## Exploiting Vibrational Strong Coupling to Make an Optical Parametric Oscillator out of a Raman Laser

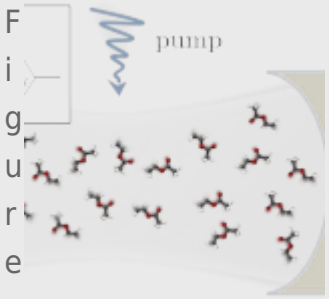


Figure: Sketch of the system. The blue arrow depicts the pump beam, while the green and red arrows show the two emitted laser beams. The inset shows the energy structure, with the vibropolaritons (purple) separated by the Rabi splitting.

Article: published in Physical Review Letters by Javier del Pino, Francisco J. Garcia-Vidal and Johannes Feist, Department of Theoretical Condensed Matter Physics and IFIMAC researchers.

Raman scattering is a nonlinear optical process in which a photon is converted into two new excitations: a photon of a different color, as well as a material excitation such as a phonon (a quantum of vibrational energy). In a Raman laser, stimulated Raman scattering is exploited to create sources of coherent light with a wide range of achievable output wavelengths. This concept can be implemented in a variety of configurations, such as under pulsed or continuous operation, and using a wide range of nonlinear media such as optical fibers, nonlinear crystals, gases, or semiconductor materials. However, all these devices share the same drawback: Only the photon resulting from Raman scattering is used, while the material excitation is “lost” and its energy deposited in the form of heat.

In a theoretical study published in Physical Review Letters, a group of researchers from the Departamento de Física Teórica de la Materia Condensada and the Condensed

Matter Physics Center (IFIMAC) at the Universidad Autónoma de Madrid present a proposal to circumvent this drawback of conventional Raman lasers. The crucial step is to modify the vibrational excitations in the material, converting them into hybrid light-matter excitations that have a photonic component, so-called vibropolaritons. This can be achieved through so-called “strong coupling”, which occurs when the coupling between the vibrational excitations and a resonant confined light mode becomes faster than the decay of either constituent. The resulting vibropolaritons then decay most efficiently through emission of light, which has two important consequences in the context of Raman lasers: First, the heat deposition is strongly reduced, as this energy is now emitted as photons. Second, and even more importantly, the Raman laser with only a single output beam is converted into a different type of nonlinear optical device, a so-called optical parametric oscillator with two output beams. These beams are coherent and have a stable phase relation, as well as providing possibly entangled pairs of photons with non-classical correlations.

In addition, the authors show that the coexistence of two vibropolariton modes with similar properties allows to operate the same device as an all-optical switch. Here, one (gate) pump beam can be used to switch on or off the Raman lasing of a second (signal) pump beam with slightly detuned frequency. In this mode of operation, both vibropolariton modes show stimulated emission as soon as the total pump power becomes large enough, such that the device produces three coherent output beams.

These results are an example of the great potential that hybrid light-matter states possess in manipulating light fields and providing novel light sources. The proposed setup could be used in novel solid-state microcavity devices for applications requiring mutually coherent and/or entangled beams in disparate frequency regions, such as quantum information transmission and storage. In addition, this approach could improve existing Raman lasers by lowering the operating threshold and reducing heat generation. [Full article]

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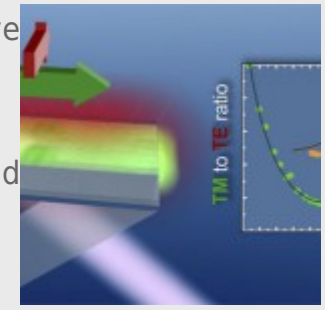
## Multi-scale Molecular Dynamics Simulations of Photoactive Molecules Strongly Interacting With Confined Light




Title: Multi-scale Molecular Dynamics Simulations of Photoactive Molecules Strongly Interacting With Confined Light.
When: Friday, January 13, (2017), 12:00.
Place: Departamento de Física de la Materia Condensada, Facultad Ciencias, Module 3, Seminar Room (5th Floor).
Speaker: Gerrit Groenhof, University of Jyväskylä, Finland.

When photoactive molecules strongly interact with confined light, for instance inside optical cavities or near (localised) surface plasmons, new hybrid light-matter states may form, the so-called polaritons or plexcitons. These polaritons are coherent superpositions (in the quantum mechanical sense) of the excitations on the molecules and the cavity photon or plasmon. Recent experiments suggest that access to these polaritons may provide a totally new paradigm for controlling chemical reactions [1]. However, to exploit strong light-matter coupling for steering chemistry we need a theoretical model that can accurately predict the effect of the coupling on the molecular dynamics. Because current models are based on phenomenological theories that are not easily inverted to design new systems, we have developed a new model based on molecular dynamics (MD) simulations, with which the dynamics of one or more photoactive molecules strongly interacting with confined light can be simulated in atomic detail. In addition to discussing this model, I will also show how the results of the simulations might explain the effect of the molecular Stokes shift on the SPP-molecule polariton emission in recent experiments [2].

References

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S. Baieva, O. Hakamaa, G. Groenhof, T. T. Heikkila, and J. J. Toppari, ACS Photonics, Article ASAP, (2016).

