Nanoscale quantum optics and nanocavity QED

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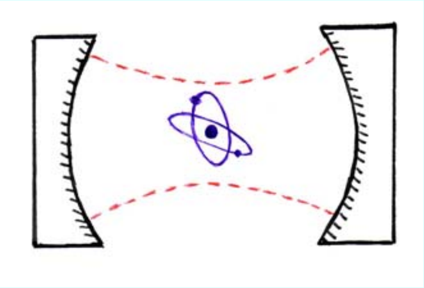
CEWQO-2008

Cavity Quantum Electrodynamics (Cavity QED):

Atomic system interacting with e.m. fields (vacuum and applied) in a confined environment



- modifications of both atomic response and field response
(spontaneous emission enhanced/inhibited, radiation diagram modified, lineshift)
- atom-field entanglement

Questions:

- Sub-wavelength size (nano-physics)
- Influence of (dielectric/metallic) material response

Outline:

1. **Introduction**
2. **Atom-light dynamics in sub-µm gas cells: “coherent Dicke narrowing”**
3. **Cavity QED in nanometric dielectric cavities**
4. **Surface-guided modes in atom-nanostructure interactions**
5. **Conclusion - Prospects**

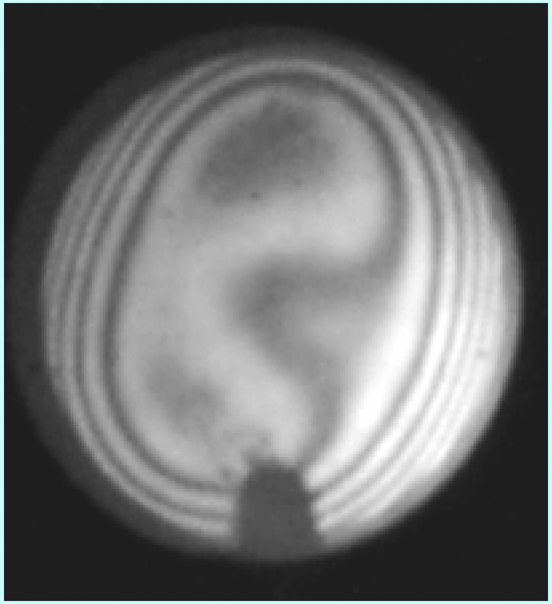
EXTREMELY THIN CELL ("nanocell")

- Thickness between windows, after evacuation

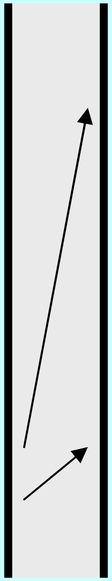
~ 20 nm - 1000 nm

- *filled with low pressure of Cs*

D. Sarkisyan ***et al.***,
Opt. Comm. **200**, 201 (2001)



A Doppler-free method



A longer interaction time for atoms flying parallel to the window
(Briaudeau,1996,….)

velocity selection

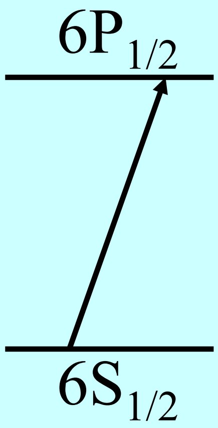
ETC : a Fabry-Perot

→ Allows a Thickness Accuracy **up to 1-2 nm**

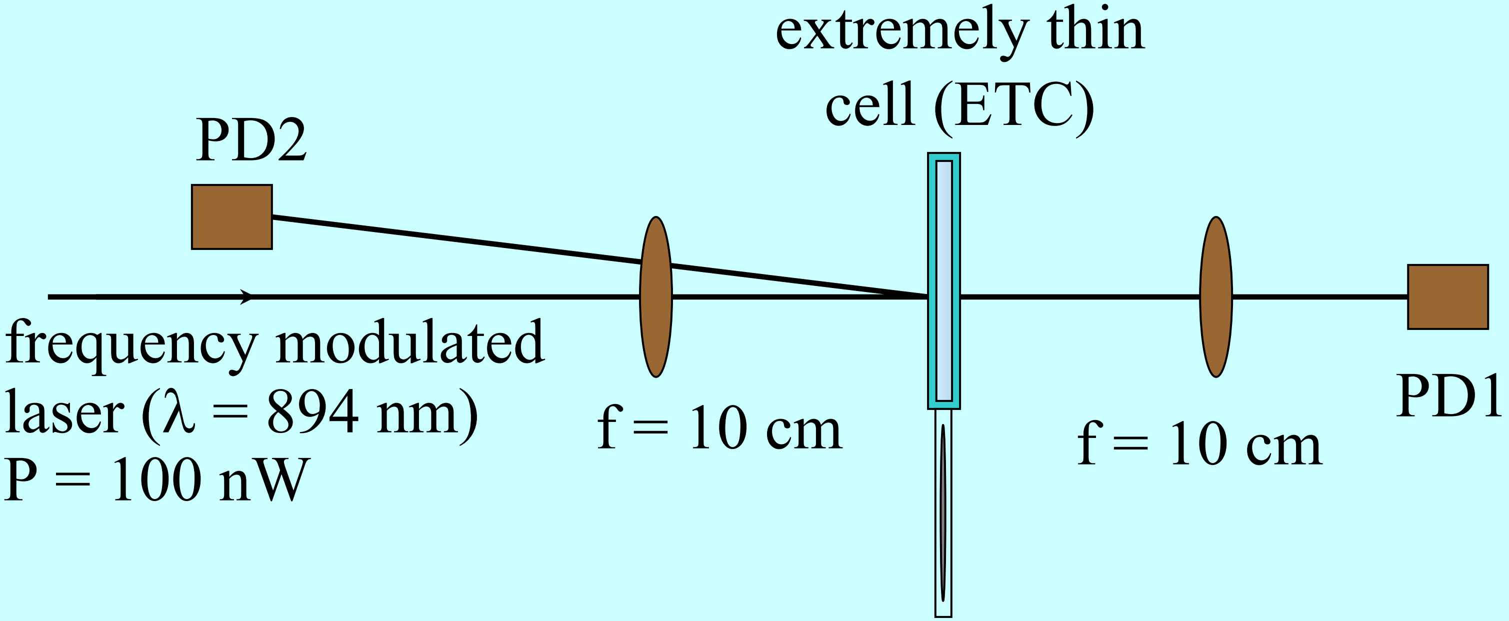
→ T and R always mixed-up

THE EXPERIMENTAL SET-UP

Excitation of Cs D1 resonance line (894 nm)



