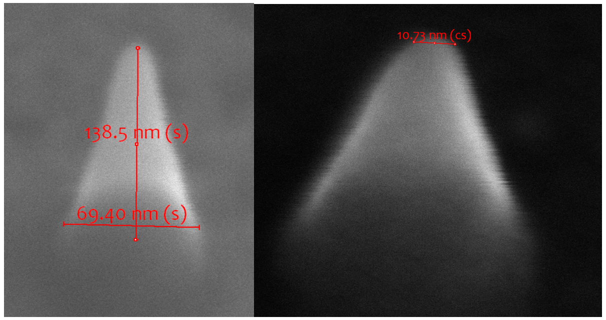


Bright field

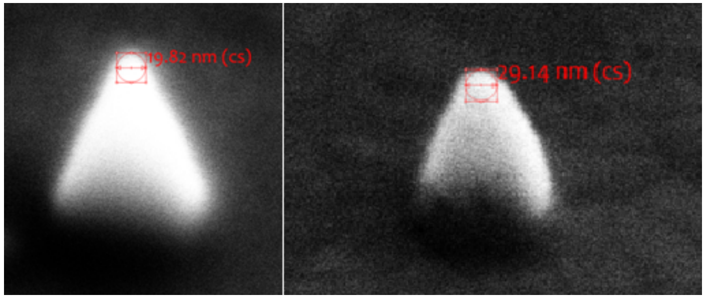


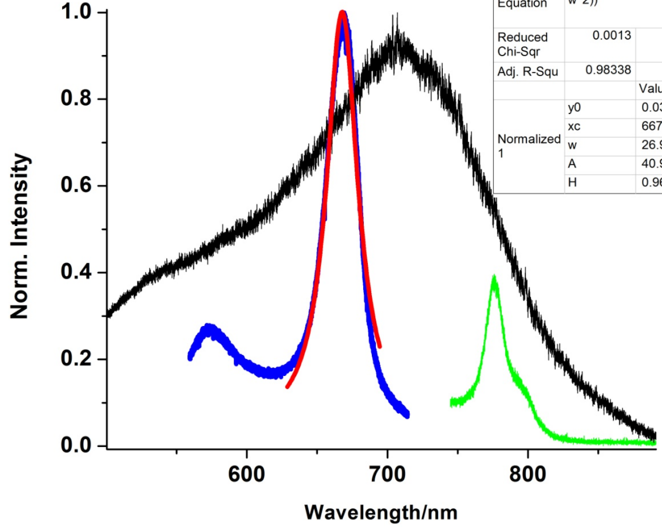
Dark field Images

Before gold deposition



After gold deposition





Model	Lorentz		
Equation	y = y0 + (2*A/PI)*(w/(4*(x-xc)^2 + w^2))		
Reduced Chi-Sqr	0.0013		
Adj. R-Squ	0.98338		
		Value	Standard E
Normalized 1	y0	0.03345	0.00201
	xc	667.841	0.01842
	w	26.9572	0.11217
	A	40.9890	0.21788
	H	0.96799	

A. Flatae in collaboration with the Nanostructures Department at IIT, Genoa, Italy

Outline

UNIVERSITÄT SIEGEN

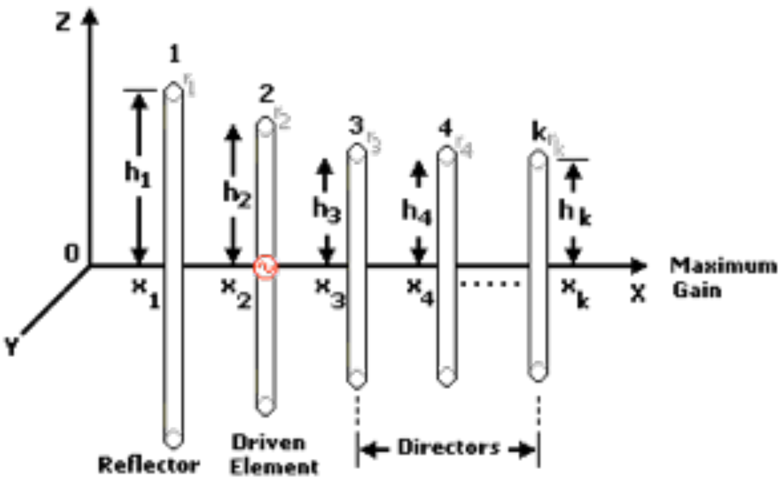
- Part I
    - Nano quantum optics
    - Metal nanostructures as optical antennas
    - Fluorescence enhancement
    - **Directional emission**
    - Quantum optics with optical antennas
- Part II
    - Quantum emitters
    - Intrinsic optical properties of diamond
    - Color centres in diamond
    - Diamond-based nanophotonics
    - Diamond electronics

Directional emission

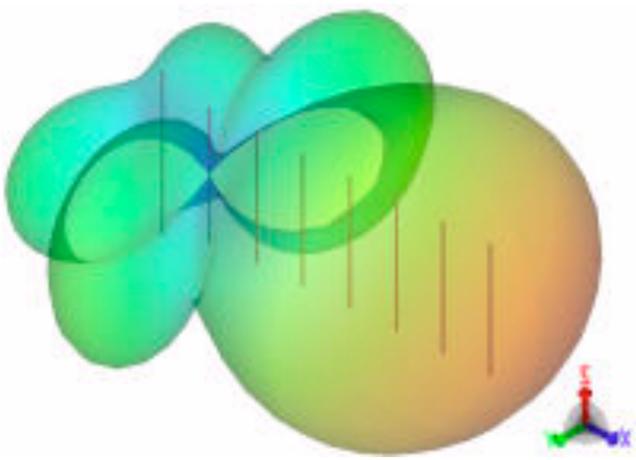
Aim: create a nanoscale 4π optical system

Antenna directivity $$D(\theta,\varphi) = \frac{4\pi}{P_{\mathrm{rad}}} P(\theta,\varphi)$$

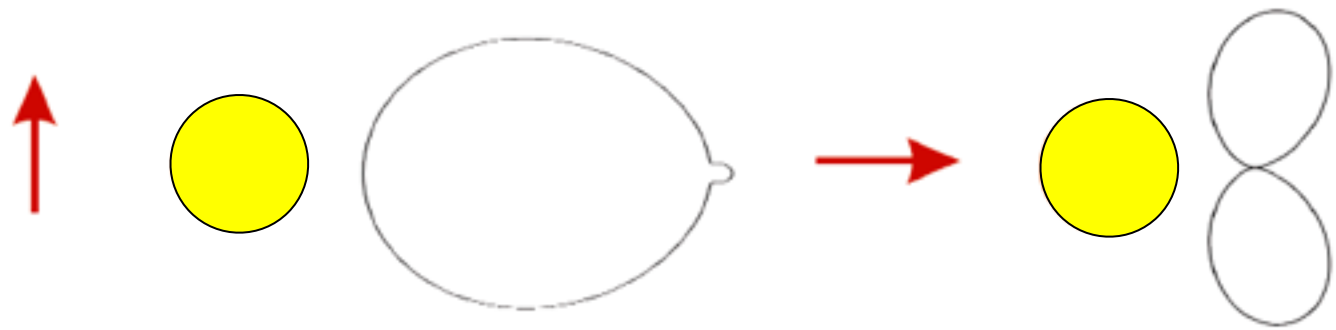
Antenna gain $$G(\theta,\varphi) = \frac{4\pi}{P} \max\{P(\theta,\varphi)\} = \eta_a \max\{D(\theta,\varphi)\}$$



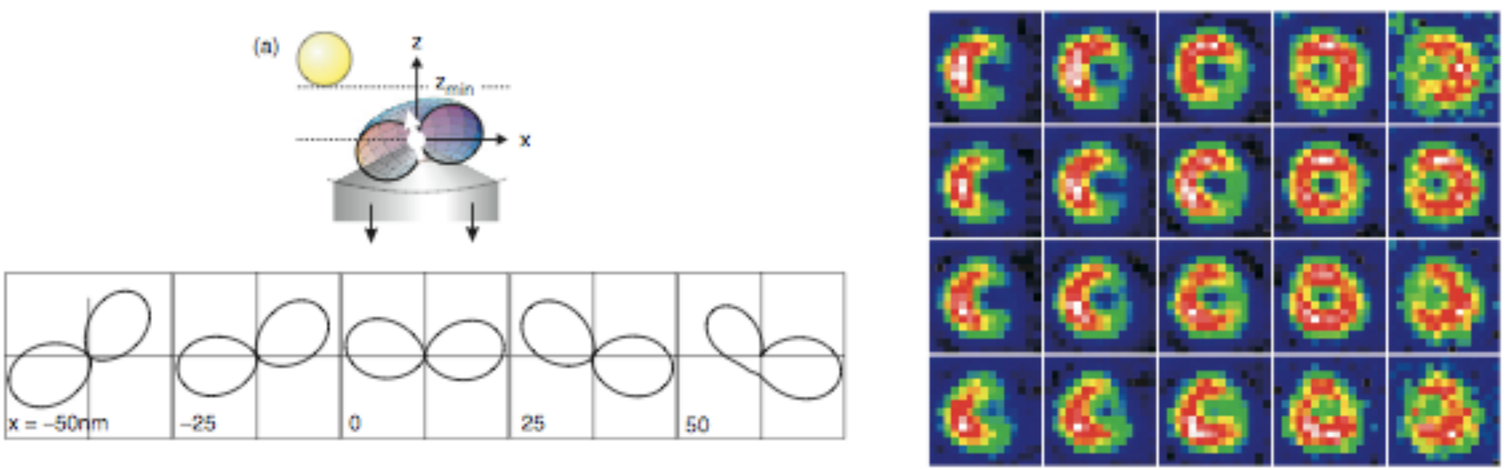
Geometry of a K elements Yagi-Uda



Redirecting light emission



Modification of the emitter radiation pattern using metal nano-particles
(L. Rogobete, PhD thesis, ETH Zurich 2006)



S. Kühn, G. Mori, M. Agio, V. Sandoghdar, Mol. Phys. (2008)

Antenna arrays

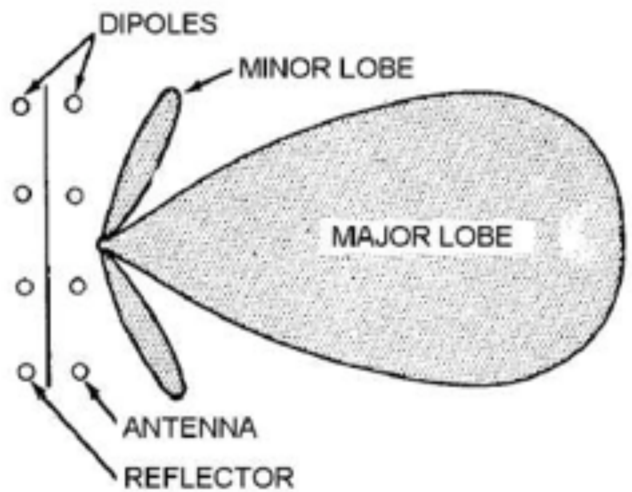
$$\mathbf{E} = \frac{Zck^2}{4\pi}\frac{e^{ikr}}{r}\sum_i e^{-ik\mathbf{n}\cdot\mathbf{d}_i}(\mathbf{n}\times\mathbf{p}_i)\times\mathbf{n}$$

$$\mathbf{S} = \frac{Zc^2k^4}{32\pi^2r^2}\mathrm{Re}\left\{\sum_{i,j} e^{-ik\mathbf{n}\cdot\left(\mathbf{d}_i-\mathbf{d}_j\right)}\left[\mathbf{p}_i\cdot\mathbf{p}_j^* - \left(\mathbf{n}\cdot\mathbf{p}_i\right)\left(\mathbf{n}\cdot\mathbf{p}_j^*\right)\right]\right\}\mathbf{n}$$

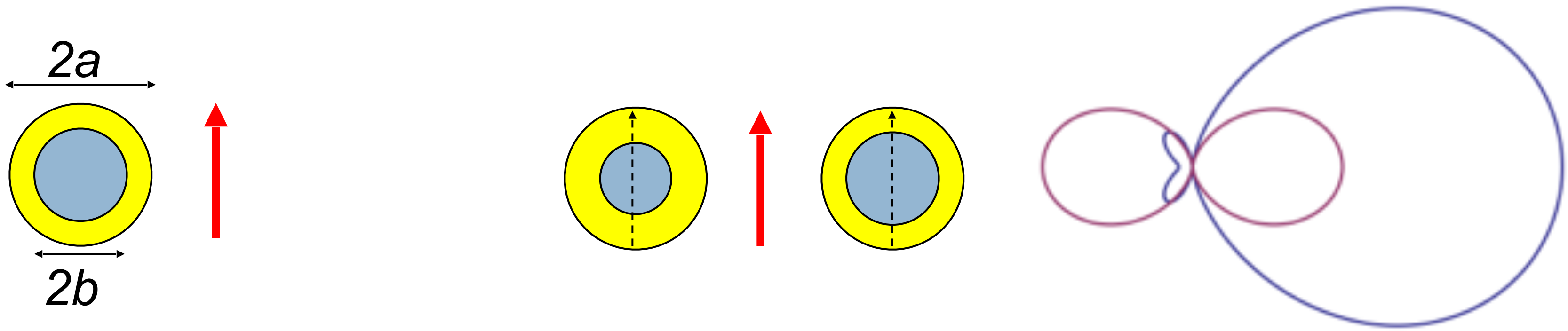
$$\overleftrightarrow{\mathbf{G}}(\mathbf{r}) = \frac{e^{ikr}}{4\pi\varepsilon r}\left[k^2(\overleftrightarrow{\mathbf{1}} - \mathbf{nn}) + (3\mathbf{nn} - \overleftrightarrow{\mathbf{1}})\left(\frac{1}{r^2} - \frac{ik}{r}\right)\right]$$

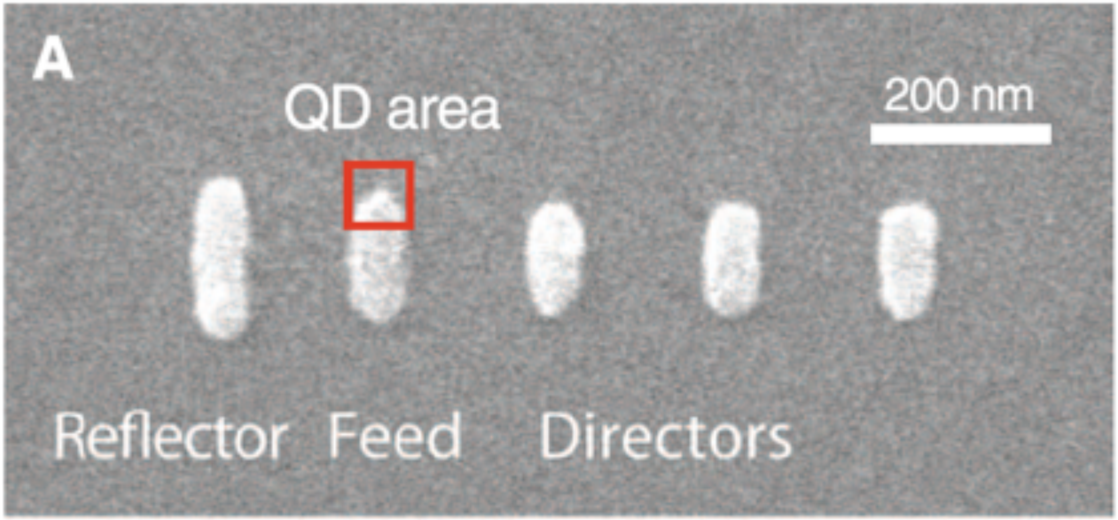
$$\mathbf{p}_i = \alpha_i\left[\sum_{j\neq i}\overleftrightarrow{\mathbf{G}}\left(\mathbf{r}_i - \mathbf{r}_j\right)\cdot\mathbf{p}_j + \overleftrightarrow{\mathbf{G}}\left(\mathbf{r}_i\right)\cdot\mathbf{p}_o\right]$$

$$\mathbf{p}_i = \alpha_i\left[\sum_{j\neq i}\overleftrightarrow{\mathbf{G}}\left(\mathbf{r}_i - \mathbf{r}_j\right)\cdot\mathbf{p}_j + \mathbf{E}_{\mathrm{inc}}\left(\mathbf{r}_i\right)\right]$$



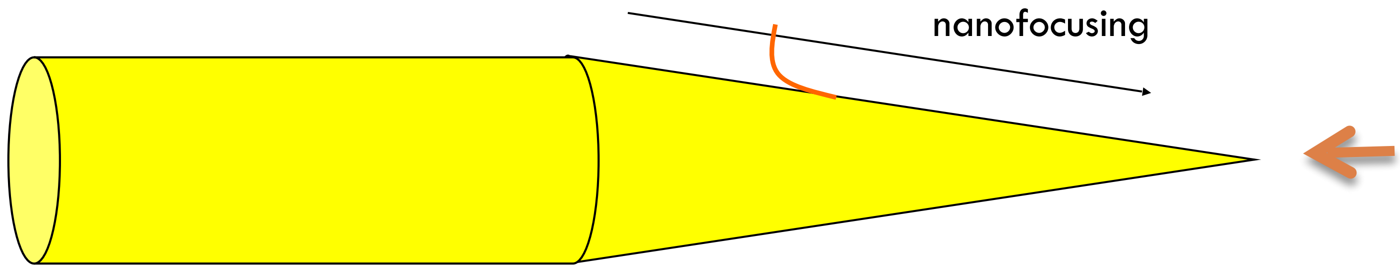
The optical Yagi-Uda antenna





J. Li, A. Salandrino, and N. Engheta, Phys. Rev. B (2007)
H.F. Hofmann, T. Kosako, and Y. Kadoya, New J. Phys. (2007)
A.G. Curto, G. Volpe, T.H. Taminiau, M.P. Kreuzer, R. Quidant, N.F. van Hulst, Science (2010)

