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Nanophotonics Research Center

FRANCE-JAPAN WORKSHOP ON NANOPHOTONICS

BOOK OF ABSTRACTS

(order in accordance to tentative schedule as of Oct 25)

- p.1 **Motoichi OHTSU** (Tokyo U.)
Nanophotonics: Dressed photon science and technology
- p.2 **Kiyoshi KOBAYASHI** (Yamanashi U.)
Dressing nature inherent to optical near fields at the nanoscale
- p.3 **Daniel BLOCH** (LPL, Paris13 U.)
Atomic spectroscopy and nano-optics
- p.4 **Emmanuel ROUSSEAU** (LCFIO-Institut d'Optique, Palaiseau)
Radiative heat transfer at the nanoscale: enhancement due to surface phonon polariton
- p.5 **Tadashi KAWAZOE** (Tokyo U. and NRC)
Room-temperature operation of nanophotonic logic gates using InAs QDs in mesa structures
- p.6 **Jean-Philippe POIZAT** (Institut Néel, CEA-CNRS-UJF Grenoble)
Subnanosecond spectral diffusion of a single quantum dot in a nanowire
- p.7 **Sébastien SAUVAGE** (I.E.F., Univ Paris 11)
Nanospectroscopy of single quantum dot ultrasmall absorption
- p.8 **Takashi YATSUI** (Tokyo U.)
Nano photonic devices using multiple quantum structures
- p.9 **Alexia AUFFÈVES** (Institut Néel-CNRS, Grenoble, France)
Quantum dots coupled to solid-state cavities : potential and applications to advanced nanophotonic devices
- p.10 **Ruggero MICHELETTO** (Yokohama City University)
Optical instabilities and blinking phenomena in the emission of InGaN quantum wells
- p.11 **Hirokazu HORI** (Yamanashi U.)
Fundamental processes of optical near-field interactions
- p.12 **Rafaelle COLOMBELLI** (IEF, Paris 11 U.)
Semiconductor plasmon sources

- p.13 **Pierre VIKTOROVITCH** (INL-Lyon U. and ECL)
Non Conventional Approaches for 3D Harnessing of Light Based on 2D Photonic Crystals
- p.14 **Agnès MAITRE / Laurent COOLEN** (INSP, Paris 6 U.)
Manipulating single-photon emission through coupling to photonic environments
- p.15 **Fabrice RAINERI** (LPN, CNRS Marcoussis)
Hybrid III-V Photonic crystal lasers on Silicon
- p.16 **Wataru NOMURA** (Tokyo U. and NRC)
Optical Near-field Signal Transfer Between 2-dimensionally Distributed CdSe Quantum Dots
- p.17 **Masaru TSUKADA** (Tohoku U.)
Theory of coherent vs dissipative electron transport of molecular junctions
- p.18 **Makoto NARUSE** (NICT Tokyo and NRC-Tokyo U.)
Information and Communications Aspects in Light-Matter Interactions at the Nanometer Scale
- p.19 **Giuseppe LEO** (MPQ- Paris7 U.)
A microcavity parametric source in the THz range
- p.20 **Naoya TATE** (Tokyo U. and NRC)
Hierarchy in nano-scale light matter interactions
- p.21 **Vasily KLIMOV** (Lebedev Physics Institute, Moscow)
Light and Matter Waves in the Presence of Negative Refraction Media: Unusual Properties and Applications
- p.22 **Jacques BAUDON** (LPL, Paris13 U.)
Coherent Atom Optics at the nano-scale
- p.23 **Ivan FAVERO** (MPQ, Paris7 U.)
GaAs Nano-Optomechanics : Nanophotonics meets Optomechanics
- p.24 **Andréï STASHKEVICH** (LPMTM, Univ Paris 13)
Magneto-optical interactions of light with magnons in nano-structured ferromagnetic media: magnonic and photonic aspects
- p.25 **Brahim LOUNIS** (CPMOH, Bordeaux U.)
Optical detection and spectroscopy of individual absorbing nano-objects
- p.26 **Anne DÉBARRE** (L.A.C., Univ Paris 11)
Two-photon excited photoluminescence of gold spherical particles dispersed in solution studied by Single Molecule Spectroscopy
- p.27 **Irène BURGHARDT** (Dép. Chimie, E.N.S. Paris)
Quantum dynamics of photoprocesses in extended molecular systems: coherence and dissipation at the nanoscale
- p.28 **Hiroyuki TAMURA** (Tohoku U.)
Quantum Dynamics of Excitons in molecular aggregates: The explicit modeling of electron-phonon coupling
- p.29 **Marc LAMY DE LA CHAPELLE** (CSPBAT, Paris13 U.)
Optical properties of gold nanowires: application to the Surface Enhanced Raman Scattering
- p.30 **Kokoro KITAMURA** (Tokyo U.)
Phonon-assisted visible light photocatalyst using ZnO nanostructure

Nanophotonics: Dressed photon science and technology

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This presentation reviews the recent progress of *nanophotonics*, which was proposed by the author in the year 1993[1,2]. The key is to utilize optical near-fields in order to realize novel nanometric device operation, fabrication, and energy conversion, etc., by the control of an intrinsic interaction between nanometer-sized materials. The nature of optical near-fields was studied by regarding the optical near-field as an electromagnetic field that mediates the interaction between nanometric materials. As a result, the physically intuitive concept of a *dressed photon* was established to describe optical near-fields, *i.e.*, the interaction between nanometric materials is mediated by exchanging dressed photons. The principles of device operation are reviewed considering the excitation energy transfer via the optical near-field interaction and subsequent relaxation. The operations of logic gates, an optical nano-fountain, a nano-coupler, a pulse generator, a phonon-assisted light emitter, etc. are described as well as their single photon emission and extremely low power consumption capability. Experimental results using quantum dots at the room temperature are described. Using a systems-perspective approach, the principles of content-addressable memory for optical router, a multilayer memory retrieval system, etc. are demonstrated.

This presentation also reviews nanophotonic fabrications based on phonon-assisted processes triggered by optical-near-field interactions. These processes represent *qualitative* innovation in photochemical vapor deposition and photolithography, suggesting that large, expensive ultraviolet light sources are no longer required, although they are indispensable for conventional adiabatic methods. A prototype of the commercial lithography system has been produced in collaboration with industry, and has been used for fabricating a diffraction grating and a Fresnel zone plate for the soft X-ray. Furthermore, phonon-assisted photochemical etching and sputtering were developed for realizing ultra-flat surfaces of glass and polycrystalline ceramics, respectively, in a self-organized manner. Phonon-assisted deposition will be also reviewed in order to increasing spatial homogeneity of the mol fractional ratio of In in a light emitting InGaN film.

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Dressing nature inherent to optical near fields at the nanoscale

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Recent experimental progresses have revealed a variety of curious and intriguing near-field optical phenomena at the nanoscale [1]. Such phenomena are now going to be applied to unique nanofabrication and nanophotonic devices leading to a novel functional system, where theoretical studies are expected to play a fundamental role. In this talk, we present an example of such intriguing near-field optical phenomena, and discuss a comprehensive picture to understand and formulate them, where dressing nature of photons is emphasized in the elementary process for a nanoscopic finite system interacting with “localized photons”, not “free photons”.

As a typical application of the phenomena, we discuss a new nanofabrication method with size and position of nanostructure controlled, paying a special attention to molecular vibrational excitation due to a phonon-vibration coupling of optical near field interactions [2]. It is noted that such molecular vibrational excitations are optically forbidden, i.e., prohibited by a far field.

As another example, we discuss the spin and excitation energy transfer between quantum dots driven by optical near fields, which results in a fundamental block of nanophotonic devices [3]. This kind of excitation transfer is also inherent in near-field optical interactions, and assisted by coherent/incoherent phonon excitations leading to the up-conversion [4].

Formulation and analysis of the dynamics driven by the near-field optical interactions are still under way; it is essential how to describe relaxation processes. We point out the importance of a structured reference system to discuss the constrained dynamics with relaxation, which might open a possibility of controlling the relaxation processes, creation of reaction fields associated with collective phenomena, and manifestation of a new functionality and system [5].