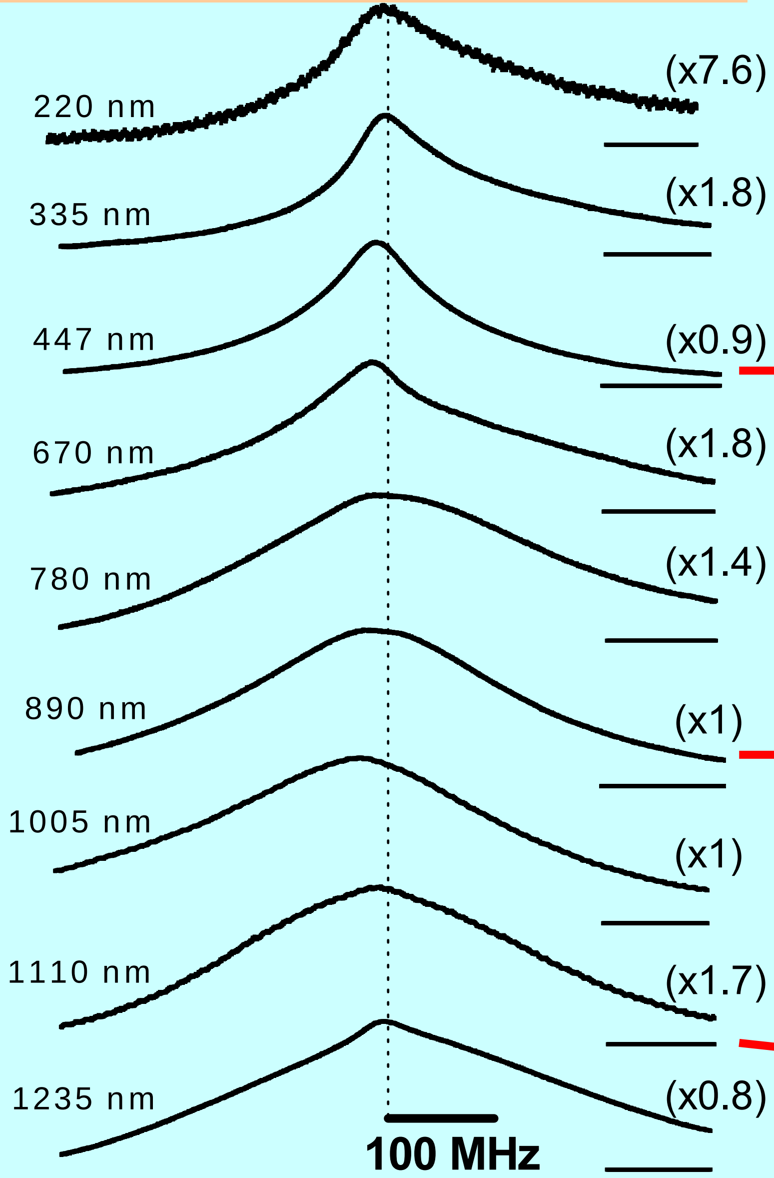
The set-up



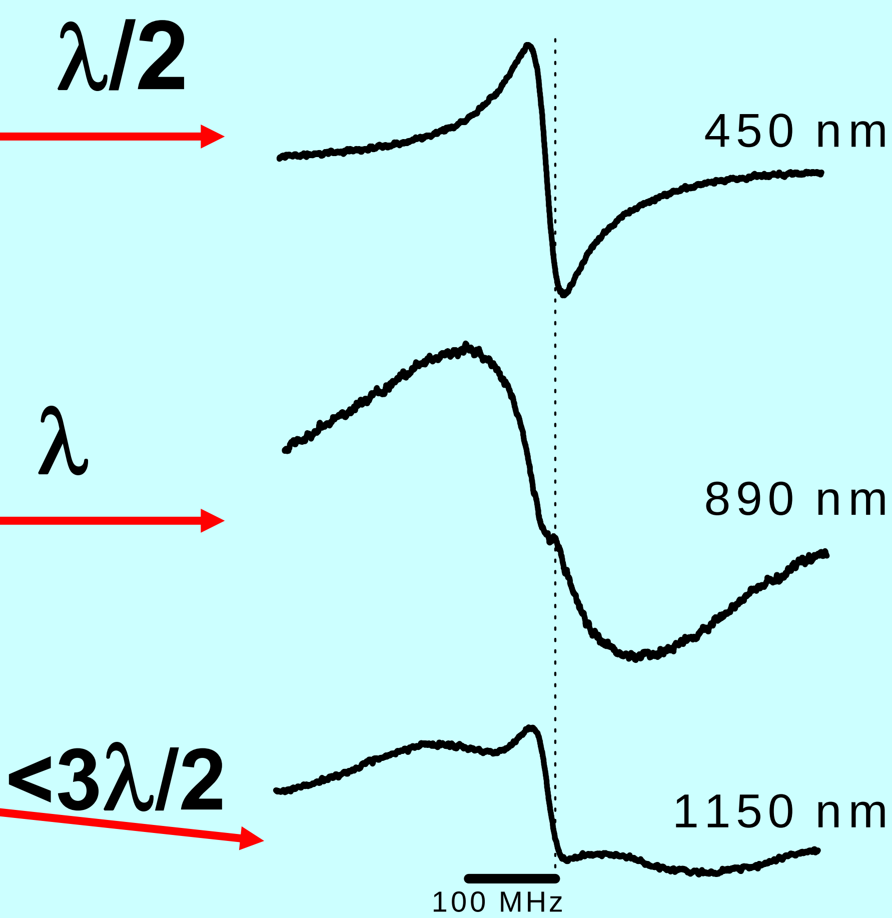
Optical transmission versus cell thickness

- J. Opt. Soc. Am. B, **20**, 793 (2003)
- Europhysics Letters, **63**, 35 (2003)

Direct Cs(D_1) Transmission



FM Transmission

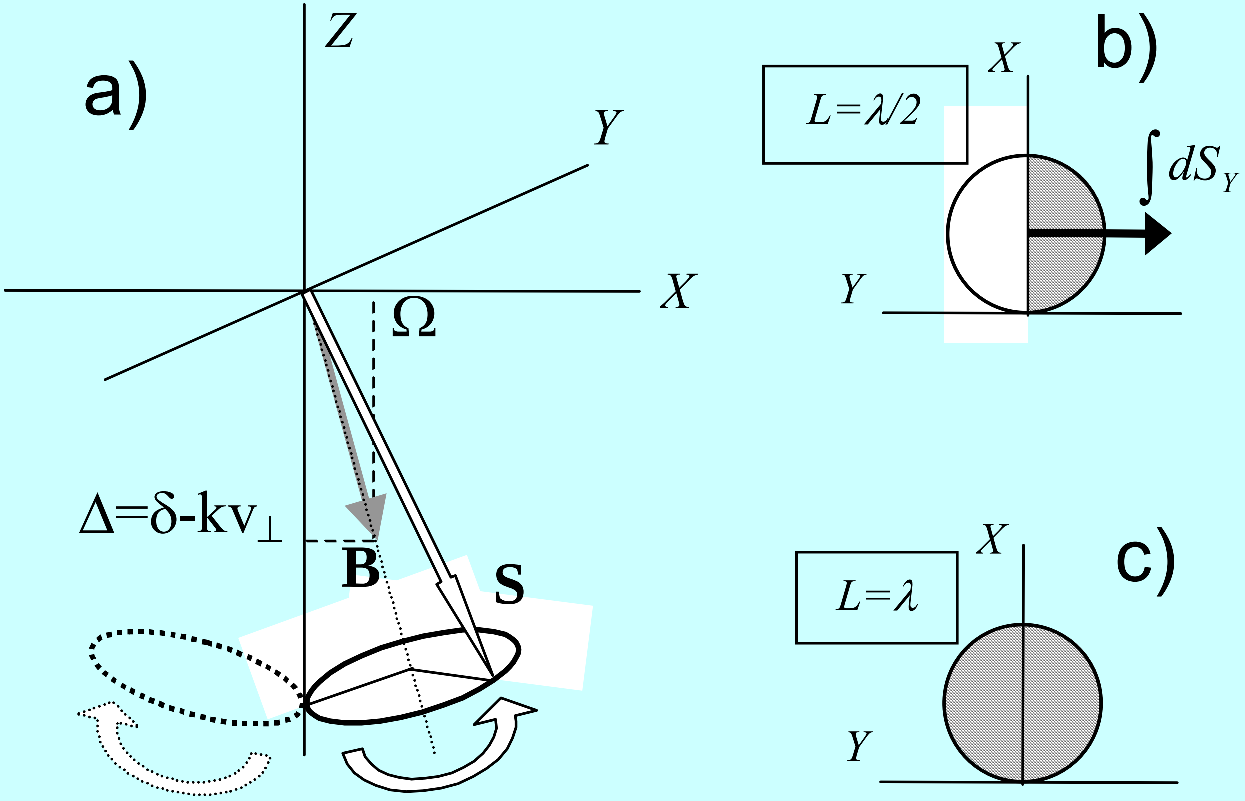


The Bloch vector model for "Dicke narrowing"

Sarkisyan *et al*, Phys Rev A **69**, 065802 (2004)

$$\frac{d\vec{S}}{dt} = \vec{S}\times\vec{B}\ ,\qquad \vec{B} = \begin{pmatrix}\Omega\\ 0\\ \delta-kv\end{pmatrix}$$

For $\delta=0$: S rotates around B at angular velocity ~ **kv** (if $\Omega\to0$) up to a duration **τ = L/v**



(Coherent)
Transmission

$$\int S_y dz$$

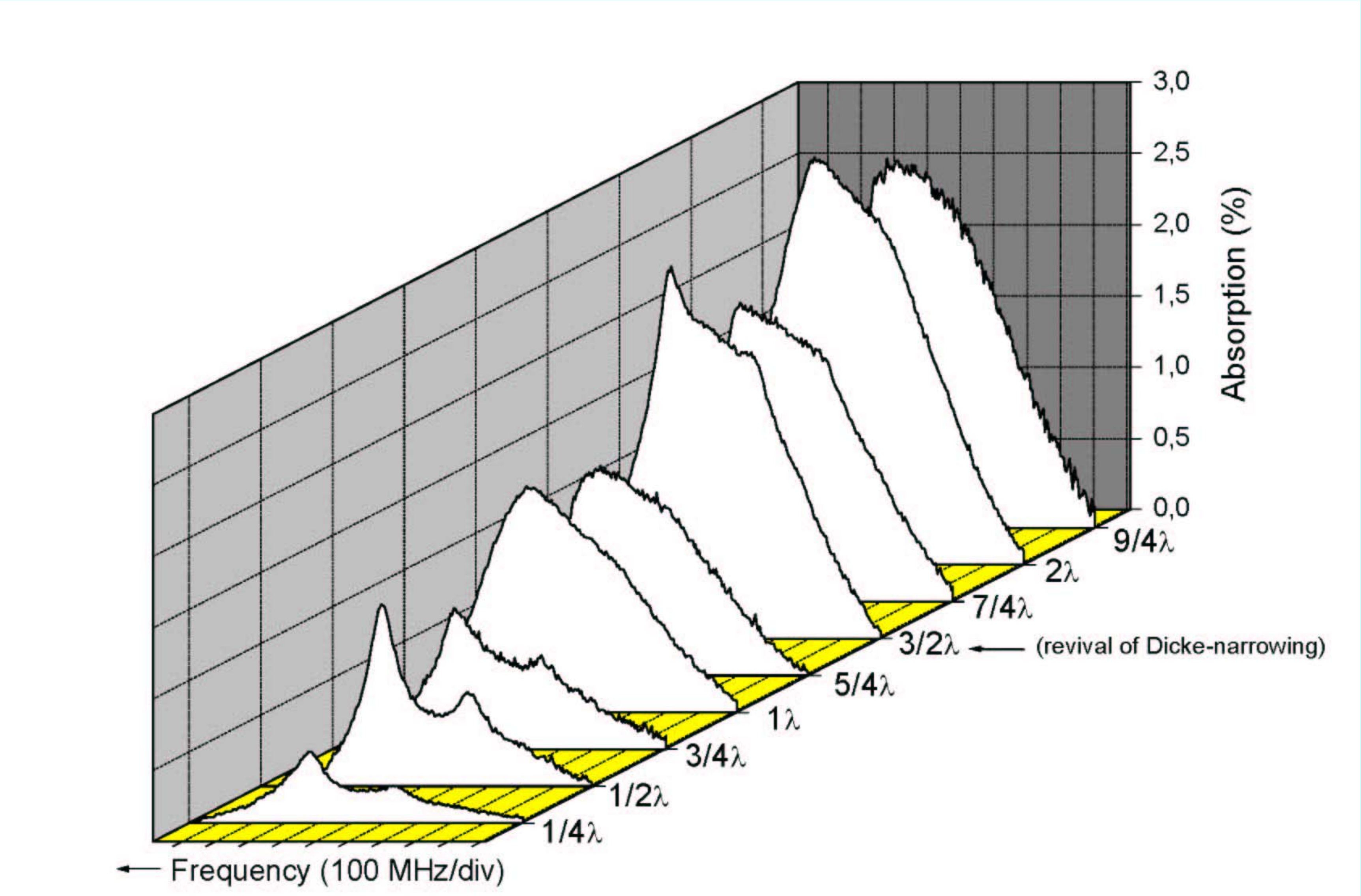
(Incoherent)
Fluorescence

$$\int S_z dz$$

Transmission: **Dicke narrowing** and revival

Rb nanocell ; λ = 780nm ; I = 0.4 mW/cm2 ; transitions F=2 → F=1, 2, 3

Sarkisyan *et al.,* Phys Rev A **69**, 065802 (2004)