Monolithic directional antennas

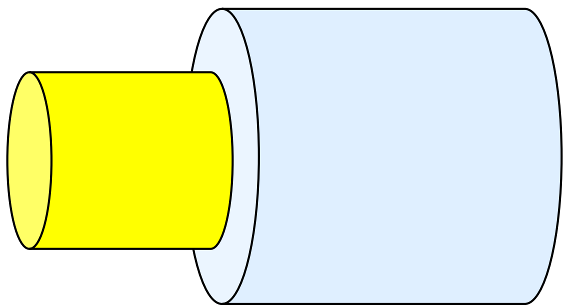


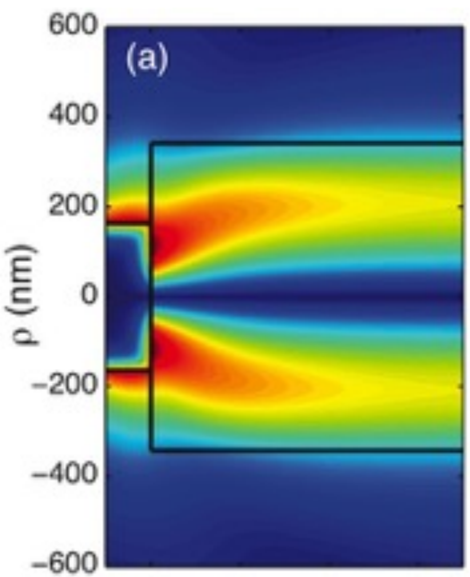
F. Keilmann, J. Microscopy (1999)
A.J. Babadjanyan, N.L. Margaryan, and Kh.V. Nerkarayan, J. Appl. Phys. (2000)
M.I. Stockman, Phys. Rev. Lett. (2004)

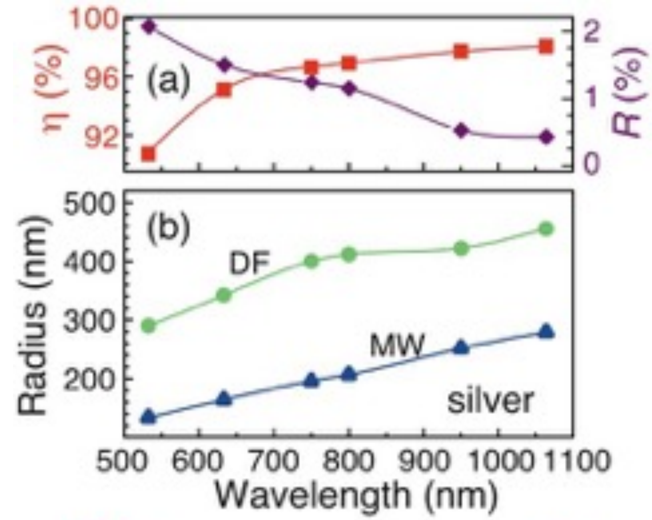
metal nanowire

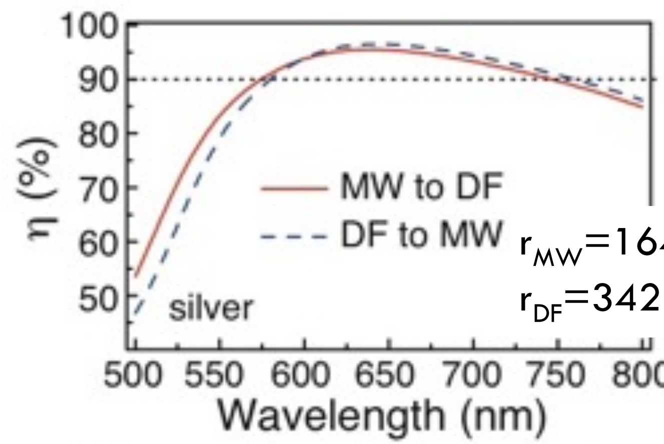
D.E. Chang, A.S. Sørensen, P.R. Hemmer, and M.D. Lukin, Phys. Rev. Lett. (2006)

Ag nanowires to SiO_2 fibers





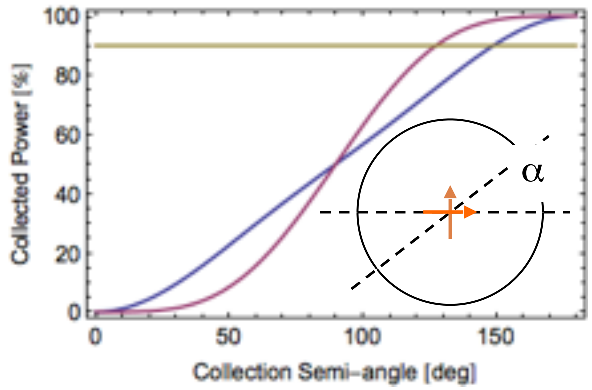


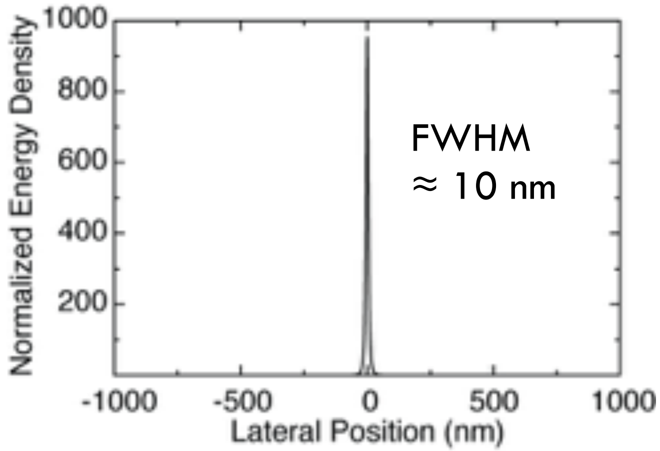


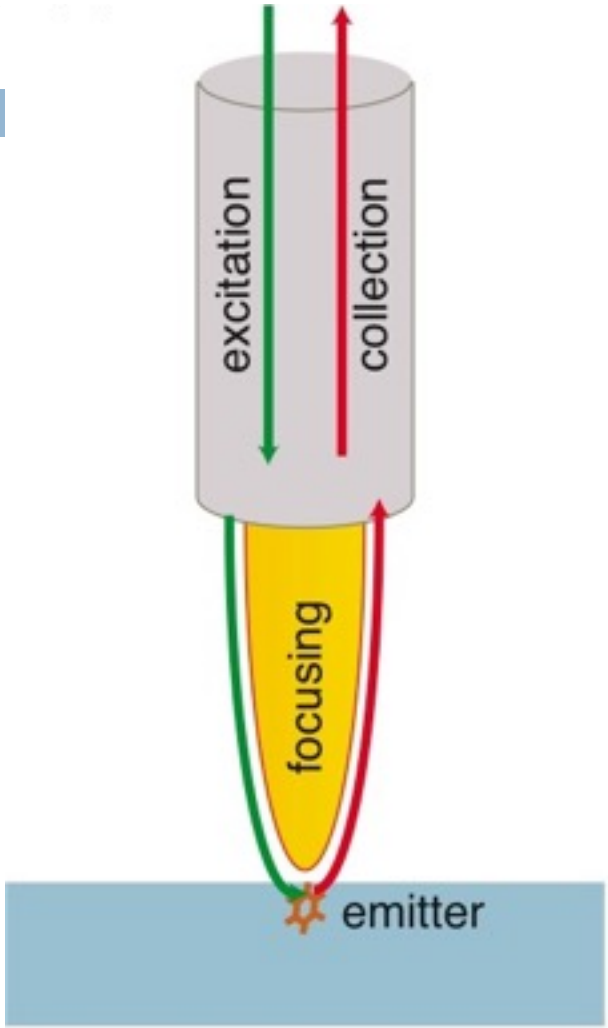
r_{MW}=164 nm
r_{DF}=342 nm

X. Chen, V. Sandoghdar, and M. Agio, Nano Lett. (2009)

High-throughput SNOM

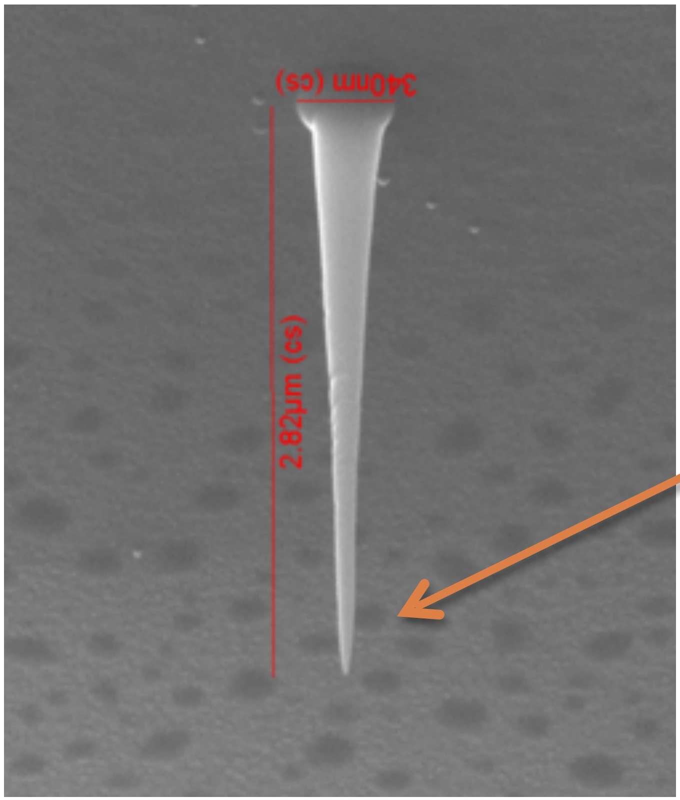


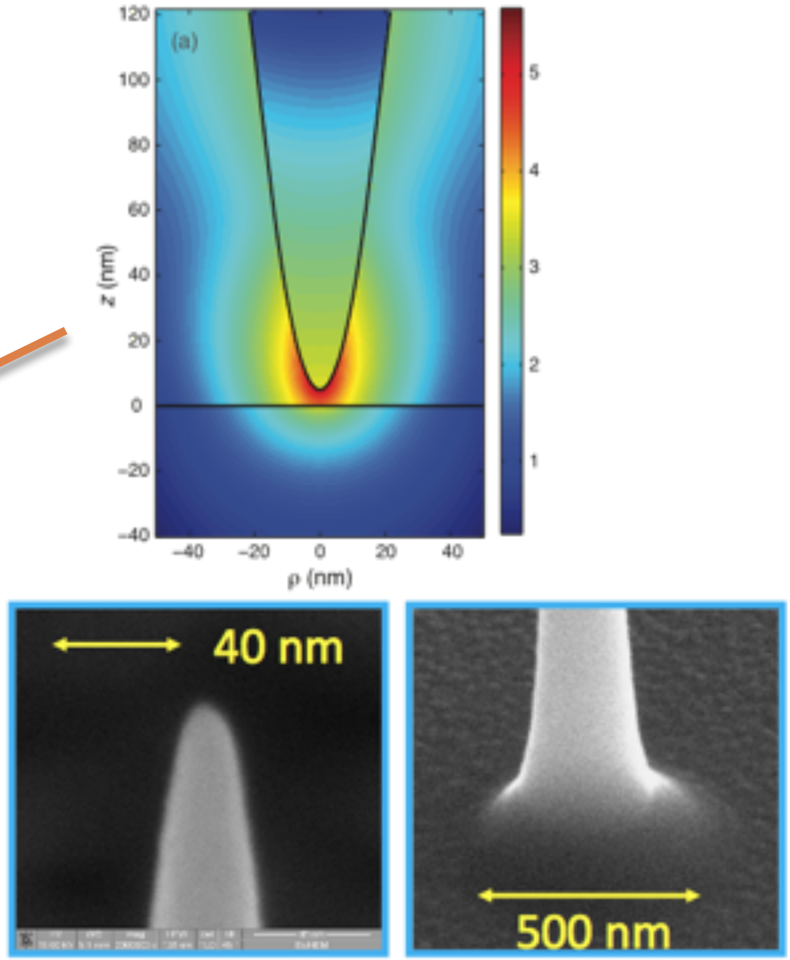




X. Chen, V. Sandoghdar, and M. Agio, Nano Lett. (2009)

Nanofabricated optical antennas



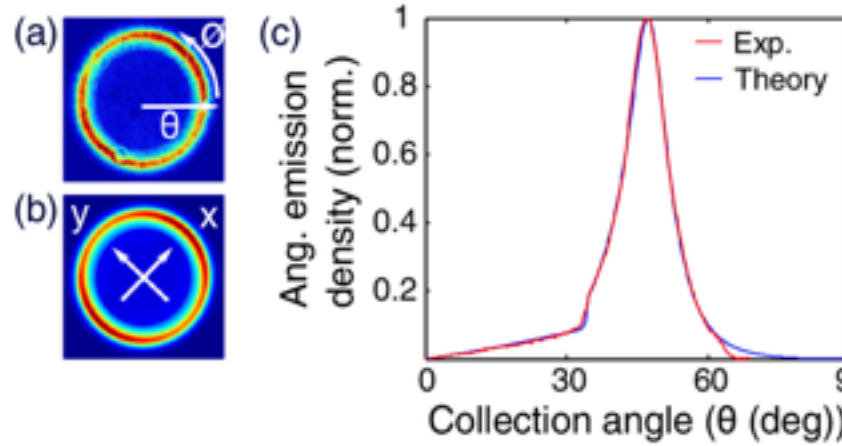


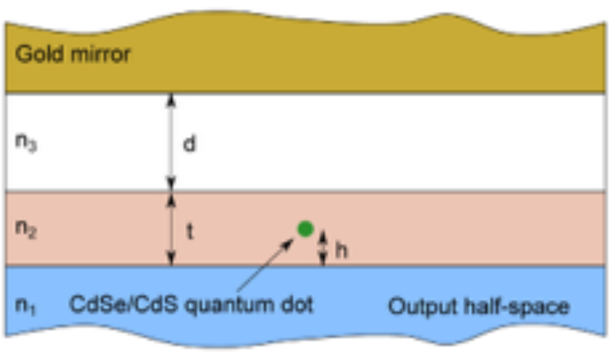
Courtesy of Prof. E. Di Fabrizio (IIT, Italy)
F. De Angelis, et al., Nano Lett. (2008)
F. De Angelis, et al., Nat. Nanotech. (2009)

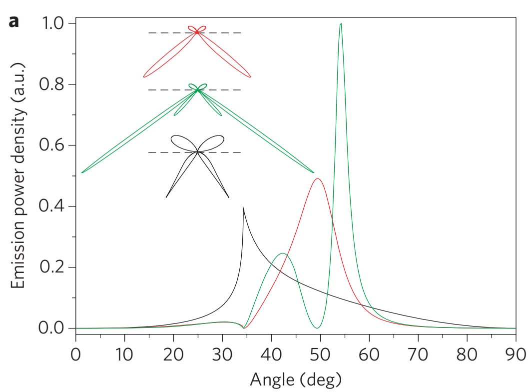
Gold & silver available

Nearly 100% collection efficiency

G.K. Lee, X.W. Chen, H. Eghlidi, P. Kukura, R. Lettow. A. Renn, V. Sandoghdar, S. Götzinger, Nat. Photonics (2011)
X.-L. Chu, T. J. K. Brenner, X.-W. Chen, Y. Ghosh, J. A. Hollingsworth, V. Sandoghdar, S. Götzinger, Optica (2014)







Research Article
Vol. 4, No. 1 / January 2017 / *Optica* 71
optica

Experimental realization of an absolute single-photon source based on a single nitrogen vacancy center in a nanodiamond

Beatrice Rodiek,[1] Marco Lopez,[1] Helmuth Hofer,[1] Geiland Porrovecchio,[2] Marek Smid,[2] Xiao-Liu Chu,[3,4] Stephan Gotzinger,[3,4] Vahid Sandoghdar,[4] Sarah Lindner,[5] Christoph Becher,[5] and Stefan Kuck[1,*]

[1]*Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany*
[2]*Cesky Metrologický Institut (CMI), Okruzni 31, 63800 Brno, Czech Republic*
[3]*Department of Physics & Graduate School in Advanced Optical Technologies (SAOT), Friedrich Alexander University (FAU) Erlangen-Nürnberg, 91052 Erlangen, Germany*
[4]*Max Planck Institute for the Science of Light, 91058 Erlangen, Germany*
[5]*Universität des Saarlandes, Fachrichtung 7.2 (Experimentalphysik), Campus E2.6, 66123 Saarbrücken, Germany*
*Corresponding author: stefan.kueck@ptb.de

