Roberto Tricarico

Tutor: Prof. Carlo Forestiere

XXXII Cycle - III year presentation

Light-Matter Interaction in Open Systems: from Nanoparticles to Atoms

Università degli Studi di Napoli Federico II

General Information

- Bachelor's degree in electronic engineering, Università Federico II (2015)
- Master's degree in electronic engineering, Università Federico II (2016)
- ITEE: athenaeum fellowship, Prof. Carlo Forestiere

Year	Courses	Seminars	Research
1°	30	9.8	21
2°	16	9.6	34.4
3°	6	0	53.2

- **7** courses, **1** language course, **6** PhD school, **12** seminars
- **6** journal papers (+1 submitted), **2** conference papers, **2** book chapters
- **Period abroad** Third year of the PhD at the Institute of Photonic Sciences in Castelldefels (Barcelona) **ICFO**, supervised by Prof. Dr. **Darrick Chang**.
- Thesis co-supervisor : Prof. Dr. Darrick Chang.

Course list (8)

- Ottica Quantistica (1st year)
- Meccanica Quantistica dei Molti Corpi (1st year)
- Teoria dei Gruppi e Applicazioni (1st year)
- Introduction to Quantum Electrodynamics (1st year)
- Fisica dello Stato Solido 2 (2nd year)
- Plasmonics and Metamaterials (2nd year)
- Elettromagnetismo e Relatività (2nd year)
- Spanish course (A1) (3rd year)

• Estimated • Achieved

	Credits year 1								Credits year 2								Credits year 3									
		1	2	3	4	5	6			1	2	3	4	5	6			1	2	3	4	5	6			TOTAL
Modules	30	8	3	11	0	0	8	30	20	0	8	4	0	0	4	16	0	3	0	3	0	0	0	6	30-70	52
Seminars	10	0.4	0	0.4	6	3	0	9.8	10	0	2.3	3.5	0	3.8	0	9.6	0	0	0	0	0	0	0	0	10-30	19.4
Research	20	3	5	3	3	6	1	21	30	8	3	3	8	7.4	5	34.4	60	7	10	6.2	10	10	10	53.2	80-140	108.6
	60	11	8	14	9	9	9	60.8	60	8	13.3	10.5	8	11.2	9	60	60	10	10	9.2	10	10	10	59.2	180	180

PhD school list (6)

- Antenna Synthesis, Napoli, (1st year)
- Ferdinando Gasparini XXI edizione, Napoli, (1st year)
- XLII Scuola estiva di Fisica Matematica, Ravello, (1st year)
- International School of Plasmonics and Nano-Optics, Cetraro, (2nd year)
- Ferdinando Gasparini XXII edizione, Napoli, (2nd year)
- Light Matter interaction in Dilute Media and Individual Quantum Systems, Les Houches, (3rd year)

• Estimated • Achieved

	Credits year 1								Credits year 2								Credits year 3									
		1	2	3	4	5	6			1	2	3	4	5	6			1	2	3	4	5	6			TOTAL
Modules	30	8	3	11	0	0	8	30	20	0	8	4	0	0	4	16	0	3	0	3	0	0	0	6	30-70	52
Seminars	10	0.4	0	0.4	6	3	0	9.8	10	0	2.3	3.5	0	3.8	0	9.6	0	0	0	0	0	0	0	0	10-30	19.4
Research	20	3	5	3	3	6	1	21	30	8	3	3	8	7.4	5	34.4	60	7	10	6.2	10	10	10	53.2	80-140	108.6
	60	11	8	14	9	9	9	60.8	60	8	13.3	10.5	8	11.2	9	60	60	10	10	9.2	10	10	10	59.2	180	180

Seminar list (12)

- Electromechanical Consequences of Violent Instabilities in Tokamaks, Vladimir D. Pustovitov
- Smart Nanodevices for Theranostics, Ilaria Rea
- Magnetic Refrigeration: Thermodynamics of novel magnetic materials for an efficient cooling technique, Vittorio Basso
- Thermodynamics in spintronics: the spin Seebeck and spin Peltier effects, Vittorio Basso
- Approssimazioni di problemi alle derivate parziali e applicazioni, Alfio Quarteroni
- Tailoring Waves at the extreme with metamaterials, Nader Engheta
- Near-zero-index photonics, Nader Engheta
- IBM Q: building the first universal quantum computers for business and science, Federico Mattei and Najla Said
- How to Produce a scientific paper, Aliaksandr Birokou and Elisa Magistrelli
- Tomografia e Imaging: Principi, Algoritmi e Metodi Numerici, Pasquale Memmolo
- The power of Trefftz Approximations: Applications in Electromagnetics, Igor Tsukerman
- Non-Asymptotic and Nonlocal Homogenization of Periodic Electromagnetic Structures , Igor Tsukerman

- Estimated
- Achieved

	Credits year 1								Credits year 2								Credits year 3									
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	60	11	8	14	9	9	9	60.8	60	8	13.3	10.5	8	11.2	9	60	60	10	10	9.2	10	10	10	59.2	180	180

Publication list

Journal papers (6)

- C. Forestiere, G. Miano, G. Rubinacci, A. Tamburrino, R. Tricarico, S. Ventre, “Material-Independent Modes of Arbitrarily Shaped Homogeneous Scatterers”, **IEEE Transactions on Antennas and Propagation**, Mar. 2018
- C. Forestiere, G. Miano, M. Pascale, R. Tricarico, “Directional scattering cancellation for an electrically large dielectric sphere”, **Optics Letters**, Apr. 2019
- C. Forestiere, G. Miano, M. Pascale, R. Tricarico, “Electromagnetic modes and resonances of two-dimensional bodies”, **Phys. Rev. B**, Apr. 2019
- C. Forestiere, G. Miano, M. Pascale and R. Tricarico, “Electromagnetic Scattering Resonances of Quasi-1-D Nanoribbons”, **IEEE Transactions on Antennas and Propagation**, Aug. 2019
- M. Pascale, G. Miano, R. Tricarico, and C. Forestiere, “Full-wave electromagnetic modes and hybridization in nanoparticle dimers”, **Scientific Reports**, Oct. 2019
- C. Forestiere, G. Miano, M. Pascale, G. Rubinacci, A. Tamburrino, R. Tricarico, and S. Ventre, "Magnetoquasistatic Resonances of Small Dielectric Objects.” Accepted by **Physical Review Research**

Publication list

Conference papers (2)

- M. Pascale, R. Tricarico, G. Miano and C. Forestiere, Full-wave mode hybridization in nanoparticle dimers, 2019 International Conference on Electromagnetics in Advanced Applications (**ICEAA 19**), Granada, Spain, 2019.
- R. Tricarico, C. Forestiere, G. Miano and M. Pascale, Field Quantization in Arbitrarily-Shaped Metal Nanoparticles, International Conference on Electromagnetics in Advanced Applications (**ICEAA 19**), Granada, Spain, 2019.

Book Chapters (2)

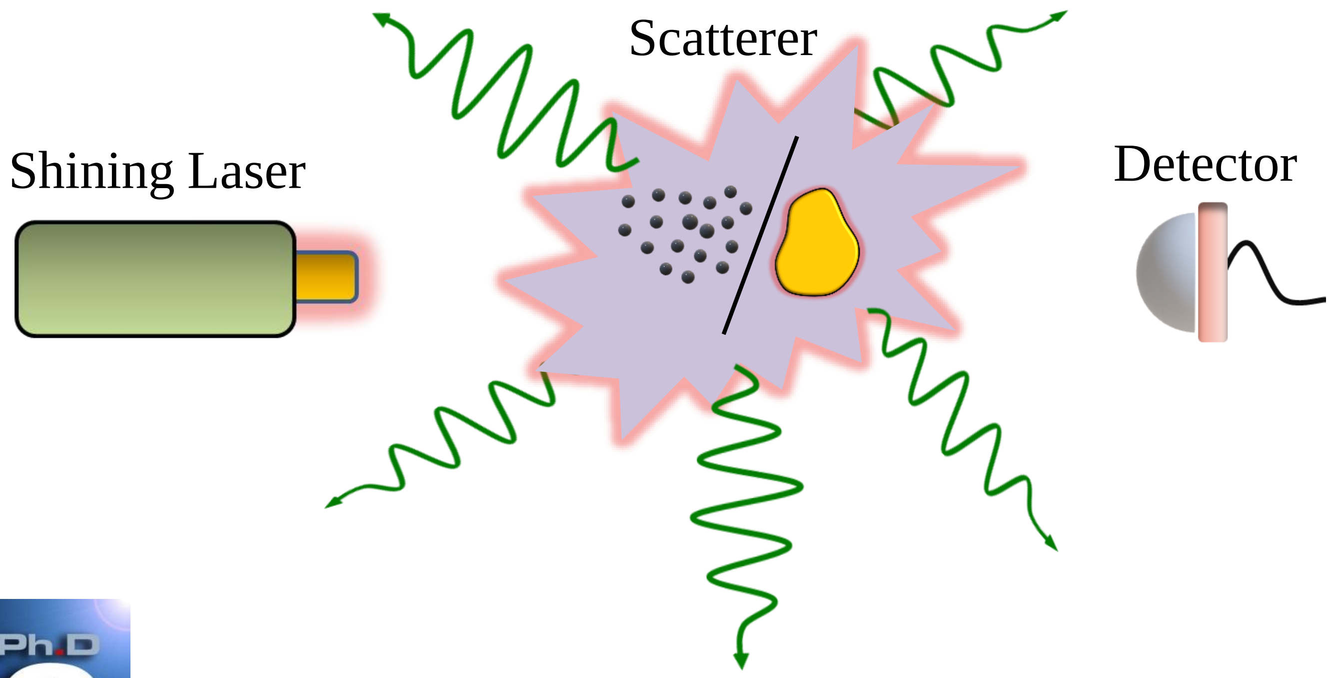
- C. Forestiere, G. Miano, M. Pascale, R. Tricarico, chapter title: “A full-retarded spectral technique for the Fano-resonance analysis in a dielectric nanosphere”, **Springer** book: “Fano Resonances in Optics and Microwaves”, pp. 185-218, Nov. 2018.
- C. Forestiere, G. Miano, M. Pascale, R. Tricarico, chapter title: “Material Independent Modes for the design of electromagnetic scattering”, **World Scientific Publishing** book: “Compendium on Electromagnetic Analysis – from electrostatics to photonics: fundamentals and applications for physicists and engineers”, out in Apr. 2020.

Submitted paper (1)

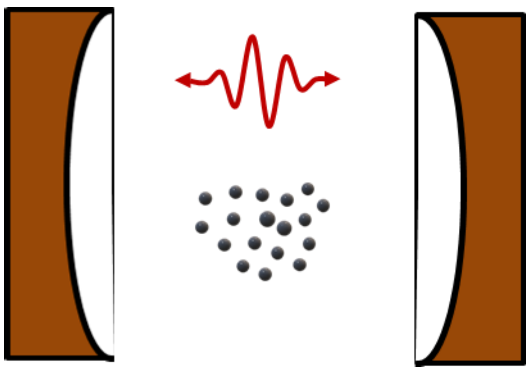
- C. Forestiere, G. Miano, M. Pascale and R. Tricarico, “Quantum theory of Radiative Decay Rate and Frequency Shift of Surface Plasmon Modes in Arbitrarily Shaped Nanoparticles”, **arXiv**, Jan. 2020.

Light-Matter in Open Systems

The study of the interaction between light and matter at the nano/atomic scale presents interesting challenges and new perspective in the case of **open systems**, i.e. systems which **lose energy** (radiation to infinity).