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ATOMIC SPECTROSCOPY AND NANO-OPTICS

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Our expertise in laser spectroscopy of atoms in the vapour phase has been mostly oriented towards the detection of atom-surface interaction (long-range van der Waals interaction) [1], and to the specificities of vapours confined in a sub-wavelength container [2].

Three different problems, partly related, are now under investigation in our group:

(i) The atom-surface van der Waals interaction can be seen as a cavity quantum electrodynamics modification of the atomic levels, owing to the modified limiting conditions for the vacuum, in the vicinity to the surface. When far infrared couplings are relevant for the considered atom or molecule, the atomic structure should depend on the vacuum temperature in an observable manner [3]. In the vicinity of the surface, these thermal effects are related to the near-field regime of a radiating blackbody [4], with a strong influence on the surface modes [5]. Our experiments aim to probe the thermal emission of the surface with a quantum (atomic) detector located at a distance ~ 100 nm, typical of selective reflection spectroscopy, in a "near-field" regime complementary to the one showing a temperature-dependent Casimir-Polder interaction with a Bose-Einstein condensate [6].

(ii) We have established that a dilute vapour confined in a narrow cell (1D confinement) exhibits sub-Doppler resonances [7] through a Dicke-type spectral narrowing [8]. We want to look for a genuine Dicke narrowing in the optical domain – instead of *rf* domain- for a vapour undergoing a 3D confinement. We are presently performing spectroscopy in the interstitial spaces of an opal of glass nanospheres [9], which is a prototype (with defects) of a photonic crystal. We expect that way to produce a generation of compact optical references that would be spectrally narrow; the theoretical understanding requires dealing with nano-optics, notably with the propagation of light within spheres of a sub-wavelength size.

(iii) Through nano-optics, or with various types of (spatially) complicated propagating fields it is possible to induce electromagnetic fields whose structure is complex on a wavelength scale, with for example strong differences between electric and magnetic energy. A well-chosen atomic or quantum probe, not restricted to the electric dipole approximation, can be a sensitive detector of these sub-wavelength structures [10]. As an example, we have recently shown [11] that a focused Laguerre-Gauss beam is not truly hollow, with a magnetic field on-axis.

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Radiative heat transfer at the nanoscale: enhancement due to surface phonon polariton

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It has been predicted theoretically that for distances smaller than the peak wavelength of the blackbody spectrum, radiative heat transfer can be increased by the contribution of evanescent waves [1].

This enhancement can reach several orders of magnitude for materials supporting resonances in the IR such as surface phonon-polariton or surface plasmon-polariton [2,3,4].

These effects have already been observed but a quantitative comparison is still missing. We have measured the heat flux between a sphere and a plate as a function of the sphere-plate distance [5]. We show that experimental and theoretical results are consistent with an accuracy better than 1% in the range 50nm-2 μ m. Particularly, the distance dependence is well reproduced by the theory.

For a sphere with a radius of 20 μ m, the heat flux exchanged with a plate is in the order of tens of nanoWatts. To measure the radiative heat transfer, a very sensitive device is needed.

The fluxmeter used in our experiment is a bimorph based on an AFM cantilever. Such fluxmeters can measure fluxes variations as small as 10 pW [6]. The bimorph bending is proportional to the flux and is measured using an optical interferometric technique based on the

device used in [7] to measure the Casimir force.

The experimental results are compared with theoretical calculations based on the Derjaguin approximation [8] commonly used to compute the Casimir force between a sphere and a plane. The transition regime between far field and near field is well reproduced with an agreement better than 1%.

These results strongly support previous theoretical works and pave the way to engineering radiative heat transfer in the mesoscopic regime. Possible applications are heat assisted magnetic recording or heat assisted lithography.

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Room-temperature operation of nanophotonic logic gates using InAs QDs in mesa structures

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We have already proposed nanophotonic logic gates that utilize energy transfer between quantum dots (QDs) caused by optical near-fields (dressed photons) and have been working on verifying the operation of AND gates and NOT gates by using CuCl QDs [1,2], ZnO QDs [3], and InAlAs QDs [2]. By using dressed photons, in addition to miniaturization and densification of elements beyond the diffraction limit of light, operation with 10^4 times less energy compared with electronic devices becomes possible [4]. Because of the light emission efficiency and uniform width of quantum dots, the operations of these elements have been restricted to no higher than liquid nitrogen temperature (77 K). We have now succeeded in depositing two layers of InAs quantum dots with favorable optical characteristics using MBE in SK-mode and achieved room-temperature operation of nanophotonic logic gates using these quantum dots, the results of which are reported here.

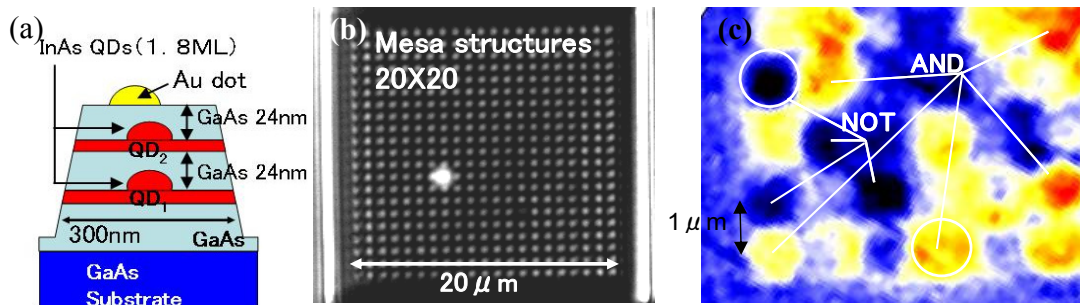


Fig. 1.

The dimensions of the elements fabricated were 300 nm on each side (Fig.1(a)), and the elements were arrayed two-dimensionally with an areal density of 10^8 elements/cm². Fig. 1(b) shows a microscope image of the high-density array of nanophotonic logic gates used for verifying the operation. The elements were arrayed at intervals of 1 μ m, and the number of elements in the array was 400. Fig. 1(c) shows the result of mapping for the change in the output signals at this time (integrated values in the wavelength range 1177–1240 nm). Dark and bright area correspond to the responses from the NOT- and AND- gates, respectively.

This paper belongs to “Innovative nanophotonics components development project” which OITDA contracted with The New Energy and Industrial Technology Development Organization (NEDO) since 2006.

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Subnanosecond spectral diffusion of a single quantum dot in a nanowire

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We have measured spectral diffusion of the photoluminescence of a single light emitter with a subnanosecond resolution. The technique we have developed [1] is based on correlations of photons emitted within a spectral window narrower than the spectral diffusion broadened line (cf fig 1(a)). Owing to the wandering of the homogeneous line, the emission energy stays a limited time within this spectral window leading to photon bunching as illustrated in fig. 1(b). The characteristic time of this effect is the spectral diffusion time and can be easily accessed by photon correlation (fig. 1(d,e)). The resolution is then only limited by the photon correlation set-up (800 ps with high efficiency avalanche photodiodes (APDs), or 100 ps with fast APDs). This is, to our knowledge, 4 orders of magnitude better than the best time resolution so far [2]. In fig. 1(f) we show the characteristic diffusion time as a function of pumping power and temperature for a CdSe quantum dot embedded in a ZnSe nanowire [3].