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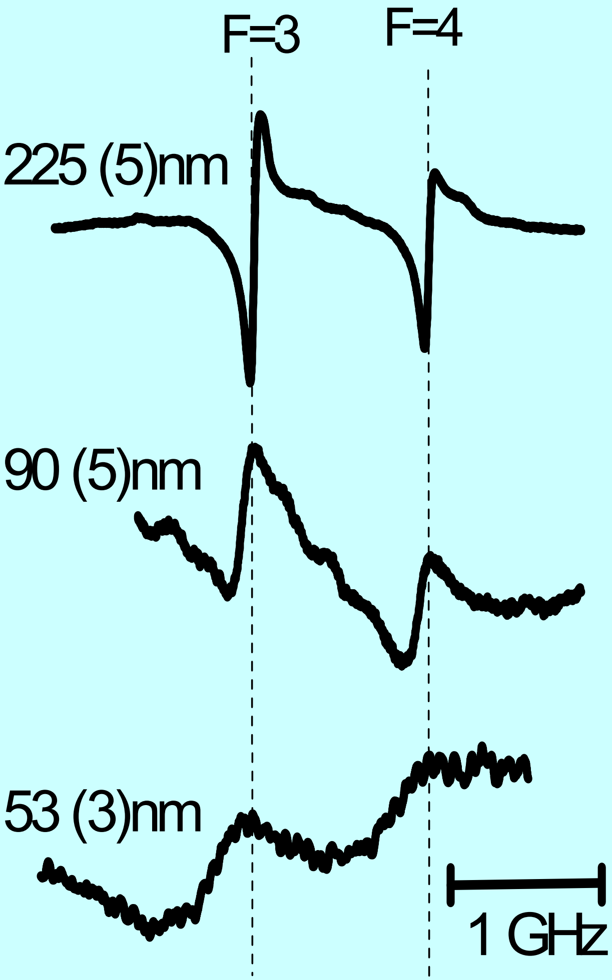
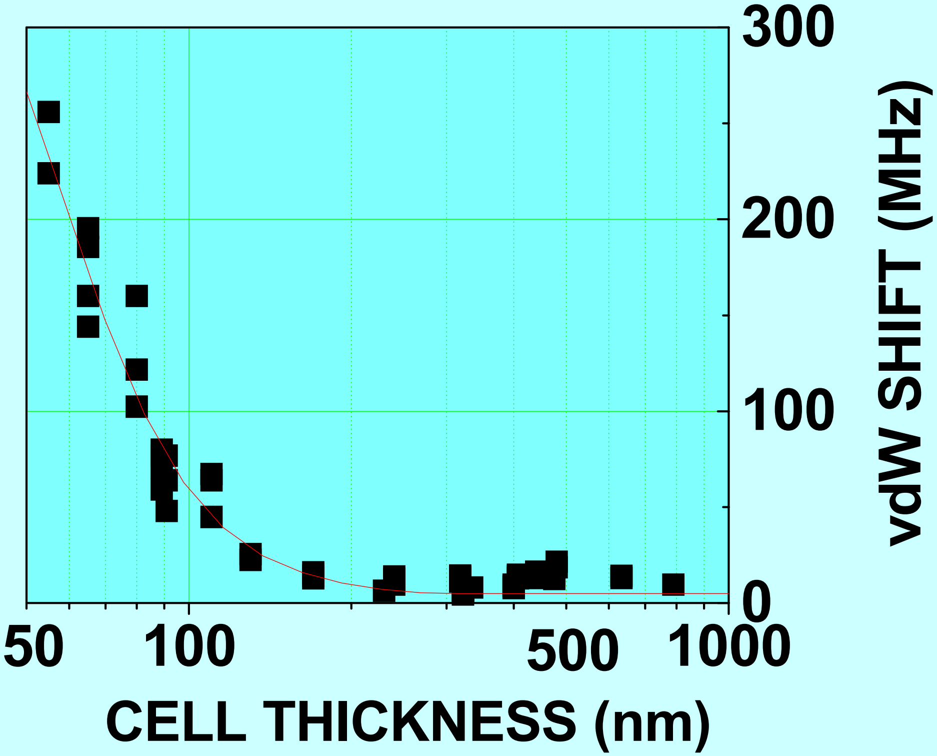
Cs (D$_1$) FM transmission for very small thickness (50-200nm) and YAG window



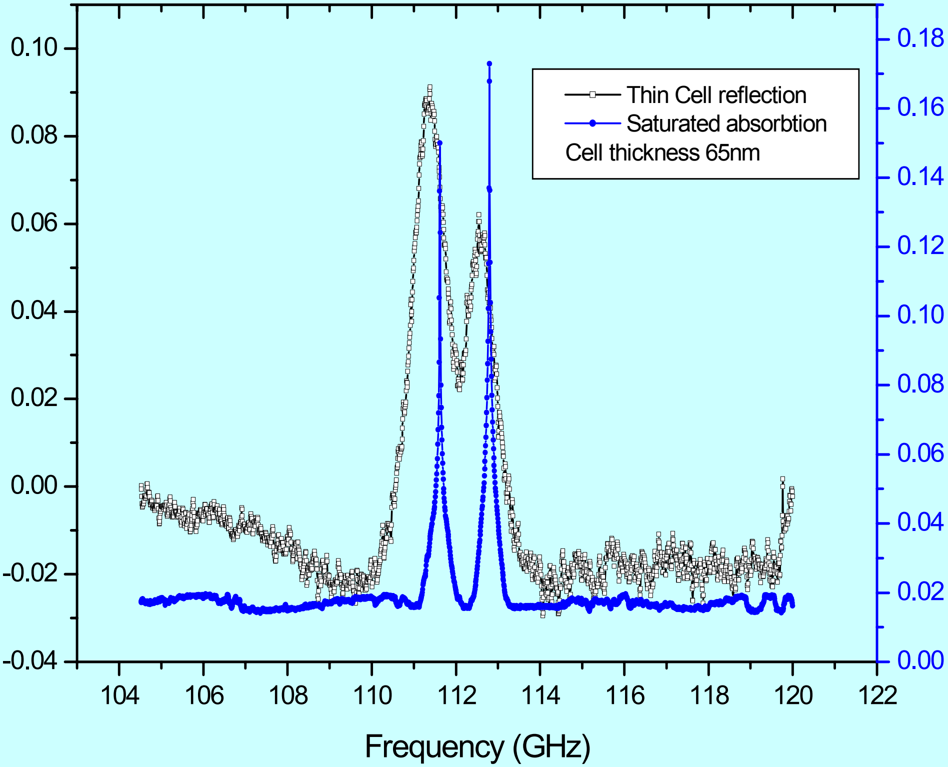


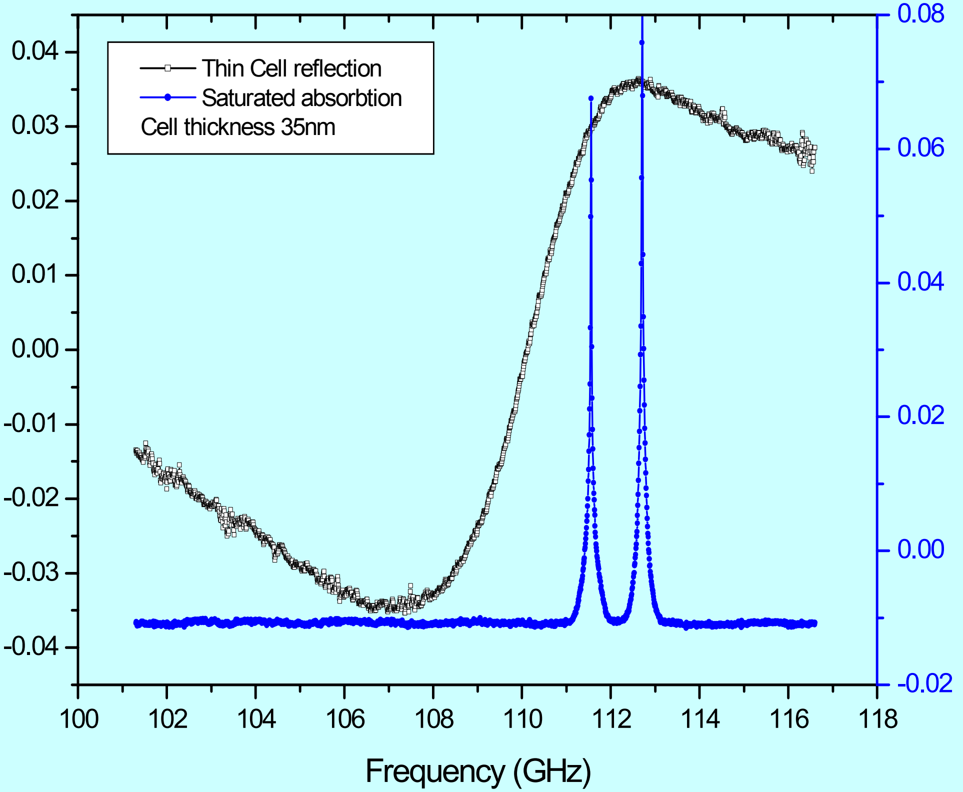
Cesium D_1 transition

Reflection spectra on vapor nano-cells (FM mode)

65nm

35 nm





Modelling transmission spectra in nanocells

- **2-wall van der Waals potential** (adding 2 walls, or multiple image modeling)