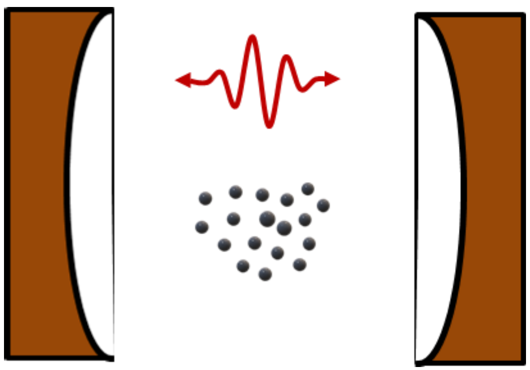


Open Systems: Atoms

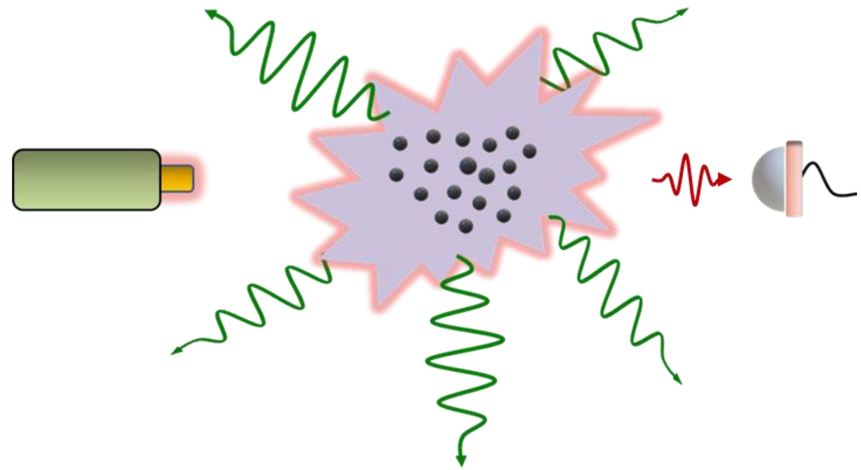


Cavity-QED: the atoms are enclosed in an optical cavity (*closed system*)

Open Systems: Atoms

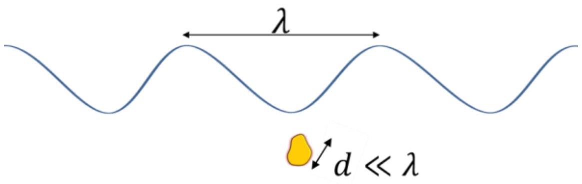


Cavity-QED: the atoms are enclosed in an optical cavity (*closed system*)



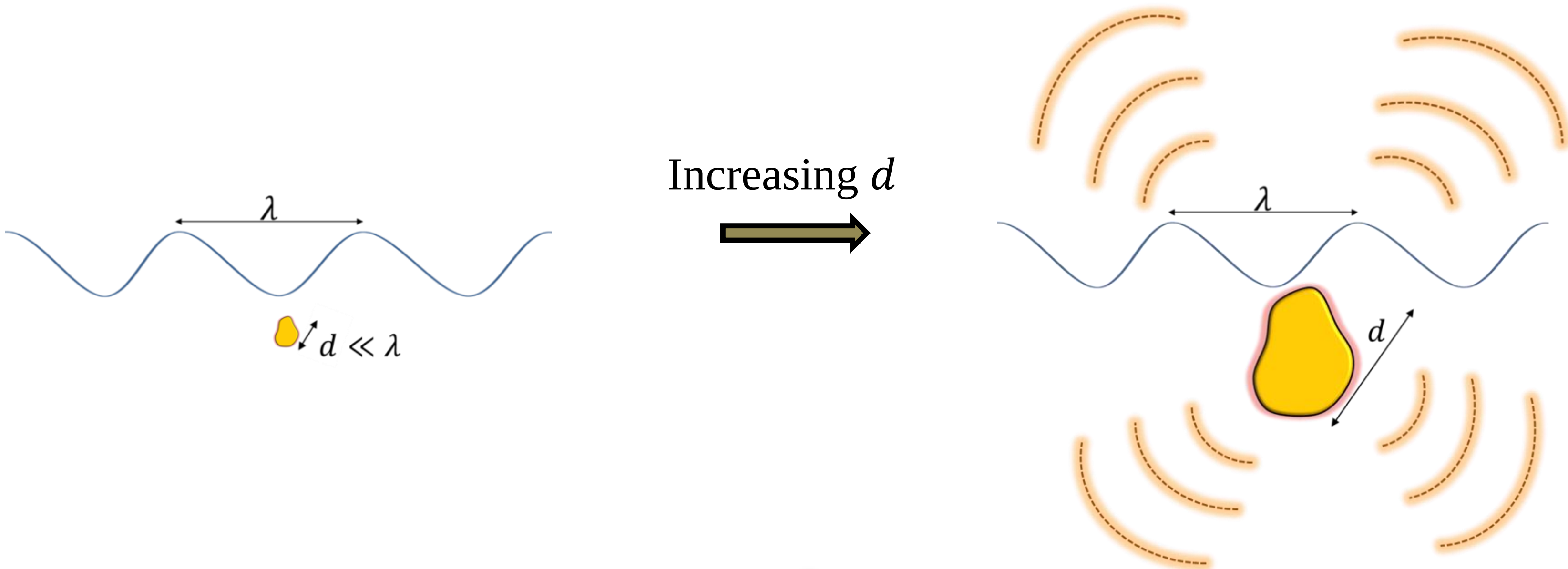
Free space QED: the atoms are in free space and scatter *uncollectable light* in 4π

Open Systems: Nanoparticles



Quasistatic limit: $d \ll \lambda$ retarded
NO RADIATION OF ENERGY TO INFINITY (*closed system*)

Open Systems: Nanoparticles



Quasistatic limit: $d \ll \lambda$ retarded
NO RADIATION OF ENERGY TO INFINITY (*closed system*)

Full-retarded limit: $d \sim \lambda$
RADIATION OF ENERGY TO AT INFINITY (*Open System*)

Light-Matter in Open Systems

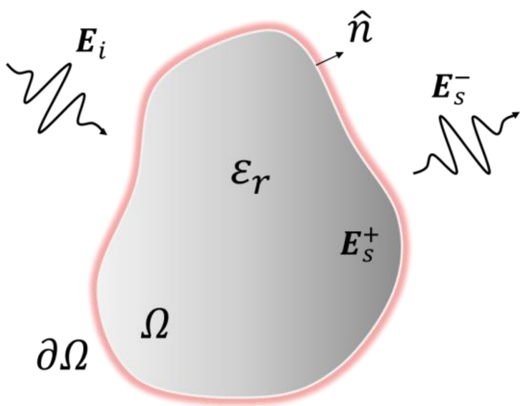
Three selected topics in Light-Matter Interaction in Open Systems

I Material Independent Modes for the Electromagnetic Scattering
NANOPARTICLES, Classical Electrodynamics

III Nonlinearities in Rydberg-EIT,
ATOMS

II Quantum theory of frequency shift and radiative decay rate in arbitrarily shaped metal nanoparticles and dimers
NANOPARTICLES, Quantum Electrodynamics

I Material Independent Modes



<u>Scattering problem</u>

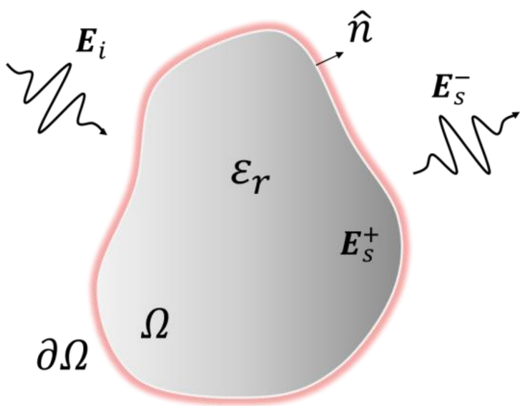
$$\nabla^2 \boldsymbol{E}_s^+ + k_0^2 \varepsilon_r(\omega) \boldsymbol{E}_s^+ = k_0^2 (1-\varepsilon_r) \boldsymbol{E}_i \text{ in } \Omega$$

+tang. comp. continuity,
and radiation

We *don't want* to solve the problem directly, but we *want* to provide a modal theory for the scattering in open systems:

- to have more insight on the physics
- for a rigorous understanding of the interference patterns (*analysis*)
- to simplify the material design of the scattered in to maximize assigned scattering features (*synthesis*)

I Material Independent Modes



Scattering problem

$$\nabla^2 \boldsymbol{E}_s^+ + k_0^2 \varepsilon_r(\omega) \boldsymbol{E}_s^+ = k_0^2 (1 - \varepsilon_r) \boldsymbol{E}_i \text{ in } \Omega$$

+tang. comp. continuity,
and radiation

Direct solution

$$x = A^{-1} b$$

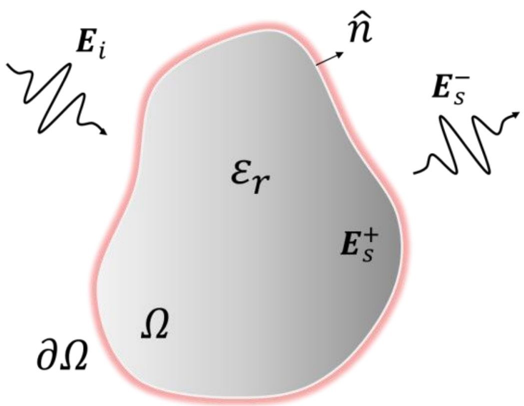
Linear algebra analogy

$$Ax = b,$$
$$A = A^+$$

Modal solution

$$A c_i = \lambda_i c_i \qquad x = \sum_i \frac{1}{\lambda_i} \frac{\langle c_i, b \rangle}{\langle c_i, c_i \rangle} c_i$$

I Material Independent Modes



Scattering problem

$$\nabla^2 \boldsymbol{E}_s^+ + k_0^2 \varepsilon_r(\omega) \boldsymbol{E}_s^+ = k_0^2 (1-\varepsilon_r) \boldsymbol{E}_i \text{ in } \Omega$$

+tang. comp. continuity,
and radiation

Auxiliary eigenvalue problem

$$-\frac{1}{k_0^2} \nabla^2 \boldsymbol{C}_r = -\gamma_r \boldsymbol{C}_r \text{ in } \Omega$$

+tang. comp. continuity,
and radiation

It is MATERIAL INDEPENDENT

Modal solution

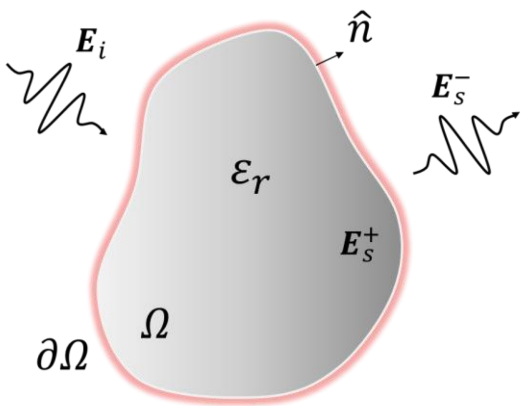
$$\boldsymbol{E}_s^+ = (1-\epsilon_r) \sum_r \frac{1}{\epsilon_r - \gamma_r} \frac{\langle \boldsymbol{C}_r^*, \boldsymbol{E}_i \rangle}{\langle \boldsymbol{C}_r^*, \boldsymbol{C}_r \rangle} \boldsymbol{C}_r$$

where $\langle \boldsymbol{A}, \boldsymbol{B} \rangle = \int_\Omega \boldsymbol{A}^* \cdot \boldsymbol{B} \, d\Omega$

Material

Geometry

I Material Independent Modes



Modal solution

$$\boldsymbol{E}_s^+ = (1-\epsilon_r)\sum_r \frac{1}{\epsilon_r-\gamma_r}\frac{\langle \boldsymbol{C}_r^*, \boldsymbol{E}_i\rangle}{\langle \boldsymbol{C}_r^*, \boldsymbol{C}_r\rangle}\boldsymbol{C}_r$$

$\boldsymbol{C}_r$ and γ_r

Depend on:

- shape of the nanoparticle
- frequency of the incident field

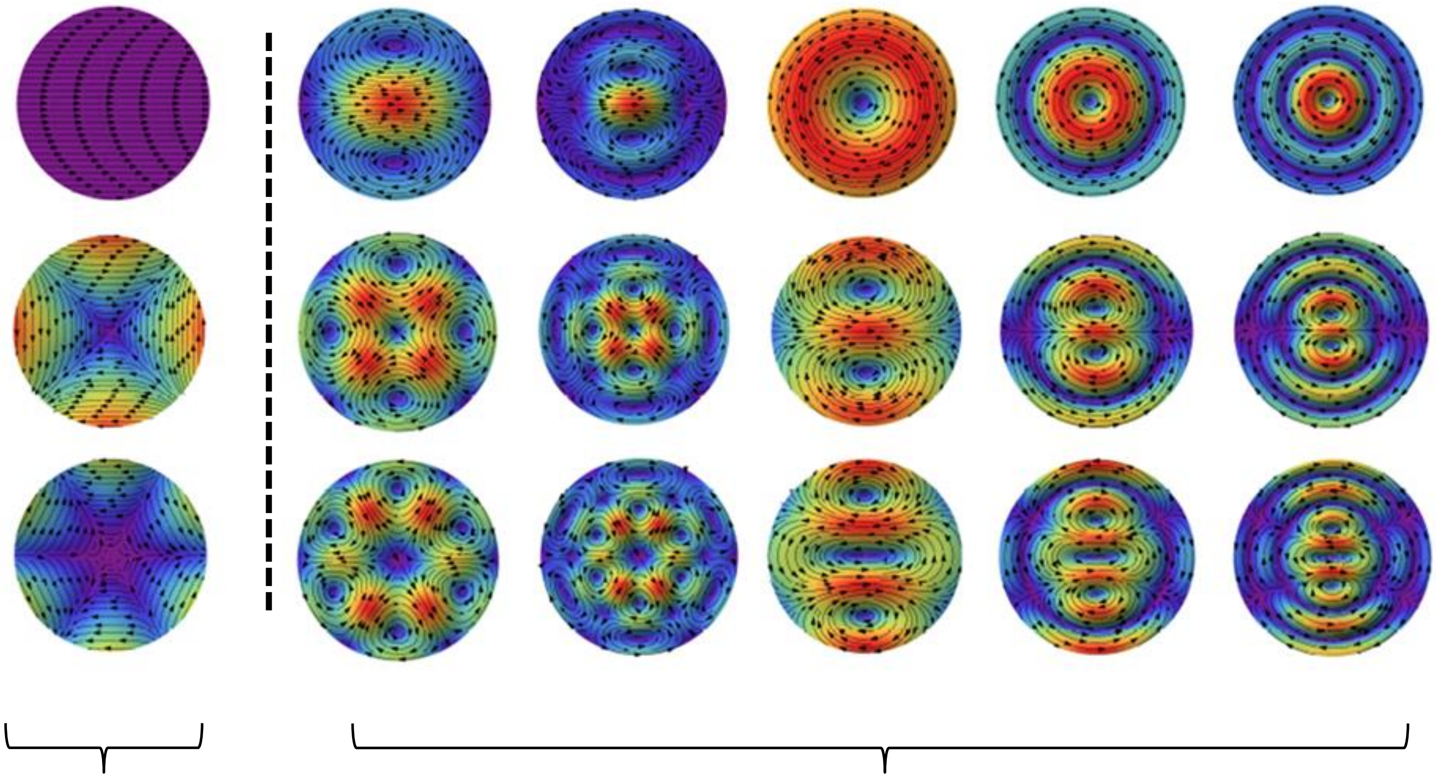
Do not depend on:

- material constituting the nanoparticle

Material Resonance → $\min|\epsilon_r - \gamma_r|$

I MIM: isolated sphere

Material Independent Modes $\boldsymbol{C}_r$



<u>Plasmonic Modes</u>

- Sources and sinks

<u>Photonic Modes</u>

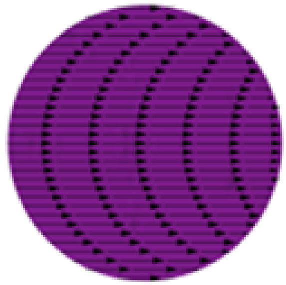
- Vortices

I MIM: isolated sphere

Eigen-permittivities

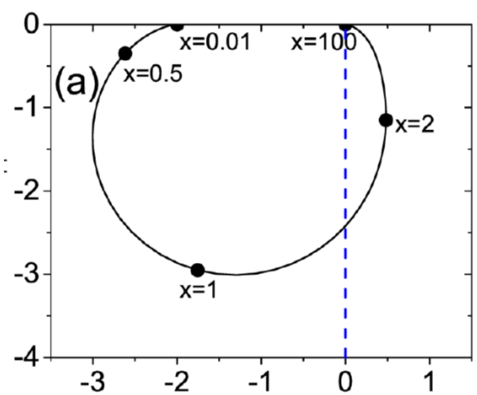
γ_r

Plasmonic Mode

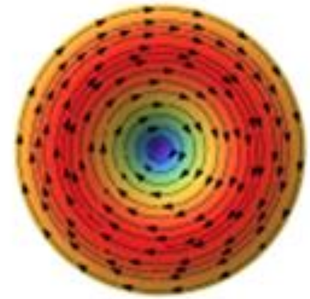


- γ in the III quadrant
- Can resonate

  in *plasmons* ($\mathrm{Re}[\varepsilon_r] < 0$)

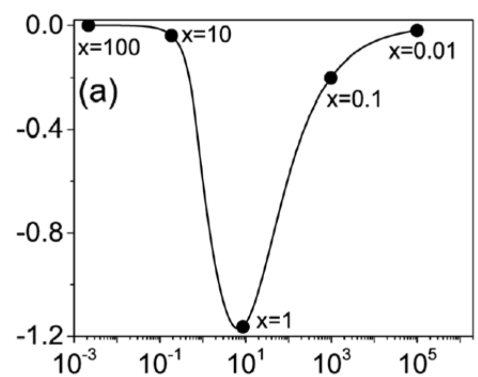


Photonic Mode



- γ in the IV quadrant
- Can resonate

  in *dielectrics* ($\mathrm{Re}[\varepsilon_r] > 0$)

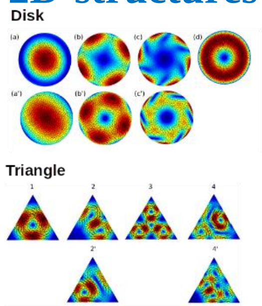


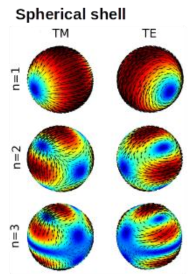
Size parameter

$$x = \frac{2\pi R}{\lambda}$$

I MIM: what done so far

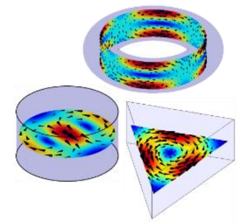
2D-structures





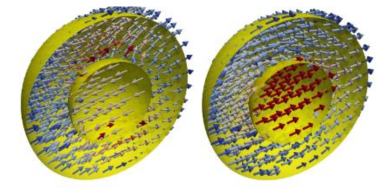
Phys. Rev. B 99, 155423 (2018)

High-index dielectrics



Physical Review Research (2020)

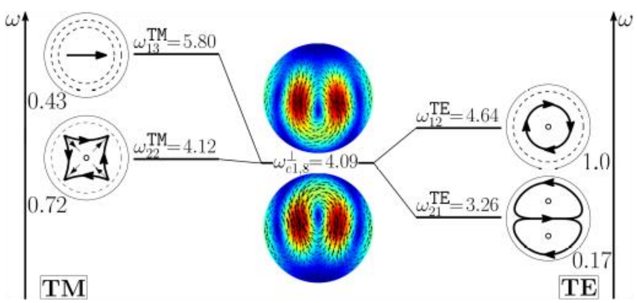
Electromagnetic coating



J. Opt. Soc. Am. B 34, 1524-1535 (2017)

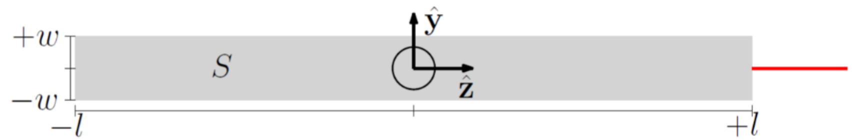
Opt. Lett. 44, 1972-1975 (2019)

Hybridization theory



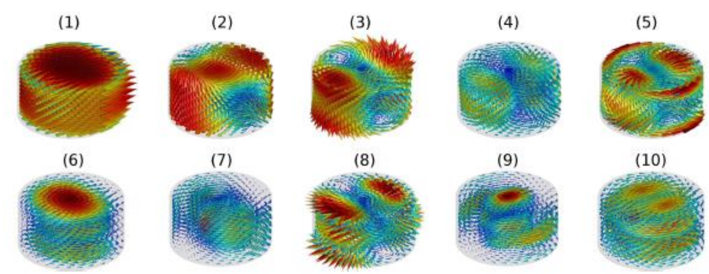
Sci Rep 9, 14524 (2019)

Quasi-1D nanoribbon



IEEE Transactions on Antennas and Propagation, vol. 67, no. 8, pp. 5497-5506 (2018)

Arbitrary shaped nanoparticle



IEEE Transactions on Antennas and Propagation, vol. 66, pp. 2505-2514 (2018)

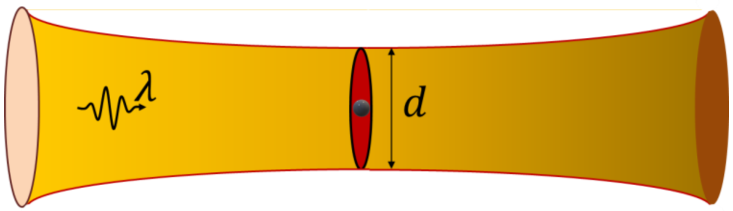
II Plamon quantization: why?

Statement

At the basis of many new applications in quantum technology, such as single-photon nonlinear optics, nonclassical states of light, photonic gates and others, there is the need of reaching an efficient atom-photon interaction.

Issue

The atoms, due to the low scattering cross section, don't like to interact with light.

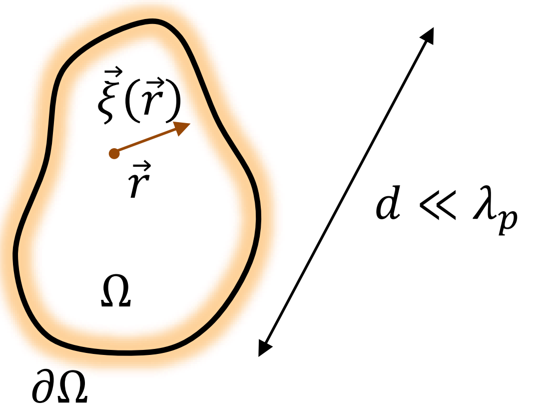


$$p \approx \frac{\sigma}{d^2} \approx \frac{\lambda^2}{d^2} \ll 1$$

Plasmonic structure can localize light beyond the diffraction limit → d ↓

A proper quantum description of the localized plasmon NPs
- is needed in some conditions (e.g. strong coupling regime in the analysis of quantum emitters)
- can give a real insight in the physics of the phenomenon

II Plamon quantization (closed/quasistatic)



$\xi(\boldsymbol{r})$ Electron displacement field

n_o electron density at rest,
m_e electron effective mass,
λ_p plasmon wavelength

$$E = \underbrace{\int_B \frac{1}{2} m_e n_o \left|\partial_t \vec{\xi}\right|^2}_{\text{Kinetic energy}} + \underbrace{\frac{(en_0)^2}{8\pi\epsilon_0} \int_S \int_S \frac{\xi_n(\vec{r})\, \xi_n(\vec{r}')}{|\vec{r} - \vec{r}'|}}_{\text{Coulomb energy}}$$

$$\vec{\xi}(\vec{r}, t) = \sum_m q_m(t) \vec{U}_m(\vec{r})$$

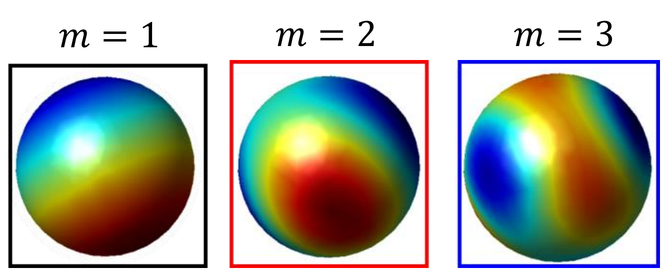
$(p_m = m_e \dot{q}_m)$

$$H(q_m, p_m) = \sum_n \left(\frac{p_m^2}{2M_m} + \frac{1}{2} M_m \Omega_m^2 q_m^2 \right)$$

Plasmon modes

$$\nabla \times \vec{U}_m = 0$$
$$\nabla \cdot \vec{U}_m = 0$$
$$\vec{U}_m \cdot \hat{n} \Big|_S \neq 0$$

Plasmon frequency	Ω_m
Plasmon mass	$M_m = m_e n_o V_m$
Plasmon volume	$V_m = \langle \vec{U}_m, \vec{U}_m \rangle$