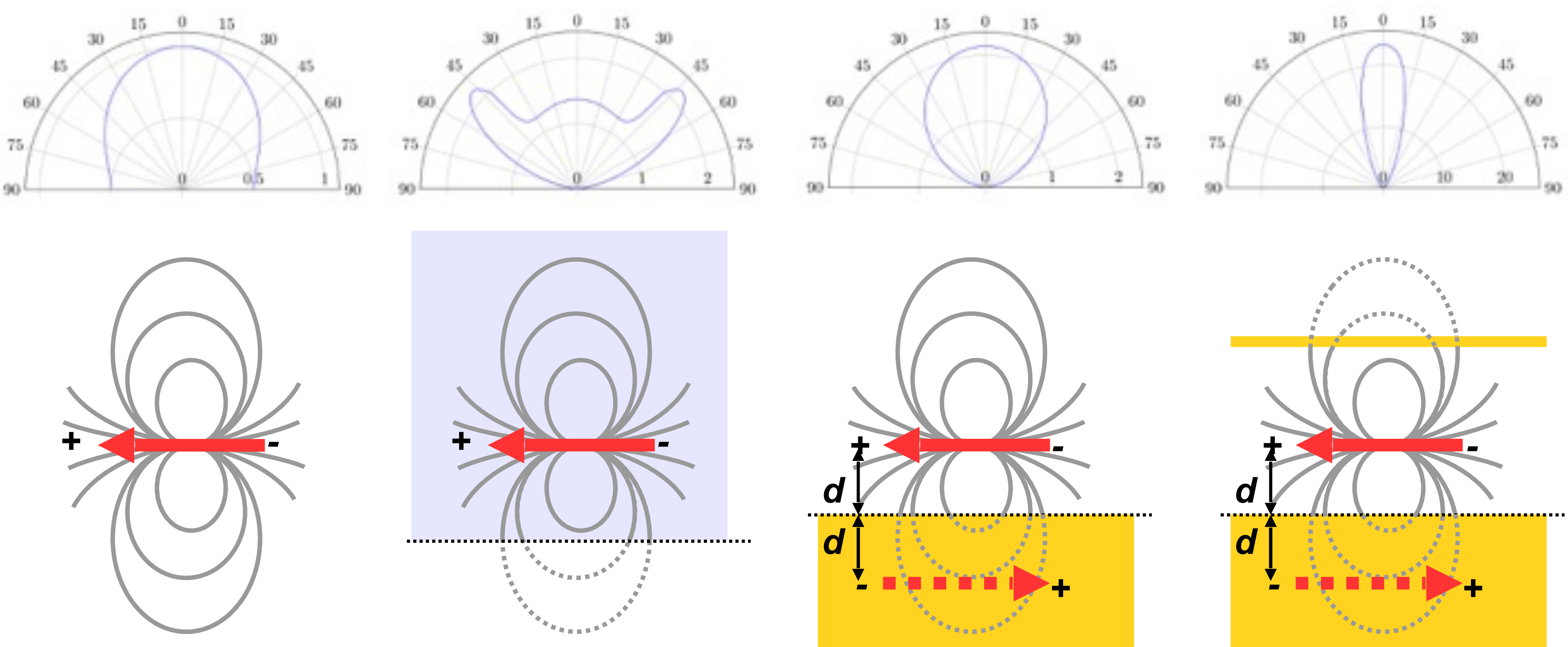
A planar directional antenna

Homogeneous medium

Glass cover slip

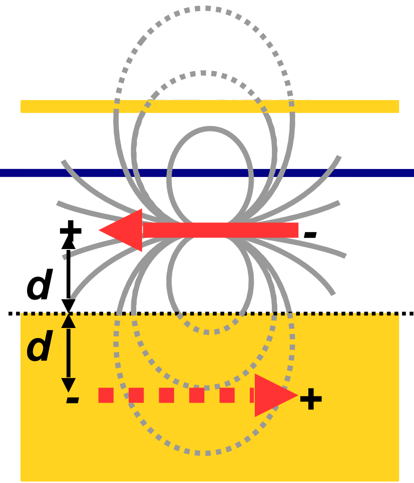
Reflector

Reflector, director

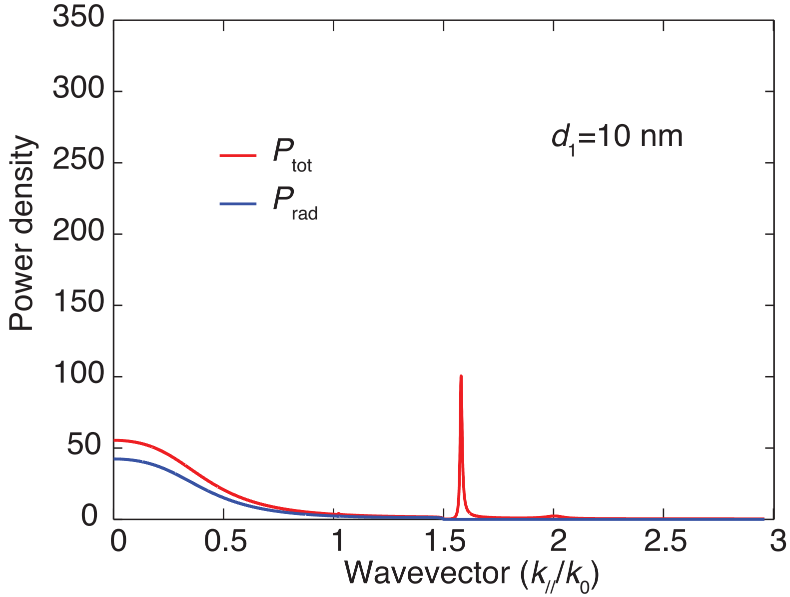


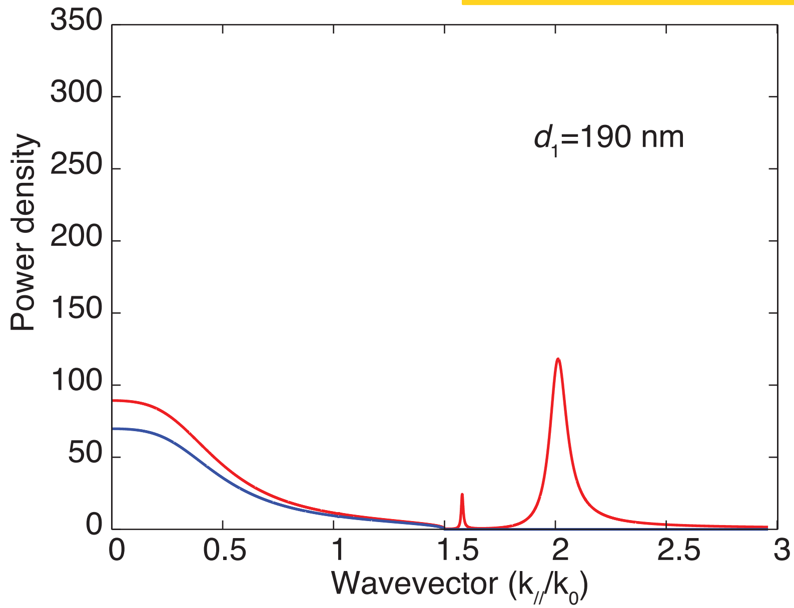
S. Checcucci, P.E. Lombardi, S. Rizvi, N. Gruhler, F.B.C. Dieleman, F.S. Cataliotti, W.H.P. Pernice, M. Agio, C. Toninelli, Light: Science & Applications (2017)

Coupling to surface plasmon polaritons



SPP are responsible for limiting the outcoupling





k_p = in-plane wavevector

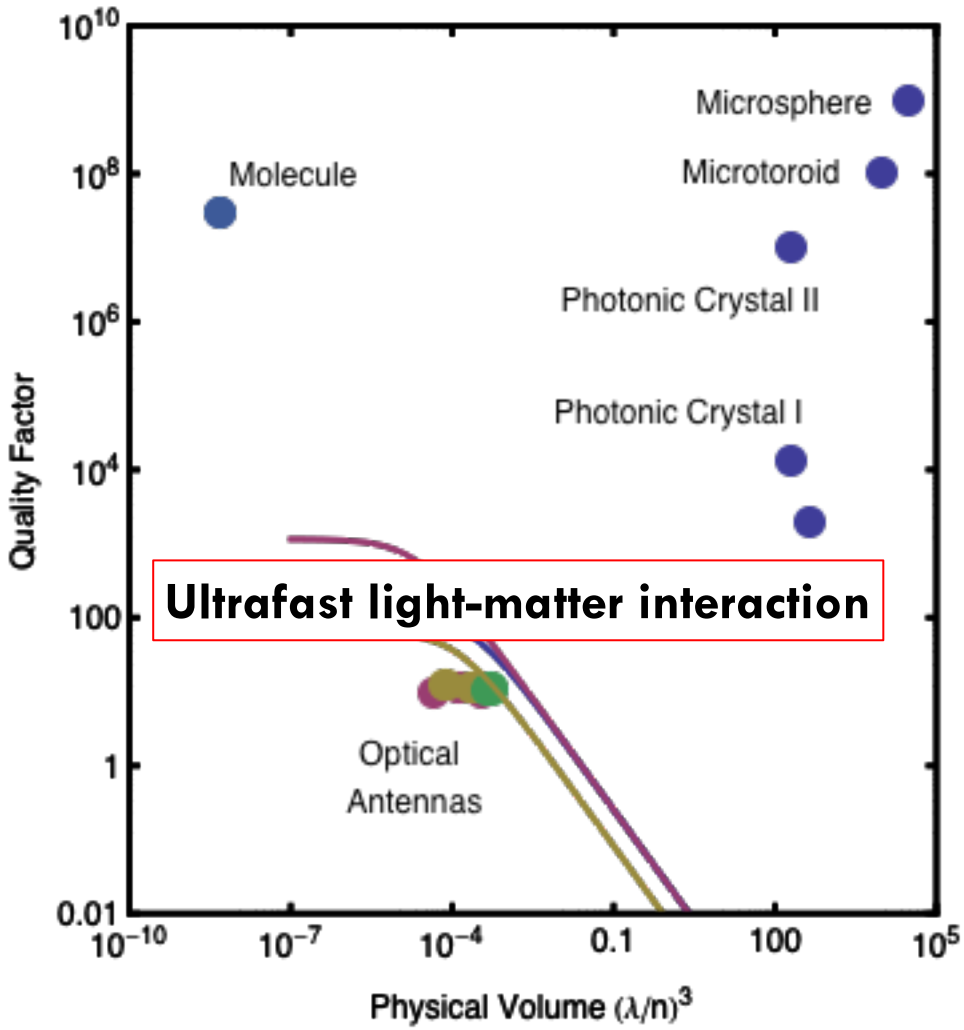
S. Checcucci, P.E. Lombardi, S. Rizvi, N. Gruhler, F.B.C. Dieleman, F.S. Cataliotti, W.H.P. Pernice, M. Agio, C. Toninelli, Light: Science & Applications (2017)

Outline

- Part I
  - Nano quantum optics
  - Metal nanostructures as optical antennas
  - Fluorescence enhancement
  - Directional emission
  - **Quantum optics with optical antennas**

- Part II
  - Quantum emitters
  - Intrinsic optical properties of diamond
  - Color centres in diamond
  - Diamond-based nanophotonics
  - Diamond electronics

Quality factor

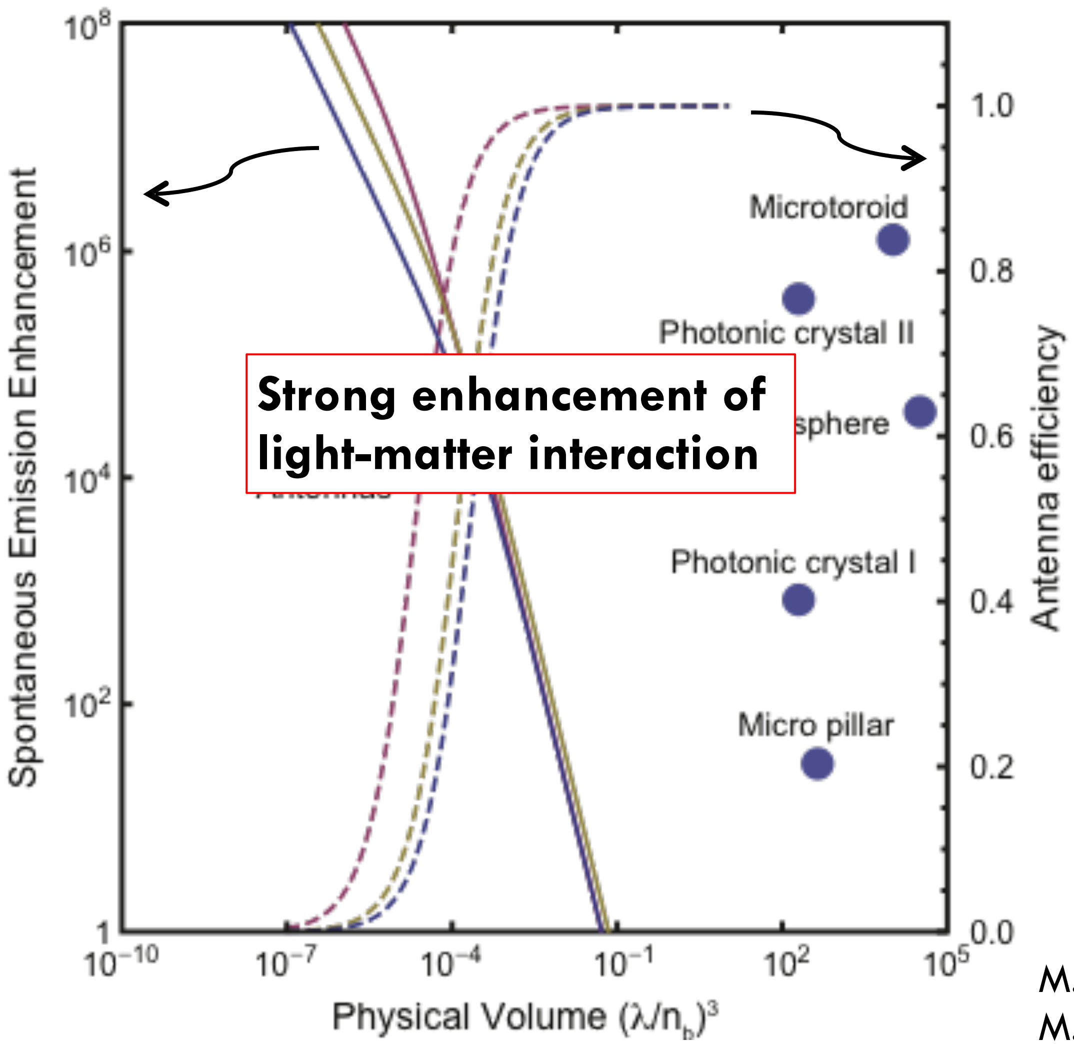


Chu-Harrington fundamental limits of electrically small antennas

$$Q \propto \eta_a \frac{1}{V_{ph}}$$

M. Agio, Nanoscale (2012)

Enhancement of spontaneous emission

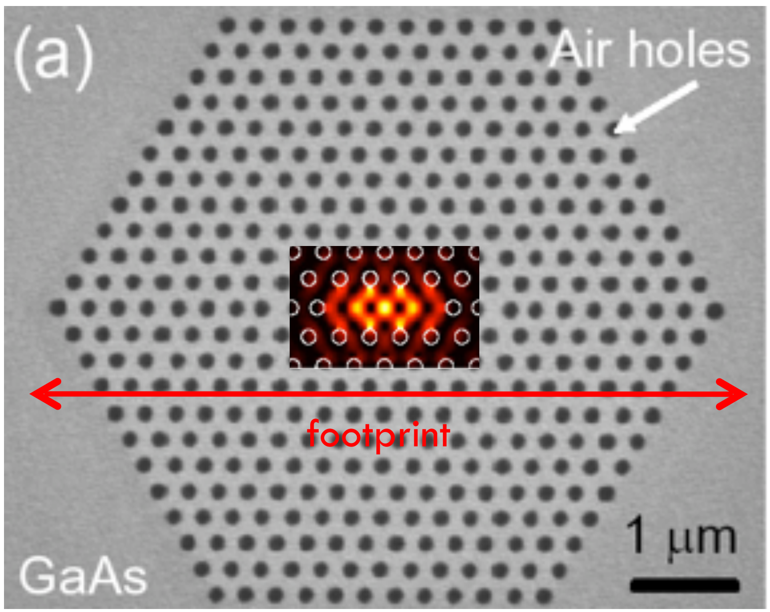


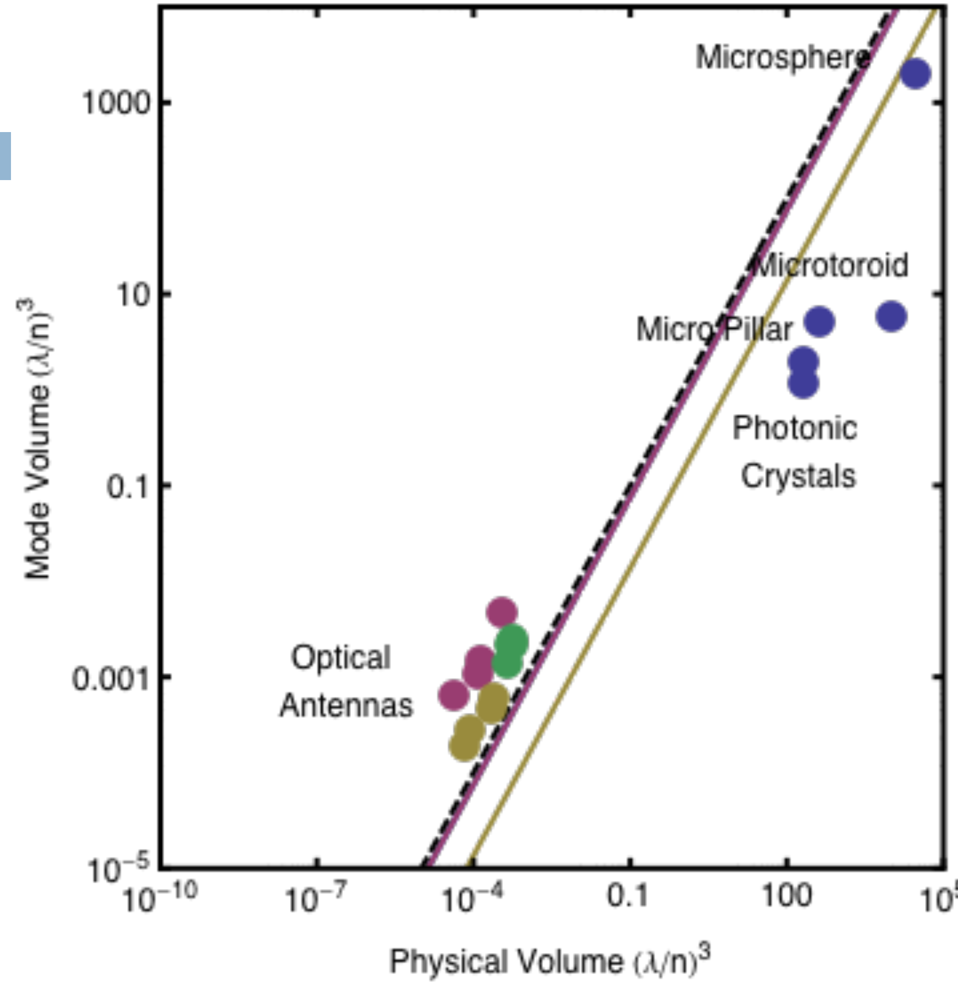
$$\eta_a = \frac{1}{1 + C\frac{1}{V_{ph}}}$$

