

Introduction to Quantum Nanophotonics

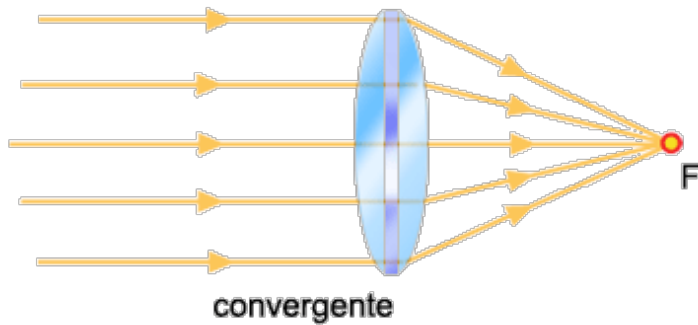
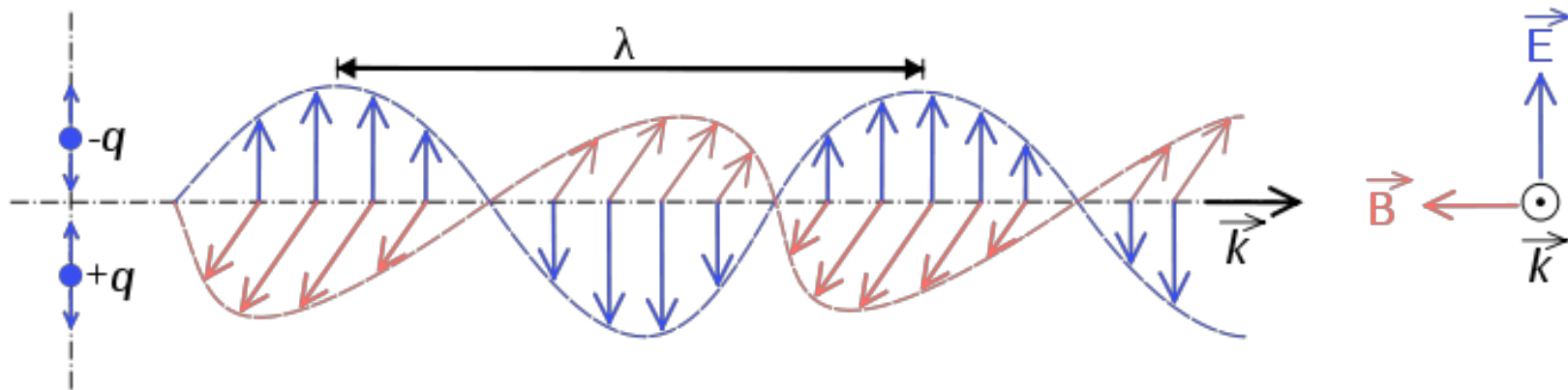
Luis Martín-Moreno

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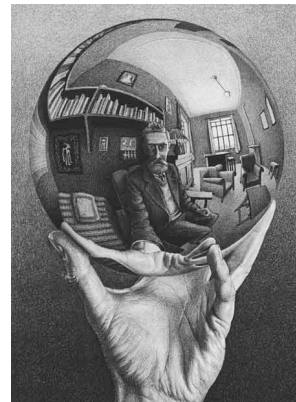
LMM@UNIZAR.ES

Photonics and Extreme Light
7/02/2019

Light and ways to control it.



Lenses



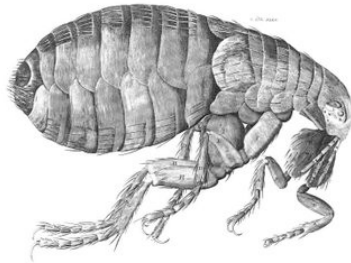
Mirrors



Waveguides

Optical Instruments.

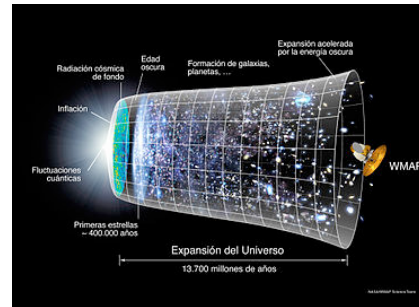
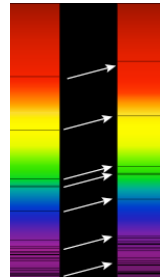
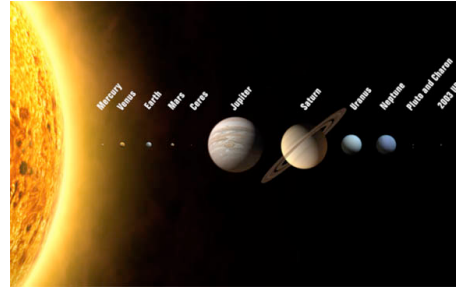
~1660



1665. Hooke:
cork cells

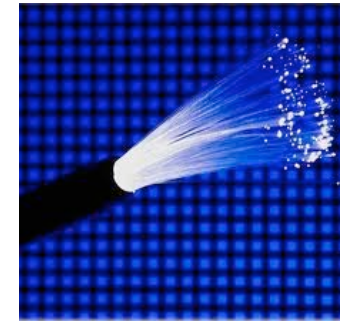
1670-80's van Leeuwenhoek:
protozoa, bacterias,
spermatozoids

~1590



Heliocentrism,
expansion of universe,
extrasolar planets,
dark energy&matter...

1950

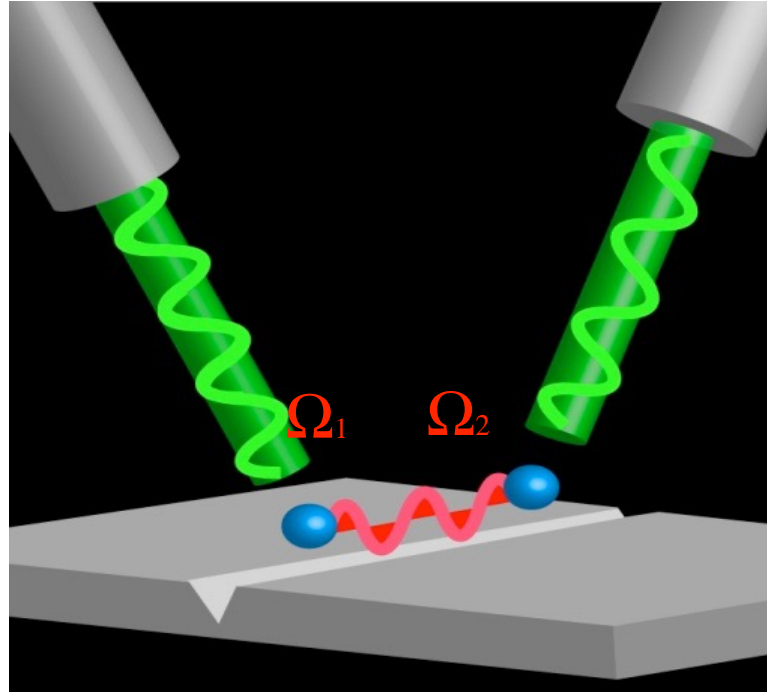


Communication,
Computation,
Medicine, ...

Nanophotonics

Why study nano-photonics?

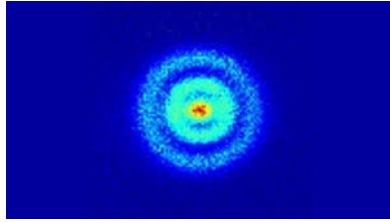
- **Light** can transport information “in parallel”
- The **energy** of light coincide with that of **electrical and vibrational excitations** in matter



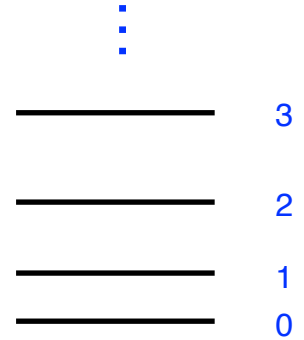
- qubit-qubit effective interactions
- photon-photon effective interactions
- nonlinear physics with minimum power
- single photon transistor / detector / emitter
- single molecule spectroscopy, photochemistry,

Light-Matter Interaction

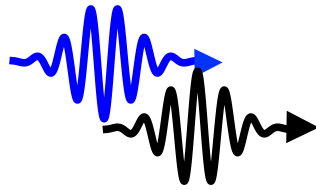
MATTER = ATOMS



$$H = \sum_i \frac{p_i^2}{2m_e} + E_{e-nucleus} + E_{e-e}$$



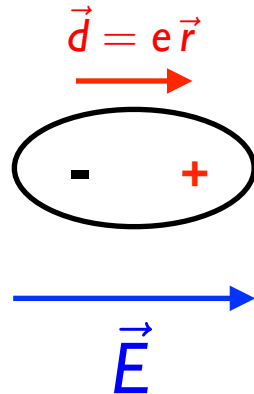
LIGHT = PHOTONS



$$H = \epsilon_0 |\vec{E}|^2 + \mu_0 |\vec{H}|^2$$

$$\omega_{\vec{k}} = c |\vec{k}|$$

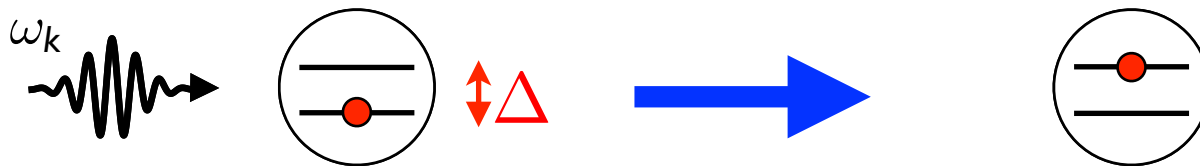
ATOMS + PHOTONS



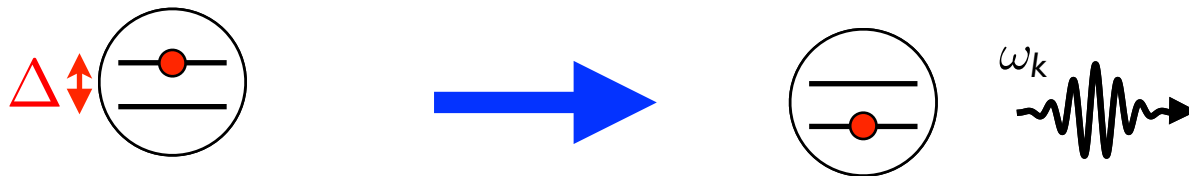
$$V \sim -q \vec{E} \cdot \vec{r} \rightarrow H_{int} = -\vec{d} \cdot \vec{E}(t)$$