- **Spatial integration** of transient interaction regimes in the nano-cell

- **Velocity distribution** *i.e.* distribution over interaction time

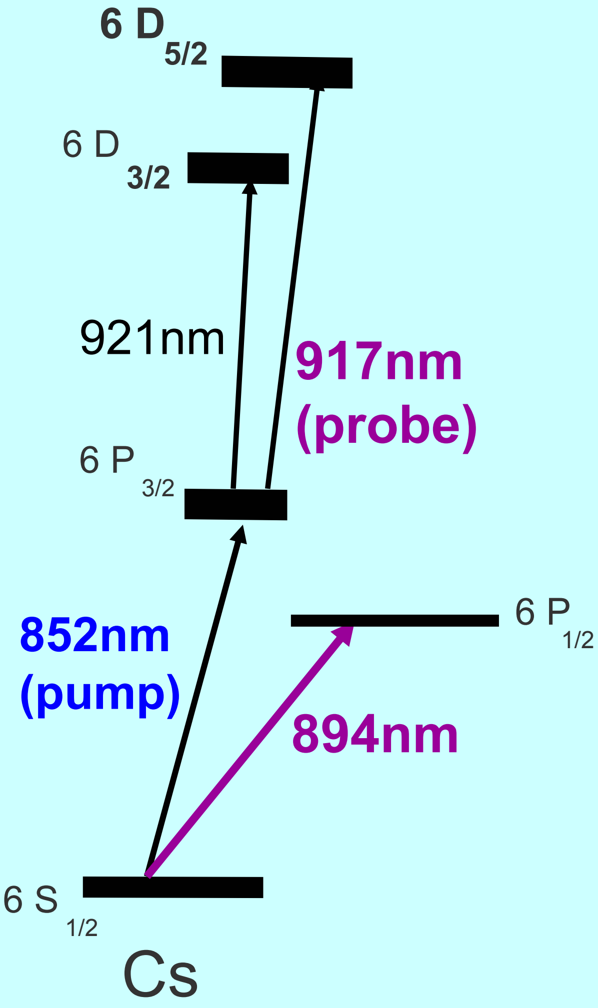
\+ *Fabry – Perot effects on the incident light*

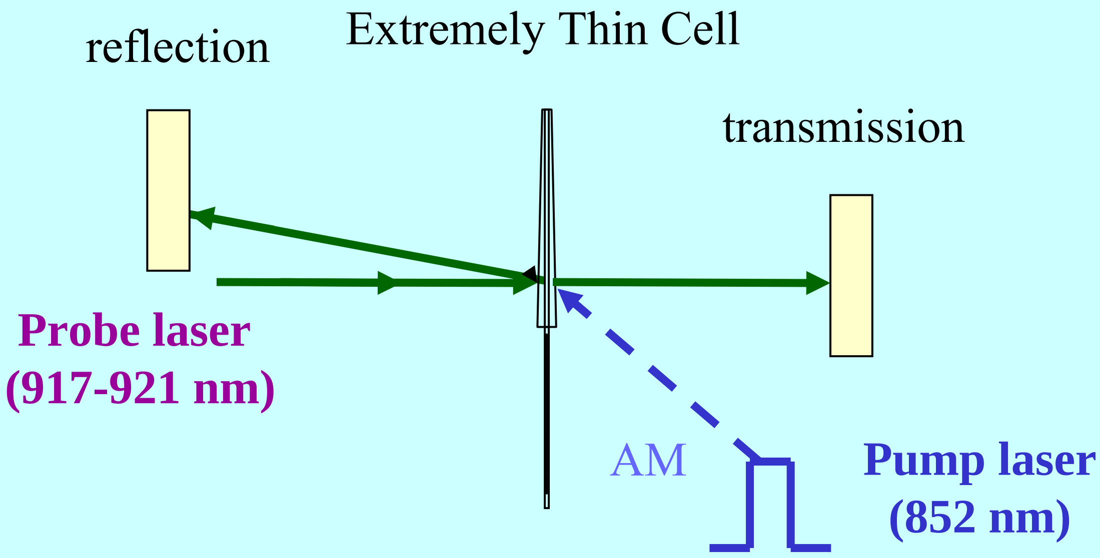
\+ **Pressure effects (**broadening, shift)

Transmission and Reflection spectra:

2 linearly **independent combinations** of absorptive and dispersive properties

Experimental Principle

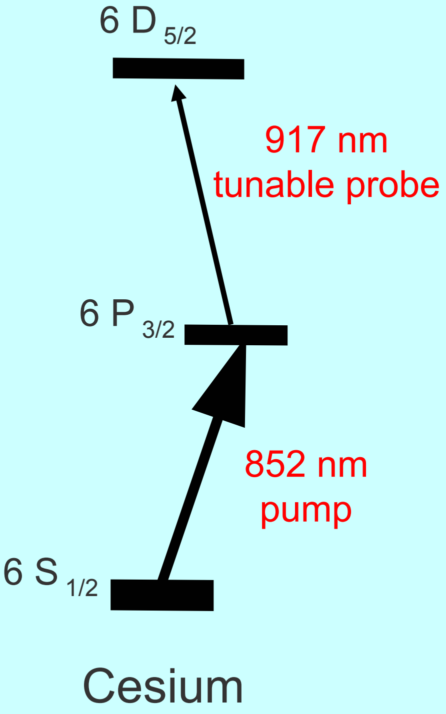




YAG/Cs thin cell transmission and reflection

strong red shift

L=50 nm
λ=917 nm
T=220°C,



6 $D_{5/2}$

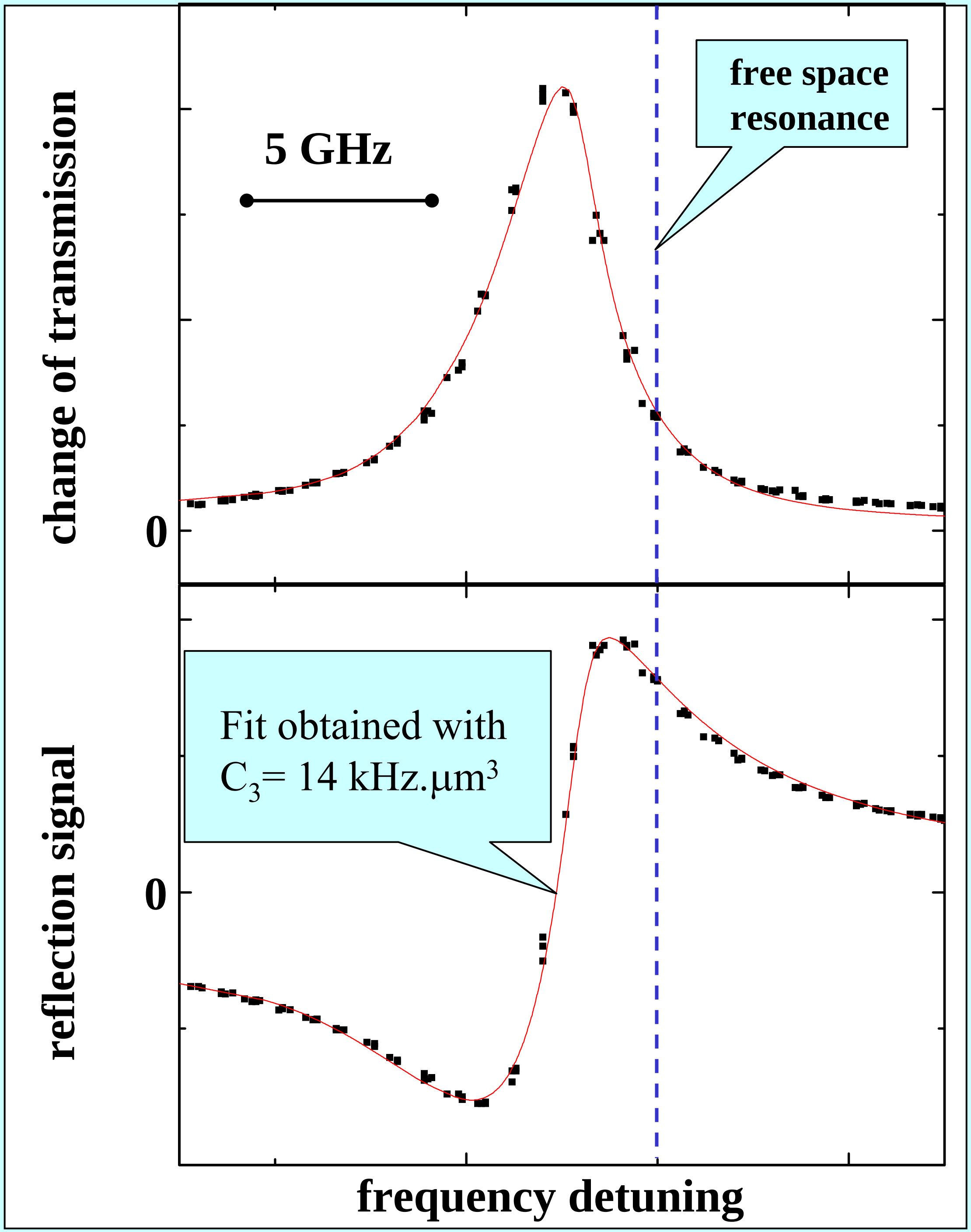
917 nm
tunable probe

6 $P_{3/2}$

852 nm
pump

6 $S_{1/2}$

Cesium

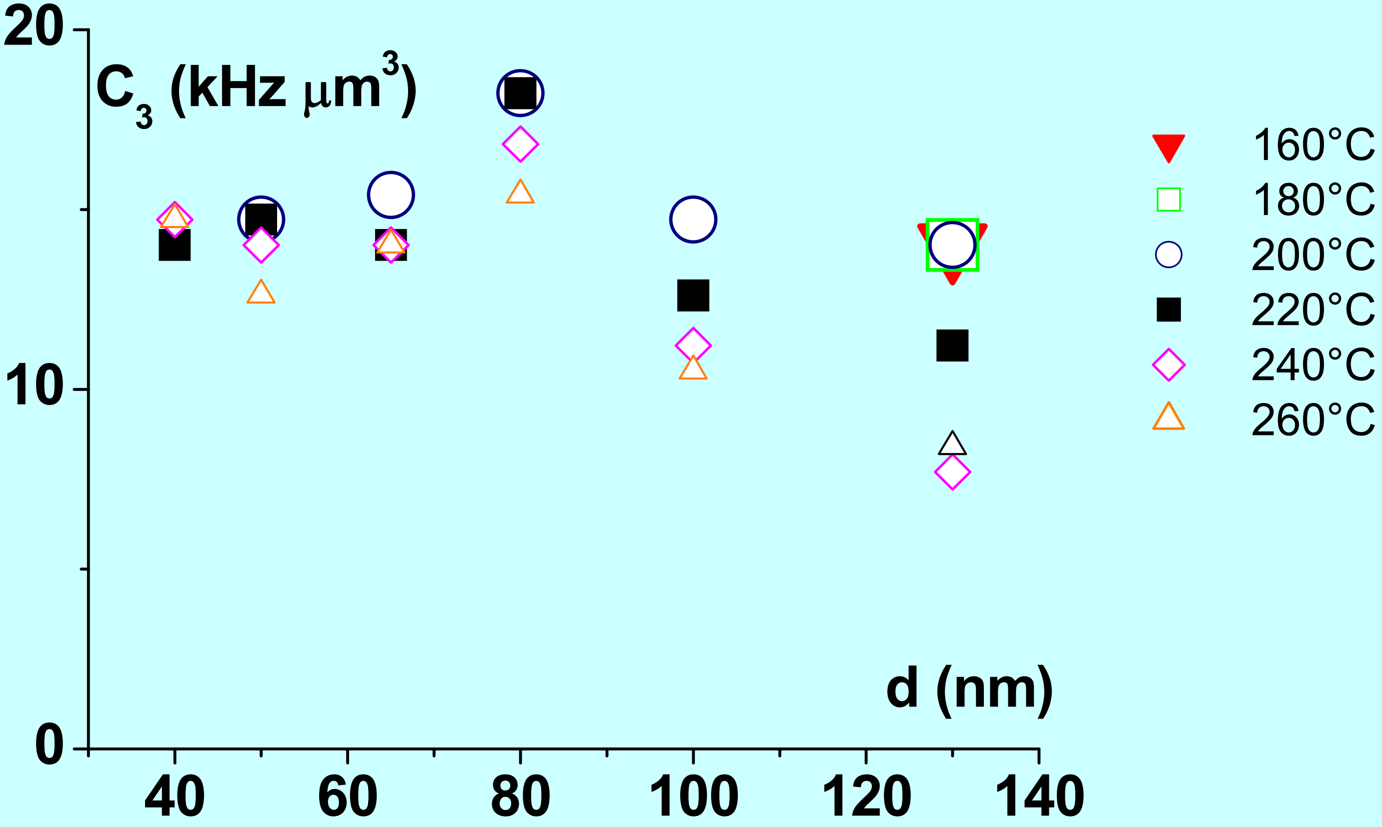


free space resonance

Fit obtained with C_3= 14 kHz.µm^3

YAG/Cs cell ; $6P_{3/2}$-$6D_{5/2}$ experiment

$\lambda = 917$ nm



$\nu = \nu_0 - C_3 / z^3$

C_3 is found to be independent of thickness

M. Fichet *et al*, Europhysics Letters <u>**77**</u>, 54001 (2007)

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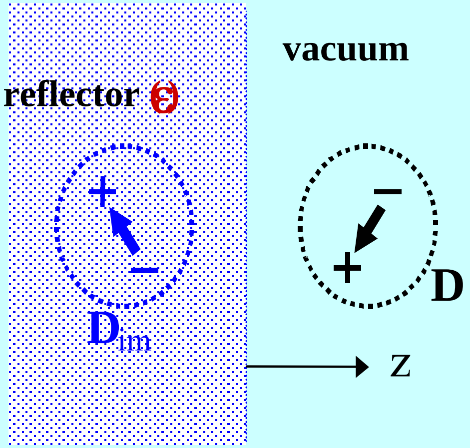
ATOM-SURFACE INTERACTION : *dielectric interface*

The model of Electrostatic images

vW potential : an interaction between **dipole** *and* **dipole-image**

$$H_{vw} = - \frac{\varepsilon - 1}{\varepsilon + 1} \frac{D^2 + D_z^2}{16 z^3}$$



A summing over virtual transitions ω_{ij}

$$\langle i|D^2|i\rangle = \sum_j r(\omega_{ij})\langle i|D|j\rangle \langle j|D|i\rangle$$

For $\varepsilon(\omega_{ij})$ complex (*i.e.* absorption)

How the image coefficient r behaves ?

REFLECTION COEFFICIENT for DISPERSIVE DIELECTRICS

How (ε-1)/ (ε+1) is transformed ?

Virtual absorption

$\omega_0 \geq 0$

|g> Mac Lachlan or
Mavroyanis 1963

$$r(\omega_0)=\frac{2}{\pi}\int_0^\infty \frac{\varepsilon(iu)-1}{\varepsilon(iu)+1}\,\frac{\omega_0}{\omega_0^2+u^2}\,du$$

$0 \leq r \leq 1$

Virtual emission

$\omega_0 \leq 0$

(*concerns only*
***excited** atom*)

Wylie & Sipe 1984-85,
Fichet *et al* 1995

$$r(-|\omega_0|)=-\frac{2}{\pi}\int_0^\infty \frac{\varepsilon(iu)-1}{\varepsilon(iu)+1}\,\frac{|\omega_0|}{\omega_0^2+u^2}\,du$$

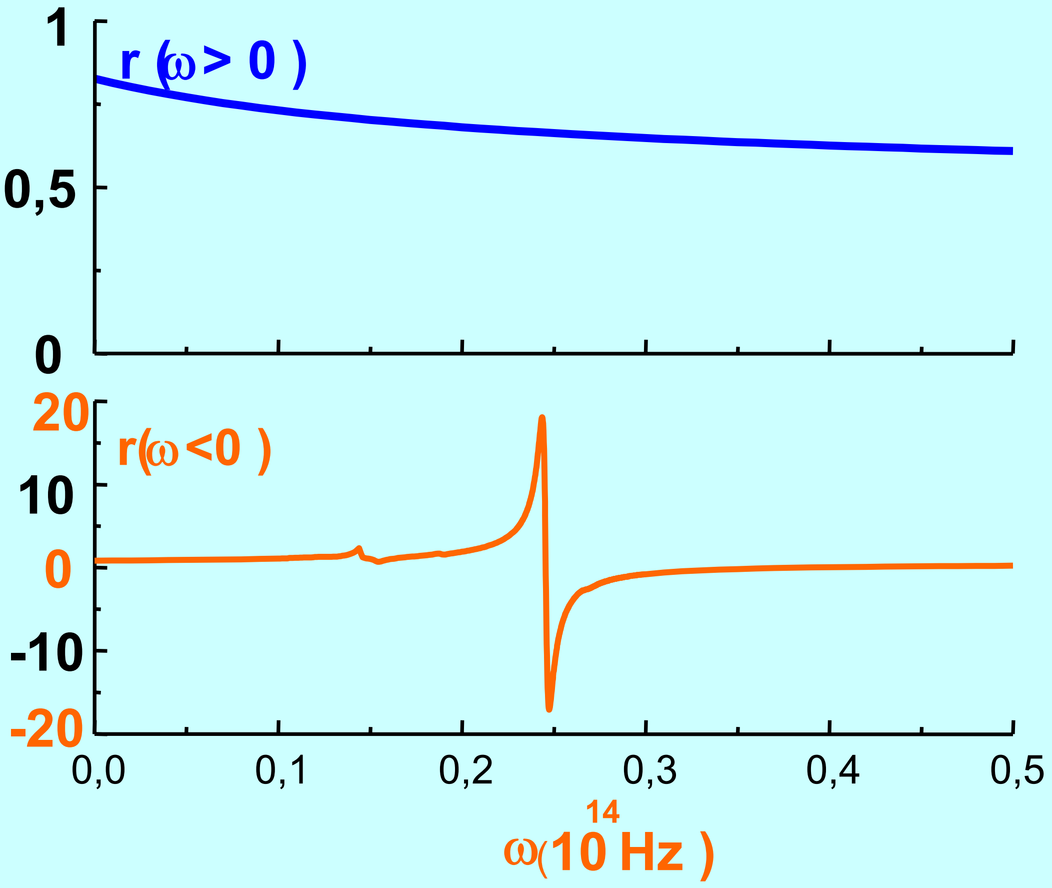
$$+2\,\mathrm{Re}\frac{\varepsilon(|\omega_0|)-1}{\varepsilon(|\omega_0|)+1},$$

r not bounded, r ≥0 or r ≤ 0

pole of [ε(ω)+1] → resonant coupling with surface mode (polariton)

THE DIELECTRIC IMAGE COEFFICIENT

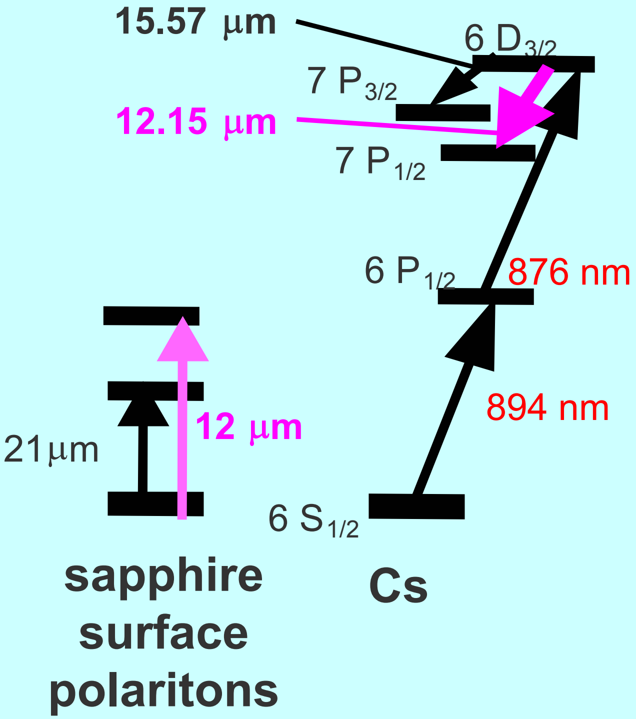
The case of **SAPPHIRE**



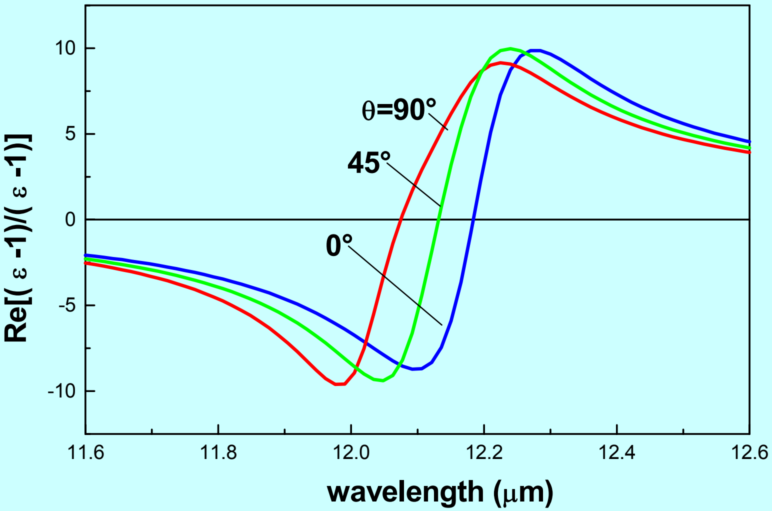
Virtual ABSORPTION of the atom (always **non resonant – at zero T**)