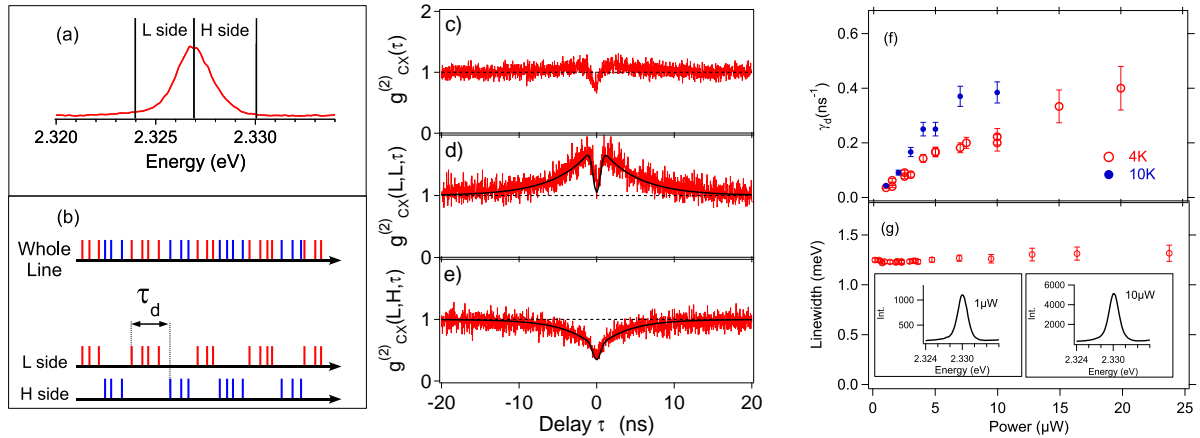


Figure 1. (a), Spectral profile of the emission of the charged exciton transition of a single QD integrated during 1 s. The two halves of the profile are labeled L and H. (b), Scheme of the photon time distribution in the two halves of the profile illustrating the bunching on the autocorrelation on one half (see (d)) and the antibunching for the cross-correlation between the two halves of the profile (see (e)). The minimum time delay between photons is of the order of $\tau_{CX} = 600ps$ and leads to the narrow antibunching dip in (c) and (d). (c), Auto-correlation of the whole profile. (d), Auto-correlation of one half of the profile. (e), Cross-correlation between the two halves of the profile. All data (c-e) shown in this figure have been obtained on the same QD with the same excitation power. (f), Diffusion rate as a function of exciting power at $T=4K$ and $T=10K$. (g), Corresponding linewidth as a function of exciting power. Insets: example of two spectra taken at different powers showing that not only the linewidth but also the shape is unchanged.

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Nanospectroscopy of single quantum dot ultrasmall absorption

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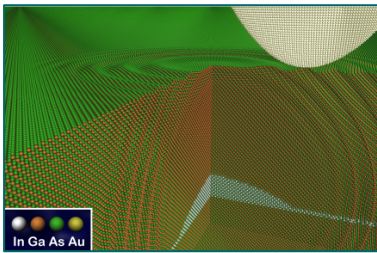
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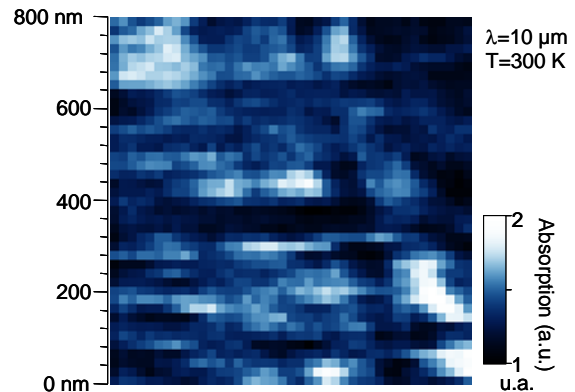
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The spectral and spatial resolution of semiconductor *absorption* at the nanometer scale is a fundamental step for the exploration of the semiconductor nanomaterial properties. Absorption nanospectroscopy is particularly challenging in the infrared spectral range because single nanostructure absorption occurs well below the diffraction limit and leads to very small expected transmission variations (10^{-9}). All optical techniques like near field optical measurements in the infrared or non-linear

susceptibility sensitive detection in the visible have been carried out on large absorption single semiconductor or metallic nanostructures. We show that both spectral and nanometric spatial resolution of single semiconductor quantum dot *ultrasmall* absorption can be reached with unmatched sensitivity by detecting locally the deformation resulting from the absorption of incident resonant optical pulses. That corresponds to a local measurement mimicking - at the nanometer scale - early 1880 A. G. Bell's photoacoustic experiments based on a photothermal effect. In practice absorption nanospectroscopy is achieved by coupling at room temperature an atomic force microscope (AFM) with a picosecond laser excitation as developed at the free electron laser facility CLIO. In InAs/GaAs self-assembled quantum dots, infrared absorption corresponds to the optical manipulation of a confined electron between discrete confined states originating from the conduction band. The local deformation originates from the emission of phonons by the buried *n*-doped nanostructures in the strong electron-phonon coupling regime. Spatial resolution as small as a few tens of nanometers is obtained for wavelengths as large as 10 μm and 25 μm ($\lambda/500$), enabling to work at the single quantum dot level even in buried nanostructure high density samples. Spectral resolution of model S-P or S-D intersublevel transitions, obtained by using the tunability of the pumping laser or by analyzing the two-dimensional absorption imagery, unravel the homogeneous line widths at room temperature at the single nanostructure level. The absorption nanoscopy is compared to the imagery and spectroscopy of macroscopic materials with calibrated absorptions such as SiO_2/Si microdisks or GaAs bulk substrates.[2] These explorations open a new investigation direction of single semiconductor quantum dots considered as localized thermal and acoustic nanosources with phonons emission controlled by optical excitation (nanophononics).



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Nano photonic devices using multiple quantum structures

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To realize nano-scale photonic device, i.e., nanophotonic device, we proposed nanophotonic signal devices (NPD) that consist of semiconductor quantum dots (QDs) [1]. These NPD devices operate using excitons in QDs as the signal carrier, due to optical near-field interactions between closely spaced QDs. To realize nanophotonic device, we use ZnO quantum structures because ZnO has large exciton binding energy than thermal energy. I will present recent works about nanophotonic devices using (1) ZnO nanorod quantum-well-structures (QWs) and (2) colloidal ZnO QDs.

(1) ZnO nanorod QWs

We fabricated ZnO quantum-well-structures (QWs) on the end of ZnO nanorod, in which well width and the separation of QWs was controlled with atomic scale controllability. Through time-resolved near-field spectroscopy of ZnO/ZnMgO nanorod double-QWs, we observed AND-gate operation by controlling the exciton excitation in the dipole-inactive state via an optical near-field [2]. Furthermore, we successfully observed a superradiant effect from multiple-QWs. The excitation power dependence of the time-resolved photoluminescence of ZnO nanorod MQWs revealed increased emission intensity with a shorter decay time compared to that of single-quantum-well structures when the excitation power was increased. This indicates cooperative emission from the MQWs.

(2) Colloidal ZnO QDs

We developed a self-assembly method that aligns nanometre-sized QDs into a straight line along which photonic signals can be transmitted by optically near-field effects [3]. ZnO QDs were bound electrostatically to DNA to form a one-dimensional QD chain. The photoluminescence intensity under parallel polarization excitation along the QDs chain was much greater than under perpendicular polarisation excitation, indicating an efficient signal transmission along the QD chain.

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Quantum dots coupled to solid-state cavities : potential and applications to advanced nano-photonic devices

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In this talk I will give an overview of our research framework, namely the study of cavity quantum electrodynamics effects (CQED) for solid-state emitters like quantum dots (QDs) coupled to semi-conducting cavities. Thanks to technological progresses in the field of solid-state physics, a wide range of quantum optics experiments previously restricted to atomic physics, can now be implemented using QDs and semi-conducting cavities. Still, a QD is far from being an isolated atom. As a matter of fact, solid state emitters are intrinsically coupled to the matrix they are embedded in, leading to decoherence and phase relaxation that unavoidably broaden any transition between the discrete states of such artificial atoms. At the same time, very high quality factors and ultra small modal volumes are achieved for state of the art cavities. These new conditions open an unexplored regime for CQED so far, where the emitter's linewidth can be of the same order of magnitude, or even broader than the cavity mode one.

Under proper conditions, this broadening can safely be modeled by a simple pure dephasing channel in the master equation describing the dynamics of the system. Because of its simplicity, the scheme of a two-level system undergoing pure dephasing can be regarded as an appealing tool to explore this new regime of CQED, as well as a useful effective model to describe solid-state emitters. First we have computed the spectrum emitted spontaneously by a quantum dot coupled to an arbitrarily detuned single mode cavity, taking into account pure dephasing processes [1]. We have shown that if the emitter is broad, the cavity can efficiently emit photons with its own spectral characteristics even if the two systems are detuned. This effect opens unique opportunities for the development of devices exploiting both cavity quantum electrodynamics effects and pure dephasing, such as wavelength stabilized single photon sources robust against spectral diffusion. This motivates an important experimental work currently under progress in our group with the financial support of the ANR ("CAFE" project).