Due to the interaction term...
atomic levels are NOT eigenstates of the TOTAL Hamiltonian

ABSORPTION



SPONTANEOUS EMISSION



Dipole-field interaction for a SINGLE photonic mode

$$H_{int} = -\vec{d} \cdot \vec{E} = g \underbrace{(\sigma^+ + \sigma^-)}_{\sigma_x} (a^+ + a)$$

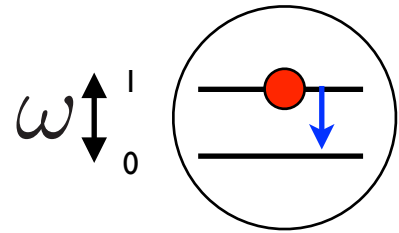
$$\sigma^+ \begin{array}{c} \text{---} \\ \bullet \\ \text{---} \end{array} = \begin{array}{c} \bullet \\ \text{---} \\ \text{---} \end{array}$$

$$= \underbrace{g(\sigma^+ a + \sigma^- a^+)}_{H_{RW}} + \cancel{\underbrace{g(\sigma^+ a^+ + \sigma^- a)}_{H_{CR}}}$$

The atomic excited state is not an eigenstate:

$$H_{int} |excited\rangle \otimes |0_{phot}\rangle = |ground\rangle \otimes |1_{phot}\rangle$$

Spontaneous emission in vacuum



$$P_I(t) = e^{-t/\tau_0}$$

$$\tau_0 = \frac{3c^2}{4n} \frac{1}{\alpha} \frac{1}{|d_{01}|^2} \frac{1}{\omega^3}$$

Excitation lifetime

Fine structure constant = 1/137

dipolar moment $d_{01} = -e \langle 1|x|0 \rangle$

??

$\omega^3 \sim$ Density of states for the outgoing photon (in 3D space)

Can we change the density of photon states?
what happens then?

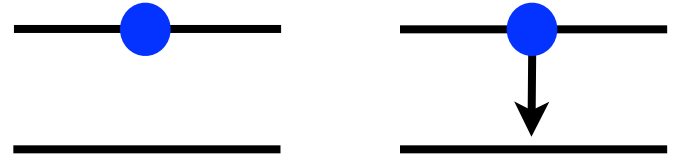
Tayloring Spontaneous Emission Rates.

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_{\nu} = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2) \text{ sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7 \text{ sec.}^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V \sim a^3$, and if δ is the skin-depth at frequency ν , $f \sim \lambda^3/a^2\delta$. For a non-resonant circuit $f \sim \lambda^3/a^3$, and for $a < \delta$ it can be shown that $f \sim \lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7 \text{ sec.}^{-1}$.

Phys. Rev. 69, 681 (1946)

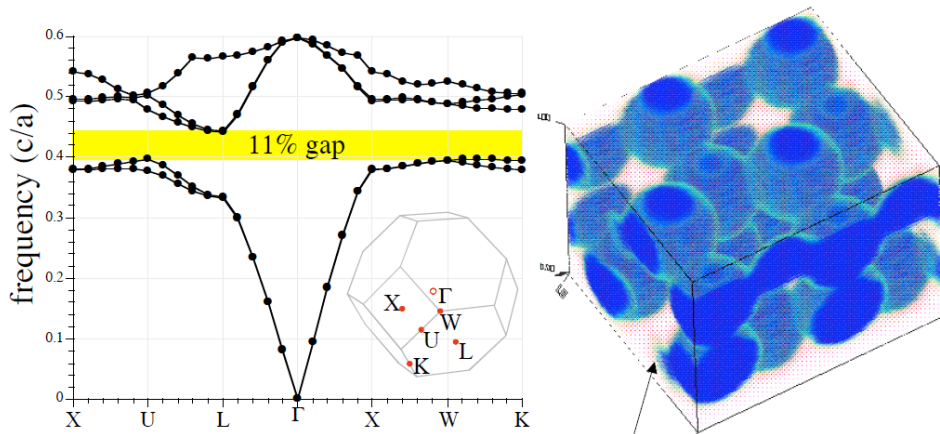


The spontaneous emission rate depends on the EM environment

Photonic crystals

The First 3d Bandgap Structure

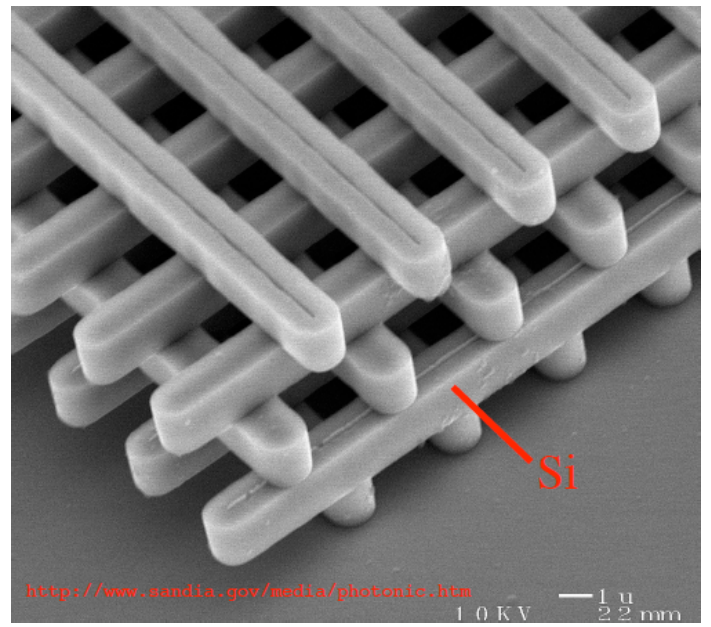
K. M. Ho, C. T. Chan, and C. M. Soukoulis, *Phys. Rev. Lett.* **65**, 3152 (1990).



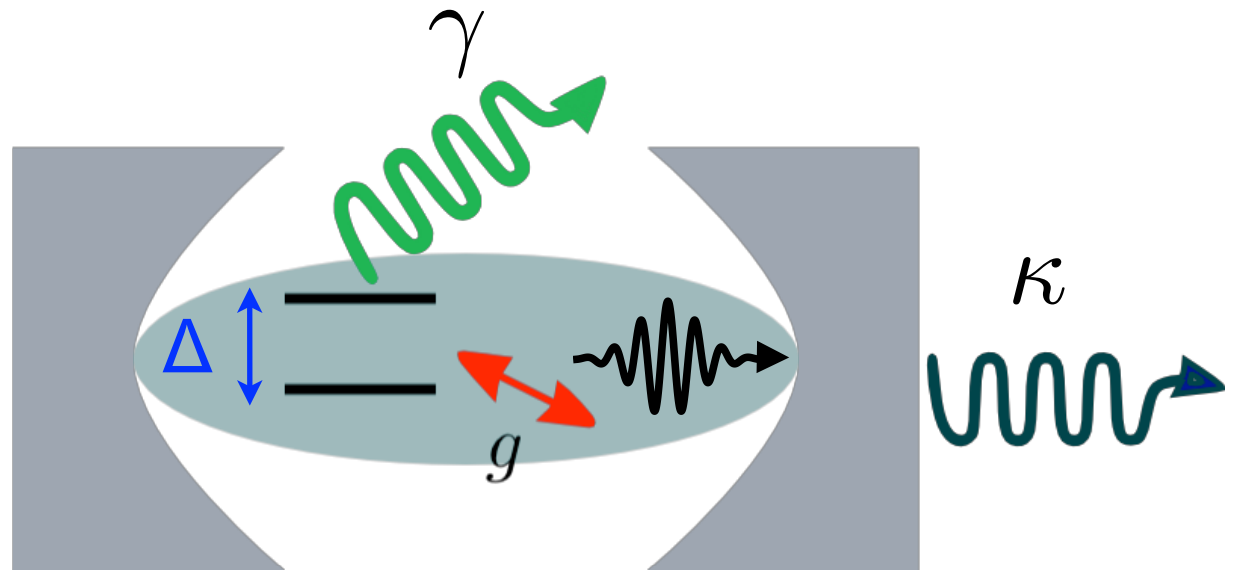
for gap at $\lambda = 1.55\mu\text{m}$,
sphere diameter $\sim 330\text{nm}$