$$F \propto \eta_a \frac{1}{V_{ph}^2}$$

M. Agio, Nanoscale (2012)
M. Agio, D. Martín-Cano, Nat. Photon. (2013)

Mode volume

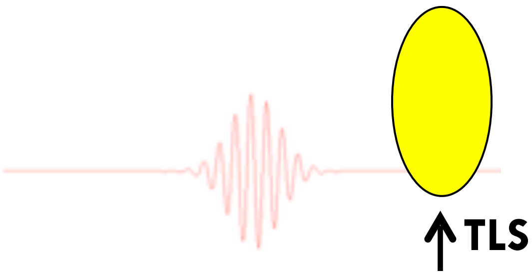




$$V_{\mathrm{m}} = \frac{L}{(1-L)^2} V$$

M. Agio, Nanoscale (2012)

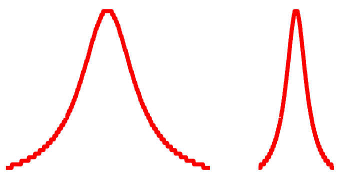
Pulsed excitation

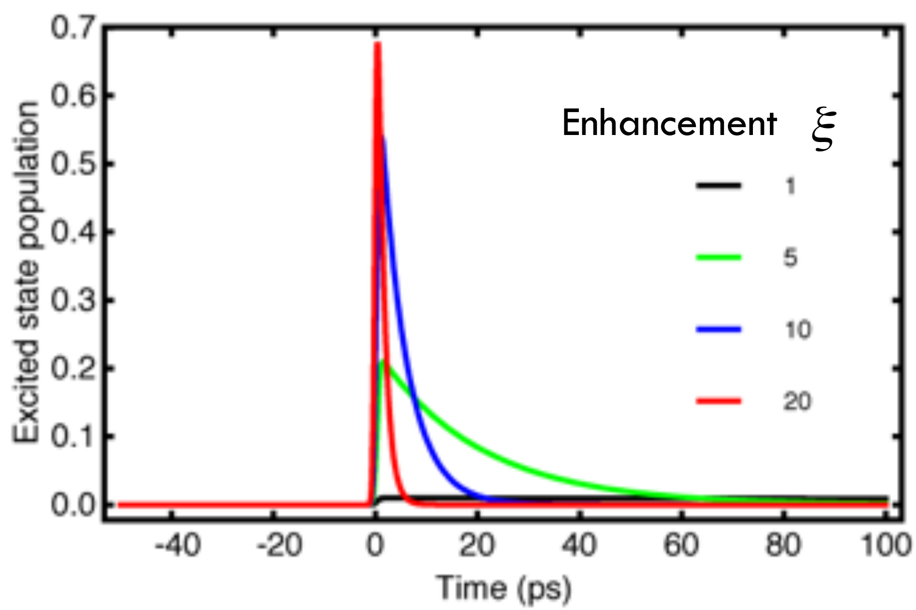


Ultrafast & few-photon nonlinearities

Energy of 10 photons at 750 nm (~2.6 aJ)
focused down to the diffraction limit
T_1=1 ns, T_2=1.6 ps (100 GHz), η_a=0.5
1 ps Gaussian pulse

1





2

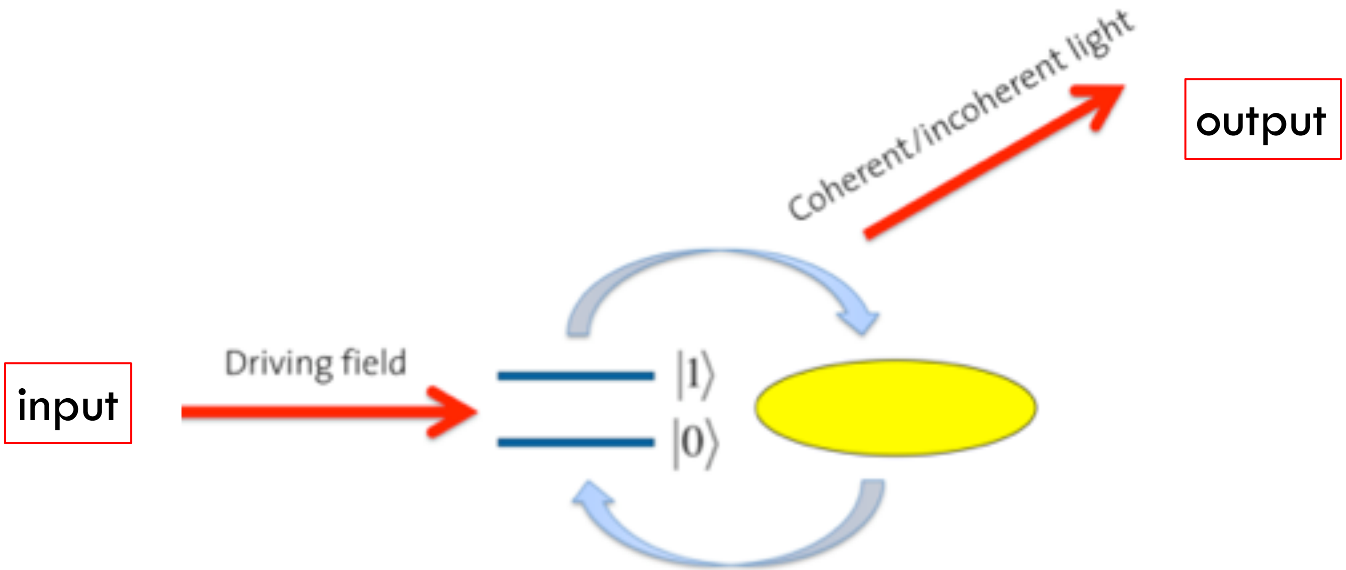
$$\sigma = \frac{T_2(\xi^2,\eta_a)}{2T_1(\xi^2,\eta_a)}\sigma_{\mathrm{TLS}}$$

3

$$\Omega(t) = \xi\Omega_o(t)$$

X.-W. Chen, A. Mohammadi, A.H. Ghasemi, M. Agio, Mol. Phys. (2013) – New Views Article

Quantum optics at the nanoscale



$$H = \int \mathrm{d}^3 r \int \mathrm{d}\omega\, \hbar\omega\, \underline{f^{+}(r,\omega) f(r,\omega)} + \frac{1}{2}\hbar\omega_o - \left(\sigma^{+}\mathbf{E}^{(+)}(r_A)\cdot \mathbf{d} + \mathrm{h.c.}\right)$$

$$c_A(t) = \int \mathrm{d}t\, \overline{K}(t-t')c_A(t') + 1 \quad (c_A(0) = 1)$$

H.T. Dung, L. Knöll, D.-G. Welsch, Phys. Rev. A (2000)

Scattered electric field operator

Nanostructure + Two-level emitter dynamics: macroscopic QED

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar}{\pi\varepsilon_0}}\frac{\omega^2}{c^2}\int d^3\mathbf{r}'\sqrt{\varepsilon''(\mathbf{r}',\omega)}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\hat{\mathbf{f}}(\mathbf{r}',\omega)$$

Heisenberg equations of motion (Markovian and rotating wave approx)

$$\hat{E}_i^{(+)}(\mathbf{r},t) = |g_i(\mathbf{r})|e^{\imath\phi_i(\mathbf{r})}\hat{\sigma}(t), \quad \propto \hat{\sigma} = |g\rangle\langle e|$$

QE coherence

where the complex electric field amplitude

$$g_i(\mathbf{r}) = \frac{\mathcal{P}}{\pi\varepsilon_0}\int_0^\infty d\omega\,\frac{\omega^2}{c^2}\frac{\mathrm{Im}\{G_{ij}(\mathbf{r},\mathbf{r}_\mathrm{E},\omega)\}d_j}{\omega_\mathrm{E}-\omega} + \imath\frac{\omega_\mathrm{E}^2}{\varepsilon_0 c^2}\mathrm{Im}\{G_{ij}(\mathbf{r},\mathbf{r}_\mathrm{E},\omega_\mathrm{E})\}d_j.$$

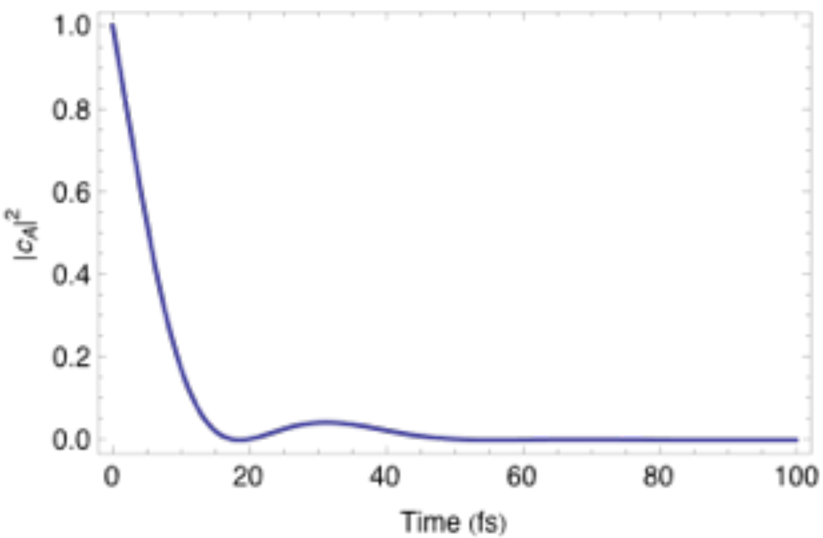
Green's tensor

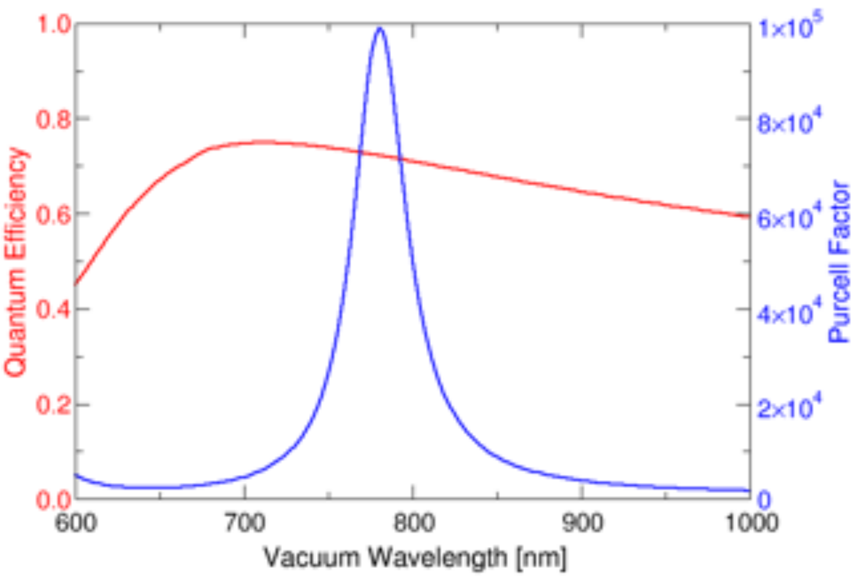
Dung H.T., Knöll L., Welsch D.-G., Phys. Rev. A 64, 013804 (2000)

Ultrafast Rabi oscillations

$$c_A(t) = c_A(0)e^{-\Gamma_{\text{pl}} t/2} \cosh\left(\sqrt{\Gamma_{\text{pl}}{}^2 - \Omega^2}\, t/2\right)$$

$$\Omega = \sqrt{2\Gamma_{\text{pl}}\Gamma_{\text{sp}}} \qquad \Gamma_{\text{pl}} = \frac{\gamma}{2} + C\omega_{\text{pl}}^4$$

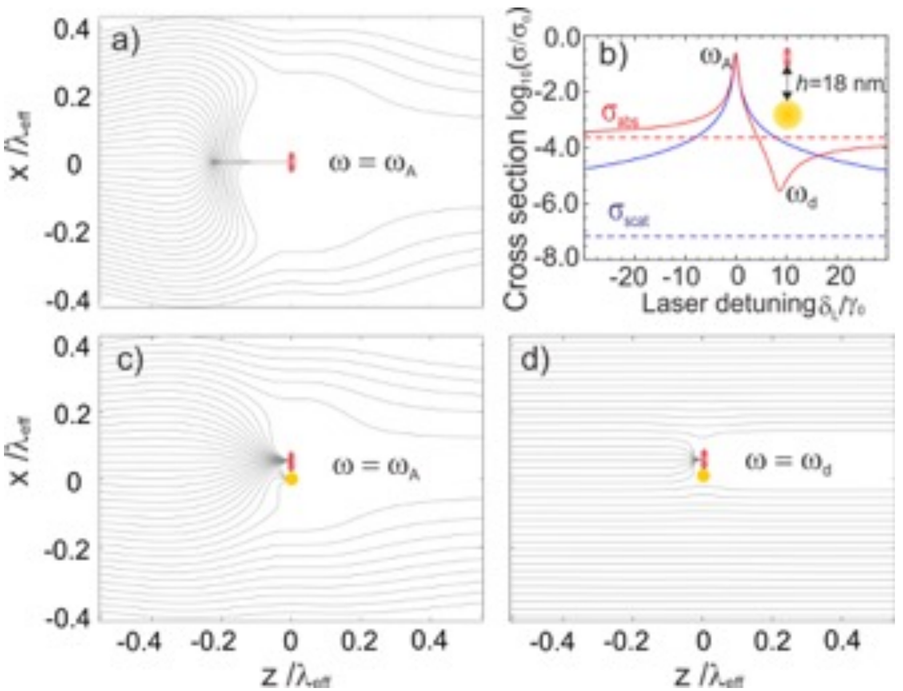


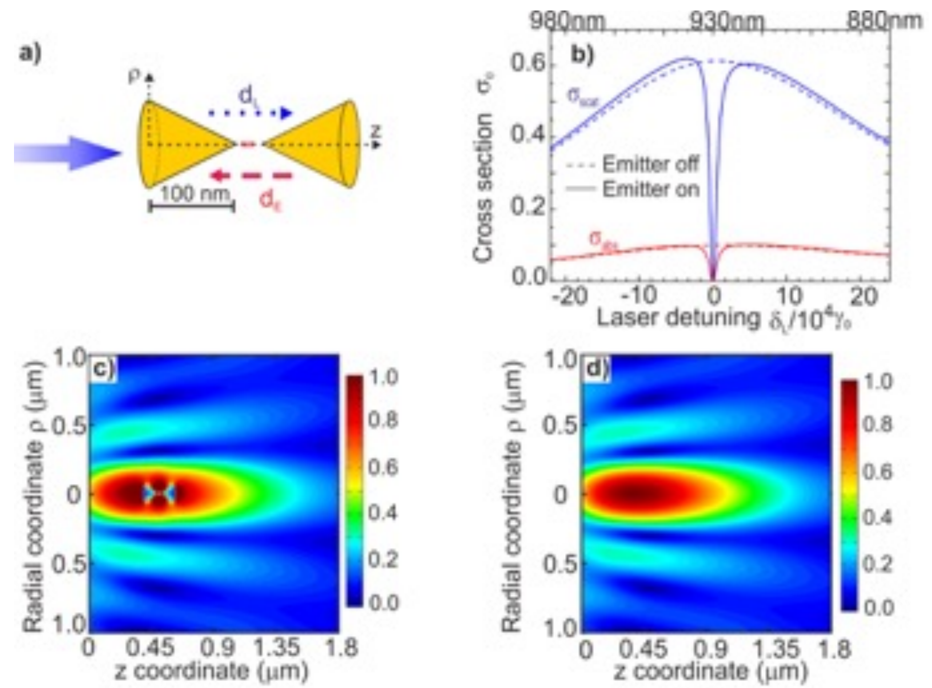


M. Agio, unpublished

Coherent effects

Beyond: Weak excitation, Rate equations, Classical light





X.-W. Chen, V. Sandoghdar, M. Agio, Phys. Rev. Lett. (2013)

What is squeezed light?