Virtual EMISSION of the atom : possibility of a **resonant COUPLING** to an **ABSORPTION in a SURFACE POLARITON** MODE

REPULSIVE vW INTERACTION



SAPPHIRE surface response



birefringence axis: normal to the interface

off- normal to the interfac

parallel to the interface

Observation by ***selective reflection*** (*i.e.* 1-wall coupling; probing at ~λ/2π)

H. Failache *et al,* Phys. Rev. Lett. **83**, 5647 (1999) ; Eur. Phys. J **D 23**, 237 (2003)
M. P. Gorza *et al,* Eur. Phys. J **D 15**, 113 (2001)

van der Waals energy shift at non-zero temperature

$$\delta E_a = -\frac{1}{12z^3}\sum_n \left|< a\left|D\right|n >\right|^2 \quad (\mathbf{r}_1{}^{\mathbf{an}} + \mathbf{r}_2{}^{\mathbf{an}} + \mathbf{r}_3{}^{\mathbf{an}})$$

$$r_1(\omega_{na}, T) = -2\mathrm{Re}\left[\frac{\varepsilon(\omega_{na})-1}{\varepsilon(\omega_{na})+1}\right]\frac{e^{-\frac{\hbar\omega_{na}}{K_B T}}}{1-e^{-\frac{\hbar\omega_{na}}{K_B T}}}$$

virtual absorption contribution

null for **T=0**

$$r_2(\omega_{an}, T) = +2\mathrm{Re}\left[\frac{\varepsilon(\omega_{an})-1}{\varepsilon(\omega_{an})+1}\right]\frac{1}{1-e^{-\frac{\hbar\omega_{an}}{K_B T}}}$$

virtual emission contribution
increases with T

[stimulated emission ~<N>]

$$r_3(\omega_{na}, T) = +\frac{4k_B T}{\hbar}\sum_p^{\infty}{}' \frac{\varepsilon(i\xi_p)-1}{\varepsilon(i\xi_p)+1}\frac{\omega_{na}}{\omega_{na}^2+\xi_p^2}$$

non-resonant contribution

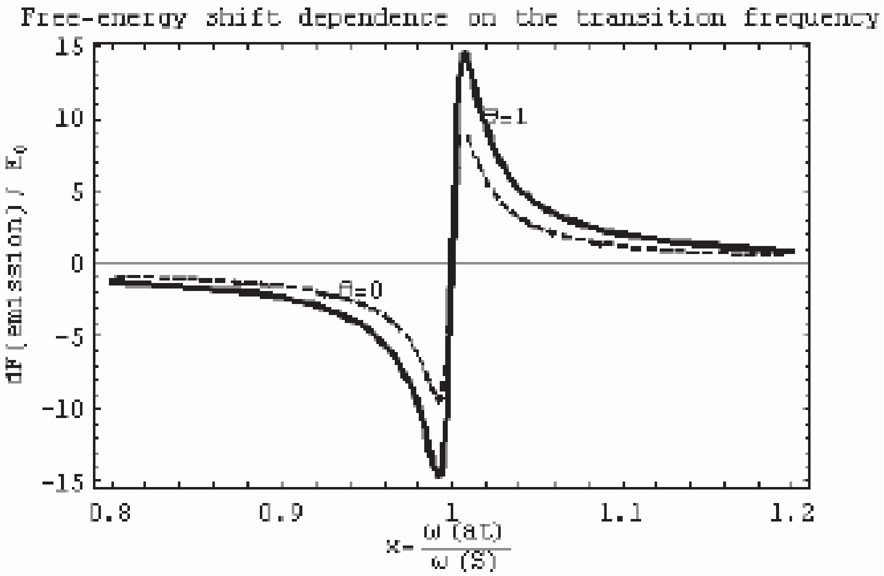
(known since Mac Lachlan 1963)

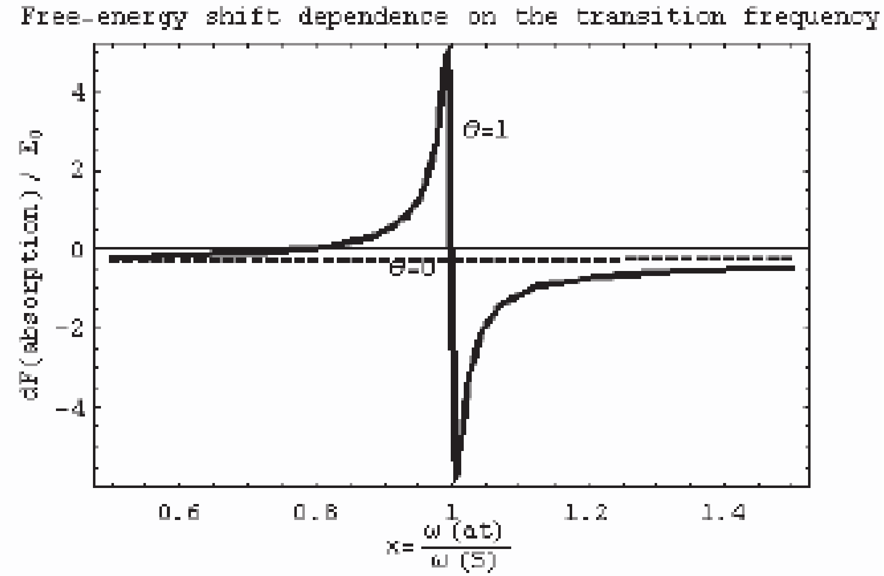
with $\xi_p = 2\pi k_B T/\hbar$

← **Matsubara frequency**

M-P Gorza & M Ducloy, Eur. Phys. J. D ***40****, 343 (2006)*

$$\theta = k_B T / \hbar\omega_S$$





Influence of surface thermal excitations

M-P Gorza & M Ducloy, Eur. Phys. J. D ***40****, 343 (2006)*

SURFACE RESONANCES in ATOM - STRUCTURE INTERACTION

1/ Dielectric microstructure

- High-Q morphology dependent resonances (MDR):

Ex.: Mie resonances, or Whispering Gallery Modes (WGM) of microspheres

Braginsky *et al* 1987
Haroche *et al.* 1992 …

- Influence on atom-surface interaction :
  Vacuum Rabi splitting

Klimov *et al,* J. Mod. Opt. 1997

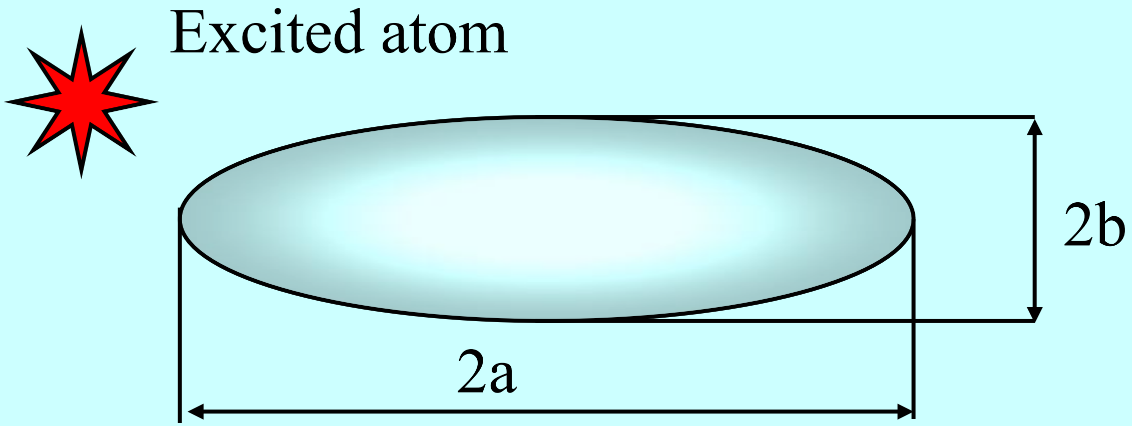
For nano-bodies, no propagation ⇒ no resonances

2/ Dispersive dielectric / metal ⇒ **surface polariton**

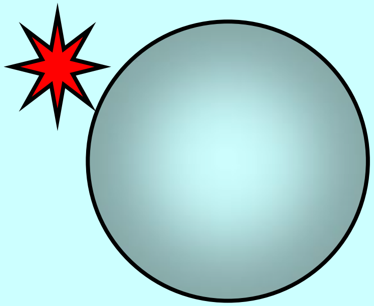
Polariton frequency tuning *via* :

- Material birefringence (*c*-axis orientation)
- Surface temperature
- Form factor in surface response:
  - * plane : $(\varepsilon-1)/(\varepsilon+1)$
  - * nanosphere : $(\varepsilon-1)/(\varepsilon+2)$
  - * nano-ellipsoid…