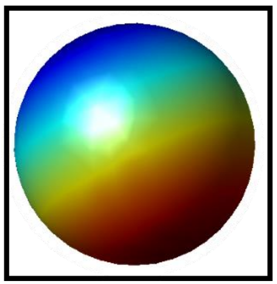
Surface charges

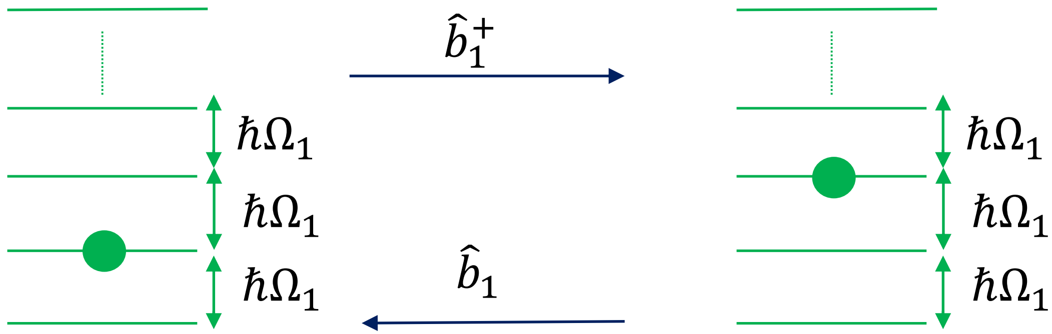
II Plamon quantization (closed/quasistatic)

$$H = \sum_n \left(\frac{p_m^2}{2M_m} + \frac{1}{2} M_m \Omega_m^2 q_m^2 \right) \quad \Longrightarrow \quad \hat{H}_M = \sum_m \hbar\Omega_m \left(\hat{b}_m^+ \hat{b}_m + \frac{1}{2} \right)$$

Canonical Quantization

$m = 1$

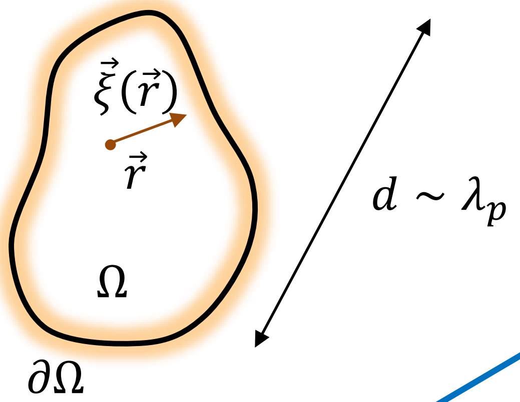




Definition of plasmon
A quantum of energy in the collective motion of electron.

- Infinite lifetime
- Energy $\hbar\Omega_\mathrm{m}$

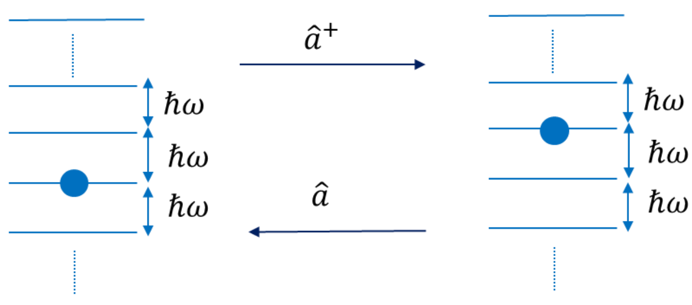
II Plamon quantization (open/retarded)

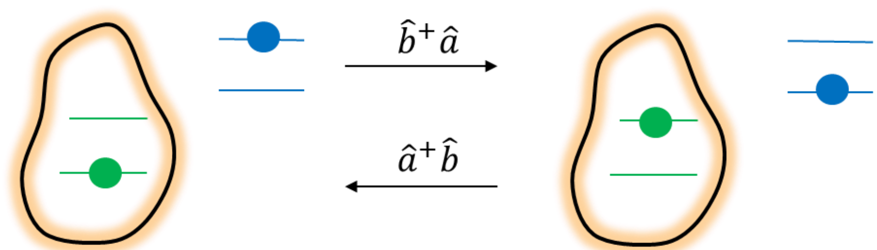


$$\hat{H} = \hat{H}_M + \hat{H}_{EM} + \hat{H}_I + \hat{H}_{II}$$

$$\int_{R_3} \frac{d^3\vec{q}}{(2\,\pi)^3} \sum_{s=1,2} \hbar\omega_{\vec{q},s} \left(\hat{a}^+_{\vec{q},s} \hat{a}_{\vec{q},s} + \frac{1}{2} \right)$$

$$\sum_m \int_{R_3} \frac{d^3\vec{q}}{(2\,\pi)^3} \sum_{s=1,2} [V^m_{\vec{q},s} (\hat{b}^+_m - \hat{b}_m)\, \hat{a}_{\vec{q},s} + h.c.]$$





II Plamon quantization (open/retarded)

Approximate plasmon-photon interaction (independent modes)

$$\hat{H} = \hat{H}_M + \hat{H}_{EM} + \hat{H}_I$$

- The plasmon excitations decay in time $\tau_m = 1/\Gamma_m$
- The plasmon energies shift $\hbar\Omega_m \rightarrow \hbar\Omega_m + \hbar\Delta\Omega_m$

$$\Gamma_m = -Im\left[\Omega_m \sqrt{1 + \frac{2}{\hbar}\frac{\Psi(\Omega_m)}{\Omega_m}}\right] \qquad \Delta\Omega_m = -Im\left[\Omega_m \sqrt{1 + \frac{2}{\hbar}\frac{\Psi(\Omega_m)}{\Omega_m}}\right]$$

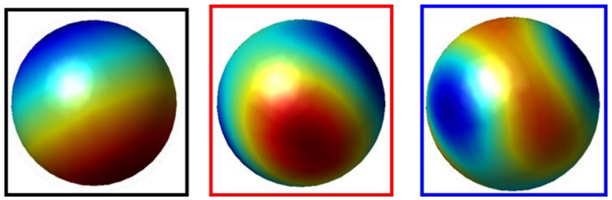
Full-retarded quantities

$$\Psi(\Omega_m) = \frac{1}{\hbar}\int \frac{d^3\vec{q}}{(2\,\pi)^3} \sum_{s=1,2} \left|V_{\vec{q},s}\right|^2 \left[p.v.\frac{1}{\Omega_m - \omega_q} - \frac{1}{\Omega_m + \omega_q} - i\pi\delta(\Omega_m - \omega_q)\right]$$

Non-perturbative

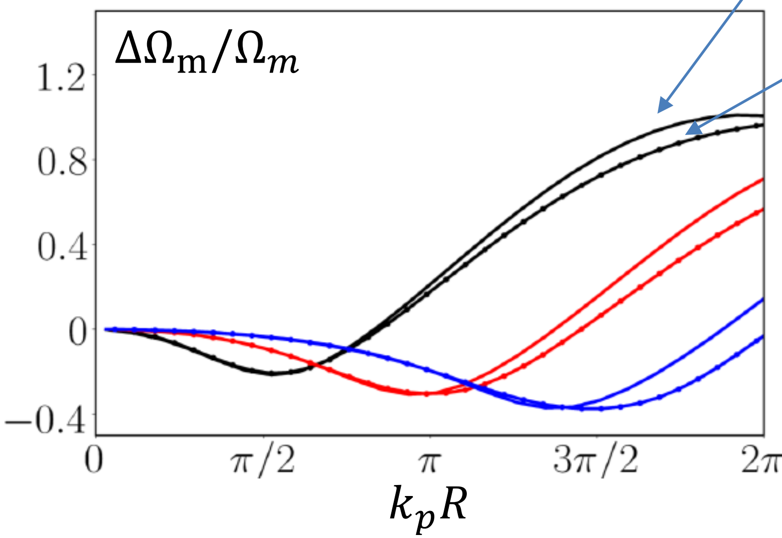
quasistatic computation

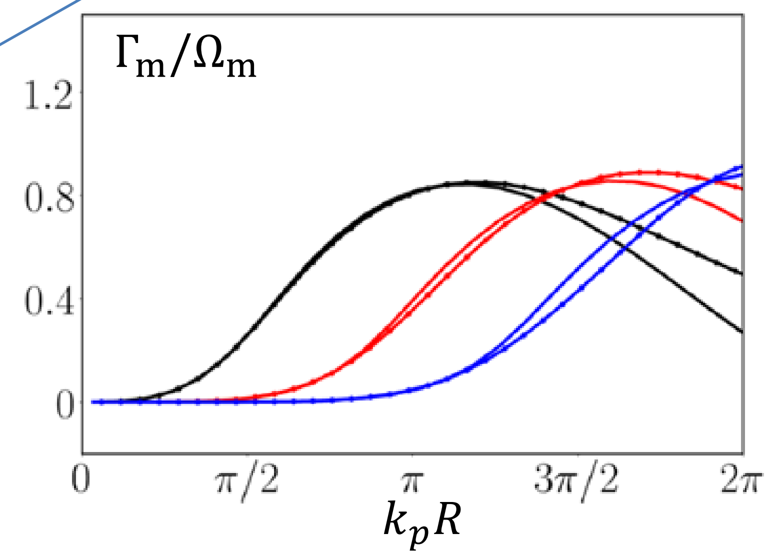
II Plamon quantization (open/retarded)



Exact solution

Approximate solution





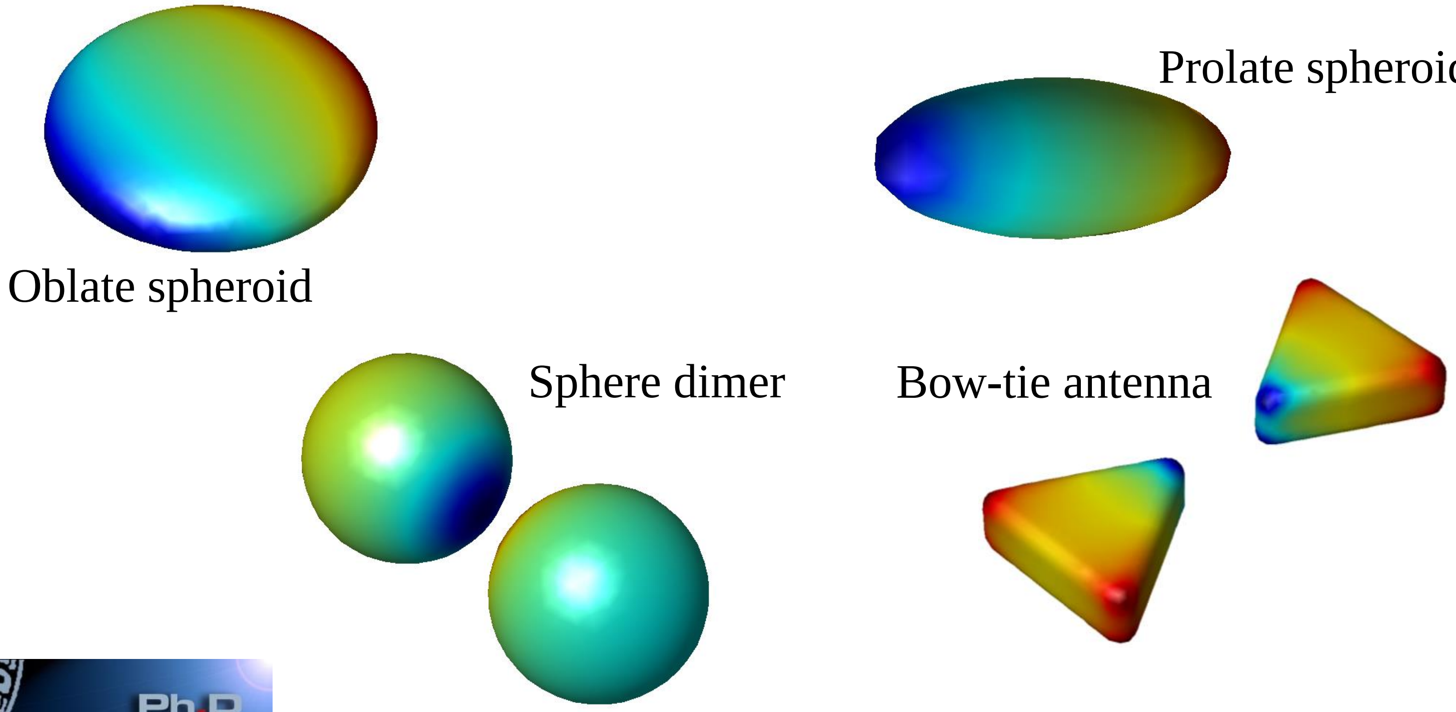
$k_p = 2\pi/\lambda_p$

It is essentially exact for $k_pR < \pi$

Ex. Gold $k_pR = \pi$ means $R = 144$ nm
Silver $k_pR = \pi$ means $R = 230$ nm

II Plamon quantization (open/retarded)

This theory has been applied to compute the *radiative decay rate* and the *frequency shift* of the plasmon oscillations in arbitrary shaped nanoparticles and dimers.