overlapping Si spheres

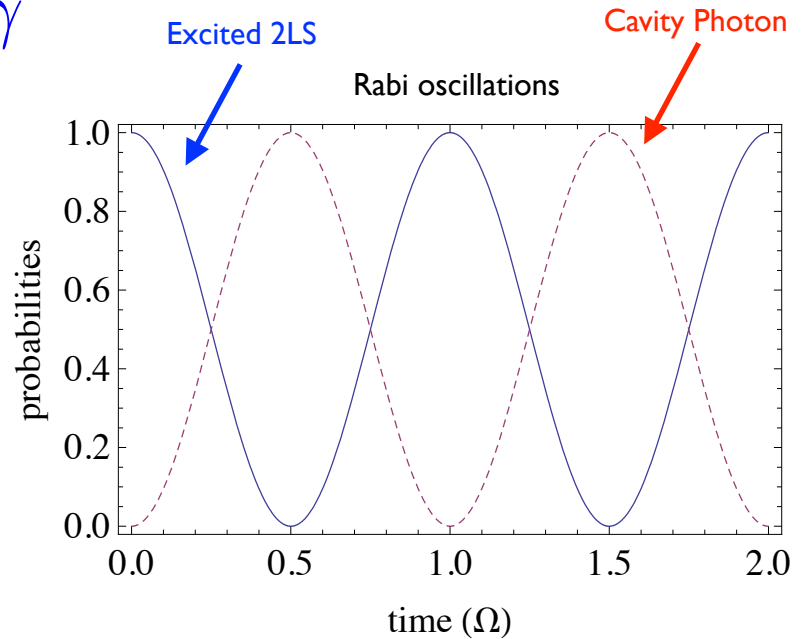
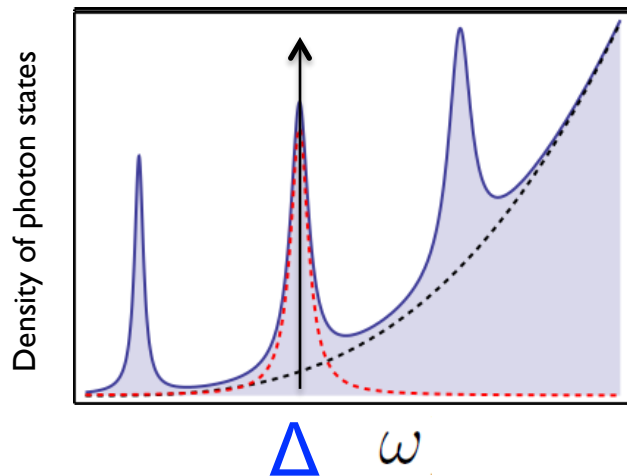
MPB tutorial, <http://ab-initio.mit.edu/mpb>

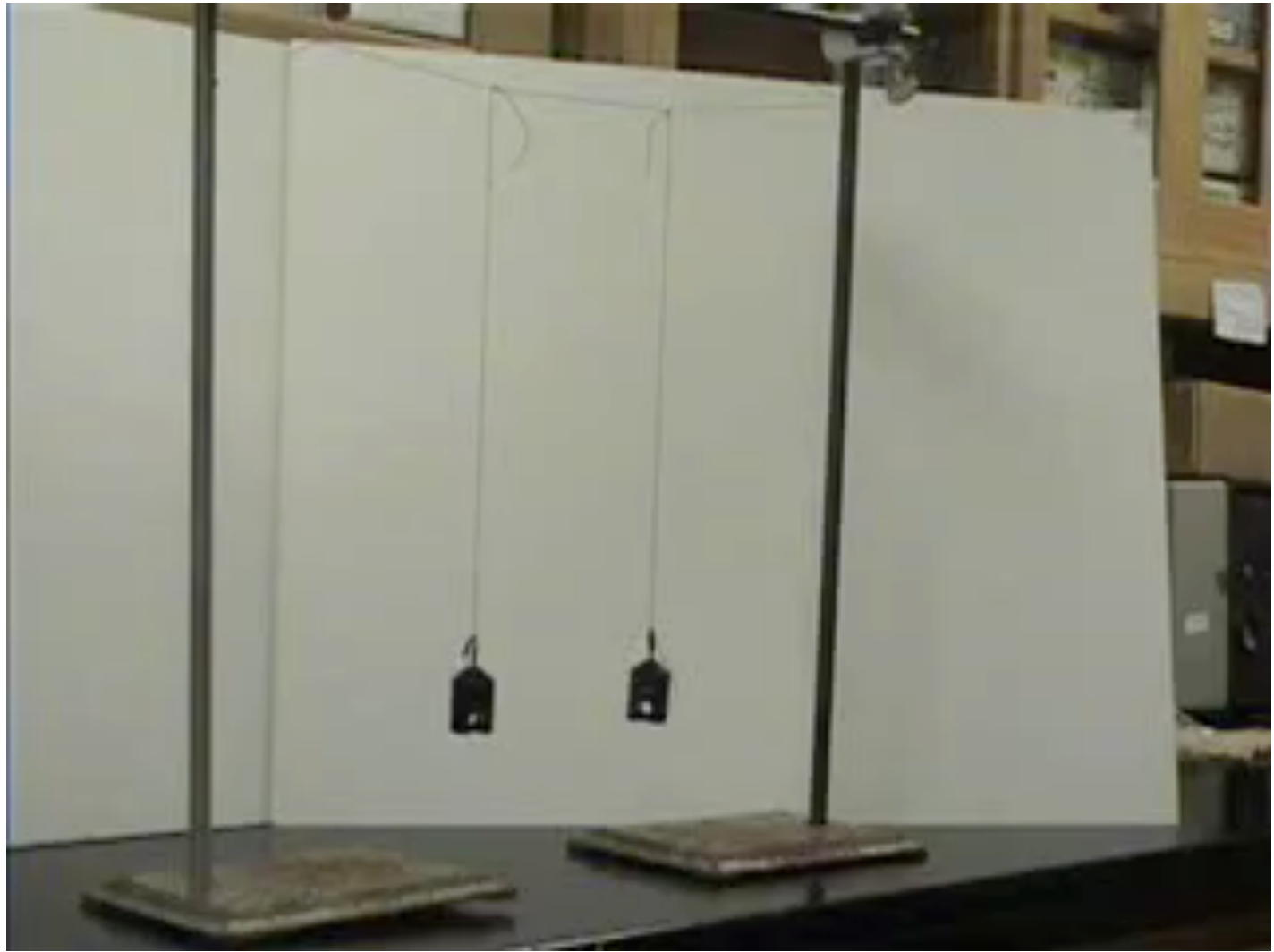


Cavity QED



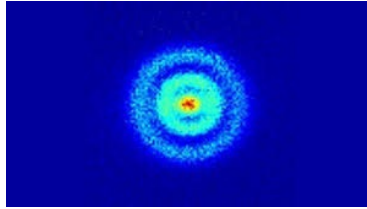
Strong Coupling : $g \gg \kappa, \gamma$



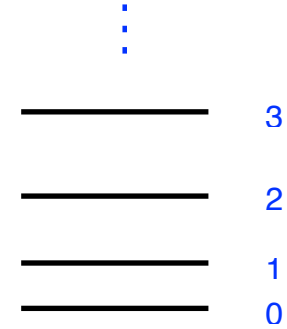


Light-Matter Interaction

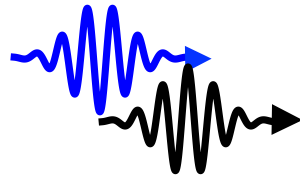
MATTER =



$$H = \sum_i \frac{p_i^2}{2m_e} + E_{e-nucleus} + E_{e-e}$$



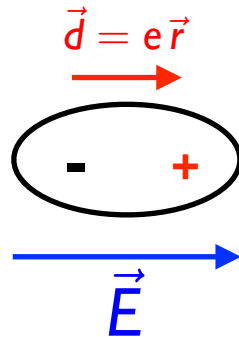
LIGHT =



$$H = \epsilon_0 |\vec{E}|^2 + \mu_0 |\vec{H}|^2$$

$$\omega_{\vec{k}} = c |\vec{k}|$$

ATOMS +

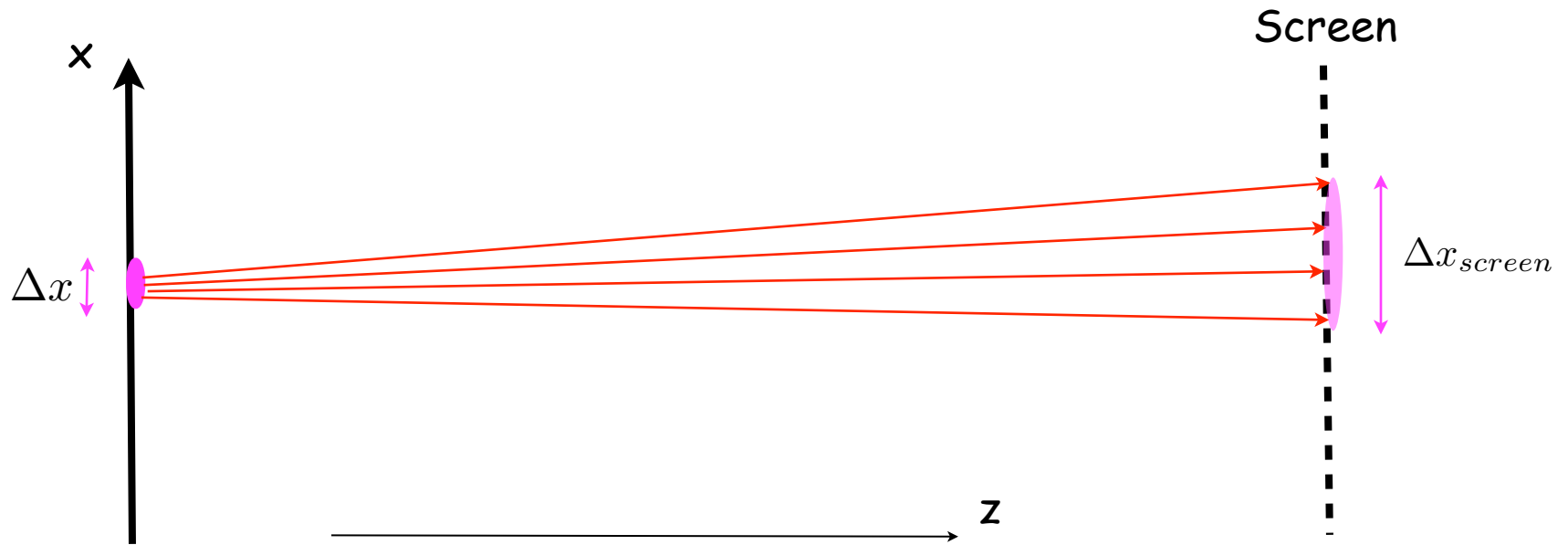


$$V \sim -q \vec{E} \cdot \vec{r} \rightarrow H_{int} = -\vec{d} \cdot \vec{E}(t)$$