Quantum electromagnetic field quadrature

$$E(\bar{r},t) = E^{+}(\bar{r},t) + E^{-}(\bar{r},t)$$ *satisfying* $$\left[E^{+}(\bar{r},t), E^{-}(\bar{r},t)\right] = 2C$$

Or *more general, as a combination of both parts with a phase*

$$E_{\theta}(\bar{r},t) = E^{+}(\bar{r},t)e^{i\theta} + E^{-}(\bar{r},t)e^{-i\theta}$$

Then a squeezed state of light corresponds to such that: $\left\langle \Delta E_{\theta}^{2}(\bar{r},t)\right\rangle < C$

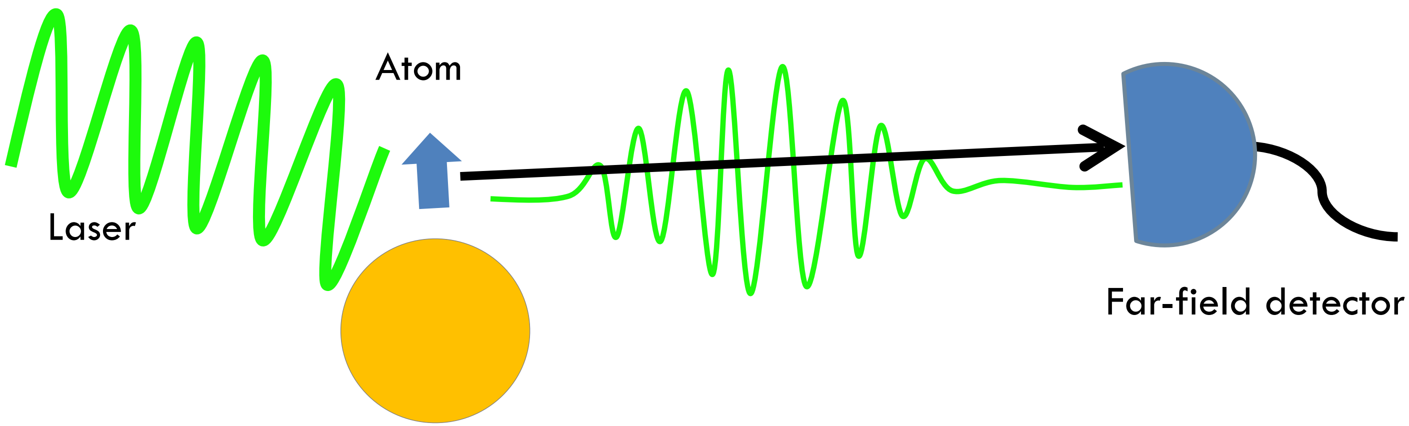
Normal ordering

Or *alternatively, since* $\left\langle \Delta E_{\theta}^{2}(\bar{r},t)\right\rangle = C + \left\langle : \Delta E_{\theta}^{2}(\bar{r},t) : \right\rangle$ → $\left\langle E^{-}E^{-}...E^{+}E^{+}\right\rangle$

$$\left\langle : \Delta E_{\theta}^{2}(\bar{r},t) : \right\rangle < 0$$

Scully, M.O., Zubairy, M.S., Quantum Optics (Cambridge Univ. Press, 1997)

Squeezing in resonance fluorescence



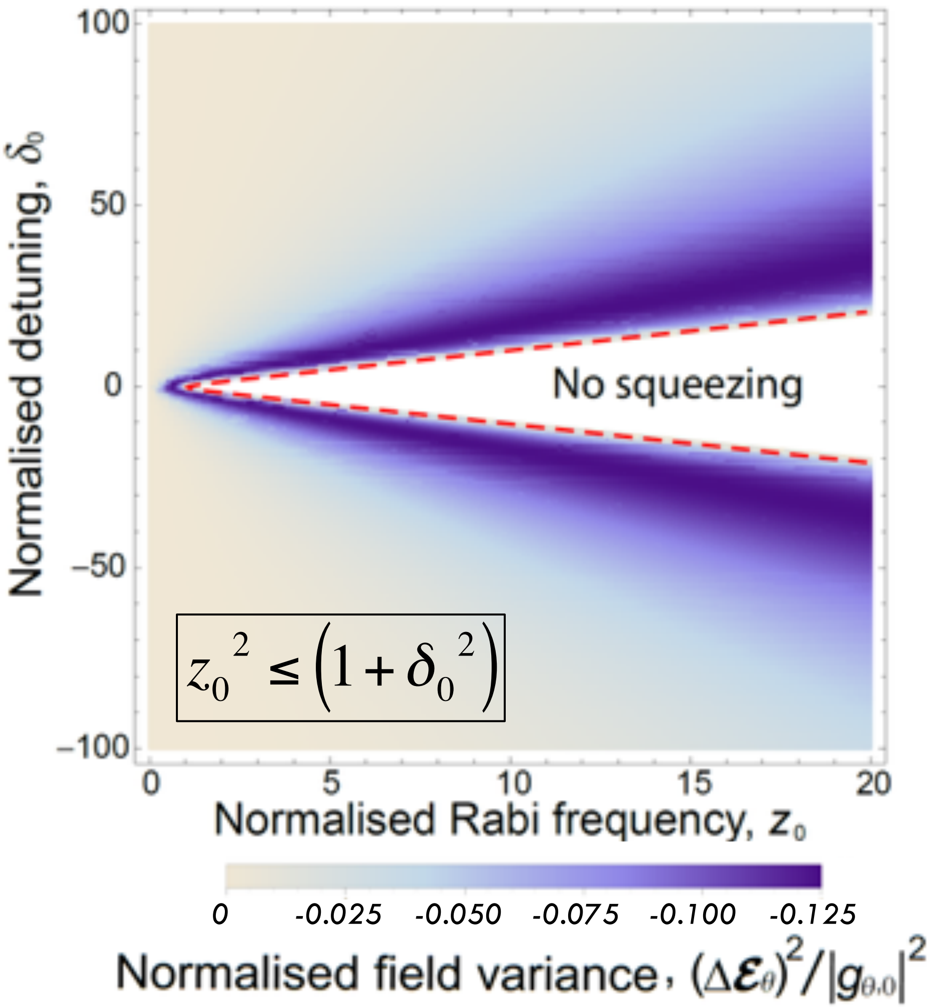
$$\left\langle : \Delta E_{\theta}^{2}(\bar{r},t) : \right\rangle < 0$$

Not verified with a single atom in vacuum
(Small collection efficiency)

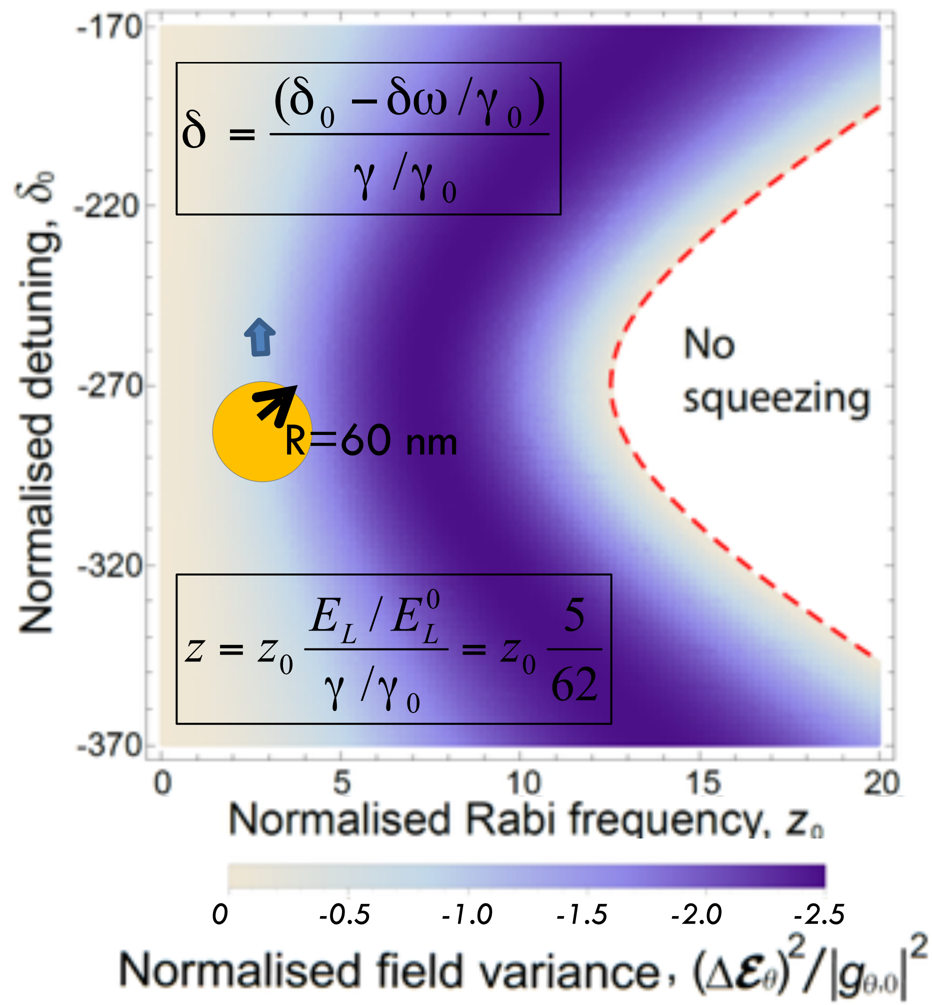
D.F. Walls, P. Zoller, Phys. Rev. Lett. (1981)
C.H.H. Schulte, J. Hansom, A.E. Jones, C. Matthiesen, C. Le Gall, M. Atatüre, Nature (2015)

Free space versus nanoparticle

Absence of nanosphere

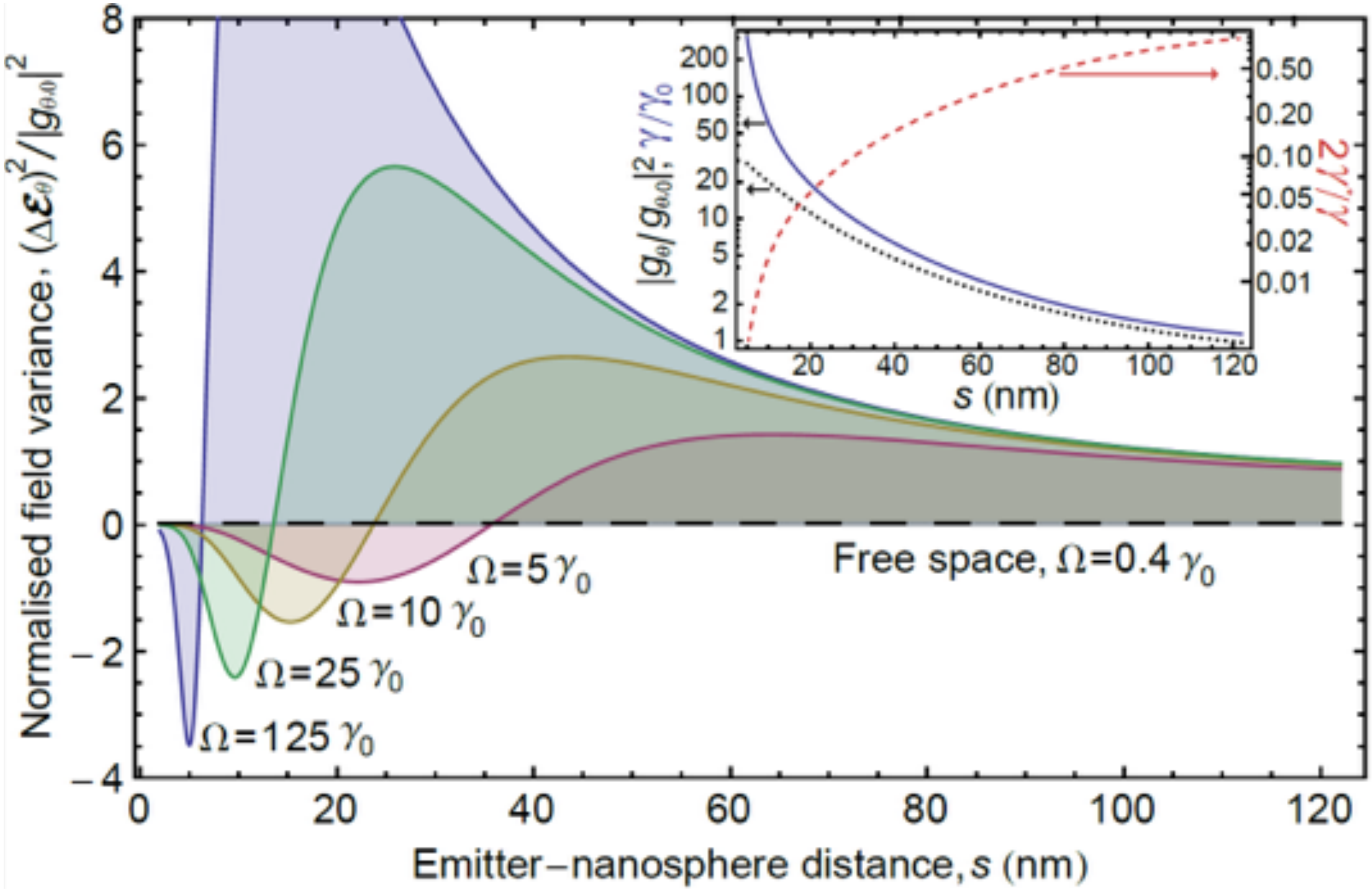


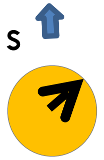
Nanosphere case



D. Martín-Cano, H.R. Haakh, K. Murr, M. Agio, Phys. Rev. Lett. (2014)
D. Martín-Cano, H.R. Haakh, M. Agio, J. Opt. (2016)

The role of a nanostructure

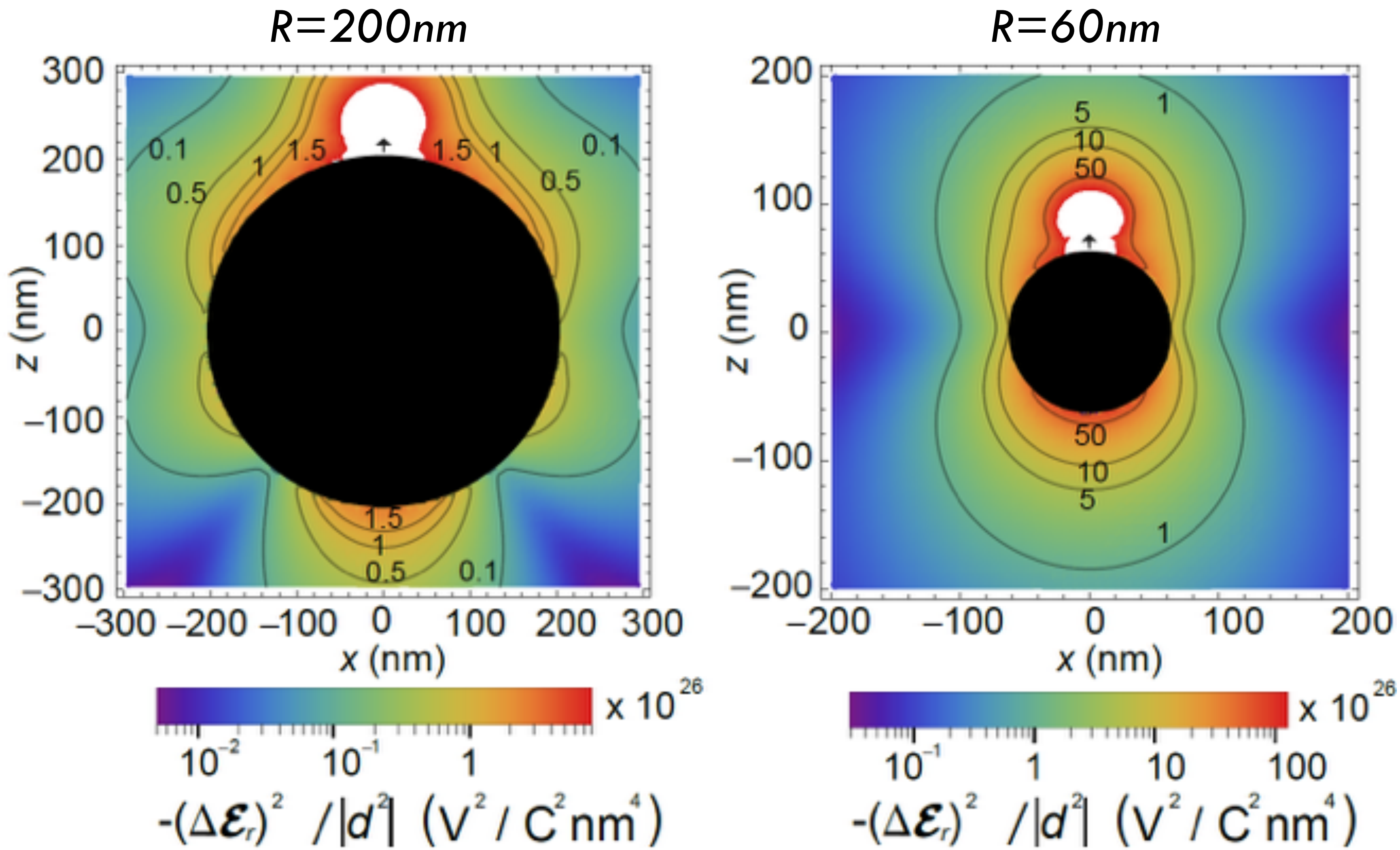




$2\,\gamma^{*}=\gamma_0$

A nanostructure may assist the creation squeezed light in emitters, which do not generate squeezing in free space

Near field maximum degree of squeezing



A factor 30 larger for the small nanoshere compared to the large one, and 10^9 respect to the far field case

Outline

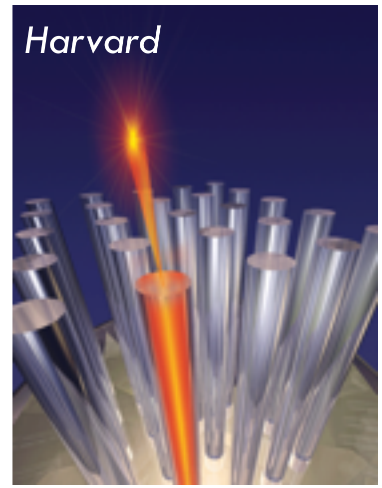
UNIVERSITÄT SIEGEN

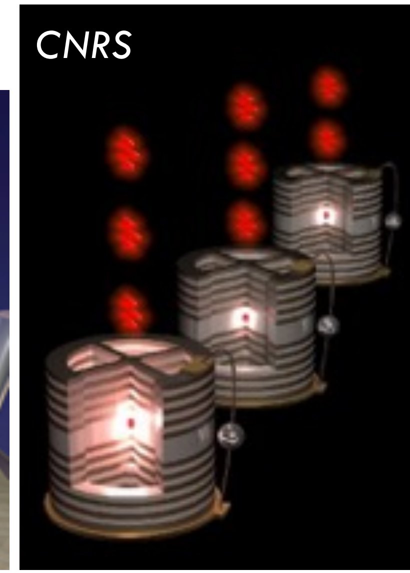
- Part I
  - Nano quantum optics
  - Metal nanostructures as optical antennas
  - Fluorescence enhancement
  - Directional emission
  - Quantum optics with optical antennas

- Part II
  - **Quantum emitters**
  - Intrinsic optical properties
  - Diamond color centres in diamond
  - Diamond-based nanophotonics
  - Diamond electronics

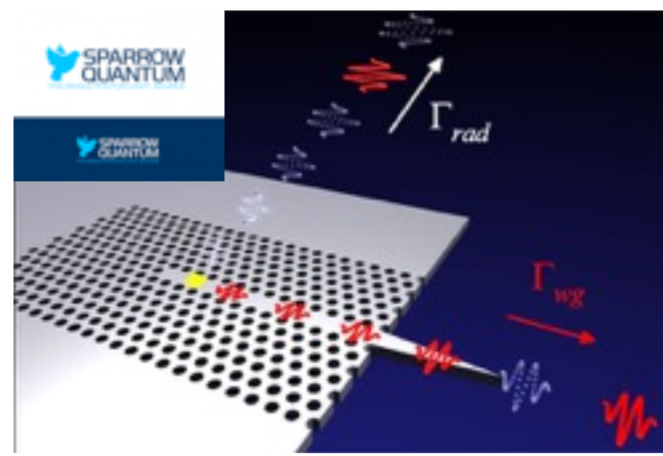
Why single-photon sources (SPSs)?