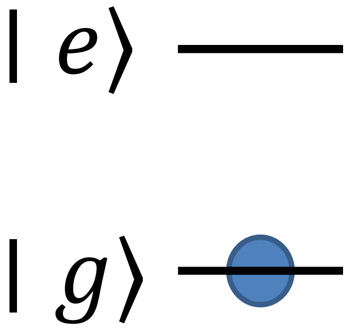
arXiv:2001.11926 (2020)

III Photon-photon interaction

Photons in linear media do not interact

→

Effective photon-photon interaction exploiting the *nonlinearity of the atoms*



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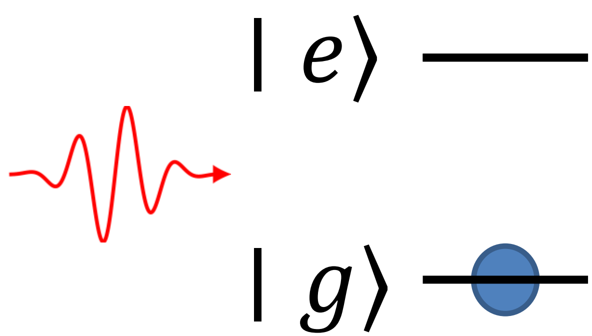

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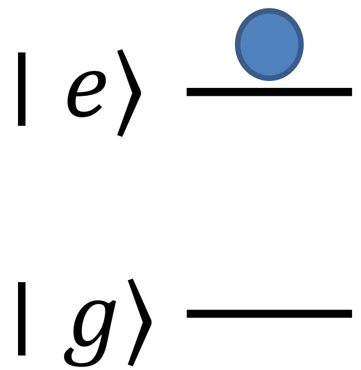

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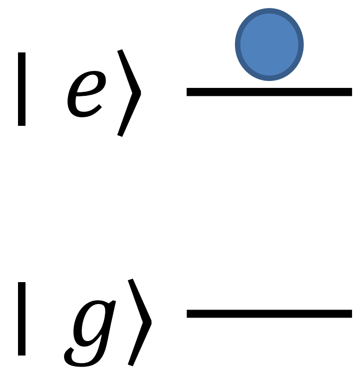

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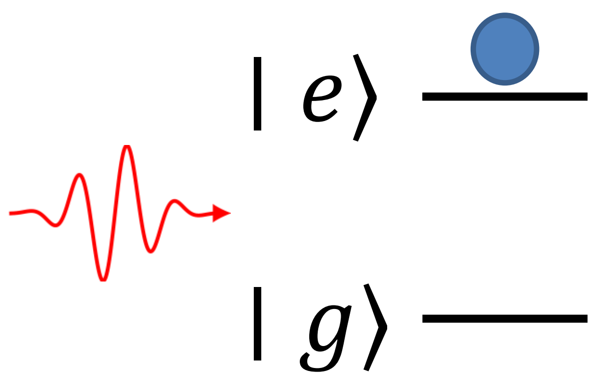

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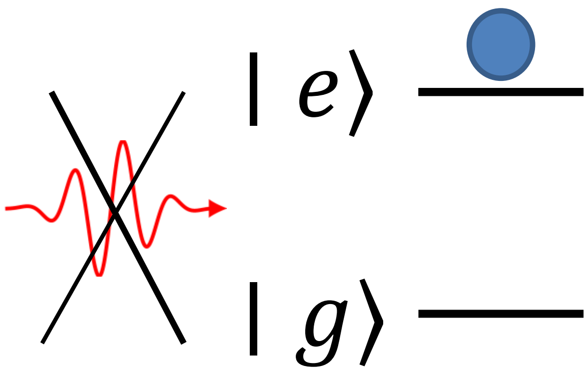

Blockade phenomenon

Another photon cannot be absorbed!

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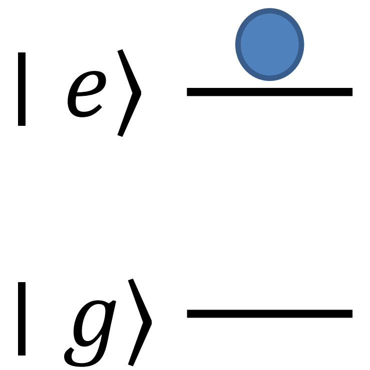
But in real life the probability of interaction is too small!



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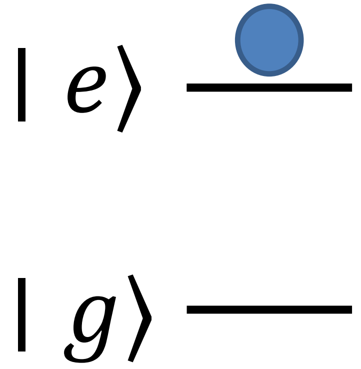

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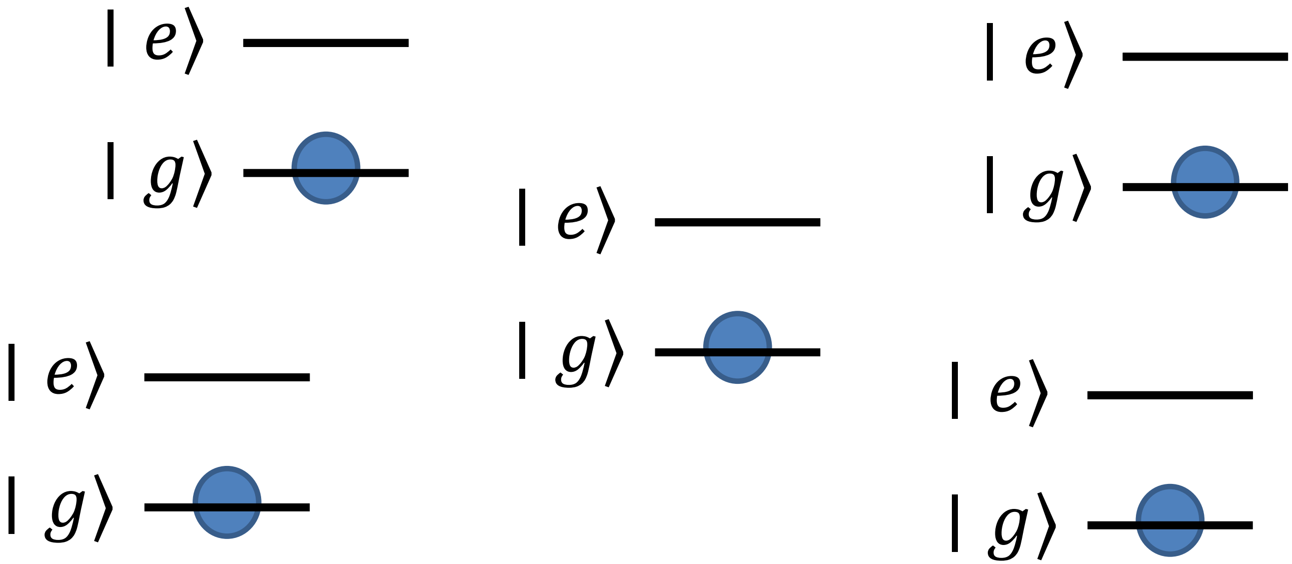

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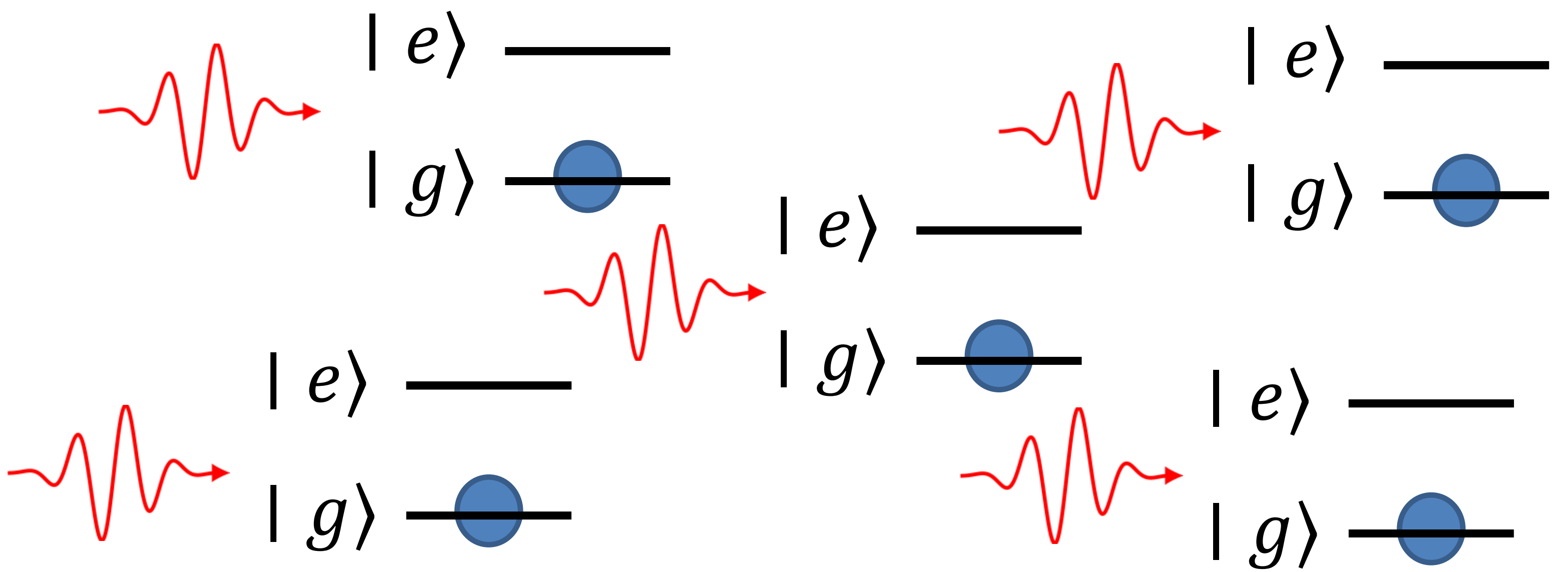

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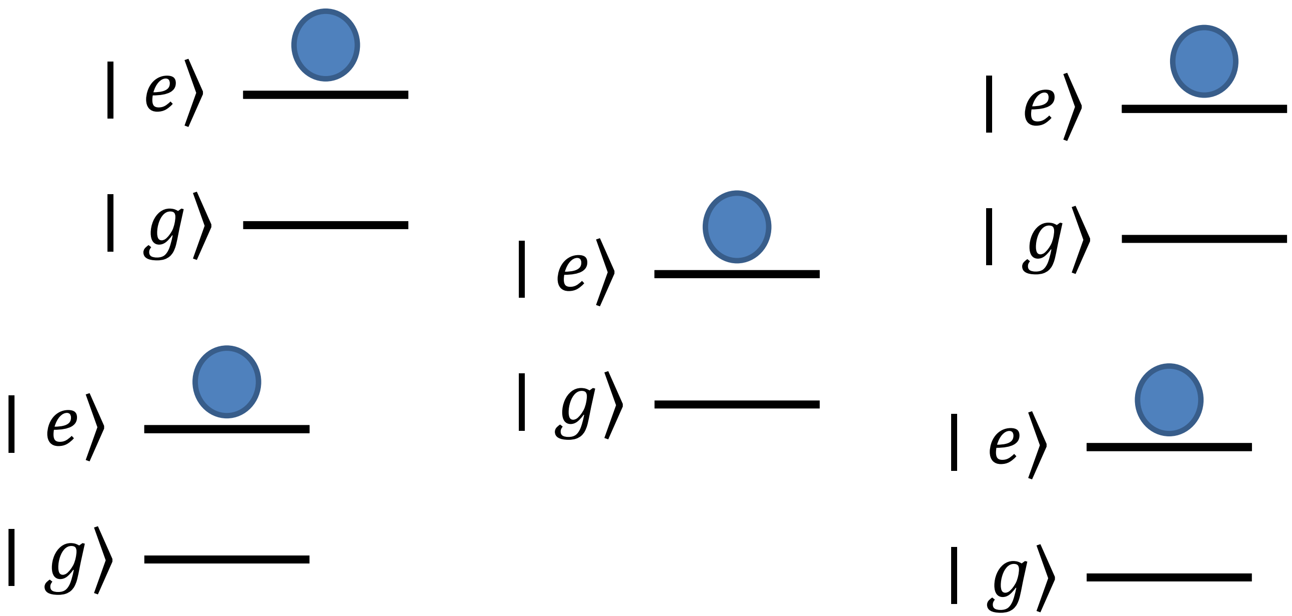