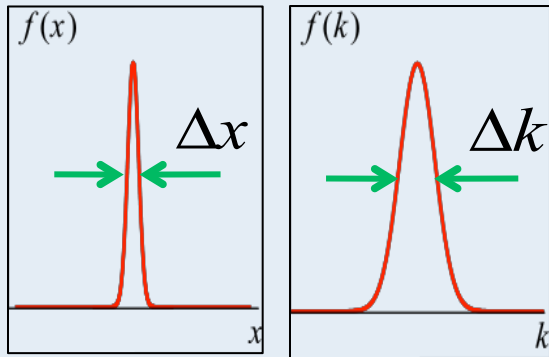
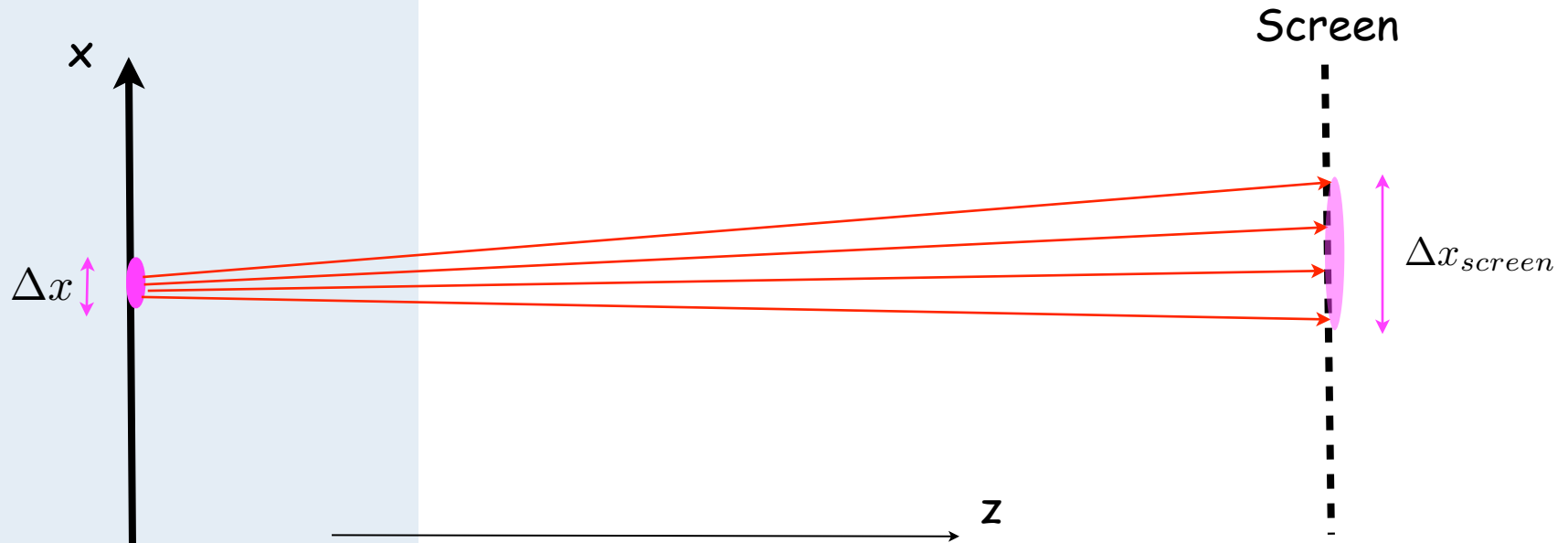
Idea:

Enhance coupling by focusing E
how much can E be confined?

The diffraction limit.



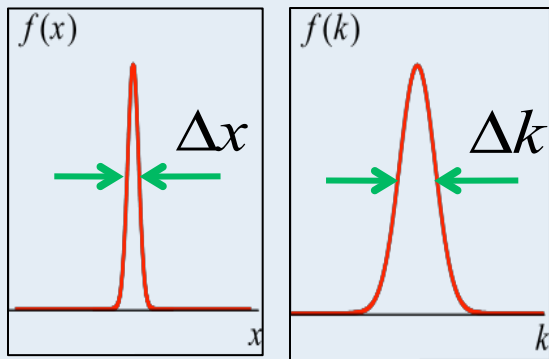
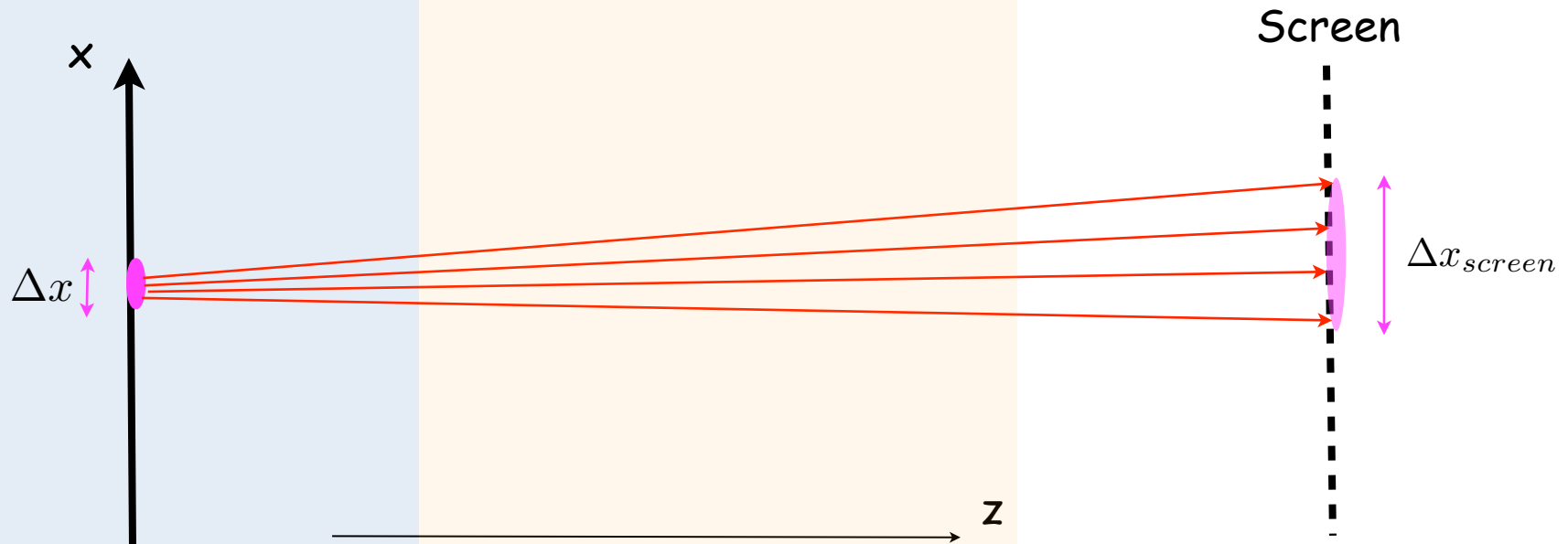
The diffraction limit.



$$f(x) = \int f(k) e^{ikx} dk$$

$$\Delta k \Delta x \approx 2$$

The diffraction limit.