We have also studied the influence of pure dephasing on the dynamics of the coupling between a two-level atom and a cavity mode. We have derived an effective atom-cavity coupling rate that is shown to be a key parameter in the physics of the problem, allowing to generalize the known expression for the Purcell factor to the case of broad emitters, and to define strategies to optimize the performances of broad emitters-based single photon sources. Moreover, we have evidenced that pure dephasing is able to restore lasing in presence of detuning [2]. These effects clearly show that decoherence, far from being a drawback, is a fundamental resource in solid-state cavity quantum electrodynamics, offering appealing perspectives in the context of advanced nano-photonic devices.

I will also mention another important axis of research of our group, namely the experimental study of a single QD weakly coupled to a directional microcavity like a micropillar, or embedded in a photonic nanowire. Such a system provides a solid-state realization of a so-called "one-dimensional atom", which has been shown to be a giant optical non-linear medium, sensitive at the single photon level [3]. This device opens promising perspectives in the frame of integrated photonic computation. This work has motivated the building of a consistent set of measurements of the Purcell factor experienced by a QD coupled to a micropillar [4].

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Optical instabilities and blinking phenomena in the emission of InGaN quantum wells

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We observed intriguing optical instabilities in InGaN quantum well devices. The transition to maximum photoluminescence of InGaN single quantum wells is a phenomenon that has time constants in the range of few seconds. Using a systematic illumination/darkening procedure we found that these characteristics are related to previous stimulations as if the sample has a memory of past illumination events. Choosing opportune time sequences, time constants were observed to vary more than 100%. These facts suggest the presence of carrier trapping/de-trapping processes that act beyond the single illumination event, accumulating over time in a complex effect.

A second type of observation is the presence of confined optical random-like blinking instabilities in the photo-luminescence. We investigated this by a modified scanning near-field optical microscope (SNOM) and studied the time/intensity signal of this phenomenon in confined 100nm domains. The dynamics of the blinking optical emission was time-resolved and its behavior studied with Fourier analysis. Despite the intensity oscillations were found to have a chaotic term (correlation coefficient is about 0.63), we found a characteristic Fourier peak in the signal in the range of 5 seconds. This long duration value suggests the presence of diverse recombination processes that emerge in intriguingly slow dynamics. We will present details about these observations, show movies with recordings of the actual photoluminescence instabilities and discuss possible models to interpret the mechanism involved.

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Fundamental processes of optical near-field interactions

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Study of optical near-field interactions between nanometer-sized isolated electronic systems reveals number of basic problems remain unsolved related to electrodynamics and quantum mechanics. One of the most important issues is the optical excitation transfer via optical near-field interactions. Due to the nature of strong light-matter coupling, the optical properties of matter and the nature of electromagnetic fields are strongly modified from the mean-field properties by the environmental structures and the excitation/dissipation processes. Accordingly, the physical quantities conserved during the near-field interaction turn out to be dependent on the environmental conditions. For instance, the pseudo-momentum and pseudo-angular momentum transfer processes have been experimentally demonstrated in our previous studies of optical near-field spectroscopy of atoms^{1,2)} and optical near-field light scattering due to nano-particles³⁾. In order to treat the problems of optical near-field excitation transfer, we should employ theory of electromagnetic interaction based on the half-space problems where evanescent electromagnetic waves play the dominant role. Several problems related to elementary processes have been solved on the bases of angular-spectrum representation of scattered fields and second quantization of electromagnetic fields involving evanescent waves employing the triplet-mode descriptions related to the optical sources and detectors^{4,5)}. For further developments towards theoretical description of nano-to-macro coupling and evaluation of optical functions of sub-wavelength space, we should include the hierarchical properties of optical near-field interactions⁶⁾, the partly coherent and partly dissipative processes of near-field interactions reflecting the local environment, the natures of non-equilibrium open system related to the source-to-detector problem, and so on. In this study, we consider the physical meaning of function in nanometer space and discuss on the fundamental processes of optical interactions of nanometer scale from these view points.

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NON CONVENTIONAL APPROACHES FOR 3D HARNESSING OF LIGHT BASED ON 2D PHOTONIC CRYSTALS

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The flow of innovations whose threshold has been initiated in the late 1980 by the introduction of the concept of Photonic Crystals (PC) is still very close to its source and will inflate in the future to an extent which is certainly beyond our full consciousness. 1D-2D PC semiconductor membrane structures have proved to be central actors in that respect, since this seminal event. Most of recent innovations in the field of 2D Micro-Nano-photonic integration, where devices are restricted to operate in the wave-guiding configuration, are based on this essential building block.

Major extensions of 2D planar photonic integration have been proposed recently by our group:

- In the so-called 2.5D Micro-Nano-Photonics approach, to be considered as an intermediate step between 2D and full 3D Micro-Nano-Photonics, multi-layered 1D-2D PC membranes provide a quasi-3D harnessing of the light at the wavelength scale, via the controllable interplay occurring between wave-guided confined photons and radiated photons propagating through the planar multilayer structure. Very generic and versatile photonic building-blocks can result from this approach: examples of innovative devices produced at INL along this line will be presented.
- Another generic approach for 3D light harnessing based on coupling engineering between 1D-2D Photonic Crystal membranes and 0D photonic structures has been proposed by our group: in this new approach, unconventional use of 1D-2D Photonic Crystal membranes as a universal photonic platform is proposed for light addressing of 0D photonic structures. As an illustration, efficient addressing of high Purcell factor metallic nano-antenna, using a large quality factor PC membrane resonator will be presented.

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Photonic Crystal based Nanophotonic Integration

Tutorial: 14th ECIO'08, Eindhoven, the Netherlands (11-13 June 2008)

A. BELAROUCI, T. BENYATTOU, X. LETARTRE, P. ROJO-ROMEIO, TAIPING ZHANG, P. VIKTOROVITCH

3D light harnessing based on coupling engineering between 1D-2D Photonic Crystal membranes and 0D photonic structures, **ICTON 2010, Munich, Germany (June 27 - July 1, 2010)**

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Manipulating single-photon emission through coupling to photonic environments

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Our group studies the modification of the fluorescence of a nano-emitter depending on its environment. We consider colloidal nanocrystals as the nano-emitter, and couple them to various photonic environments : 2D or 3D photonic crystal, micropillar ...

Semiconductor colloidal nanocrystals are spheres of a few nanometers obtained by chemical synthesis. In these structures, the fluorescence emission wavelength is determined by the nanocrystal size (quantum confinement), which can be controlled by the synthesis parameters. By use of a confocal microscope, a single nanocrystal can be observed. It can then be shown (even at room temperature) that a nanocrystal emits single photons : it emits photons one by one, and never two at the same time.

We have considered the fluorescence of single nanocrystals in the vicinity of a gold surface [1]. We measured lifetime modifications up to sevenfold and modifications of detected intensities up to 2.5-fold. By using an analytical model, we found a good agreement with the experimental results and we distinguished the importance of the various effects of the gold surface (interferences, plasmons and other non radiative decay channels...).

We also considered the fluorescence of an ensemble of nanocrystals infiltrated in a silica opal fabricated at the ISSP in Russia [2]. Opals are 3D photonic crystals obtained by self-assembly of colloidal spheres of a silica or polymer colloidal spheres. We were able to evidence a 10 % photonic-crystal effect on the nanocrystals fluorescence decay time. We are now developing an opal fabrication activity at the INSP.

We are presently collaborating with the LAAS in Toulouse and the LPN in Marcoussis in order to fabricate respectively some cavities inside planar photonic crystals and some micropillars. These structures are usually designed to work in the infrared ; new materials must be used to work in the visible range. We are especially interested in “deterministic” fabrication procedures, where the position and the resonance of the cavity can be adjusted to match the emitter.