- Quantum technologies:
  - Quantum key distribution
  - Quantum cryptography
  - Quantum computing
  - Quantum enhanced measurements
  - Radiometry

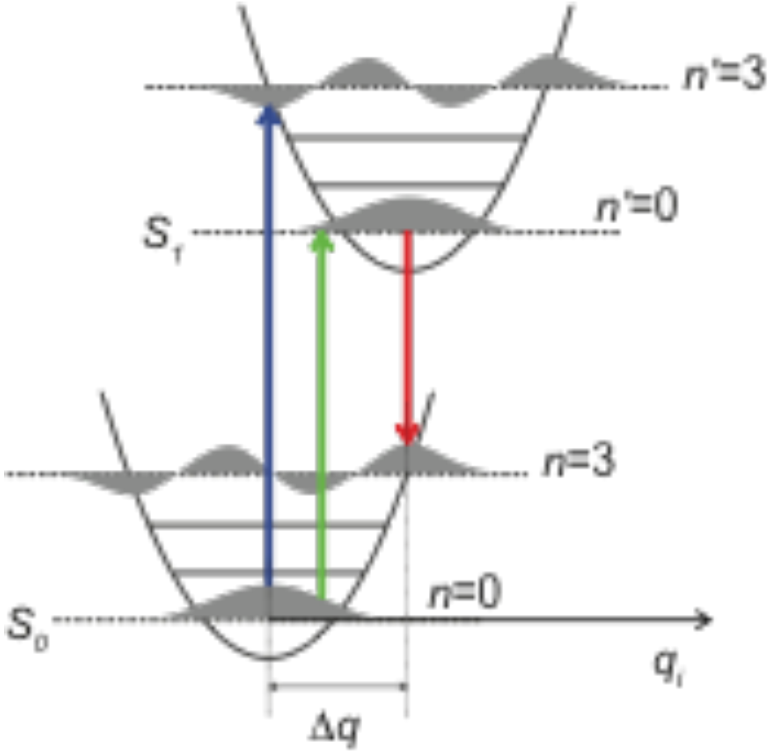




- Available solutions:
  - Attenuated lasers
  - Heralded SPS
  - Quantum emitters:
    - QDs, color centers, molecules

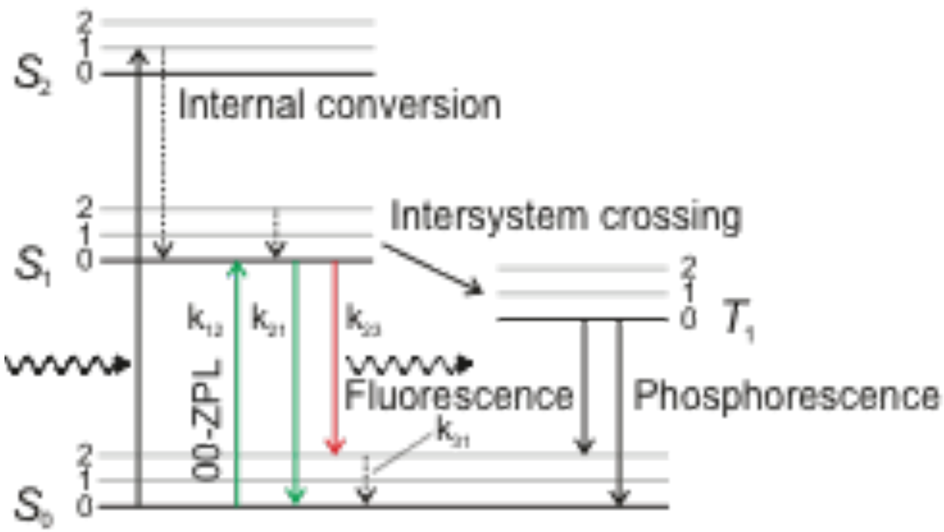


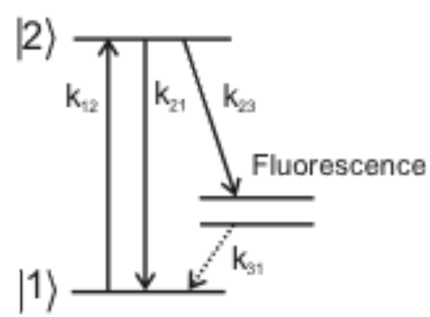
Single-molecule level scheme



Level scheme and Frank-Condon principle

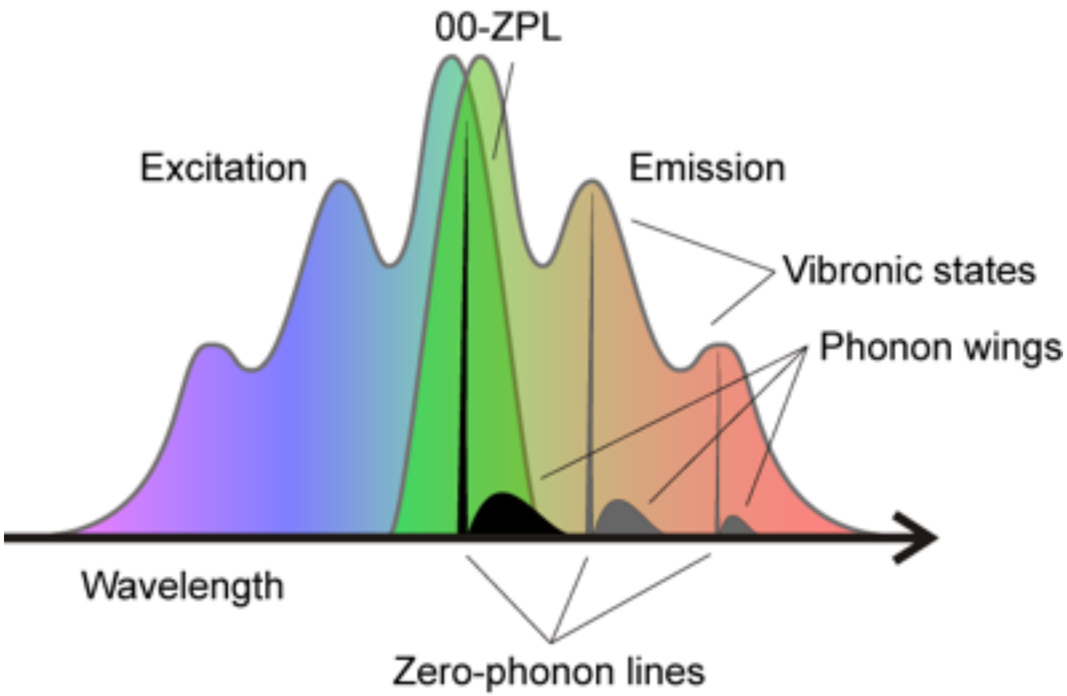
M. Agio, Habilitation Thesis (2011)

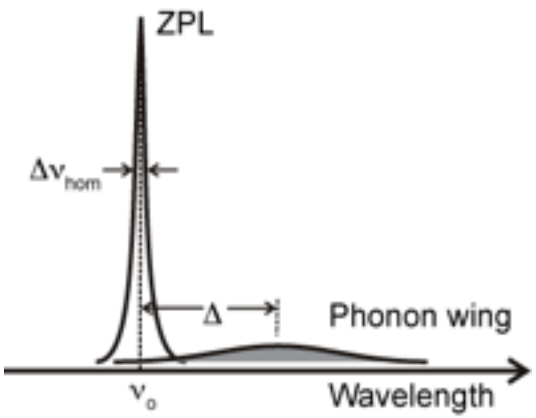




Jablonsky diagram

Optical properties of single molecules





Typical SM spectrum

M. Agio, Habilitation Thesis (2011)

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Diamond growth

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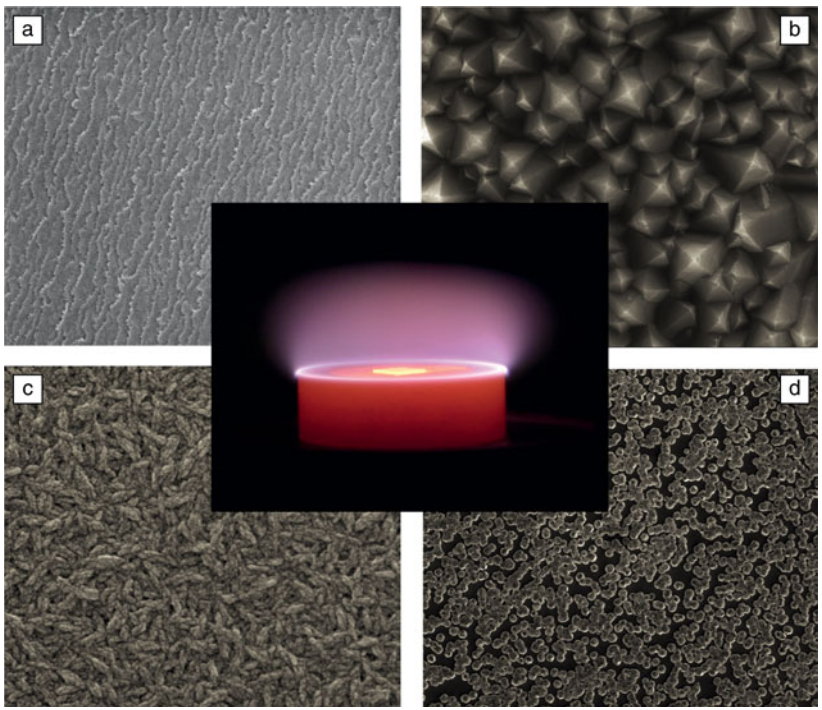
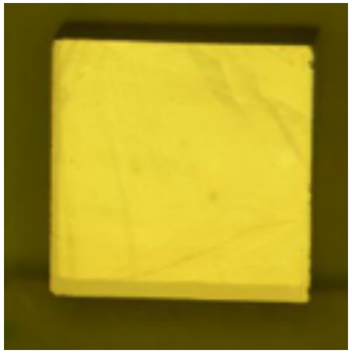
Single crystal *Polycrystalline*



Nano crystal *B-doped crystal*

Discovered about 30 years ago, the use of hydrogen in plasma-enhanced chemical vapor deposition (CVD) has enabled the growth and coating of diamond in film-form on various substrate materials.



CVD diamond sample

Optical and electronic applications of diamond

- Properties
  - Highest thermal conductivity of any material, up to 2000 W/mK
  - Low absorption coefficient allows higher power outputs to be transmitted through the window without suffering damage or distortion.
  - Widest transmission spectrum from visible to far IR — from 220 nm to >50 μm—x-ray, infrared, terahertz and microwave
  - Hardest material known to science
  - Highly chemically inert
  - Wide band-gap semiconductor material
  - Electric insulator with high breakdown field strength
- Diamond optics & photonics
  - Lenses and diffractive elements
  - Raman lasers
  - Bioimaging
- Diamond junction devices
  - LEDs
  - Power electronics
- Quantum optical technologies
  - Magnetometers
  - Single-photon sources
  - ...

R.P. Mildred & J.R. Rabeau (eds), Optical engineering of diamond (Wiley, 2013)

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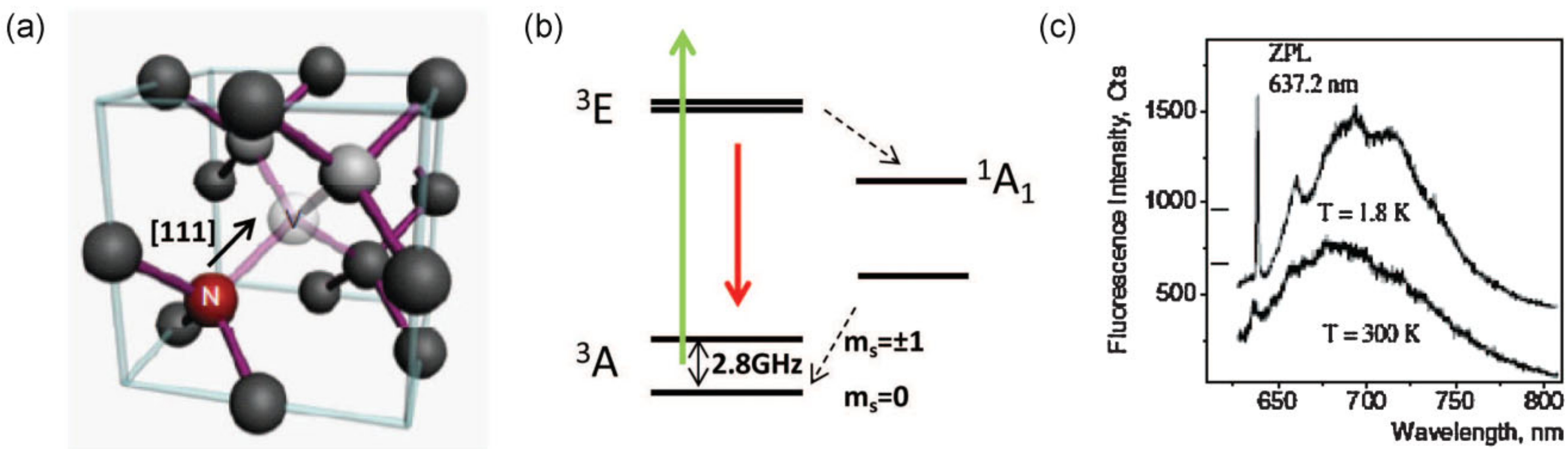
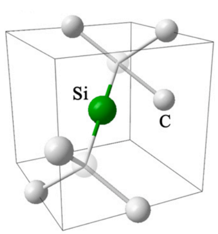
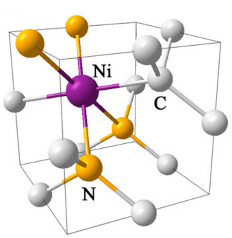
Near IR color centers in diamond

Diamond is optically transparent and can host hundreds of different point defect centers, called color centers, that emit light.





SiV center

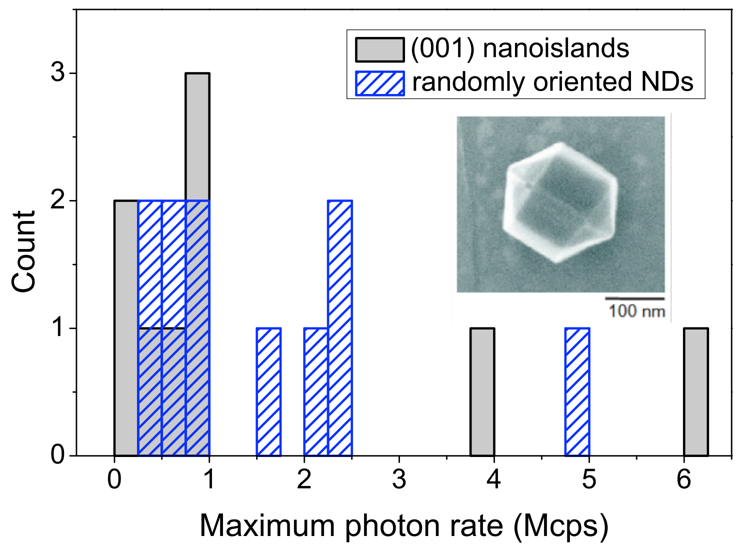


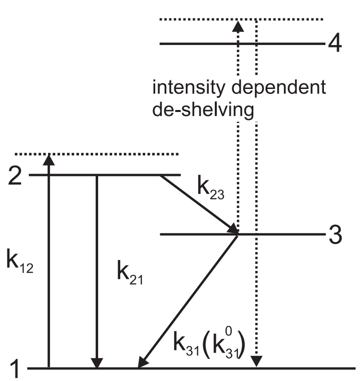
NE8 center

NV - interesting for sensing (magnetometer)
SiV, NE8 - interesting for single-photon emission

A.M. Zaitsev, Optical properties of diamond (Springer, 2001)
I. Aharonovich, *et al.*, Rep. Prog. Phys. (2011)

Photophysics of the SiV color centre



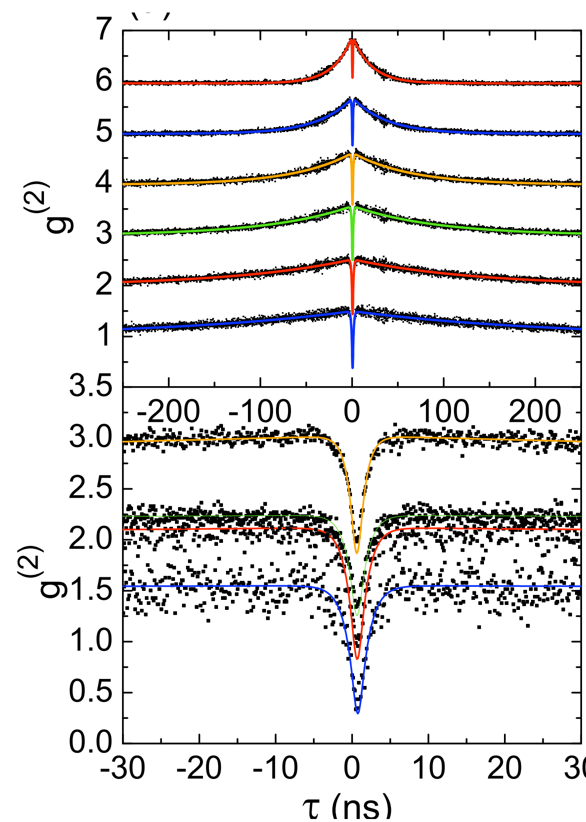


$$\tau_{1,2} = 2/(A \pm \sqrt{A^2 - 4B})$$

$$A = k_{12} + k_{21} + k_{23} + k_{31}$$

$$B = k_{12}k_{23} + k_{12}k_{31} + k_{21}k_{31} + k_{23}k_{31}$$

$$a = \frac{1 - \tau_2 k_{31}}{k_{31}(\tau_2 - \tau_1)}$$



$$g^{(2)}(\tau) = 1 - (1+a)\,e^{-|\tau|/\tau_1} + a\,e^{-|\tau|/\tau_2}$$

E. Neu, M. Agio, C. Becher, Opt. Express (2012)

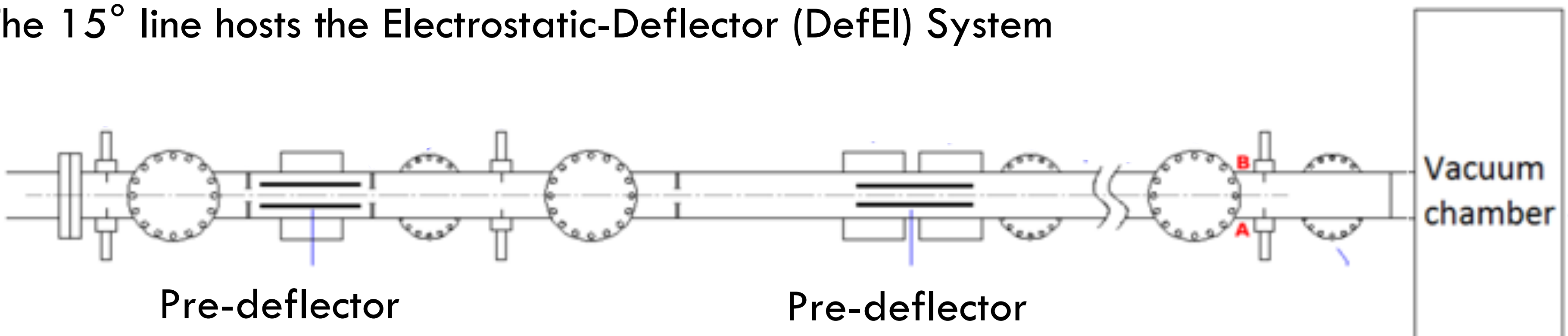
Implantation of Si at LABEC, Florence

At the Tandetron 3MV accellerator facily of the LABEC INFN Florence we can accelerate a vast veriety of ion species