$$\vec{E}(\vec{r}) = \vec{e}_\sigma e^{ik_x x} e^{ik_z z}$$

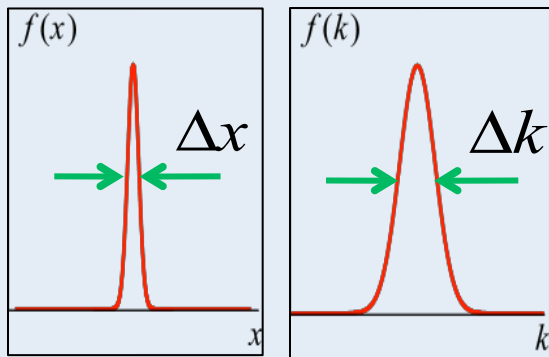
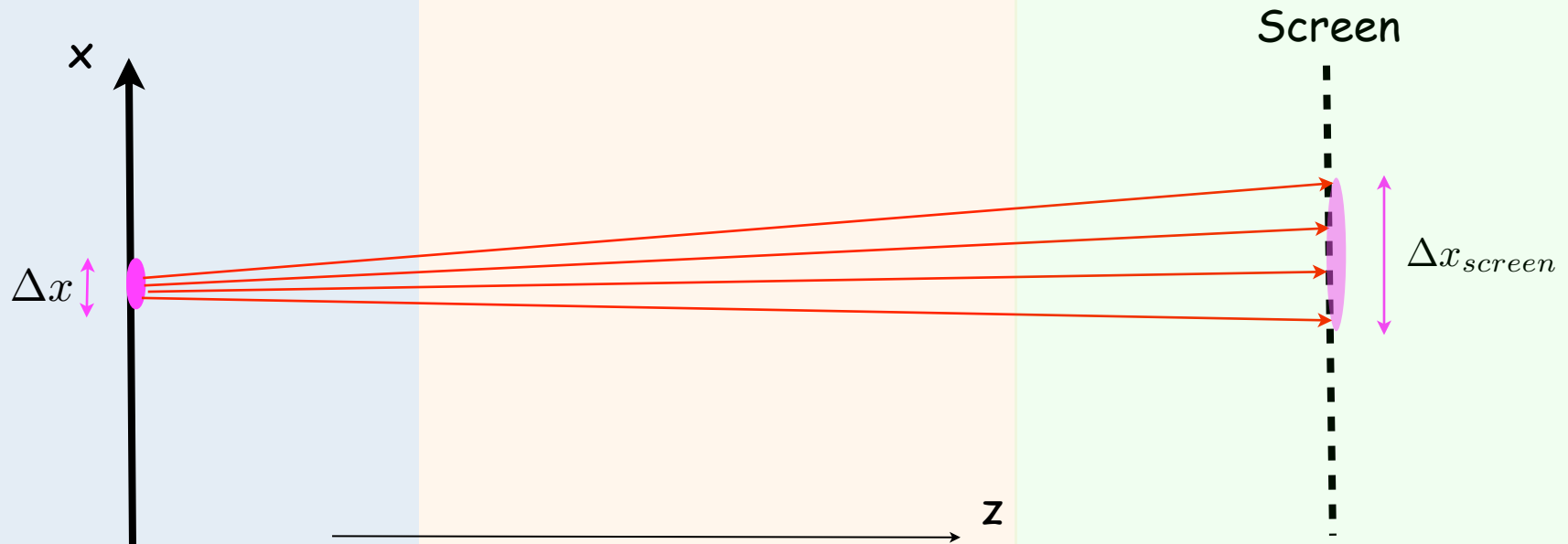
$$|k| = \sqrt{k_x^2 + k_z^2} = \omega/c$$

$$k_z^2 \geq 0 \rightarrow |k_x| < |k|$$

$$f(x) = \int f(k) e^{ikx} dk$$

$$\Delta k \Delta x \approx 2$$

The diffraction limit.



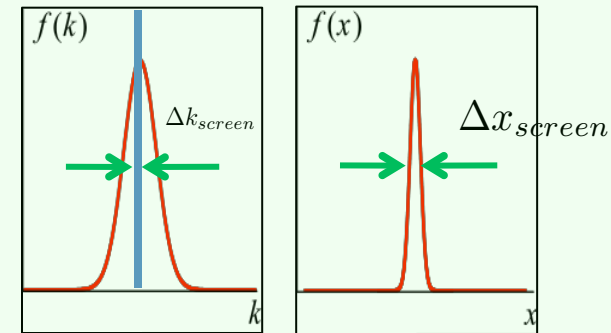
$$\vec{E}(\vec{r}) = \vec{e}_\sigma e^{ik_x x} e^{ik_z z}$$

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$$k_z^2 \geq 0 \rightarrow |k_x| < |k|$$

$$f(x) = \int f(k) e^{ikx} dk$$

$$\Delta k \Delta x \approx 2$$



$$\Delta k_{\text{screen}} = 2 \frac{\omega}{c}$$

$$\Delta x_{\text{screen}} \approx \lambda/2$$

$$\Delta x \geq \frac{\lambda}{2}$$

Minimum **lateral** size of light
(and this is only confined in 1D!)

- For nm resolution we need λ in the nm range -> X rays
- X rays are intensively used for this reason, but have problems (too much energy, so they damage matter)

With light λ is 500-900nm, so $\Delta x \approx 250\text{nm}$

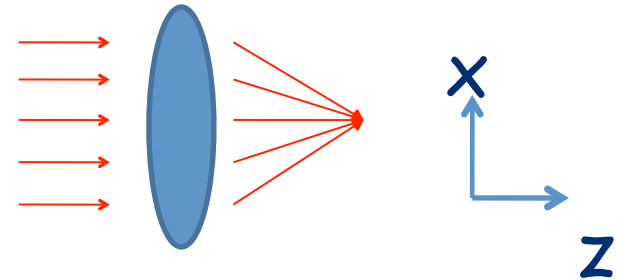
Poor resolution

It is not possible to address single molecules separately

Revisiting the diffraction limit (I)

Beating the diffraction limit.

$$\omega^2 = c^2 k^2 = c^2 (k_x^2 + k_z^2)$$



However, if we could play with $k_z^2 < 0$...

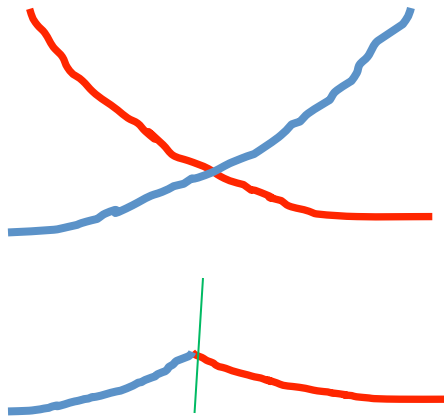
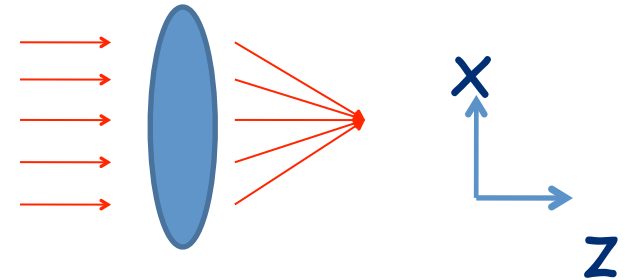
$$k_x^2 = k_\omega^2 - k_z^2 = k_\omega^2 + |k_z^2| \quad \text{as large as we want}$$

$\rightarrow \Delta x$ **arbitrarily small**

$$k_z^2 < 0 \longrightarrow k_z = \pm i |k_z| \quad \rightarrow \quad E(\vec{r}) = \vec{e}_\sigma e^{ik_x x} e^{ik_z z}$$

Beating the diffraction limit.

$$\vec{E}(\vec{r}) = \vec{e}_\sigma e^{ik_x x} e^{\pm|k_z|z}$$



These waves can not exist
in uniform media, but...

exist if there are interfaces

Problem:

the field is only intense close to the interfase

-> near-field optics

Another way to change the
density of photon states:

Metals

Surface Plasmons

$$H_y^{SP}(x, z, \vec{k}) = H_o e^{i \operatorname{Re}(k_x)x} e^{-\operatorname{Im}(k_x)x} e^{i \operatorname{Re}(k_z)z} e^{-\operatorname{Im}(k_z)z}$$