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Hybrid III-V Photonic crystal lasers on Silicon

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In the quest for efficiency, integrated optics has recently turned its attention to development of sub-wavelength, high-contrast photonic components. Preferably based upon a Silicon platform, these would allow one to envision low-cost and high component density. To achieve these ambitious goals Si technology takes advantage of processing know-how from the electronics industry. Mature CMOS processing technology renders Silicon unsurpassed in terms of fabricated device quality. Furthermore, the ability of low-loss Si wire waveguides to be bent with tight radii of curvature predisposes them to small device footprints and thus large scale integration. Beyond purely passive features such as guiding and filtering, the inclusion of active components in the Si platform would vastly enhance the portfolio of optical functions. For this we require devices capable of emitting, modulating and detecting light, ideally with low power expenditure. Heterogeneous integration of III-V materials onto Silicon could offer an efficient solution, providing tailor-made optoelectronic properties. Patterning this active material on the wavelength scale also permits control of its photonic properties. In this context 2D Photonic Crystals (PhCs) allow us to achieve ultra-small components and can be configured to substantially augment the light-matter interaction, thus reducing laser thresholds and switching energies. Initially, our effort was concentrated on the exploration of the possibilities offered by III-V PhCs bonded onto unpatterned Si wafers. High contrast modulation was achieved using a BCB bonded InP-based surface emitting PhC [1]. Subsequently, graphite lattice PhC structures were integrated onto a Si wafer using AuIn eutectic bonding yielding continuous wave (CW) laser operation. This was due to the 5-fold higher thermal conductivity of the SiO₂ cladding [2]. Rapid modulation of these PhC lasers integrated on Si was also demonstrated by dynamic measurements of the pulse turn on/off [3]. These demonstrations pointed the way to all-guided hybrid systems, which is now our primary concern. The enhancement of silicon photonics by III-V PhC based optical functions would make for a versatile integrated photonics platform.

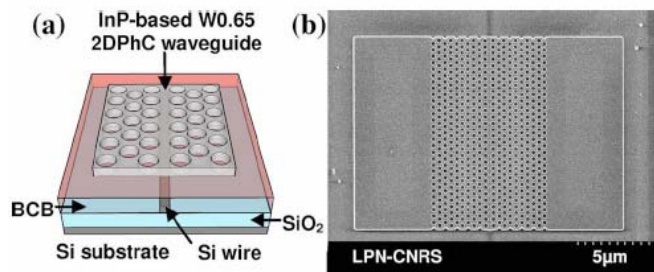


Fig. 1. a) Schematic of the sample. B) SEM image of the sample

With this in view, we study the integration of a PhC membrane containing InGaAsP/InGaAs quantum well material and SOI wire waveguides, using die-to-die bonding (as shown in Fig. 1) [4]. For this heterogeneous integration, we choose adhesive bonding as it is a rapid, convenient alternative to molecular wafer bonding. We demonstrate laser emission at telecom wavelength at room temperature from InP-based PhCs heterogeneously integrated to SOI wires. The SOI circuitry is used both for collecting the laser emission from, as well as, for channelling the optical pump to, the 2DPhC. Excellent performance was obtained and pump threshold energy as low as 1pJ was obtained [5].

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Optical Near-field Signal Transfer Between 2-dimensionally Distributed CdSe Quantum Dots

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Optical near-field interactions have allowed realization of not only integration below the diffraction limit, but also various new functions (e.g., nanophotonic devices) [1]. In order to utilize optical excitation transfer via optical near-field interactions in devices and systems, an understanding of the signal transfer between quantum dots (QDs) is essential. In this research, optical near-field signal transfer array dependency of QDs was theoretically and experimentally analyzed with respect to the basic properties of signal transfer using optical near-field interactions.

We observed optical signal transfer between 2-dimensionally distributed CdSe/ZnS core-shell QDs layers using optical near-field microscope. Figure 1 (a) shows measured results of three samples, whose heights of QDs layer H were 10, 20 and 50 nm. We evaluated the optical signal transfer distance T_0 from these results, and found that T_0 increased with an increase of H . As shown in Fig.1 (b), the trend in the shows good agreement with the theoretically calculated T_0 using a rate equation.

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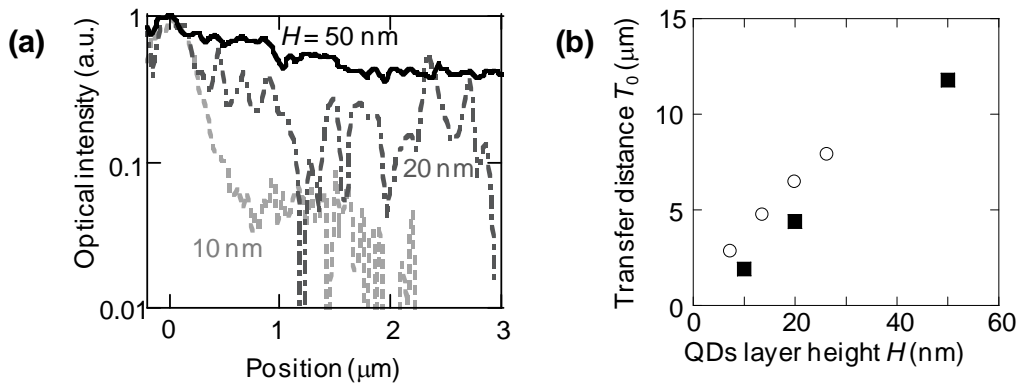


Fig. 1 (a) Optical signal profile of distribution between CdSe/ZnS QDs layers. (b) Transfer distance T_0 of 2D QDs layer. Closed squares and open circles obtained by experimental result and theoretical calculation, respectively.

Semiconductor plasmon sources

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Surface plasmons (SPs) are electromagnetic waves originating from electrons and light oscillations at metallic surfaces. They propagate in a direction parallel to the metal-dielectric interface. Three main techniques are available for the excitation of SPs: total internal reflection *via* prism coupling; scattering from a topological defect (small holes in a thin film); use of periodic corrugations in the metal's surface (gratings).

A compact electrical device capable of SP generation and injection into passive components would represent an important step towards “plasmonic electronics”. The task is very challenging, since SPs cannot be optically excited unless the missing momentum between free photons and SPs of the same frequency is provided.

We will present a new method for the generation of SP via a semiconductor laser operating in the mid infrared ($\lambda = 7.7\mu\text{m}$), whose metallic top cladding has been structured [1,2]. We *directly* prove the generation of surface-plasmon polaritons by reproducing - in the near-field and with electrically generated surface-plasmons - the slit-doublet experiment. The demonstration is based on scattering-type scanning near-field optical microscopy (s-SNOM), which reveals the plasmonic waves on the device top metallic surface [3]. Furthermore, operating at longer wavelengths allows one to demonstrate the principle and – simultaneously – to take advantage of reduced plasmonic losses.

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Theory of coherent vs dissipative electron transport of molecular junctions

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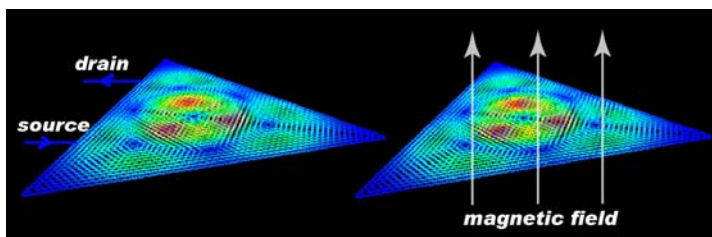
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Mechanisms of electron/exciton transport through molecular junctions as well as molecular layers sandwiched between electrodes are fundamental issues of molecular electronics and molecular photonics. In spite of a number of theoretical studies performed on these problems, comprehensive understanding of the transport through molecular junctions has not been given yet. In the present talk, we will introduce some fundamental features of the coherent transport and mechanism of de-coherence leading to dissipating transport based on theoretical analyses.

First we will introduce a novel interpretation of the transition voltage spectroscopy (TVS), and its significance to the issue of the molecular level positions relative to the electrode Fermi level. Then some characteristic features of the resonant/off-resonant quantum tunneling effects are introduced, including an extreme sensitivity of the molecular transport on the linkage part of the molecule. Remarkable quantum nature appearing in the resonant tunneling through degenerate molecular levels is a strong loop current in molecules[1]. The relationship with the persistent current induced by the magnetic field is discussed.

When the electron couples with the atomic vibration, or the electro-magnetic environment, the electron energy is dissipated to the external systems and de-coherence is switched on. A theoretical framework is proposed to treat the competition and/or coexistence of the coherent and dissipating transport[2]. In this context, some interesting features of the electron transport are predicted as the zero bias anomaly of the I-V curve, as well as quasi-polaron nature of the transport through molecular thin layers.



Distribution of the large loop current induced by the source-drain current (Left) and the magnetic field (Right).