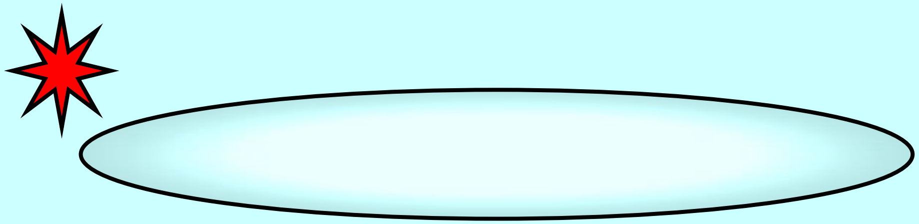
SPONTANEOUS EMISSION OF AN ATOM PLACED NEAR A NANOSPHEROID



Spherical case (b = a)



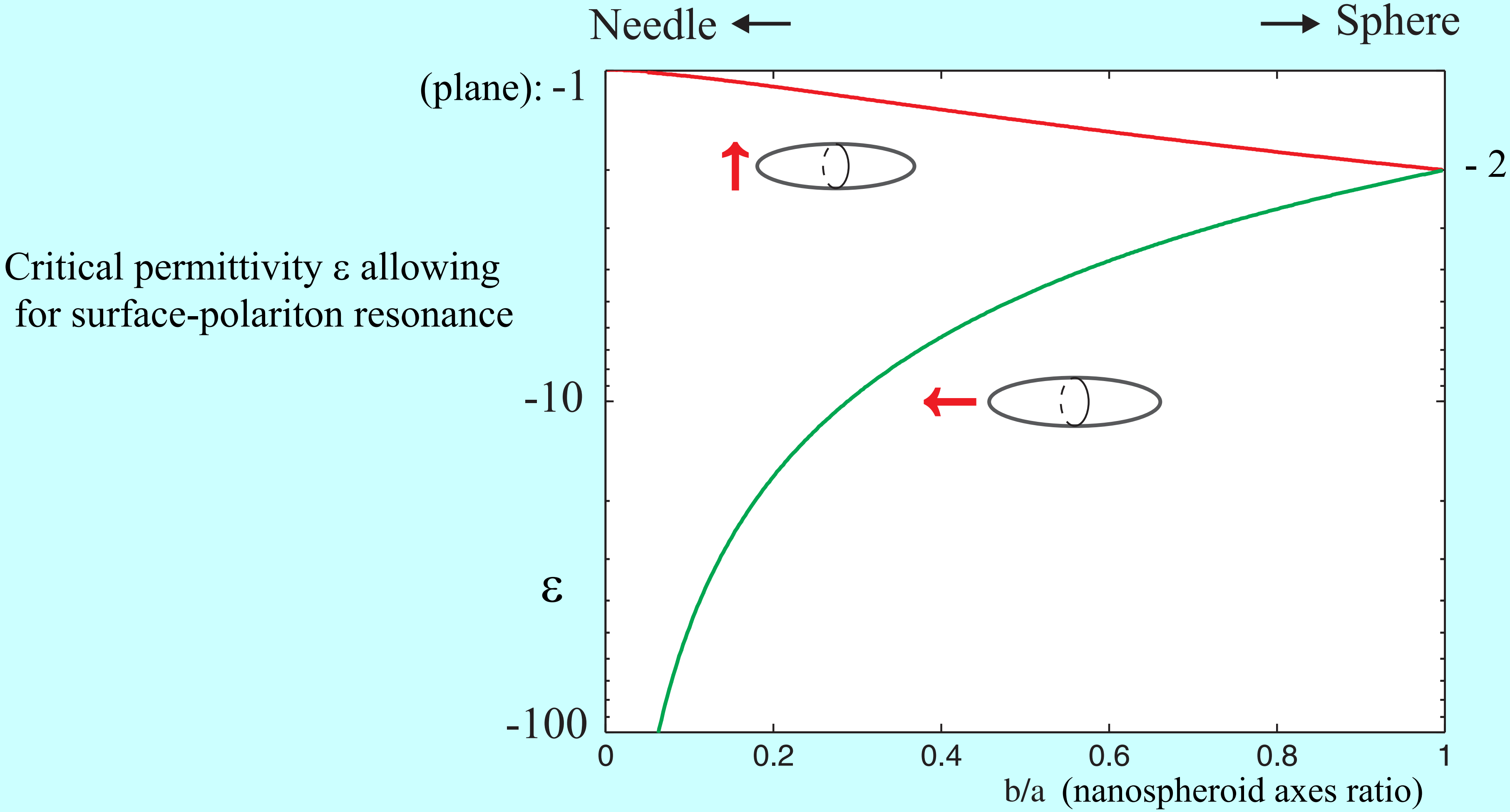
Needle or cylindrical case (b/a →0)



Eur. Phys. J., **D <u>20</u>** (2002)133

Chem. Phys. Lett., **358** (2002)192

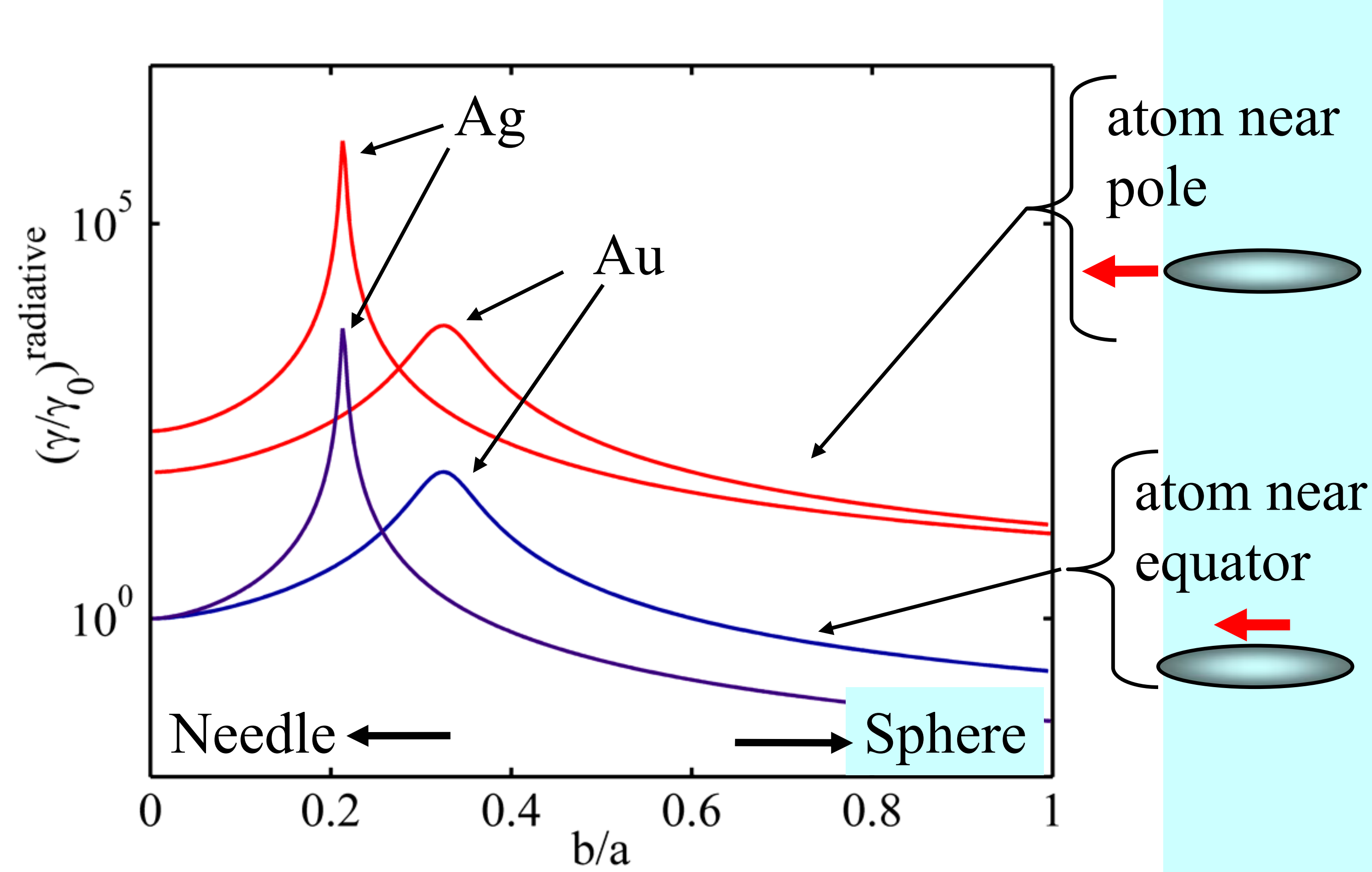
INFLUENCE OF NANOBODY SHAPE ON SURFACE MODES



Plasmon resonance conditions for different orientations of dipole momentum

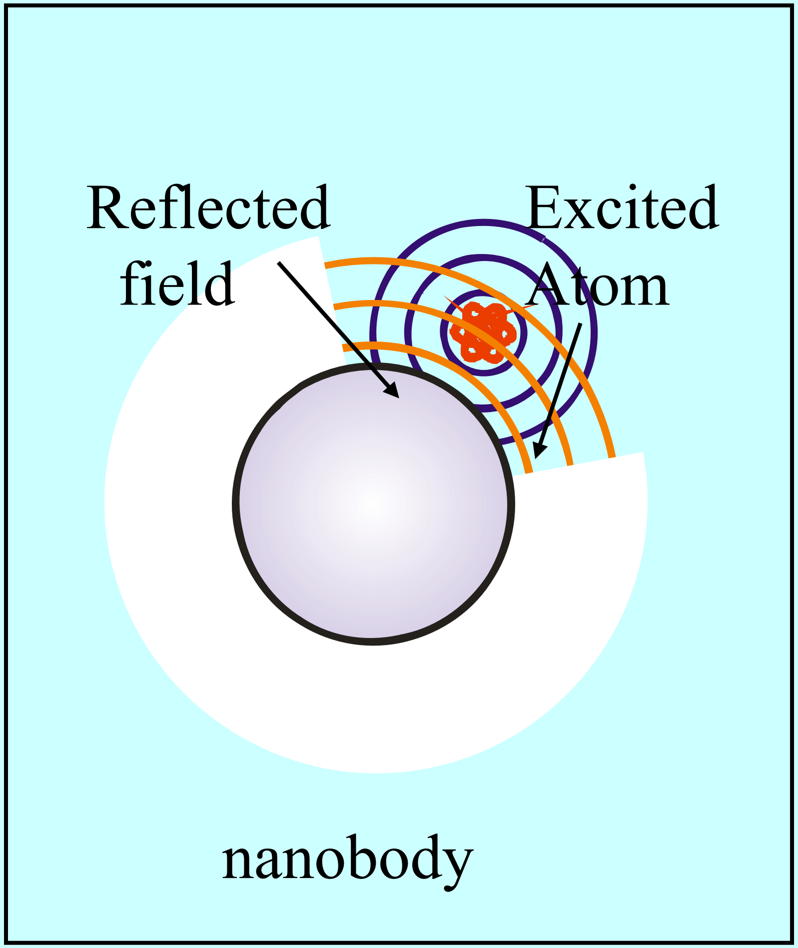
The dependence of decay rates on ratio b/a near nanospheroids made of gold (ε=-8.37+i 1.16, λ=600 nm) **or silver (** ε=-15.37+i0.231, λ=632.8 nm).