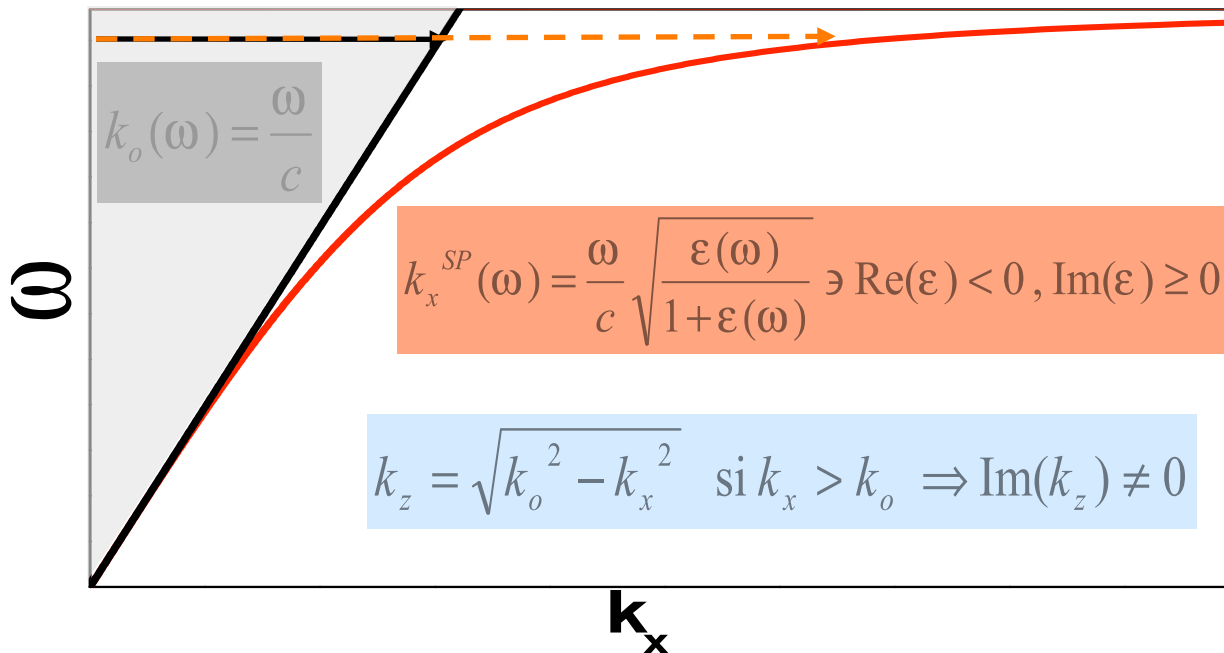
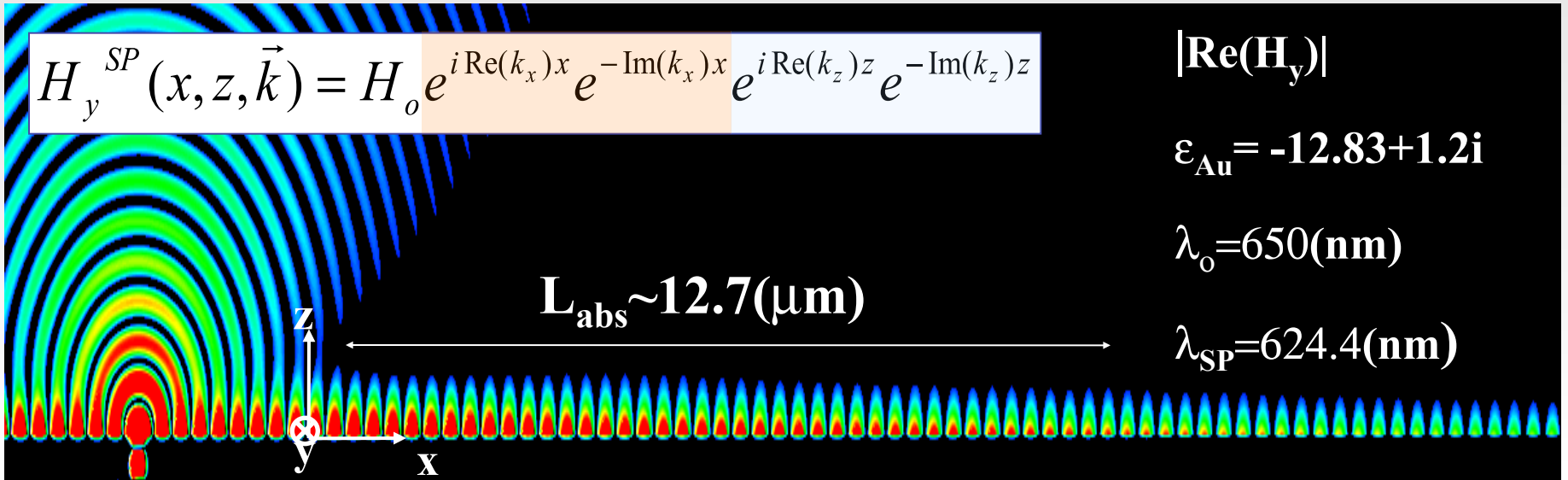
$$|\operatorname{Re}(H_y)|$$

$$\epsilon_{\text{Au}} = -12.83 + 1.2i$$

$$\lambda_o = 650(\text{nm})$$

$$\lambda_{\text{SP}} = 624.4(\text{nm})$$

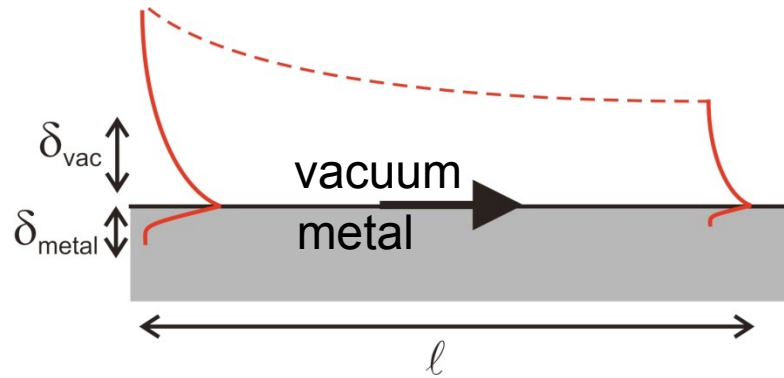
$$L_{\text{abs}} \sim 12.7(\mu\text{m})$$



Horizontal light

Small “Modal”
volume: localized
enhancement of the
EM field

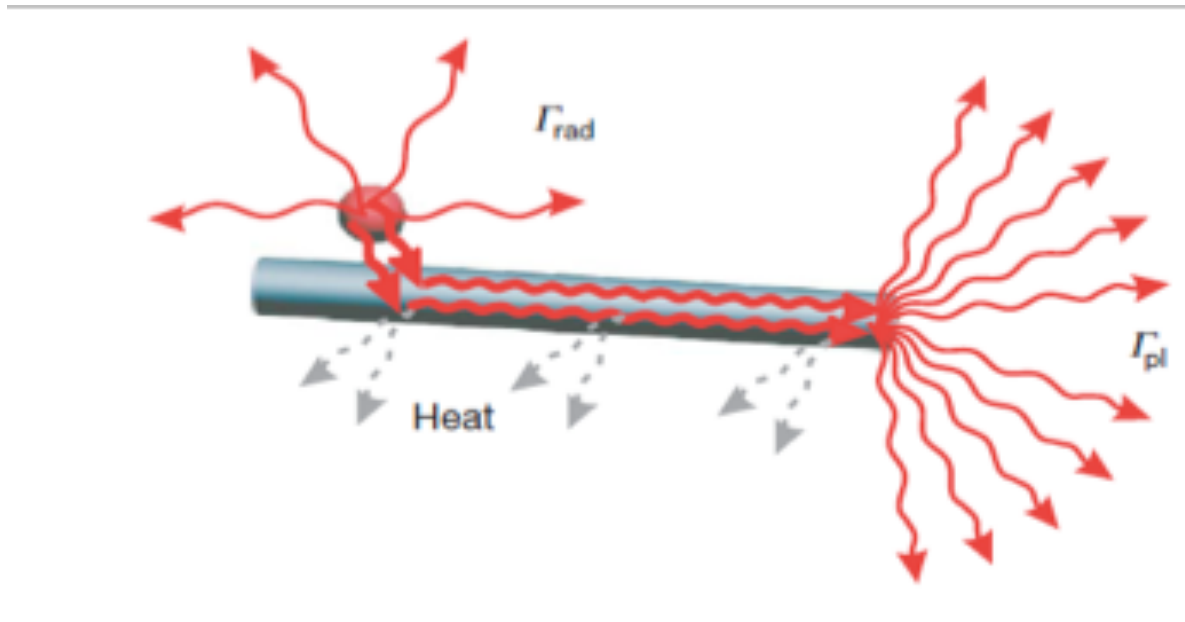
SPPs: typical numbers in Silver



$$\lambda_0 = 600 \text{ nm} \rightarrow \epsilon \approx -15 + i0.5 \left\{ \begin{array}{l} \delta_{\text{metal}} \approx 30 \text{ nm} \\ \delta_{\text{vacuum}} \approx 0.3 \mu\text{m} (0.5\lambda_0) \rightarrow \text{sub-}\lambda! \\ \ell_{\text{propagation}} \approx 40 \mu\text{m} (60\lambda_0) \end{array} \right.$$

$$\lambda_0 = 1500 \text{ nm} \rightarrow \epsilon \approx -110 + i3 \left\{ \begin{array}{l} \delta_{\text{metal}} \approx 25 \text{ nm} \\ \delta_{\text{vacuum}} \approx 2.3 \mu\text{m} (1.5\lambda_0) \\ \ell_{\text{propagation}} \approx 300 \mu\text{m} (200\lambda_0) \rightarrow \text{low loss} \end{array} \right.$$