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Information and Communications Aspects in Light-Matter

Interactions at the Nanometer Scale

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Theoretical fundamentals of optical near-field interactions at the nanometer scale have been intensively studied through the development of, for example, dressed photon models [1] and angular-spectrum analysis [2]. Experimental technologies, both in fabrication and characterization of nanostructures, have also seen rapid progress such as in geometry-controlled quantum nanostructures, advanced spectroscopy, and so forth [1]. Besides those physical insights and experimental methodologies, it is important to grasp the fundamental properties from the viewpoint of information and communication to maximize their ultimate capabilities [1-6]. We examine optical near-field interactions taking an approach from fundamental aspects of information processing and communication. In particular, we focus on two unique attributes available on the nanometer scale light-matter interactions; one is the optical excitation transfer via optical near-field interactions and the other is hierarchical properties of optical near-fields. We discuss the unique characters of signal transfer, interconnects, and computational state variables based on optical excitation transfer [1]. We also examine their tamper resistance [3], robustness to disturbances [4], and minimum energy dissipation as novel attributes associated with optical excitation transfer. The functional aspects of hierarchical nature of optical near-field interactions are also discussed [2,5] including their information theoretical analysis [6].

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A MICROCAVITY PARAMETRIC SOURCE IN THE THZ RANGE

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The last decade has witnessed an increasingly strong interest for the THz in the scientific as well as the commercial, military, and security fields. Despite the considerable demand in terms of possible applications, the “ideal” THz system (source + coherent detector) is still lacking.

In this presentation we propose a radically new CW, electrically pumped terahertz emitter, designed to operate at room temperature. Compared to existing THz sources, this source will bring together several crucial advantages that are far from being simultaneously available in any existing source today: compact size, power in the μW range, custom frequency from 2.4 to 6 THz, spectral purity, feasibility of multi-spectral array of emitters, and perspective of coherent detection schemes.

Various routes to THz generation have been explored to date in different materials, based on difference-frequency generation (DFG) or parametric generation. However, most of the existing THz nonlinear sources are passive and require external pulsed pump laser sources, making the overall systems neither compact nor practical outside research laboratories. The prospect of integrating optical pump and frequency converter greatly simplifies the packaging and alignment of devices, thus promising major improvements in yield and cost reduction. Today, the most relevant example in this direction is given by a THz source based on DFG in a dual-wavelength mid-IR QCL.¹ However, this device is severely limited by free-carrier absorption (FCA) and thermal dissipation problems, and can hardly emit a few hundreds of nW at room temperature, even in the pulsed regime.

GaAs/AlGaAs microdisks (MDs) can sustain high-Q whispering-gallery modes (WGMs) in the near IR. When quantum dots (QDs) are inserted in such MDs, their inhomogeneous gain allows simultaneous lasing of several WGMs without mode competition, even for pumping levels well above threshold. THz generation by DFG from these WGMs is possible with MD diameters above 20 μm , provided that a proper phase-matching (PM) scenario is available. In our case, two PM schemes are simultaneously used for THz DFG: modal PM of THz and near-IR modes, associated to the anomalous dispersion created by the reststrahlen band;³ and the quasi-PM associated to the effective azimuthal periodic modulation of $\chi^{(2)}$ in (100)-GaAs.⁴

Based on these grounds, our MD source is etched in a GaAs/AlGaAs double heterostructure and capped on both sides by an Au mirror, to ensure both electrical injection and a double-plasmon, vertical confinement of the THz mode. InAs QD arrays located within the GaAs layer are used as active medium. At 300 K, the homogeneous broadening of QDs is of the order of 10 meV, which will restrict typical THz emission to frequencies higher than 2.4 THz. Conversely, an upper limit of 6 THz is set by the rest-strahlen band of GaAs. Preliminary calculations predict 1 μW THz edge emission at 4.8 THz (i.e. $\lambda_3=63.4 \mu\text{m}$) for $\lambda_1=0.923\mu\text{m}$ and $\lambda_2=0.936\mu\text{m}$. With the low quality factor in the THz playing an important role in the power extraction at λ_3 , this device behaves like an almost ideal point source in the THz. We also predict that λ_3 scales with the radius, allowing e.g. 2D arrangements of THz emitters with slightly different radii, to be exploited as a practical means for obtaining a multi-spectral emission.

Thanks to the coherent nature of DFG, our concept paves the way to the fabrication of a dual THz detector, in which an impinging THz wave could couple to a QD-laser local oscillator, up-converting it through a sum-frequency-generation process. What we envisage is therefore an “ideal” system of THz integrated sources and coherent detectors, based on the same technological platform.

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Hierarchy in nano-scale light matter interactions

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Optical near-field interactions respond hierarchically at the nanometer scale, allowing unique nanophotonic functions¹. For engineering hierarchical system based on Nanophotonics, the size, shape, and composition of nanostructures are important physical entities. In this presentation, we will demonstrate these hierarchical functions numerically and experimentally by using several prototype optical elements.

Our *Nanophotonic hierarchical hologram* is proposed as one of such applications utilizing the hierarchy of nanophotonics². It works in both optical far- and near-fields, the former being associated with conventional holographic images, and the latter being associated with the optical intensity distribution based on a nanometric structure that is accessible only via optical near-fields. In principle, the phenomenon occurring at a subwavelength scale does not affect the function induced by propagating light. Therefore, the visual aspect of the hologram is not affected by such a small structural change on the surface. We propose embedding a *nanophotonic code*, which is retrievable via optical near-field interactions involving nanometric structures, *within* an embossed hologram. Due to the one-dimensional grid structure of the hologram and concentration of oscillated surface charge distribution, evident polarization dependence appears in retrieving the code.

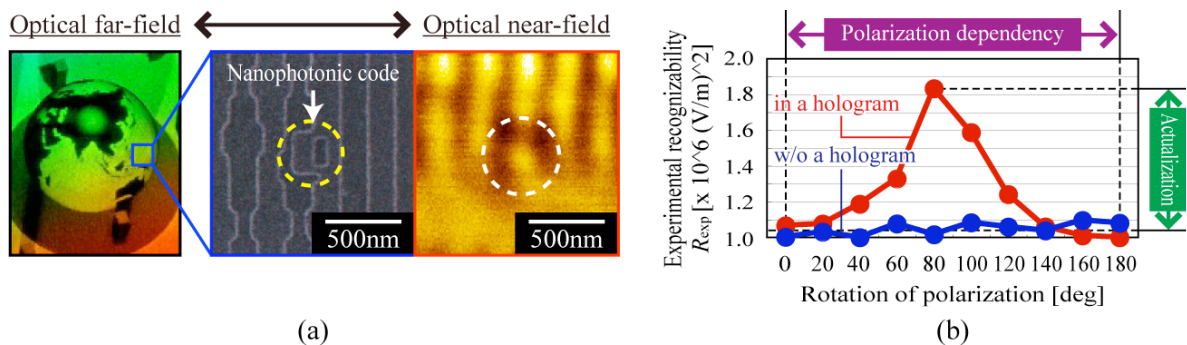


Figure 1: (a) Fabrication of a nanometric structure as a nanophotonic code within the embossed structure of hologram. (b) Experimental recognizability R_{exp} of embedded nanophotonic code and that of isolated nanophotonic code. Evident recognizability and polarization dependency were exhibited.

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Light and Matter Waves in the Presence of Negative Refraction Media: Unusual Properties and Applications

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Despite the huge interest in negative refraction media (or negative index media - NIM), where phase and group velocities have opposite directions of propagation, no clear analytical solution of this problem was found until now neither for Maxwell nor for Dirac fields. The absence of exact solutions results in many unproved and incorrect statements and application proposals. In this talk, we shall present exact analytical solutions both for matter and light wave outgoing from a point source and suffering negative refraction in the half-space region or in a slab with negative refraction. It is a unique feature that for a *lossless* negative refraction material with $n=-1$ the theory of images can be applied allowing one to find an exact solution in spatial domain as a combination of real and imaginary sources (sinks) and free propagating surface plasmon waves. In Fig.1 energy flows for light (left panel) and massless electrons (right panel) found within our analytical solutions, are shown.