More information on IFIMAC Website

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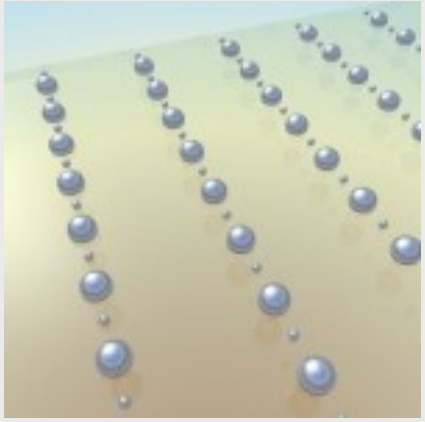
## Signatures of Quantum Condensation in a Plasmonic Nanoparticle Array

Tuesday, 21st March 2013. 12:00-13:00



*Said R.K. Rodríguez*

*Center for Nanophotonics, FOM Institute AMOLF, c/o Philips Research Laboratories, Eindhoven, The Netherlands*

ABSTRACT:

We present experimental signatures of quantum condensation in a plasmonic system. We investigate a periodic array of metallic nanorods covered by a polymer layer doped with an organic dye at room temperature. Surface lattice resonances of the array – hybridized plasmonic/photonic modes – couple strongly to excitons in the dye, and bosonic quasi-particles known as plexcitons are formed. By increasing the plexciton density through optical pumping, we observe the emergence of Bogoliubov-Goldstone excitations on top of the strongly coupled plexciton band in the light emission dispersion diagram. The Bogoliubov-Goldstone mode shows signatures of thermalization and condensation, despite the nonequilibrium character of this driven and dissipative system.

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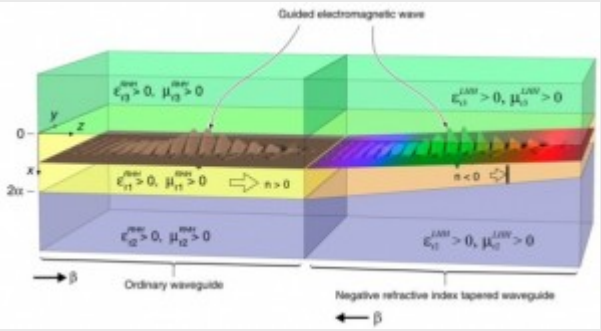
## Modelling of Active Plasmonic and Metamaterial Systems in the Time-Domain

Tuesday, 26th February 2013. 12:00-13:00




*Joachim Hamm*
*Imperial College, London*

ABSTRACT:

Advances in plasmonics and metamaterial research increasingly focus on device-functionalization with quantum-electronic materials, such as dyes, semiconductors or graphene. The field concentration around metallic nano-features serves as key mechanism to enhance light-matter interactions but also poses new modelling challenges. The presentation will highlight recent results based on the self-consistent modelling of photonic/quantum-electronic structures, such as gain-infiltrated active metamaterial structures. In addition it will offer an outlook to future work on the modelling of quantum-electronic metamaterials that include semiconductor media and graphene.

## Qantum Effects in Plasmonic Nanostructures

Thursday, 16th February 2012. 12:00-13:00

*Pablo García González*

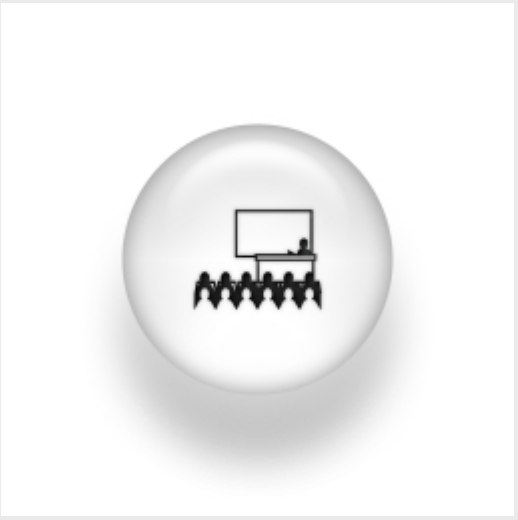
Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid

ABSTRACT:
One of the most attractive aspects of surface plasmon polaritons is their ability to collect and concentrate light into sub-wavelength volumes. The theoretical description of the relevant processes is often done by using classical local optics. However, the miniaturization in the fabrication of plasmonic devices is approaching the limit where non-local effects in the optical response of a metal cannot be neglected.
A possible way to take into account these effects is the modelling of the metal permittivity through hydrodynamical approximations [1]. Nevertheless, the predictive accuracy of such methods depends very sensitively on the details of the model permittivity. At a more fundamental level, the electron response of a system can be evaluated by using time-dependent density functional theory (TDDFT) [2]. Under this prescription, non-local and quantum-mechanical effects in the optical response are treated on the same footing.
In this informal seminar, I shall present some preliminary results of the TDDFT optical response of two metal nanowires in close proximity (sub-nanometric) to each other. A comparison with the corresponding hydrodynamic and local responses in the limit of zero separation will be presented as well.
Work done in collaboration with Lorenzo Stella and Angel Rubio (UPV/EHU), and F.J. García Vidal (UAM).
[1] A.I. Fernandez-Dommnguez, A. Wiener, F.J. Garcma-Vidal, S.A. Maier and J.B. Pendry (in press).
[2] E. Runge and E.K.U. Gross, Phys. Rev. Lett. 52, 997 (1984).