Dipole is parallel to nanospheroid axis



Klimov *et al*, Eur. Phys. J. D **20**, 133 (2002)

QUALITATIVE PICTURE OF NANOBODY INFLUENCE ON DIPOLE/QUADRUPOLE EMISSION



$$\frac{\gamma^{DIPOLE}}{\gamma^{QUADR}} \propto \left(\frac{a_{nano}}{a_0}\right)^2$$

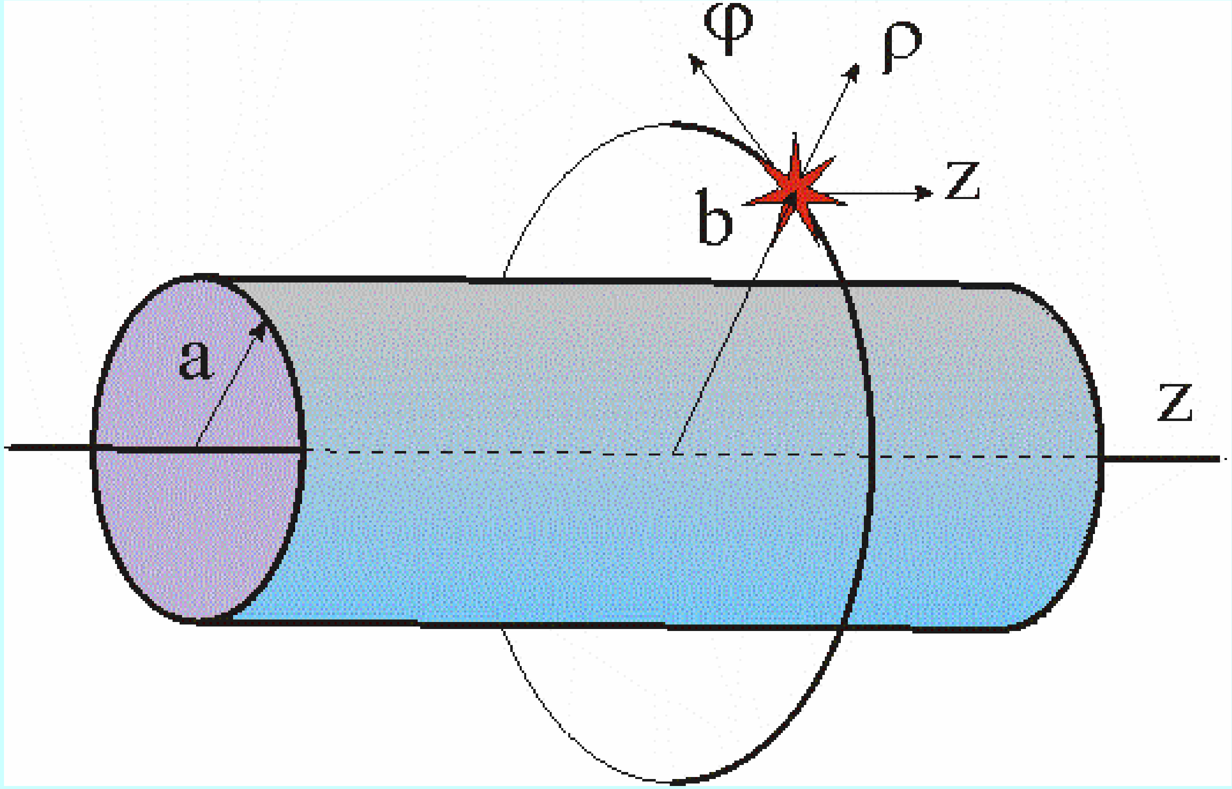
Near nano-objects

a_0 - Bohr radius
λ - radiation wavelength
a_{nano}- characteristic size of nano-object

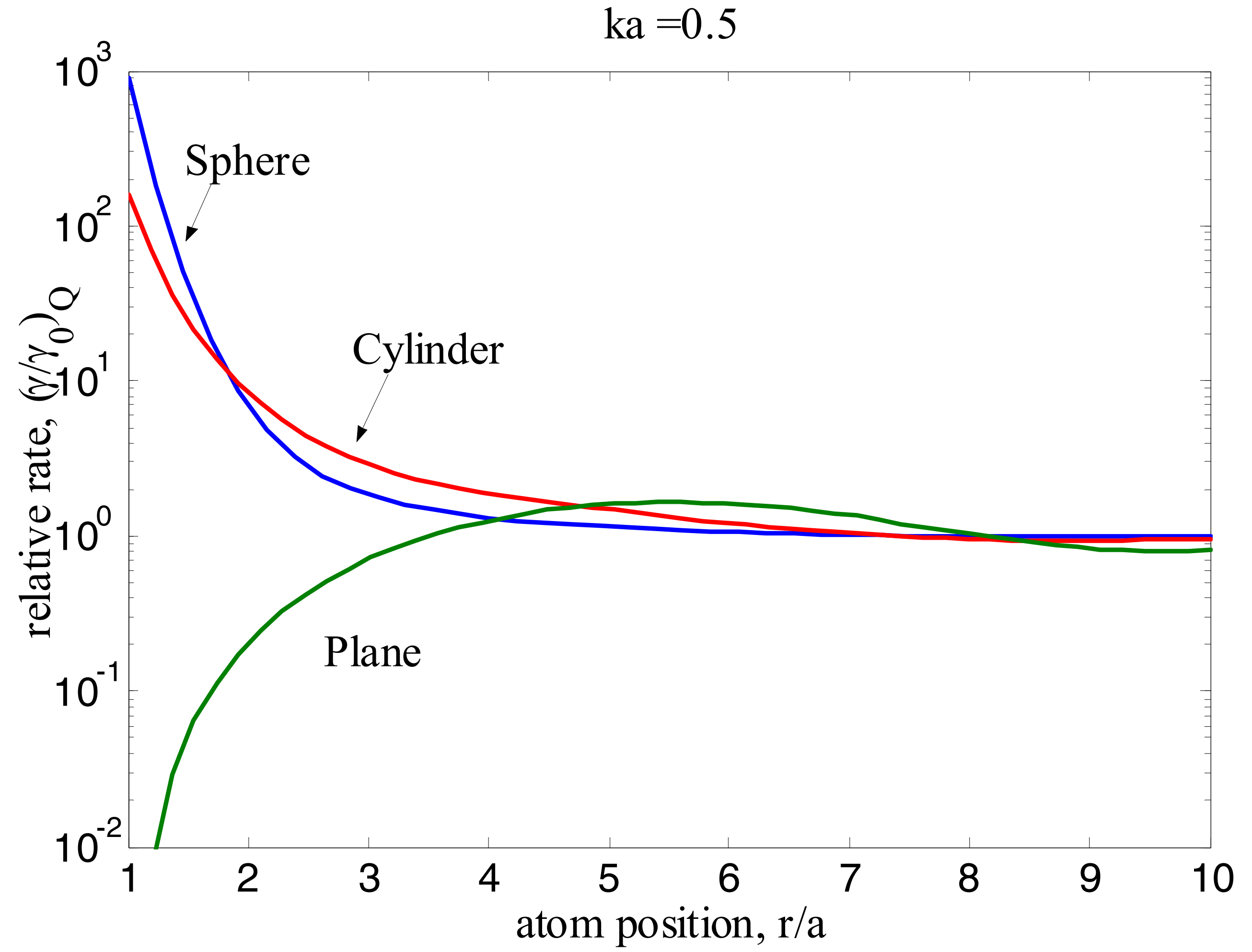
in free space

$$\frac{\gamma^{DIPOLE}}{\gamma^{QUADR}} \propto \left(\frac{\lambda}{a_0}\right)^2$$

Quadrupole transition rates near nanowire



Quadrupole decay rates for different geometries



Klimov, Ducloy, Phys. Rev. A **62**, 043818 (2000)

Conclusion

- Substantial enhancement of quadrupole transitions near nano-bodies with large surface curvature
- Their sensitivity to field gradients allows to measure nano-optical fields
- Extension of “quadrupole detection” to focused Laguerre-Gauss beams (“spiral beams”) paves the way to new approaches in Quantum Optics (work submitted for publication; see the coming talk by V. V. Klimov, Lecture Hall 2)

General conclusion

Interest of nanometer-thin gas cells for investigating:

- Quantum Electro-Dynamics in dielectric Fabry Perot nano-cavity
- Nanophysics with "*free*" atoms; atom-surface interaction probed in 20-200nm range
- Novel regime of atom dynamics and atom-light interactions (*Dicke regime*)

PROSPECTS

- Modification of atomic fluorescence, branching ratios; non-radiative decays…
- Nano-structured walls for higher-D confinement
- Nano-cavity QED in the non-zero temperature limit
- Atom-atom interaction in confined space