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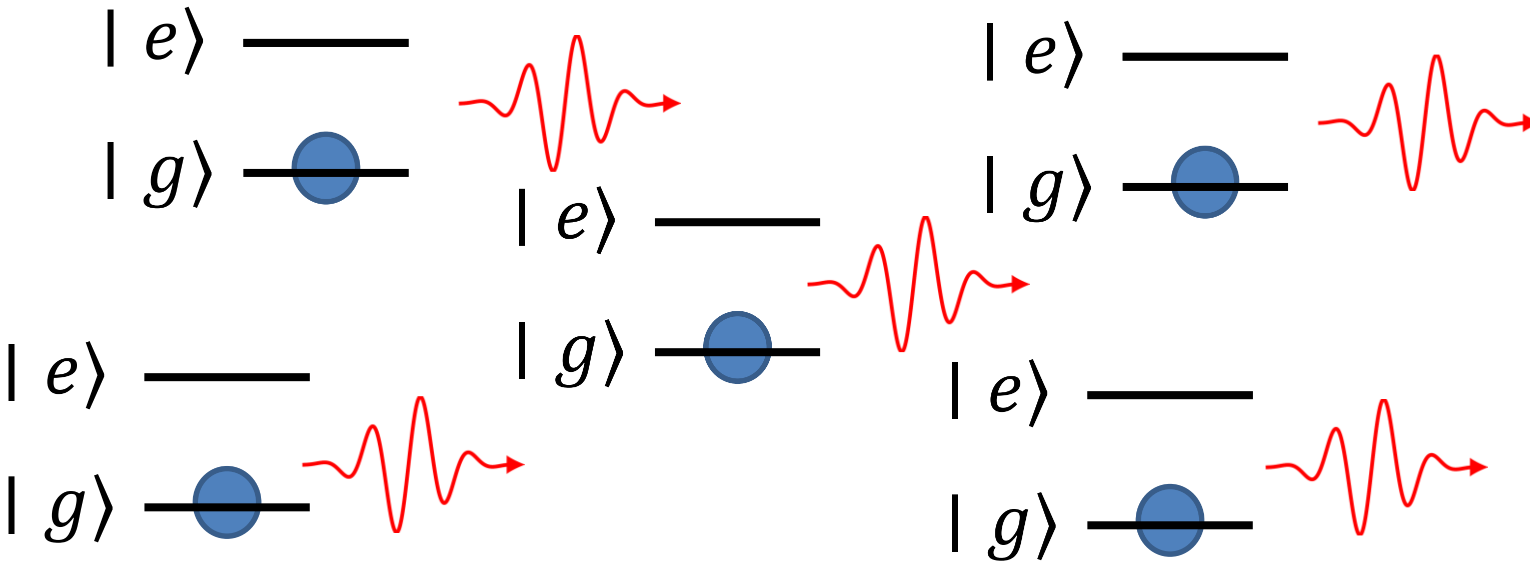

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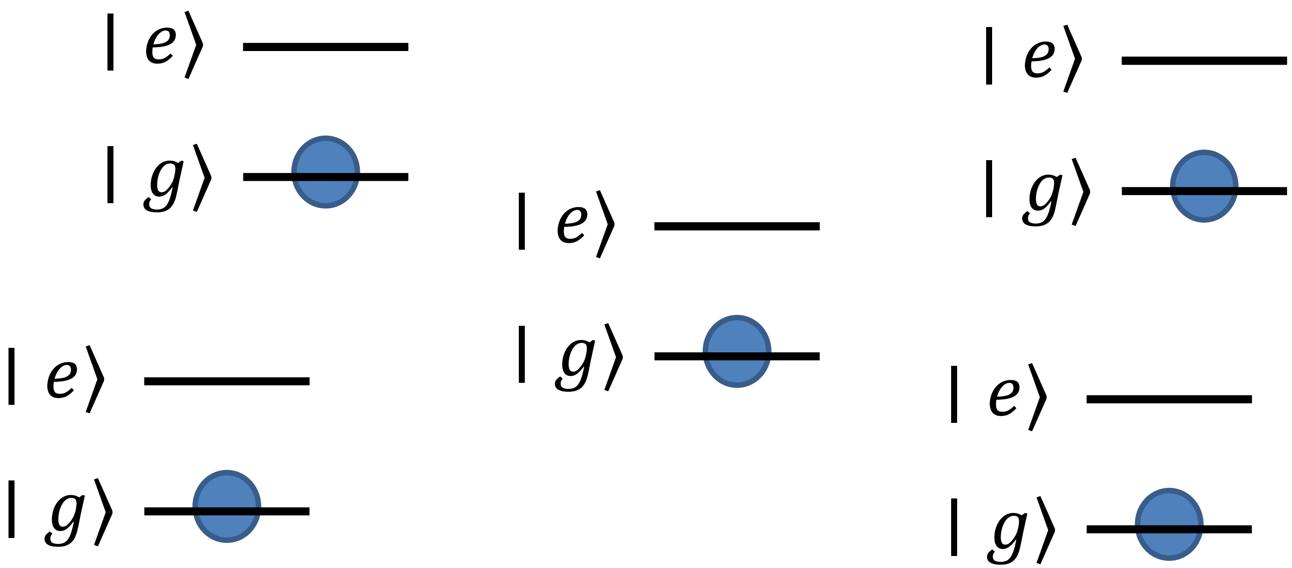

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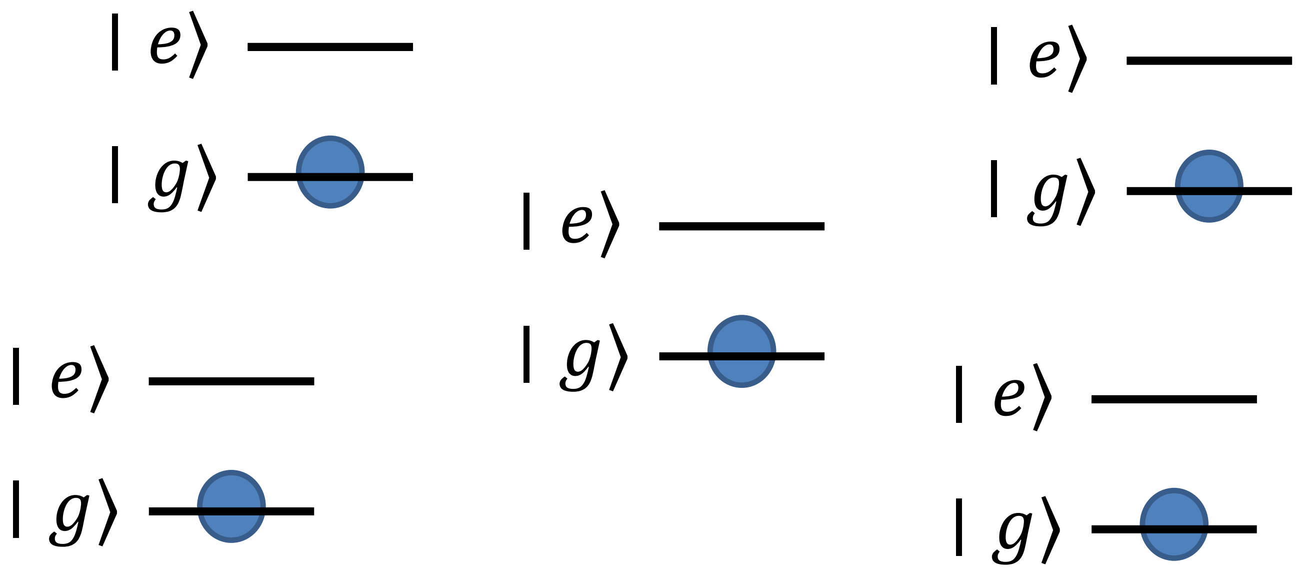

We measure a linear response!

III Rydberg atoms: why?

Even if an atom is a highly nonlinear object, the nonlinearity is very hard to be measured: it is LOCAL in space and NARROW in time.

⬇

A Rydberg atom is an atom excited to a *high principal quantum number state*. These states exhibits a very long lifetime and a huge dipole momentum, able to shift the energy levels of the surrounding atoms, within the blockade sphere.

⬇

A Rydberg atom acts as a 'super atom'. It provides a NONLOCAL blockade mechanism able to introduce an effective nonlinear photon-photon interaction.

III Rydberg atoms: why?

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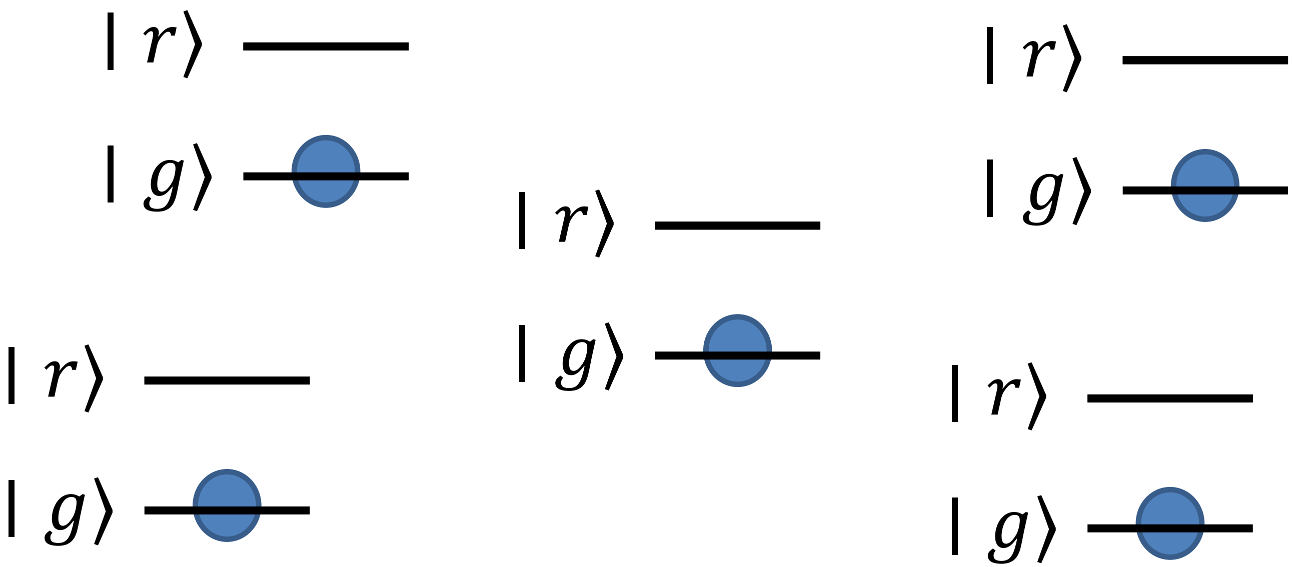

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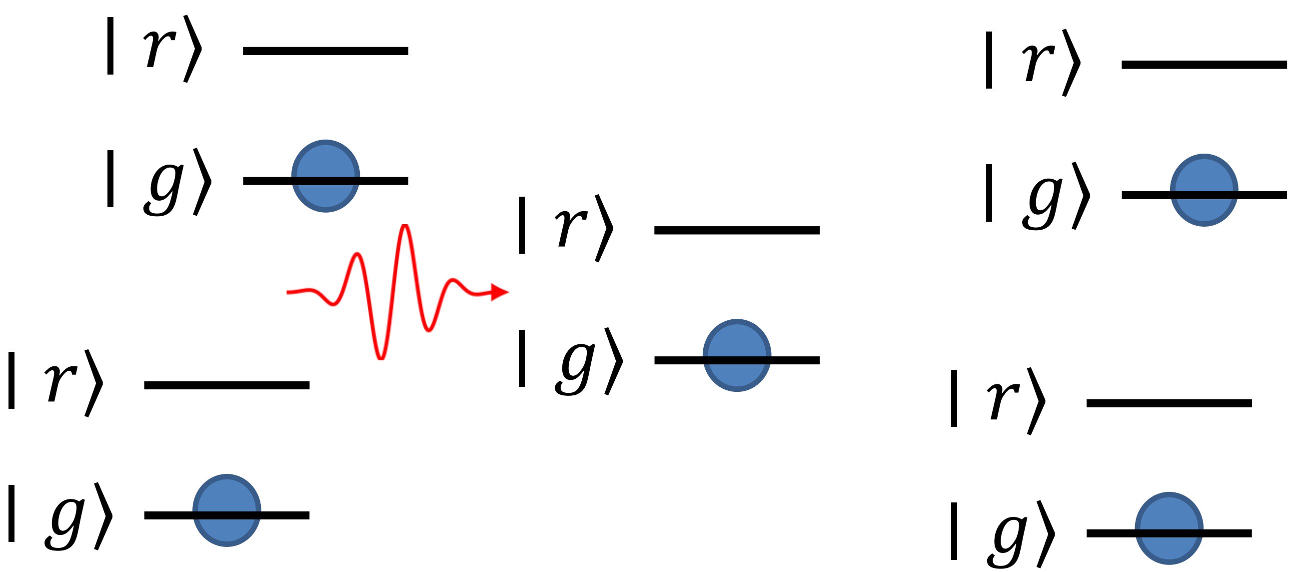