The deflectors allows to operate from the continuum regime to the single pulse, whith pulses down to 5 μs long

Possibility to implant ions over a range of fluences (implanted ions/cm^2) spanning over at least 8 orders of magnitude ($10^7 - 10^{15}$ ions/cm^{-2})

The 15° line hosts the Electrostatic-Deflector (DefEl) System

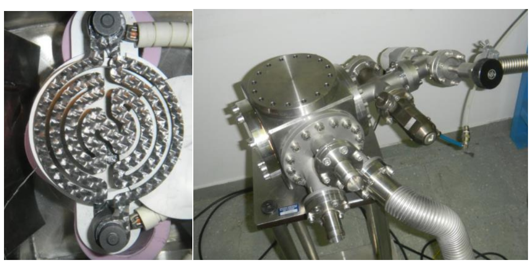


S. Lagomarsino, *et. al.*, in preparation (2017)

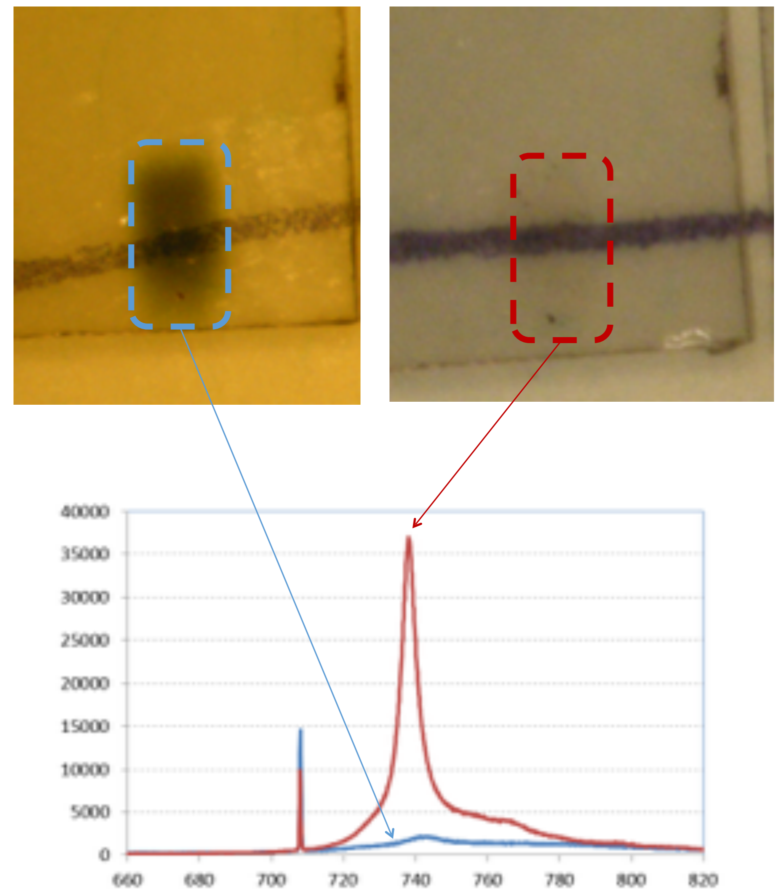
Implantation and annealing

Implanted Silicon ions take place predominantly in intestitial position, with generation of defects (~ 2 vacancies/(nm x ion))

After proper annealing at 1150°C transparency recovers



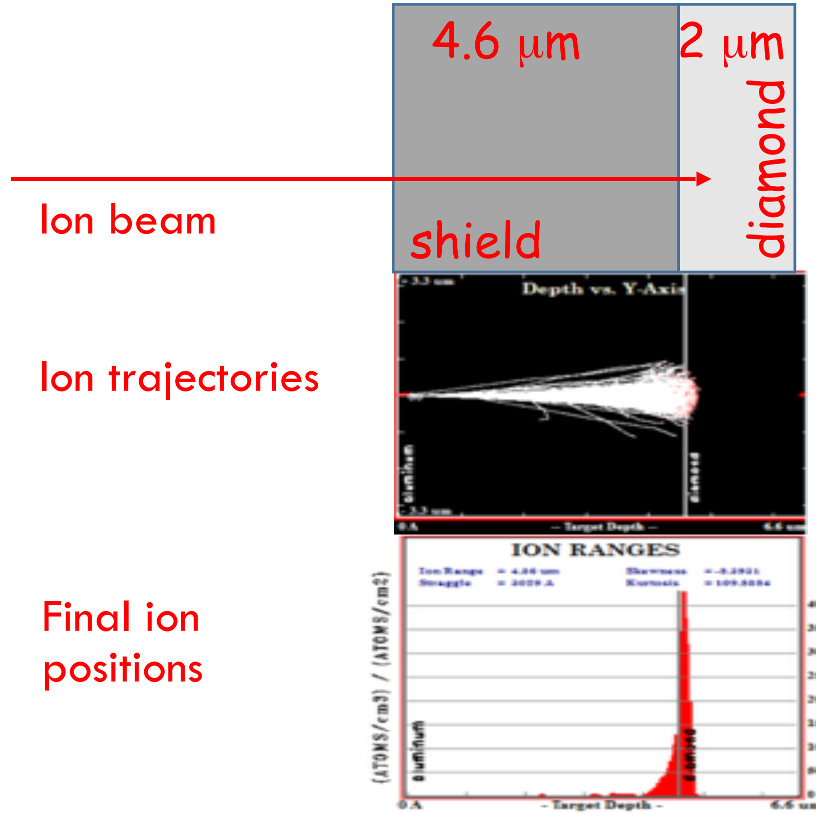
x20 increase of luminescence signal (738 nm ZPL of SiV, negligible sidebands)

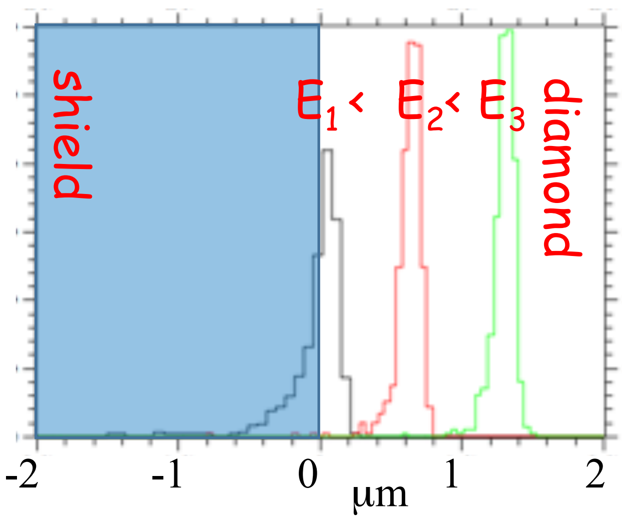


Implantation depth

The implantation depth depends on the ion energy

We modulated the implantation depth varying the energy and interposing a controlled-thickness metal shield in front of the sample.

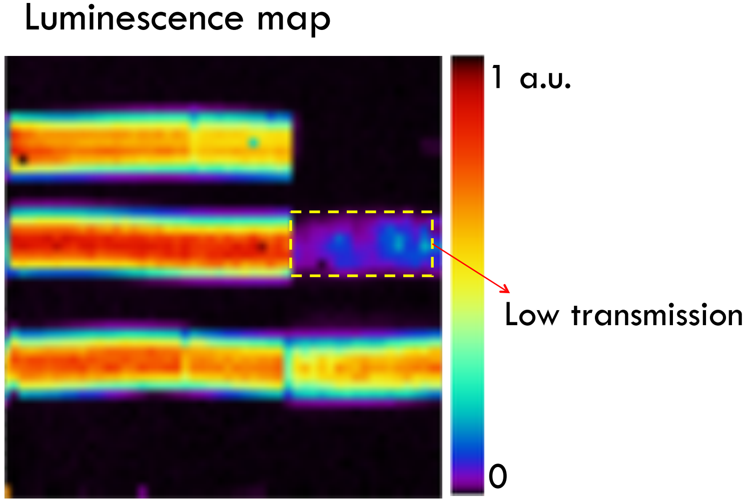


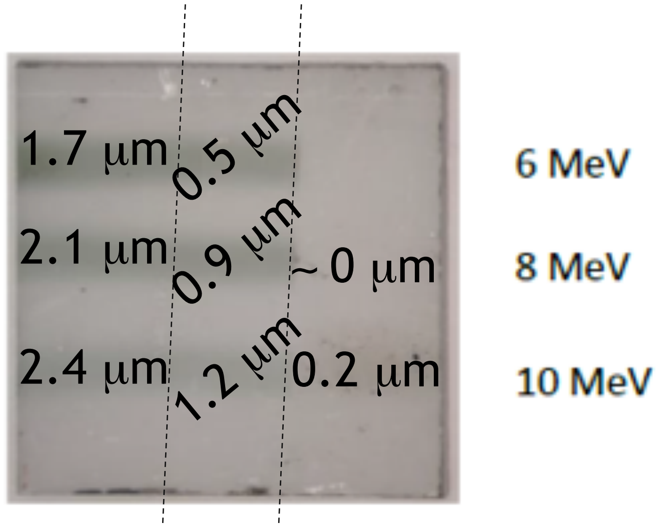


Efficiency independent of depth

The implantation depth depends on the ion energy

We implanted ions at depths from 0 to 2.4 µm

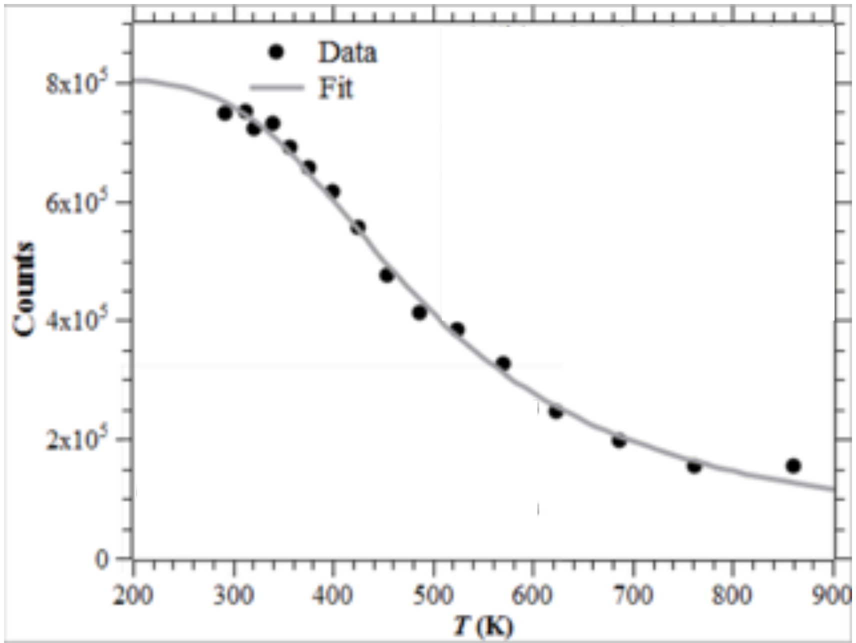


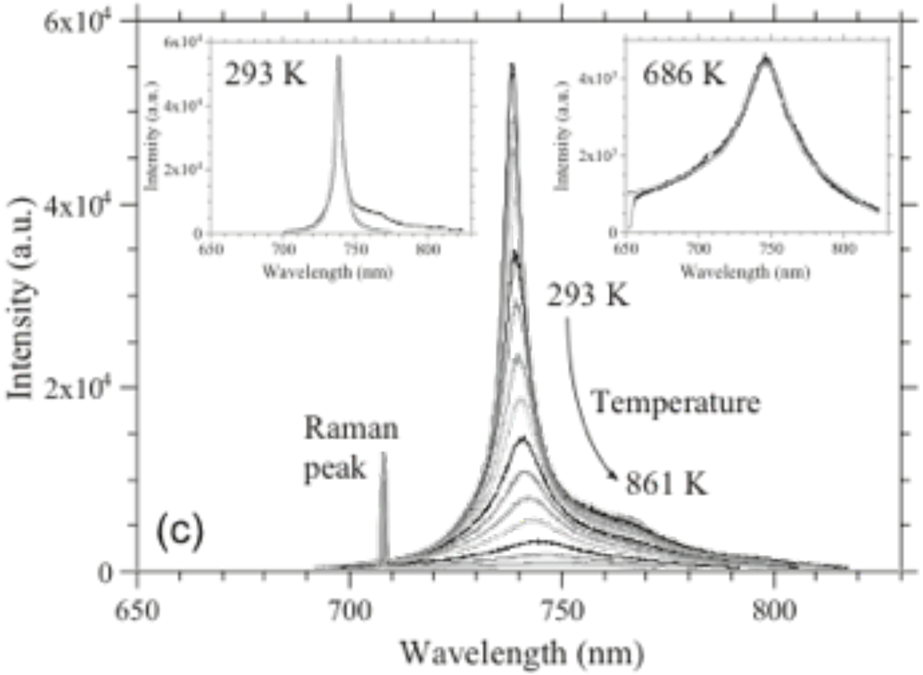


Fluence = 3.2×10^{13} cm^{-2}

The activation efficiency is substantially independent on depth

The SiV color center at large T





Slight increase (4nm) in λ_{peak} from RT to 500K

FWHM from 6 to 30 nm

Integrated intensity decreases only 40%

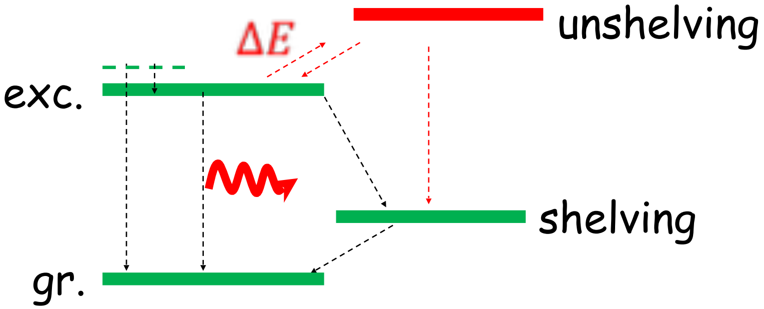
The luminescence intensity is restored coming back at room temperature.

S. Lagomarsino et al., AIP Advances (2015)

The SiV color center at large T

An unshelving state has been proposed to give account of a bounching effect at long correlation times.

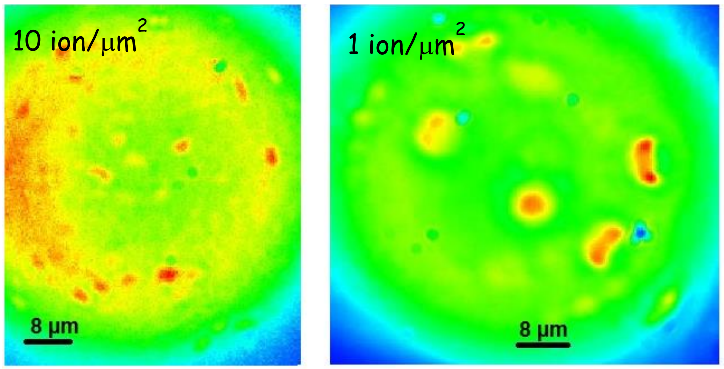
The observed decrease of luminescence with temperature is well reproduced if the unshelving state lies at about $\Delta E = 0.18$ eV from the excited one (thermal de-population of excited state).