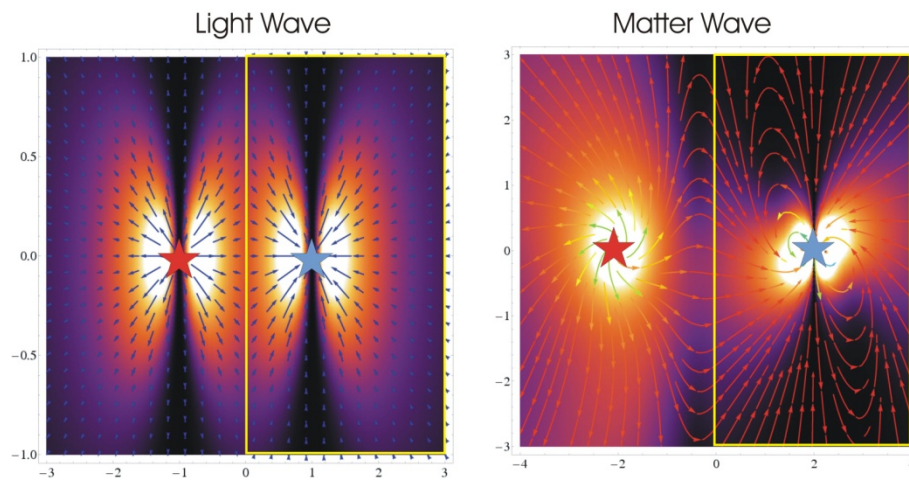


Fig.1. Energy flows in focusing of light (left panel) and matter (right panel) waves by a negative refraction half-space without losses. The yellow box indicates the negative refraction half-space. Sources and sinks are labeled by red and blue stars correspondingly.

From these figures, one can see that the behavior of energy flows differs for photons and electrons. While for the light we have straight flow lines with a sharp bend at the interface of the negative refraction half-space, for the electrons we have more complicated curves. Moreover, negative refraction for electrons, in fact, begins slightly in front of region of the negative refraction half-space. Other important feature of our solutions is the necessity to introduce singularity (sink) in the negative refraction half-space. This sink together with a source of field forms a pair that is analogous to particle-antiparticle pair explaining the Klein paradox. So, our results are another manifestation of the Klein paradox. The effects predicted can be verified more easily in the case of massless electrons in graphene.

In the case of arbitrary small losses, there are also other solutions, where the singular sink of energy is replaced by distributed sink of energy, that is by surface plasmon waves. This branch of solution corresponds to a so-called superlens where image appears as surface plasmons at the interface. It is very important that our approach allows tracing appearance of both solution with point singularities and surface plasmon waves as a result of bifurcation of unique solution valid for a source placed far enough from a negative index material slab. As a result of this bifurcation, creation of a pair of source-sink or a pair of surface plasmons propagating in opposite directions takes place.

For some other geometries (sphere, cylinder, wedge, cone etc) we have found analogous solutions.

Possible applications of our solutions (preparation of entangled pairs of atoms, effective excitation of atom or molecules) will be also discussed.

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Coherent atom optics at the nanoscale

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Recent advances in extending coherent atom optics processes in the nanometre range are reviewed. Generalising nano-optics and nano-photonics approaches to the optics of matter waves allows one to introduce novel concepts in this field. It includes atomic Fresnel bi- and poly-prisms, negative-index meta-materials specially devised for atom optics, coherent non-diffracting, atom nano-beams, etc. Special attention will be devoted to the mutual interaction of atoms and fields at the nanoscale, the richness of the corresponding field and its applications.

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GaAs Nano-Optomechanics : Nanophotonics meets Optomechanics

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The coupling of mechanical oscillators with light has seen a recent surge of interest: applications of « optomechanical systems » are rapidly evolving, from sensing to fundamental tests of quantum mechanics. For recent reviews see ^{1,2}. At the nanoscale, the optomechanical coupling increases thanks to a smaller optomechanical interaction volume and reduced mass of the mechanical oscillator ^{3,4}.

We will present GaAs nano-optomechanical resonators implemented directly on chip and which combine all assets for nano-optomechanics experiments: pg motional mass, high mechanical frequencies, nanoscale optical mode volume and high optical Qs. These resonators consist of GaAs disks evanescently coupled to a tapered Silica or GaAs nano-waveguide. We have observed record optomechanical coupling in these systems, mechanical frequencies up to the GHz, and obtained a shot-noise limited motional sensitivity of 10^{-17} m/ $\sqrt{\text{Hz}}$ in ambient conditions. These results show an example of how nanophotonics systems can bring benefits to optomechanics experiments, be it for fundamental perspectives or applications.

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Magneto-optical interactions of light with magnons in nano-structured ferromagnetic media: magnonic and photonic aspects

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Nano-structured ferromagnetic materials are interesting from several points of view. Apart from demonstrating rich physics of wave phenomena, they are regarded as potential candidates for many important applications such as high-density magneto-optic (MO) recording and microwave signal treatment. Not surprisingly, their high frequency dynamics has continuously driven technological as well as academic research.

In the framework of this talk we will concentrate on the following two major aspects. Similar oscillatory nature of both optical and spin waves (SW) dictates direct parallels in the physics of wave propagation in periodically structured media. More specifically, Bloch modes underlying wave behaviour both in photonics and magnonics are characterised by appearance of Brillouin zones and, as a result, pronounced variation of dispersion. The latter manifests itself through such features as creation of zones in the $\omega - K$ space characterised by slow light/SW propagation or by a negative group velocity. However, in magnonics these features superimpose on already existing highly peculiar SW dispersion characteristics [1].

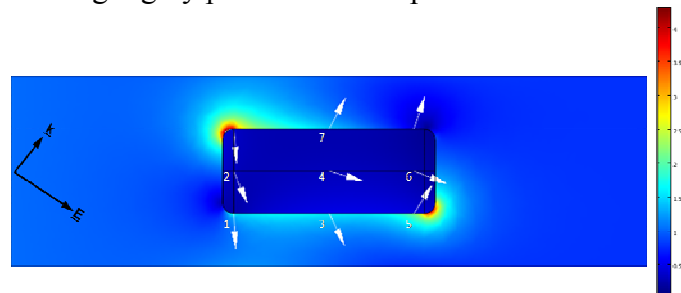


Figure. Near field pattern of the diffraction of light, leading to the magnetic cylinder being “flown over” by the optical wave, which leads to light penetrating inside from all sides(finite element ComSol simulations) .

Another point of particular interest addressed in this talk is the influence of the film structuring on the mechanisms of MO interactions especially on its cross section. The latter, as is known, can be significantly improved by resorting to photonic effects, for example localised optical Tamm states in 1D photonic crystals [2]. This part of the talk will be illustrated with the original results obtained on nano-composite ferromagnetic films comprised of Ni nano-wires imbedded in an Al_2O_3 matrix [3] (see Figure).

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Optical detection and spectroscopy of individual absorbing nano-objects

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In the fast evolving field of nanoscience, where size is crucial for the properties of the objects, simple and sensitive methods for the detection and characterization of single nano-objects are needed. The most commonly used optical techniques are based on luminescence. Single fluorescent nanoobjects have been studied on their own and are now routinely applied in various research domains ranging from quantum optics to life science. Yet, fluorescence methods allow only for short observation times due to inherent photo-bleaching. An interesting alternative relies solely on the absorptive properties of the object. In general, nanoparticles with large absorption cross sections and short time intervals between successive absorption events are likely candidates for detection with absorption methods. We have demonstrated a new two-color photothermal heterodyne technique for the detection of small absorbing nanoparticles. This photothermal method has been applied to the detection of individual metal nanoparticles, non-fluorescent quantum dots and single walled carbon nanotubes. The absorption spectroscopy of these systems at the single particle level is performed for the first time.

Two-photon excited photoluminescence of gold spherical particles dispersed in solution studied by Single Molecule Spectroscopy.

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The functionalization of gold particles is an important matter regarding the development of new hybrid probes for applications in sensing and molecular recognition. The strong activity in the field is explained by the development of robust protocols to protect gold nanoparticles and to link molecules to the gold surface by attachment to a ligand.

The main issue concerns the ability to control the emission properties of the nearby chromophore, which depends not only on the nature of the molecule itself but also on the plasmonic properties response of the particle. Most hybrid systems studied so far, are composed of a metallic core and a ligand shell, feature quenching when dispersed in solution [1]. An alternative to fluorescence to track their location is to exploit their luminescence under two-photon near IR excitation. This intrinsic optical response could offer the opportunity of bypassing the specific functionalization protocol needed to anchor chromophores and favor the use for future bioanalytical sensing of gold nanoparticles capped with usual ligands as such.