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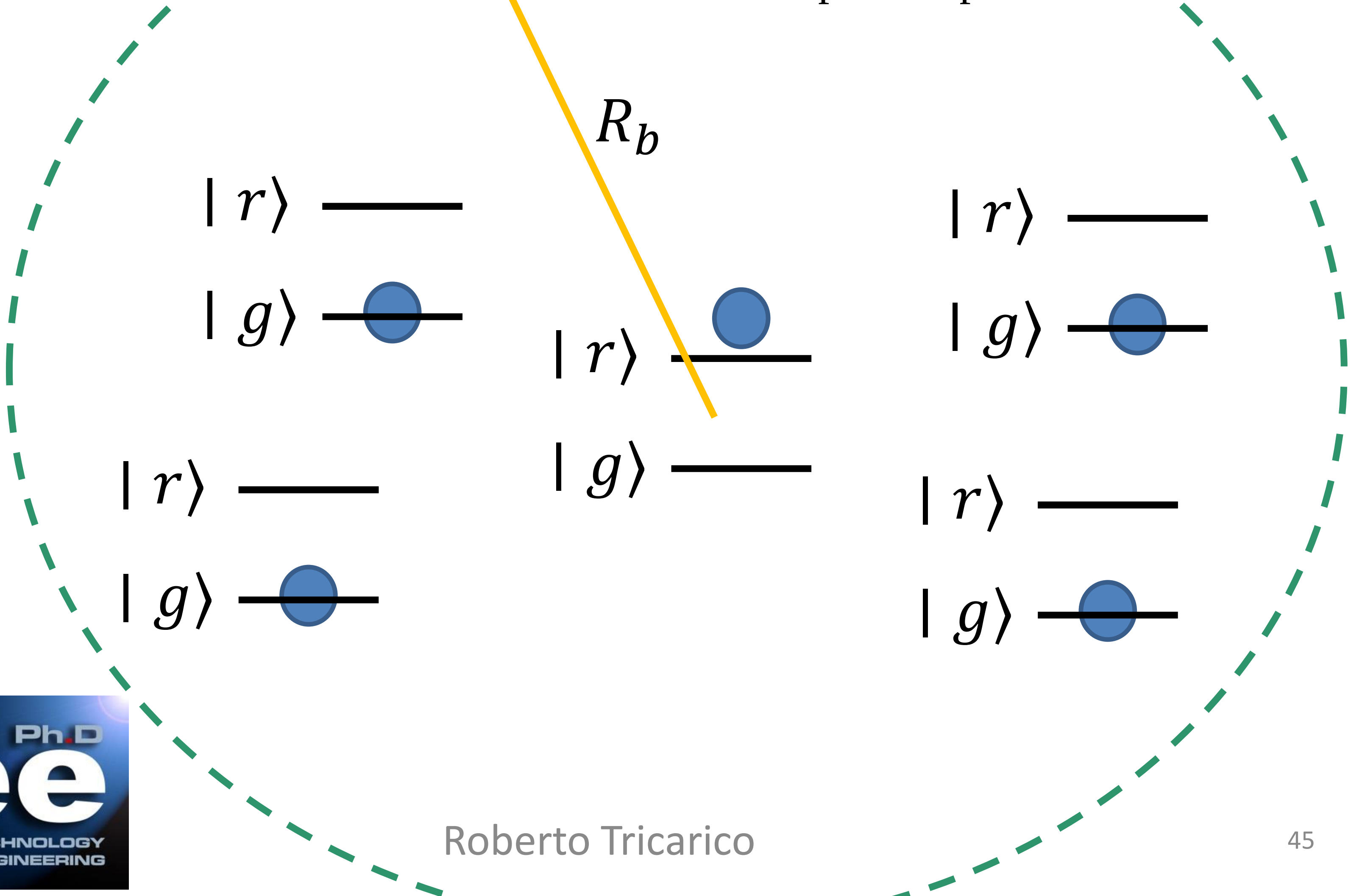

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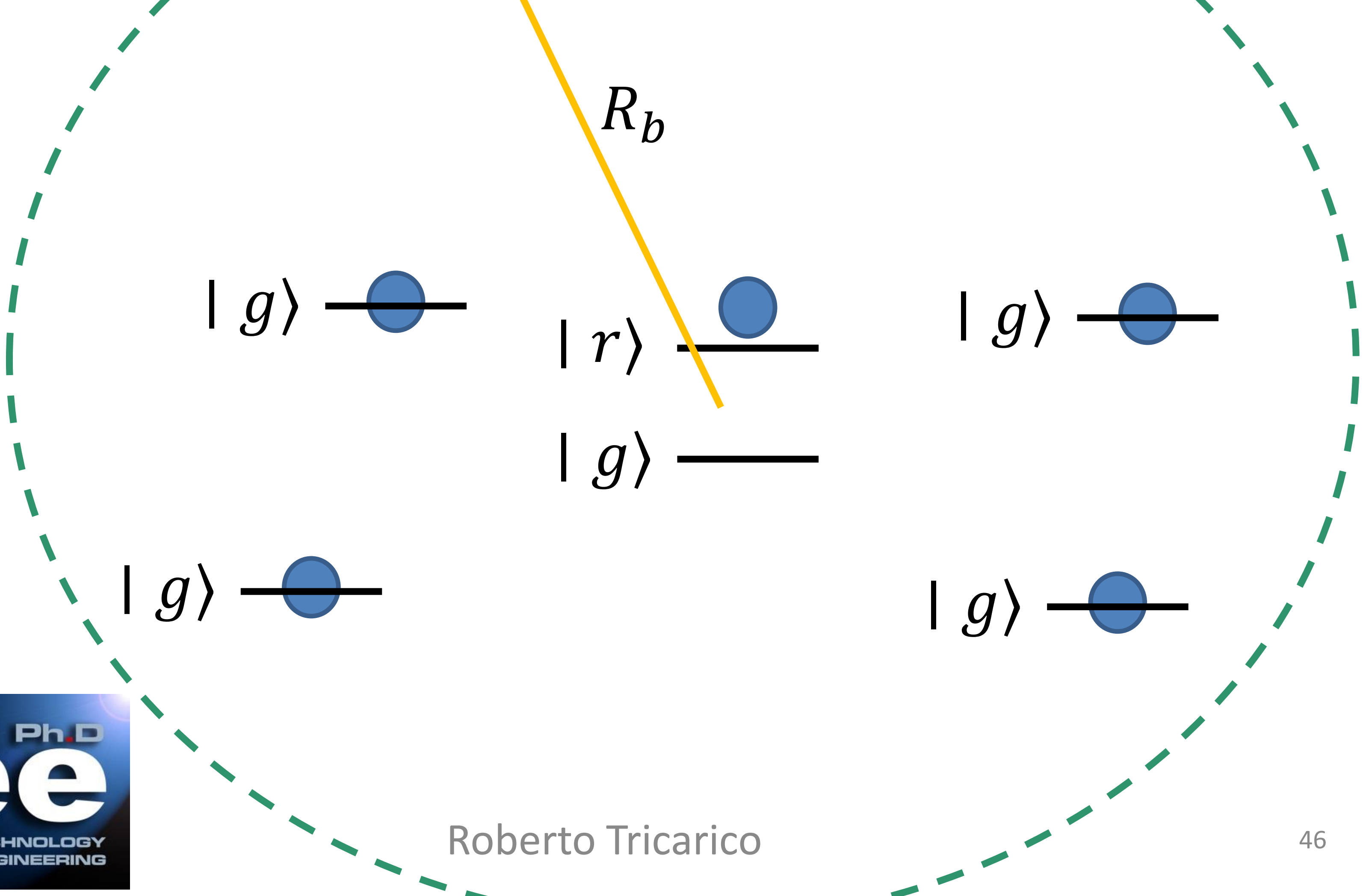

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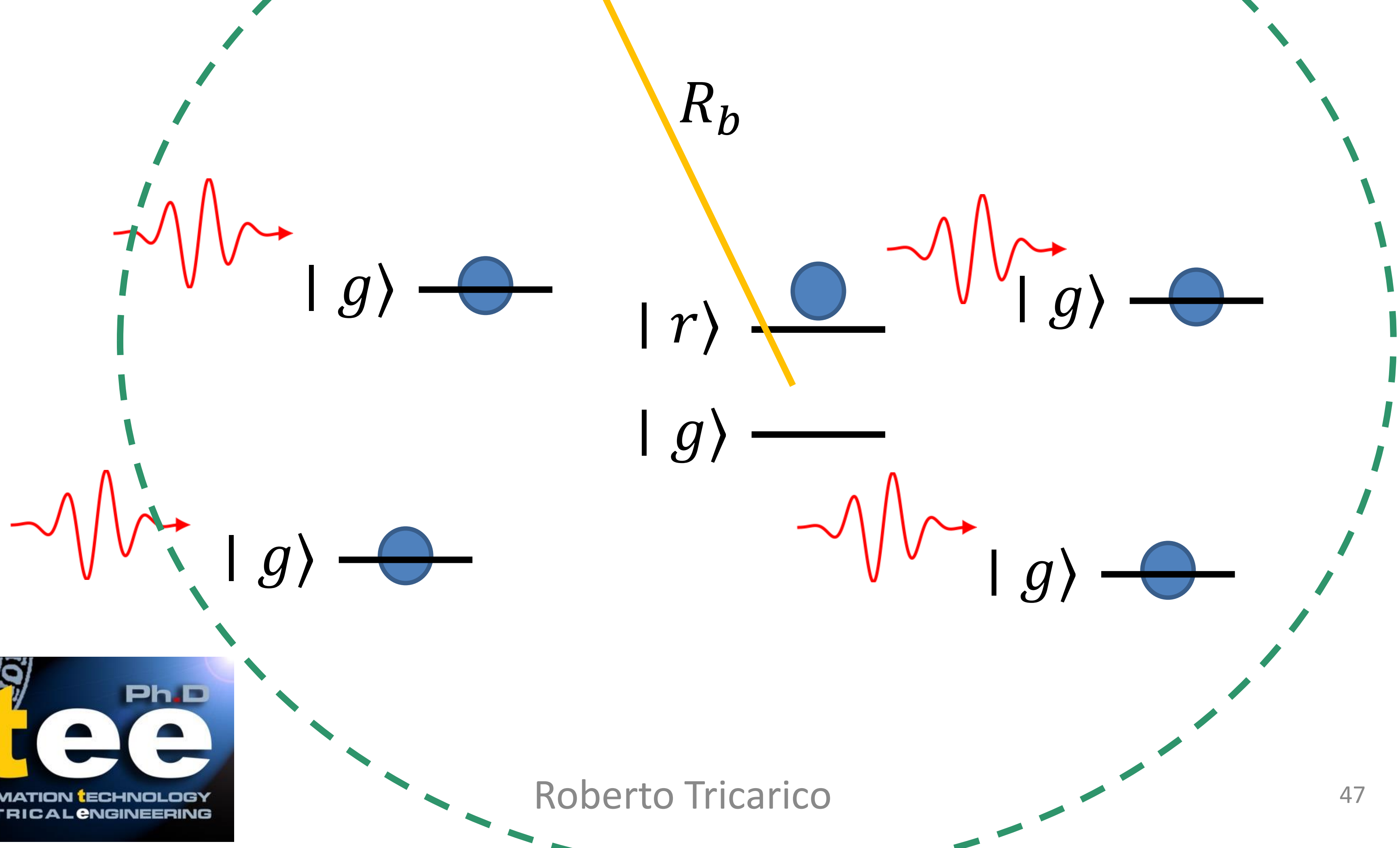