## Magnetoplasmonics: The interplay between magneto-optics and plasmonics

Wednesday, 14th december 2011. 12:00-13:00

*Antonio García-Martín*



Instituto de Microelectrónica de Madrid, CSIC

ABSTRACT:
Subwavelength composite materials constitute an interesting path towards the development of materials with “on demand” optical properties. We will present our latest results on systems composed of both noble and ferromagnetic metals, which we denote as magnetoplasmonic. While noble metals have intense and narrow plasmon resonances they lack magneto-optical (MO) activity at reasonable magnetic field intensities. On the other hand, ferromagnetic metals are MO active but their plasmon resonances are weak and broad. By combining both kinds of materials we intend to obtain systems which simultaneously exhibit plasmon resonances and MO activity. We will show that thus it is possible to both (1) enhance the magneto-optical activity of the system via surface plasmon excitation, and (2) modulate the plasmon properties via application of a magnetic field [1]. Localized surface plasmon resonances (LSPRs) greatly influence the optical [2,3] and magneto-optical (MO) [4,5,6,7,8] properties of fully metallic and metal-dielectric nanostructures. We will analyze the MO response of isolated nanodisks, where we will show how the excitation of the LSPR produces an enhancement of the MO activity [4]. The observed enhancement in the MO is attributed to the high intensity of the electromagnetic (EM) field inside the nanostructure when the LSPR occurs. Here we show how the EM profile related to the LSPR can be probed locally inside the nanostructure by measuring the MO activity of the system as a function of the position a MO active probe (a Co nanolayer) [9]. This EM field profile is the key element in the analysis of the MO activity and thus a clever engineering would make it possible to get large MO effect and low losses [10]. The same kind of structures allows the analysis of the effect of the MO activity on the plasmon properties. We will show that the wavevector of the plasmon is modified upon application of a magnetic field in the transverse configuration [11]. That modification can be used in a wide

variety of scenarios: e.g. in active microinterferometry [12,13].
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## Towards quantum plasmonics: plasmon mediated qubit-qubit entanglement

Wednesday, 2 March 2011, 12:00-13.00

*Alejandro González-Tudela*
Departamento de Física Teórica de la Materia Condensada, UAM

ABSTRACT:
The field of nanoplasmonics has received an extraordinary attention in the last few years due to the prediction of a lot of interesting physical phenomena such as extraordinary optical transmission, enhanced energy transfer, surface plasmon sensors and many more [1].
All these phenomena can be obtained from a classical description of the field, however there have been a few steps in exploring its quantum regime,i.e., generating single plasmons [2] using nanowires. Our way to quantum plasmonics started studying the dissipative dynamics of a single emitter close to a metal-semiconductor interface [3].

After the proposal of coupling two qubits through plasmonics one-dimensional nanowaveguides [4], we have studied the possibility of coupling two qubits by plasmons supported by them. The plasmons induce coherent and incoherent coupling between the qubits, dephased π/2 between them, allowing us to switch off one of the two contributions while maximising the other by altering the interqubit distance. Mainly due to the dissipative component of this coupling we could find situations of spontaneous formation of entanglement and using a laser a stationary entangled state appears for distances larger than the operating wavelength [5].

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## Resonant Optical Forces on Metallic and Dielectric Nanoparticles

Wednesday, 19 January 2011, 12:00-13.00

*Prof. Juan José Sáenz*
Dpto. Física de la Materia Condensada, UAM

ABSTRACT:
Light forces on small (Rayleigh) particles are usually described as the sum of two terms: the dipolar or gradient force and the scattering or radiation pressure force. The scattering force is traditionally considered proportional to the Poynting vector, which gives the direction and magnitude of the momentum flow. However, as we will show, when the light field has a non-uniform spatial distribution of spin angular momentum, an additional scattering force arises as a reaction of the particle against the rotation of the spin. This non-conservative force term is proportional to the curl of the spin angular momentum of the light field [1]. We will illustrate the relevance of the spin force in the

particular simple case of a 2D field geometry arising in the intersection region of two standing waves. The unusual properties of the optical forces acting on particles with both electric and magnetic response will also be analyzed [2,3].
We will also discuss the peculiar particle dynamics in the non-conservative force field of an optical vortex lattice [4]. Radiation pressure in the vortex field (arising in the intersection region of two crossed optical standing waves) plays an active role spinning the particles out of the whirls sites leading to a giant acceleration of free diffusion. Interestingly, we show that a simple combination of null-average conservative and non-conservative steady forces can rectify the flow of damped particles. We propose a "deterministic ratchet" stemming from purely stationary forces [5] that represents a novel concept in dynamics with considerable potential for fundamental and practical implications.
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