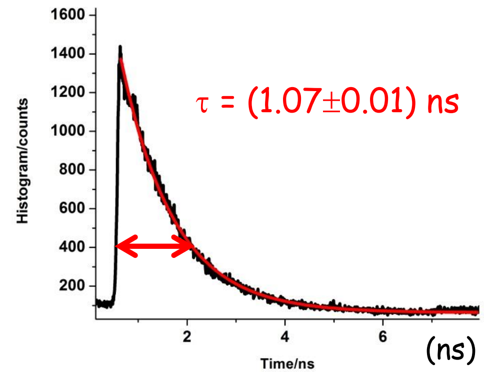


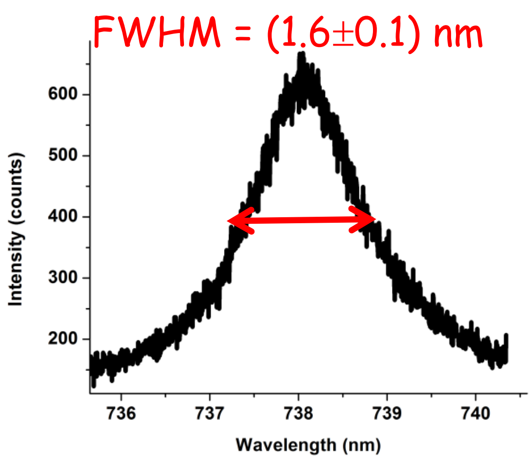
$$I = \frac{I_p}{1 + A_p \exp\left(-\frac{\Delta E}{KT}\right)}$$

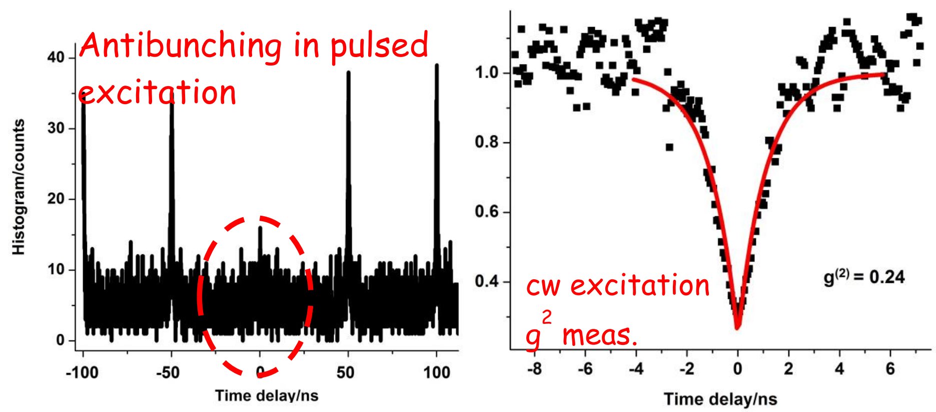
S. Lagomarsino et al., AIP Advances (2015)

The SiV color center









Work in progress, in collaboration with LABEC-INFN (Florence, Italy)
S. Lagomarsino et al., AIP Advances (2015)

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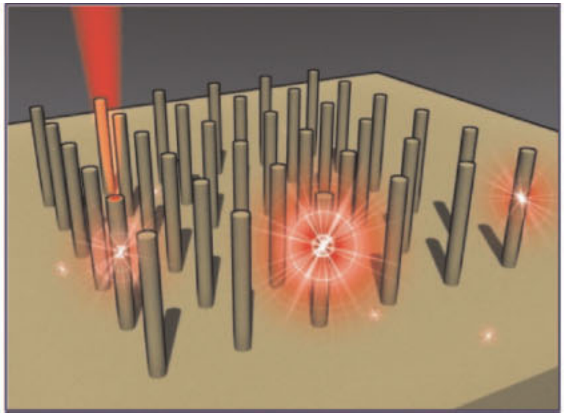
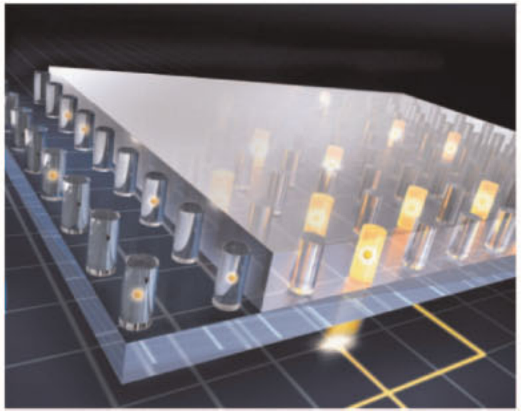
Diamond nanophotonics

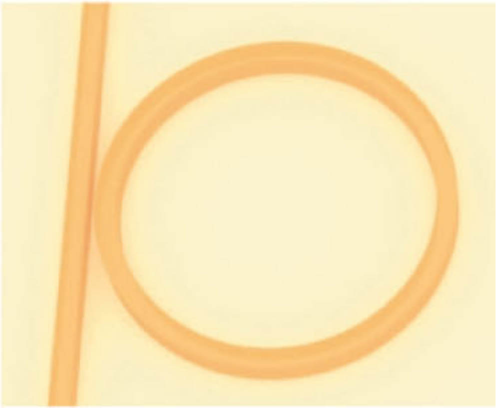
(a)

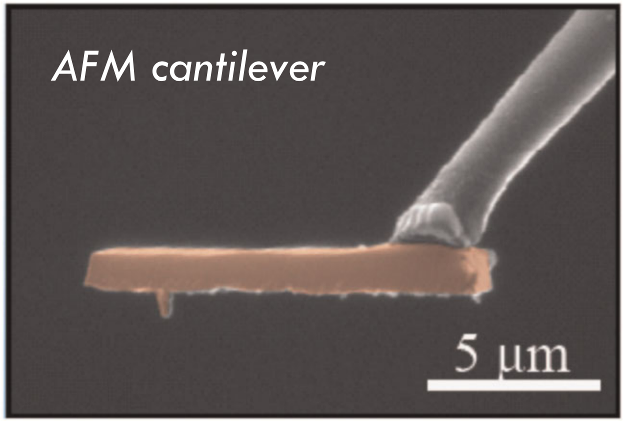


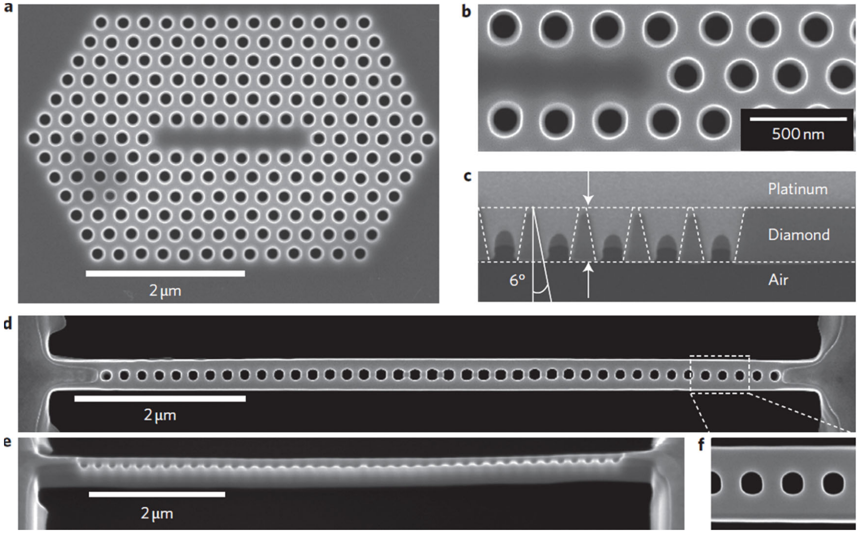
(b)



(c)







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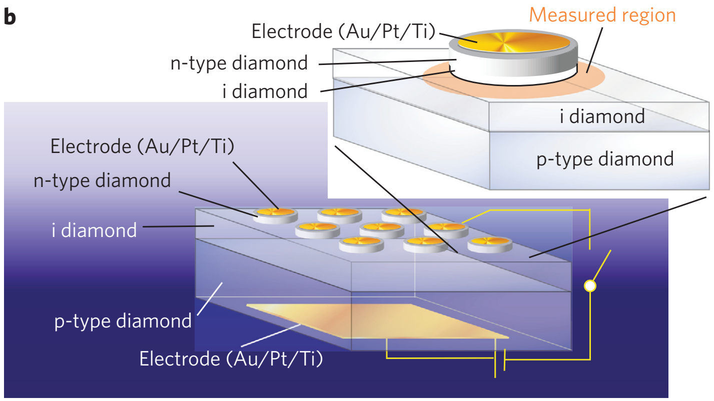
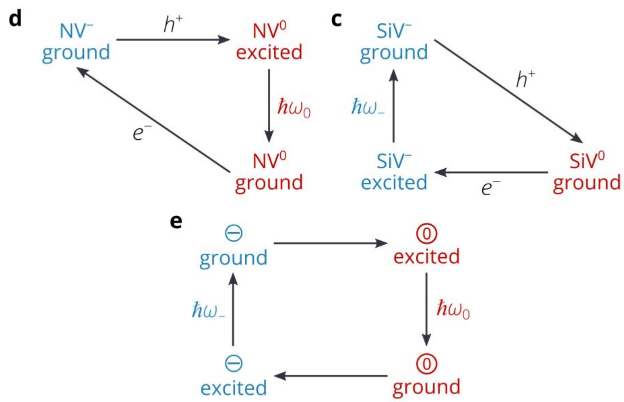
Level scheme and physical model



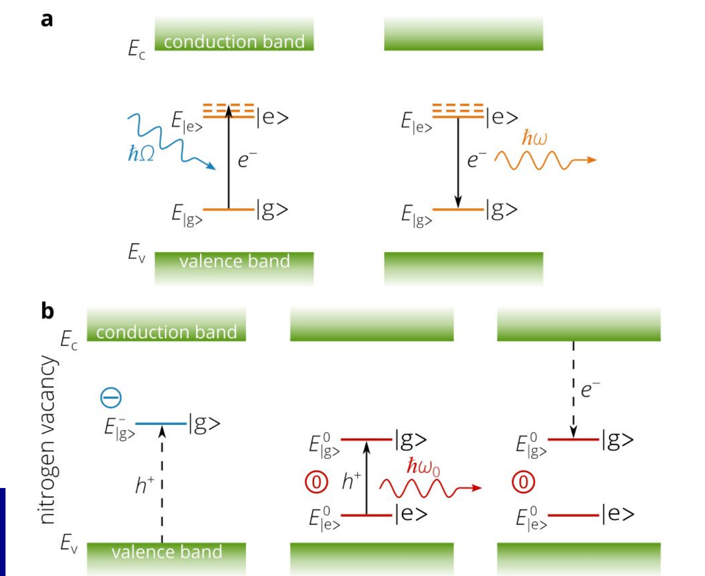
N. Mizuoki, *et al.*, Nat. Photonics (2010)

requires considerations about the emission mechanism of electroluminescent devices and its dependence on the VB and CB populations …



Emission rate (approximate)

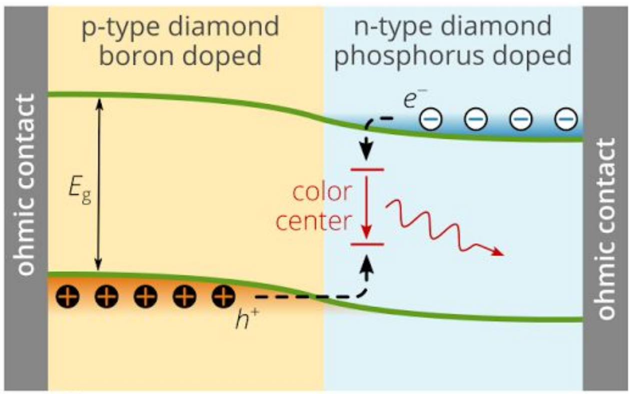
$$R_{\mathrm{ph}} = \Phi \frac{1}{\dfrac{1}{c_{\mathrm{n}} n} + \dfrac{1}{c_{\mathrm{p}} p} + \tau}$$



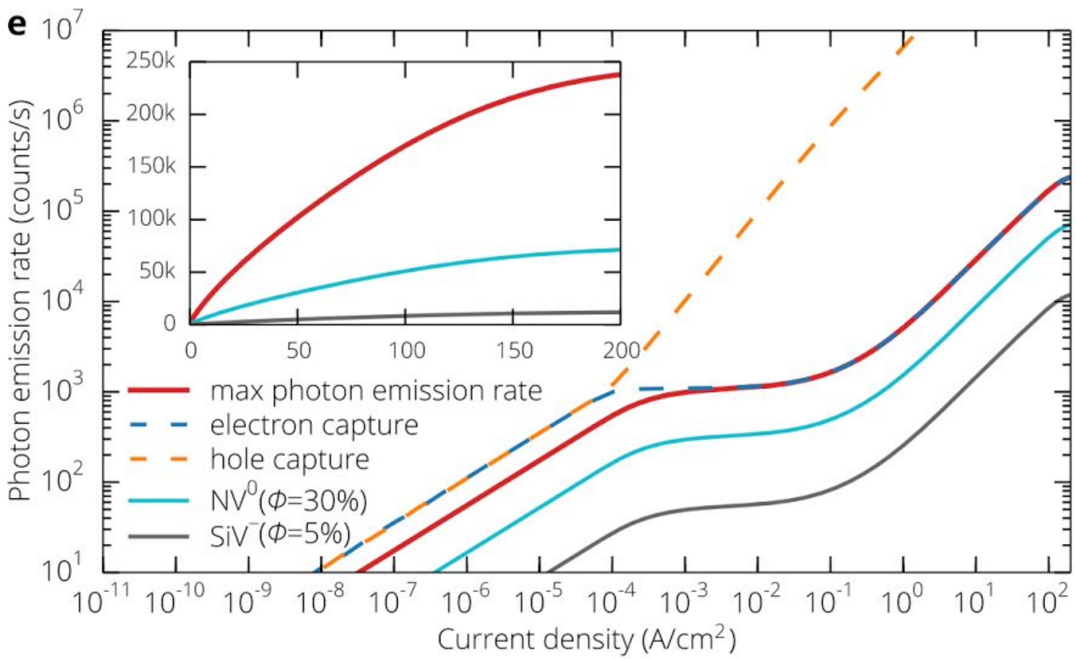
D. Yu. Fedyanin and M. Agio, New J. Phys. (2016)

Emission rate vs current density

Typical device (pn diode)



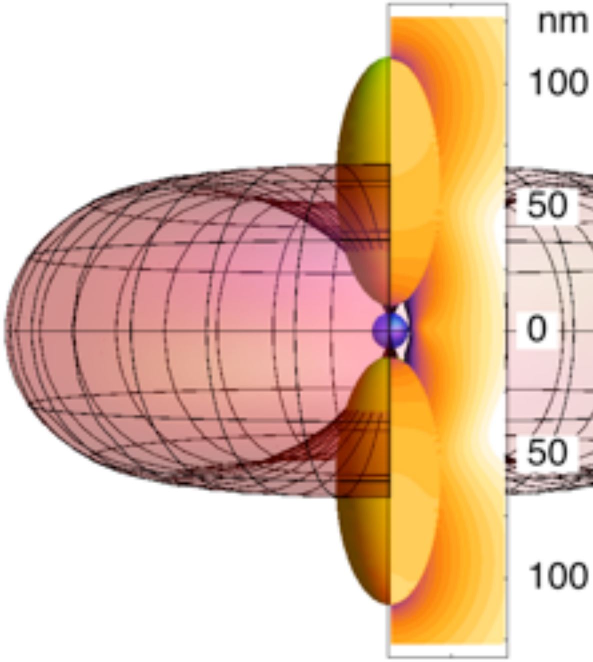
Photon emission rate



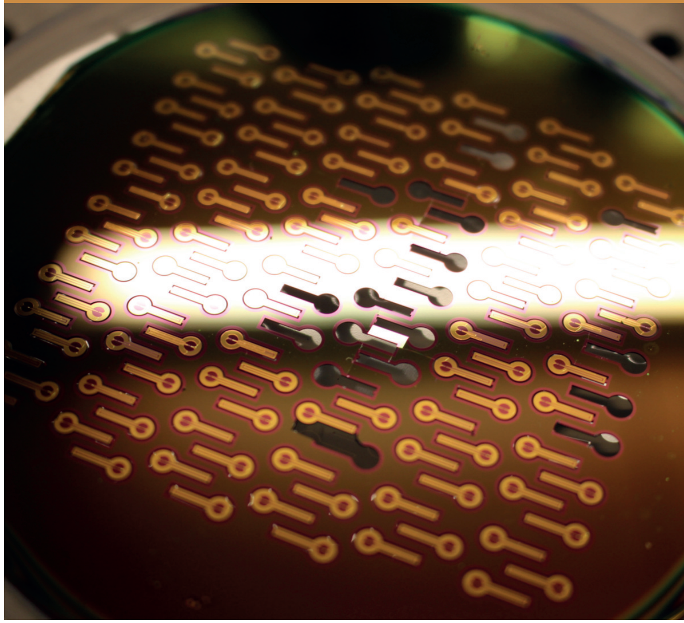
N. Mizuochi et al.
Electrically driven single-photon source at room temperature in diamond, Nat. Photon. (2012)

A.M. Berhane et al.
Electrical excitation of silicon-vacancy centers in single crystal diamond, Appl. Phys. Lett. (2015)

D. Yu. Fedyanin and M. Agio, New J. Phys. (2016)



THE BIG IMPLICATIONS
OF QUANTUM OPTICS
AT THE NANOSCALE



WWW.COST-NQO.EU

Nanoscale
Quantum Optics (NQO)

COST Action MP1403 - 12/2014 –12/2018 - www.cost-nqo.eu

Scientific workplan

<table>
<tr><th colspan="2" rowspan="2">Working Groups</th><th colspan="2">Technology Driven</th><th colspan="2">Research Driven</th></tr>
<tr><th>WG1</th><th>WG2</th><th>WG3</th><th>WG4</th></tr>
<tr><td rowspan="3">Application areas</td><td>ICT</td><td rowspan="3">Generation, detection & storage of quantum states of light at the nanoscale</td><td rowspan="3">Nonlinearities & ultrafast processes in nanostructured media</td><td rowspan="3">Nanoscale quantum coherence</td><td rowspan="3">Cooperative effects, correlations and many-body physics tailored by strongly confined optical fields</td></tr>
<tr><td>Sensing & Metrology</td></tr>
<tr><td>Energy</td></tr>
<tr><td colspan="2">Methods & Means</td><td colspan="4">Theory, Experiments and Materials Development</td></tr>
</table>

References

- Part I
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  - M. Agio, *Optical antennas as nanoscale resonators,* Nanoscale 4, 692 (2012)
  - M. Agio, A. Alù, *Optical Antennas* (Cambridge University Press, 2013)
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  - X.W. Chen, V. Sandoghdar, M. Agio, *Coherent interaction with a metallic structure coupled to a single quantum emitter: from super absorption to cloaking*, Phys. Rev. Lett. 110, 153605 (2013).
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  - S. Lagomarsino, *et al.*, *Robust luminescence of the silicon-vacancy center in diamond at high temperatures*, AIP Advances 5, 127117 (2015).
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