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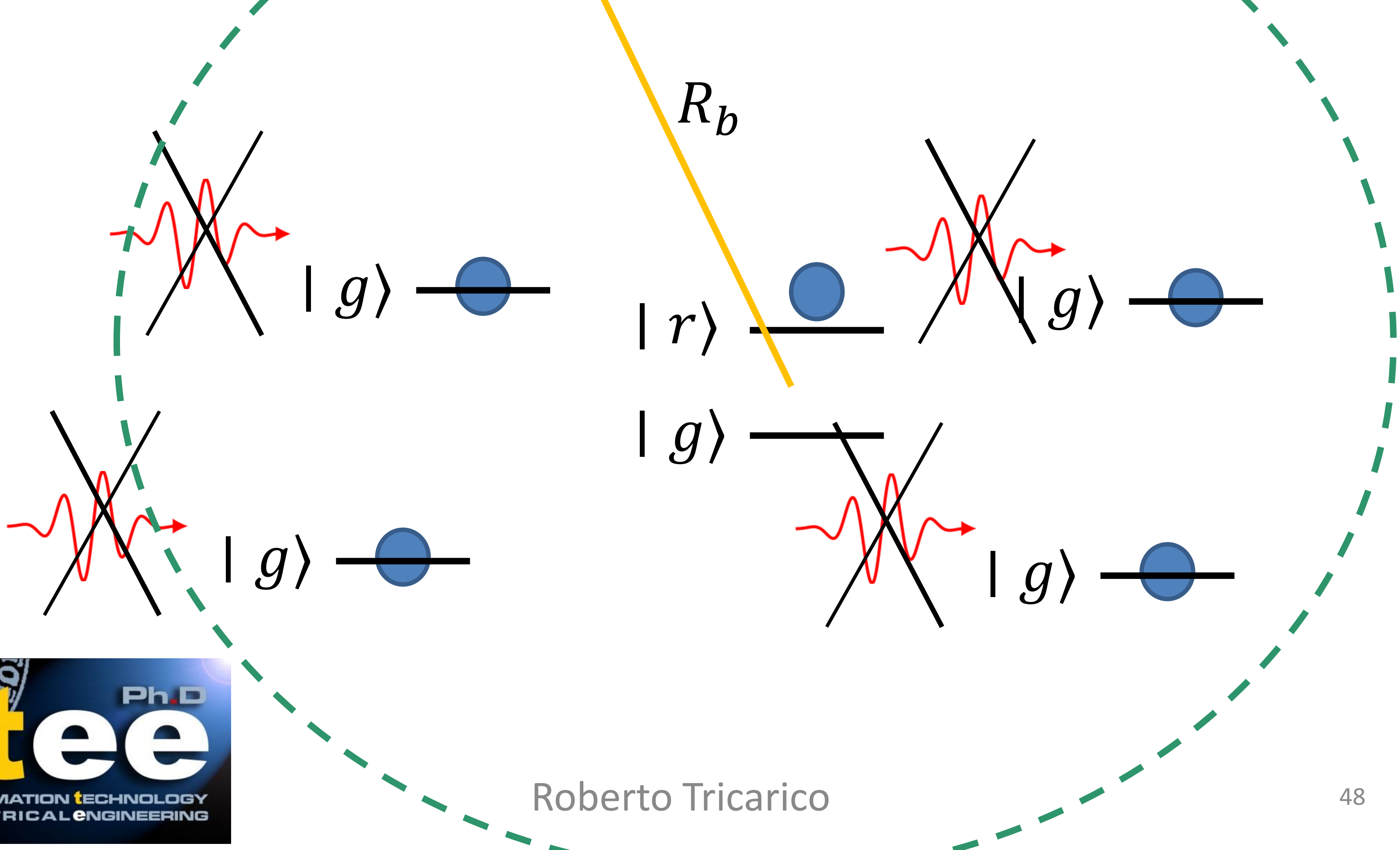
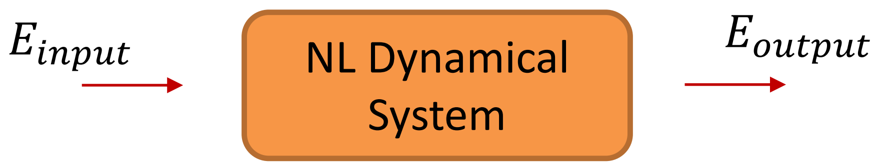


III How to quantify nonlinearity?

- Classical Point of view



Nonlinear relation between input and output
$E_{output} = f(E_{input})$

- Quantum point of view

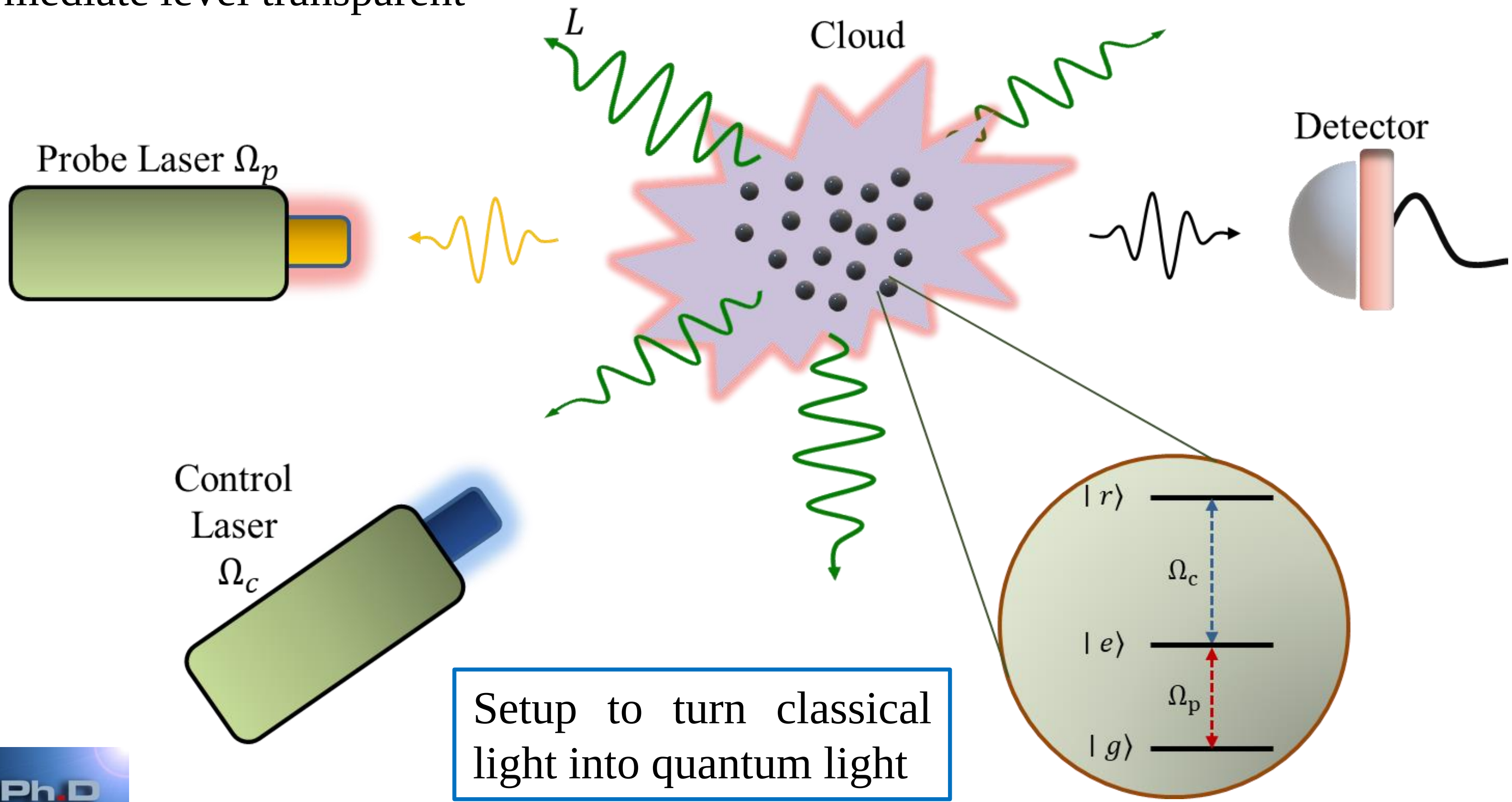
Can turn a classical field (e.g. a coherent state) into a purely quantum state of light.
The signature of the pure quantum state of light in the *antibunched* behavior, i.e. $g^{(2)}(t,t) < 1$.

$$g^{(2)}(t,t') = \frac{\langle\psi|E^{-}(t)E^{-}(t')E^{+}(t')E^{+}(t)|\psi\rangle}{\Omega^4\langle\psi|E^{-}(t)E^{+}(t)|\psi\rangle\langle\psi|E^{-}(t')E^{+}(t')|\psi\rangle}, \qquad (t' \geq t)$$

III Rydberg-EIT

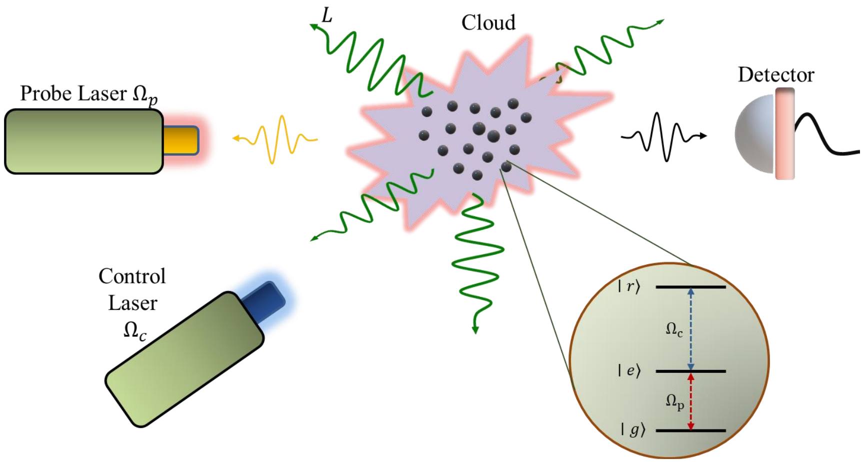
EIT= Electromagnetic Induced Transparency

It is a 3-level scheme, where an inference phenomenon between 2 shining laser makes the intermediate level transparent

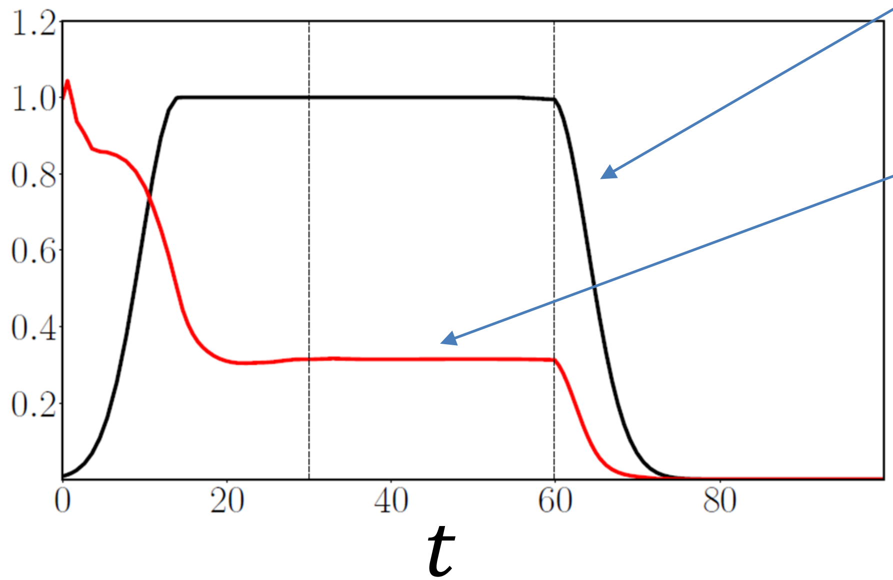


Setup to turn classical light into quantum light

III Nonlinearities in R-EIT



I theoretically modelled the experimental measurements of C. Adams' group (2019) which showed that the pulse dynamics can be more antibunched than the steady state one.



New protocols to turn classical light into quantum light

THANK YOU

Roberto Tricarico

Light-Matter Interaction in Open Systems: from Nanoparticles to Atoms