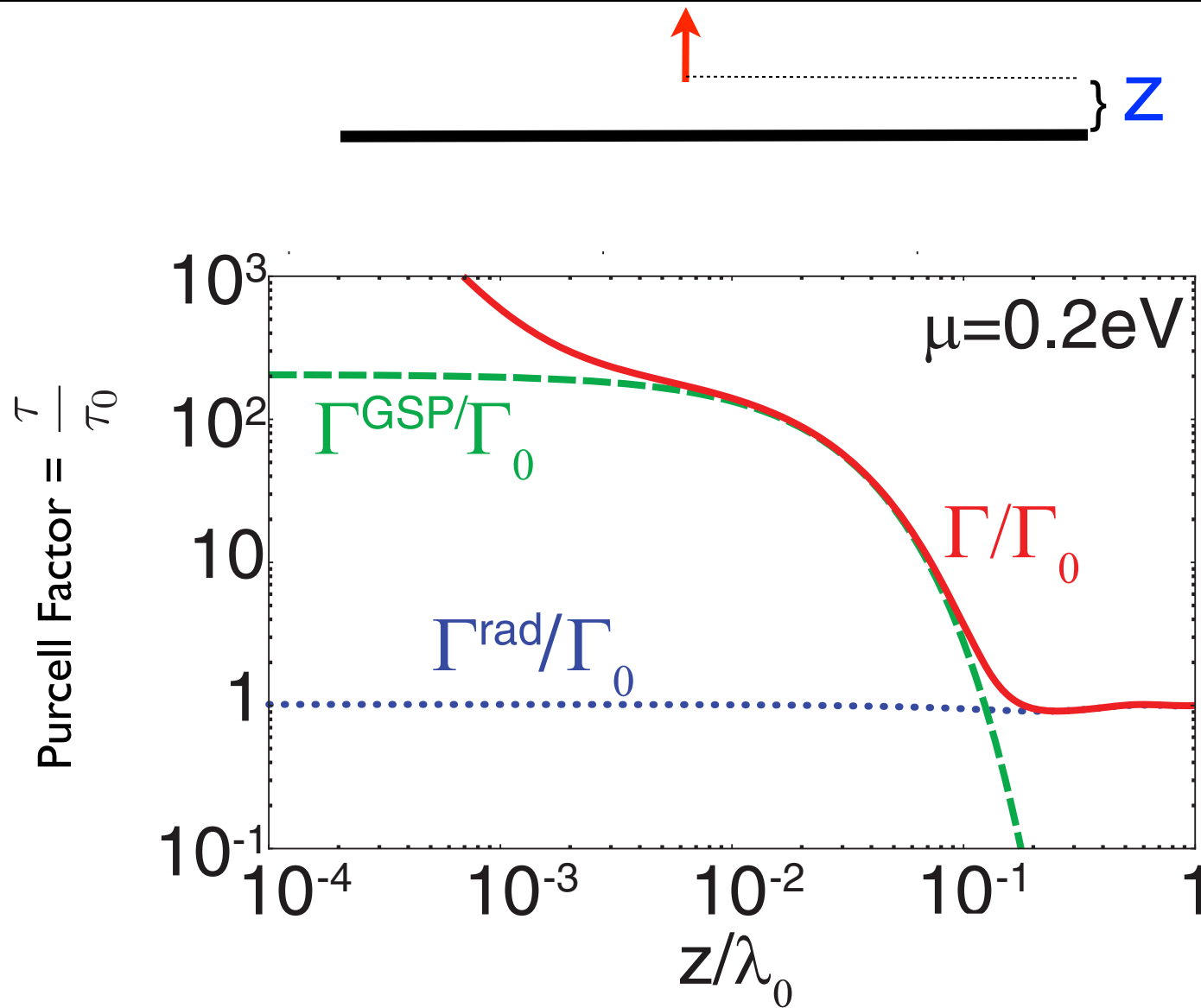
Radiation by a dipole



$$\gamma_{\text{total}} = \gamma_{\text{free radiation}} + \gamma_{\text{non-radiative (Heat)}} + \gamma_{\text{pl}}$$

If $\beta \equiv \frac{\gamma_{\text{pl}}}{\gamma_{\text{total}}} \approx 1$ — Most of the radiation is emitted as a propagating SPP!

Purcell Factor in a metallic system: Graphene



Strong coupling between SPPs and excitons in organic molecules

PHYSICAL REVIEW B 71, 035424 (2005)

VOLUME 93, NUMBER 3

PHYSICAL REVIEW LETTERS

week ending
16 JULY 2004

Strong Coupling between Surface Plasmons and Excitons in an Organic Semiconductor

J. Bellessa,* C. Bonnard, and J. C. Plenet

Laboratoire de Physique de la Matière condensée et Nanostructures, CNRS UMR 5586, Univ. Lyon-I, Villeurbanne 69622, France

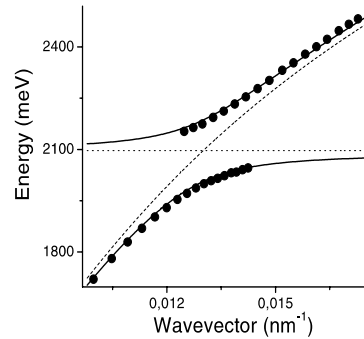
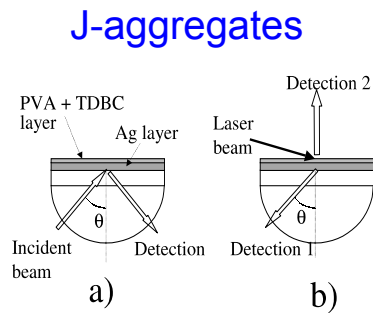
Strong coupling between surface plasmon-polaritons and organic molecules in subwavelength hole arrays

J. Dintinger,¹ S. Klein,^{1,*} F. Bustos,^{1,†} W. L. Barnes,² and T. W. Ebbesen^{1,‡}

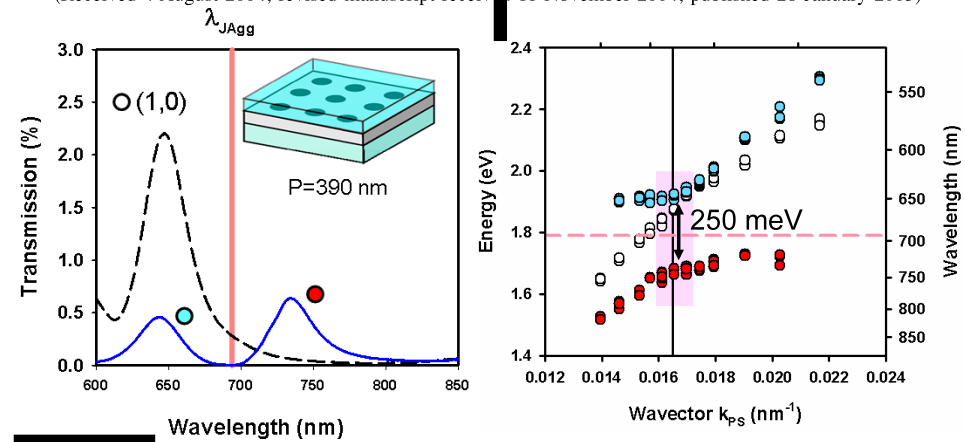
¹ISIS, Université Louis Pasteur, 8 allée G. Monge, 67000 Strasbourg, France

²School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom

(Received 4 August 2004; revised manuscript received 11 November 2004; published 28 January 2005)



$\Delta = 180 \text{ meV}$

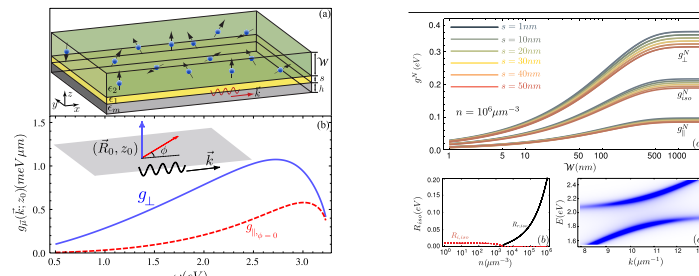


- T. K. Hakala, et al, Phys. Rev. Lett. 103, 053602 (2009).
- P.Vasa, et al, Nano Lett. 12, 7559 (2010).
- T. Schwartz, et al, Phys. Rev. Lett. 106, 196405 (2011).
- S. Aberra Guebro, et al, Phys. Rev. Lett. 108, 066401 (2012).

Semiconductor nanocrystals & Quantum wells:

- P.Vasa, et al., Phys. Rev. Lett. 101, 116801 (2008).
- J. Bellessa, et al, Phys. Rev. B 78 (2008).
- D. E. Gomez, et al, Nano Lett. 10, 274 (2010).
- M. Geiser, et al, Phys. Rev. Lett. 108, 106402 (2012).

Theory: A. Gonzalez-Tudela, P. Huidobro, LMM,
C. Tejedor and F. J. Garcia-Vidal, PRL, 110, 126801 (2013)



Transition Dipole-field interaction

$$H_{int} = -\vec{d} \cdot \vec{E} = g \underbrace{(\sigma^+ + \sigma^-)}_{\sigma_x} (a^+ + a)$$

$$\sigma^+ \begin{array}{c} \text{---} \\ \bullet \\ \text{---} \end{array} = \begin{array}{c} \bullet \\ \text{---} \\ \text{---} \end{array}$$

$$= \underbrace{g(\sigma^+ a + \sigma^- a^+)}_{H_{RW}} + \underbrace{g(\sigma^+ a^+ + \sigma^- a)}_{H_{CR}}$$

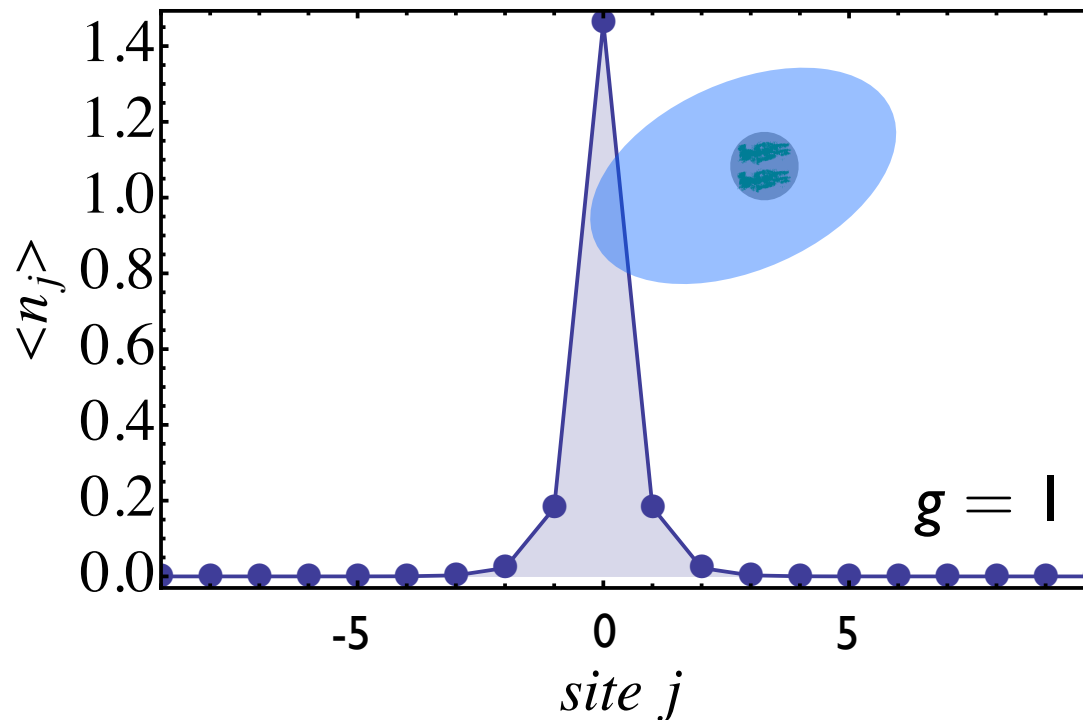
Important if $\frac{g}{\omega} \gtrsim 0.1$