The interest in the luminescence of gold nanoparticles has been recently renewed in this context [2, 3]. Whereas the two-photon excitation process and the role of the surface plasmon in the emission efficiency are almost understood, the mechanism of de-excitation is still unclear. Moreover, almost all the studies have been performed on deposited particles, and the role of the substrate cannot be discarded.

We have been investigating the properties of the luminescence of gold spheres in aqueous suspensions, at the single particle scale by two-photon Fluorescence Correlation Spectroscopy (TP-FCS). We will present recent results that demonstrate the role played by the ligands capping the particle in the luminescence, and the consequences on its excitation wavelength dependence [4].

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Quantum dynamics of photoprocesses in extended molecular systems: coherence and dissipation at the nanoscale

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The photophysics of extended systems like conjugated polymers or molecular aggregates is characterized on the one hand by the properties of the molecular building blocks and on the other hand by the delocalized nature of the electronic excitations, i.e., the formation of excitonic states. The dynamical phenomena induced by photoexcitation therefore involve an interplay of site-site interactions entailing excitation energy transfer (EET), and vibronic (electron-phonon) coupling which typically leads to ultrafast internal conversion and charge transfer processes. We propose here a molecular-level, quantum-dynamical approach as exemplified by our recent study of exciton dissociation at interfaces of semiconducting polymer phases (so-called heterojunctions) [1]. This study combines a vibronic coupling model parametrized for several relevant electronic states and 20-30 phonon modes, with accurate quantum dynamics simulations using the multiconfiguration time-dependent Hartree (MCTDH) method and a Gaussian-based variant thereof (G-MCTDH) [2]. In addition, we employ transformation techniques [1,3] by which a relevant set of effective modes is constructed which account for the short-time dynamics in high-dimensional systems involving conical intersection topologies. This approach has recently been extended to more general system-bath type models, in conjunction with a hierarchical approximation scheme for the spectral density that is tailored to non-Markovian situations [4]. Applications to EET in conjugated polymer chains involve both site-local and site-correlated electron-phonon coupling, and shed some light on recent observations of unexpectedly long-lived coherences in such systems [5]. A perspective is given on the role of static and dynamic conformational disorder.

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Quantum Dynamics of Excitons in molecular aggregates:

The explicit modeling of electron-phonon coupling

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Dynamics of exciton play a crucial role in, e.g., light-emitting diodes, solar cells, photosynthesis, bio-imaging using quantum dots and/or dye molecules, as well as various other systems. We theoretically study on exciton dynamics by using *ab initio* electronic structure calculations [1] and quantum dynamics calculations [2-4]. Based on multi-state quantum dynamics calculations, we can analyze exciton transfers and coherent excitons in molecular aggregates as well as those in assemblies of quantum dots. We have recently developed a methodology that enables explicit analyses and characterizations of electron-phonon coupling in realistic systems [2-4]. Our methodology can be widely applied for exciton dynamics, charge transfers, and radiationless decays mediated by electron-phonon coupling in various materials including semiconducting polymers [3,4]. Here, we demonstrate some illustrative quantum dynamical analyses of the exciton dynamics in aggregates of porphyrin molecules. We investigate the relaxation from the bright to dark excitons and the exciton transfers in porphyrin aggregates, where energy dissipations from the electronic to vibrational degrees of freedom play an important role.

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Optical properties of gold nanowires: application to the Surface Enhanced Raman Scattering

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We have studied the Surface-Enhanced Raman Scattering (SERS) of shape controlled metallic nanowires which can act as nano-antenna, designed through electron beam lithography and lift off techniques. We have notably studied the influence of localized surface plasmon resonance (LSPR) on the efficiency of SERS.

The deposited molecular probe used is the trans-1,2-Bis(4-pyridyl)ethylen (BPE) with two different excitation wavelengths of 632.8 nm and 676 nm. The nanowire lengths vary from 50 nm up to 1 μm (the width and the height of the nanowires is fixed). The observation of the dependence of the Raman enhancement versus the nanowire length is clearly demonstrated and remarkably the enhancement is observed to be maximum for a specific length. Moreover, we have compared our SERS results to the particle LSPR and we have determined that the best enhancement is obtained for a LSPR position close to the Raman wavelength. For such arrays, we also observe the existence of multipolar surface plasmon modes (up to 7th order) and we show clearly that multipolar LSP modes exhibits a stronger efficiency than the first dipolar order in SERS process.

Our results support the fact that the optimization of SERS efficiency, relative to the LSPR, strongly depends on the shape of the metallic nanoparticles. The enhancement strongly depends on the nanowire parameters which is a crucial point to determine the optical efficiencies of such structure and to optimise the SERS processes.

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Phonon-assisted visible light photocatalyst using ZnO nanostructure

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Since conventional photocatalyst material of TiO_2 is activated only under UV light irradiation, the effective utilization of visible light has been one of the most important objectives. Here we report visible light photocatalyst reaction using phonon-assisted optical near-field process. Schematic of this process is shown in Fig. 1. To excite the carriers using propagating light, the higher photon energy than band gap energy is required (3.3 eV for ZnO). While, the usage of the optical near-field allows us to excite the dipole forbidden phonon state, which is called as a non-adiabatic process [1]. To realize high efficiency of the non-adiabatic excitation process, we introduced ZnO nanorods grown by MOVPE, which were used for semiconductor electrodes (Fig. 2). Optical near-field generated around the material depends on the size of material. The diameter of the ZnO nanorods is controlled by the growth temperature in MOVPE process. We prepared two samples of ZnO nanorod with the diameters D of 100 nm (sample 1) and 10 nm (sample 2). Using these electrodes, water splitting reaction was performed. Current was measured under laser irradiation. The potential of the electrodes was maintained at open circuit potential under a non-irradiated condition by potentiostatically. We used bulk single crystal ZnO substrate with a flat surface as reference. Figs. 3 (a) and (b) show the dependence of current on power of UV (3.8 eV) and visible (2.6 eV) laser, respectively. Under UV laser irradiation, all samples had similar dependence. While, under visible light irradiation, more than four times higher current was measured using sample 2 than others. This result indicated that the efficient non-adiabatic excitation process due to phonon-assisted optical near field was realized by introducing the fine structure as small as 10 nm.

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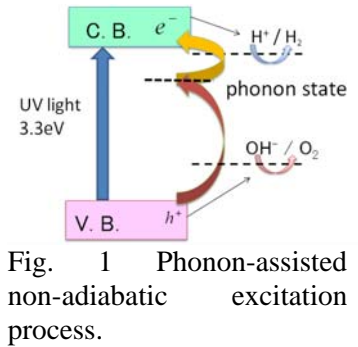


Fig. 1 Phonon-assisted non-adiabatic excitation process.

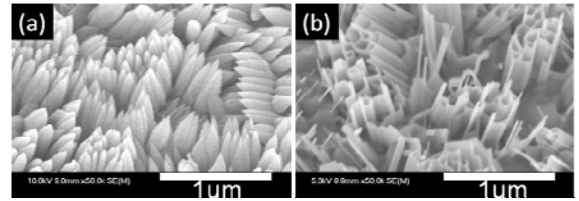


Fig. 2 SEM images of (a) Sample 1 ($D = 100\text{nm}$) and (b) Sample 2 ($D = 10\text{nm}$).

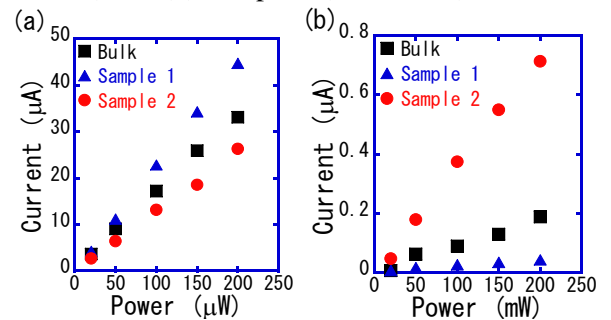


Fig. 3 Dependence of current on incident light power of (a) UV and (b) visible lasers.