When H_{CR} is relevant : Ultra-Strong Regime

A consequence



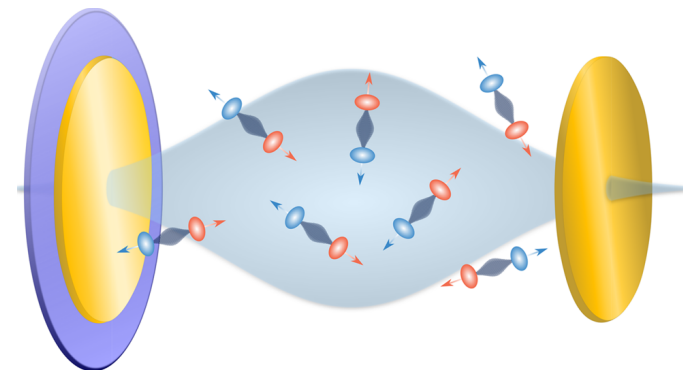
- $N = a^\dagger a + \sigma^+ \sigma^-$ is not a good quantum number
- The **ground state** is a “dressed” vacuum.



Hybrid Light–Matter States in a Molecular and Material Science Perspective

Thomas W. Ebbesen*

2016



$$\hbar\Omega_R = 2V_n = 2\mathbf{d}\cdot\mathbf{E}_0 = 2d\sqrt{\frac{\hbar\omega}{2\epsilon_0 v}} \times \sqrt{n_{\text{ph}} + 1}$$

$$\hbar\Omega_R \propto \sqrt{N/v} = \sqrt{C}$$

Interaction between molecules and EM fields in the UltraStrong regime:

↓
Hybrid modes without external illumination

↓
Change of energetics in the system

↓
change in the Thermodynamic properties:
magnetization, chemical reaction pathways, conductivity, etc...

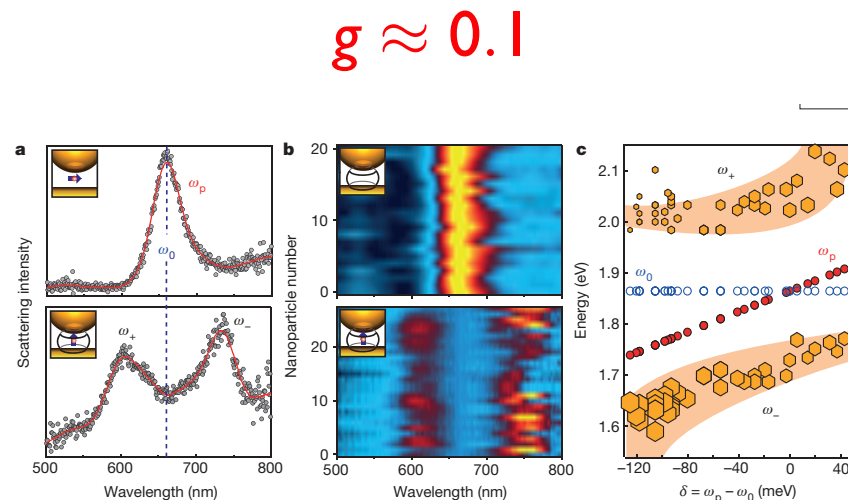
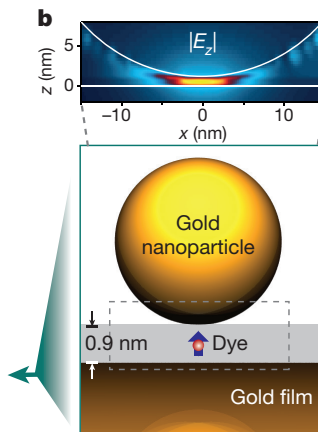
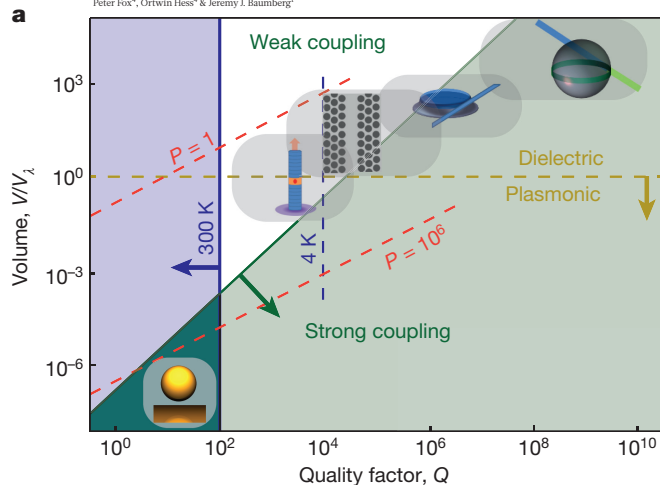
Single Molecule Strong Coupling in Cavities

LETTER

doi:10.1038/nature17974

Single-molecule strong coupling at room temperature in plasmonic nanocavities

Rohit Chikkaraddy¹, Bart de Nijs¹, Felix Benz², Steven J. Barrow³, Oren A. Scherman², Edina Rosta³, Angela Demetriadou⁴, Peter Fox⁴, Ortwin Hess⁵ & Jeremy J. Baumberg¹



ARTICLE

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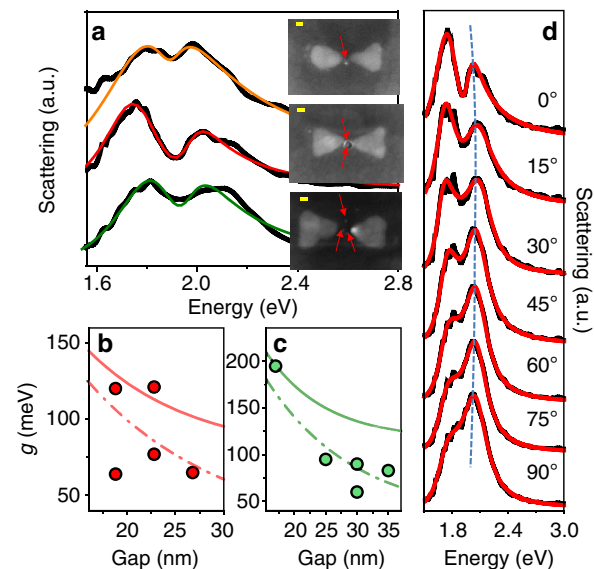
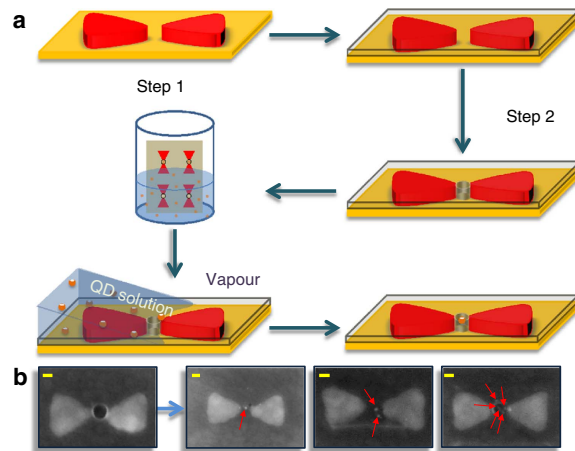
DOI: 10.1038/ncomms11823

OPEN

Vacuum Rabi splitting in a plasmonic cavity at the single quantum emitter limit

Kotni Santhosh^{1,*,†}, Ora Bitton^{2,*,†}, Lev Chuntonov^{3,*,†} & Gilad Haran¹

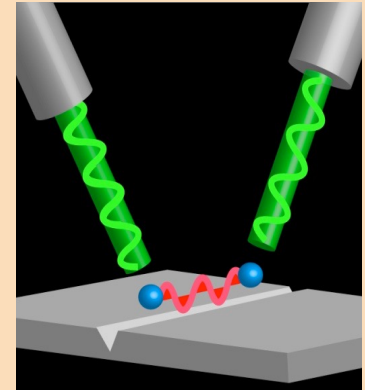
$g \approx 0.1$



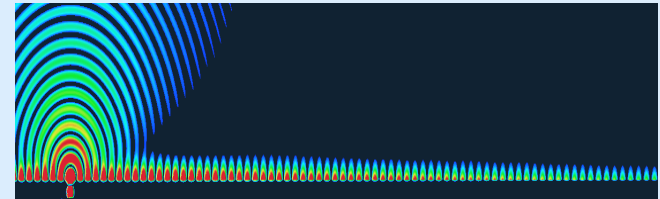
Conclusions

Nanophotonics aims at concentrating EM fields:

- Ultra-compact optical devices.
- Improved spacial resolution
- Enhanced Light-matter interaction



Metals support surface plasmons, which provide EM confinement for free



Ultrastrong coupling effects may lead to new physics:

- tailoring the vacuum
- new photochemistry (without photons!)

