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Harmonic light scattering as a tool to study molecular interactions

Thierry Verbiest

University of Leuven, Department of Chemistry, Belgium

E-mail: thierry.verbiest@fys.kuleuven.be

In this presentation we will give an overview of a recently developed harmonic light scattering technique to study molecular interactions and aggregation processes [1]. Harmonic light scattering is based on two nonlinear optical scattering techniques, *i.e.*, second- and third-harmonic light scattering. In second-harmonic light scattering two photons at frequency ω are combined into a scattered photon at the double frequency through a nonlinear optical interaction with the medium, while for third-harmonic scattering three photons of frequency ω are scattered as one photon at the triple frequency. Both processes are extremely sensitive to the symmetry of the scatterer and to interactions between the scatterers.

In this presentation we will give examples of recent measurements on the supramolecular aggregation of polymers and demonstrate that the technique is able to pick up weak interactions between molecules that are not visible to other spectroscopic techniques.

Keywords: *harmonic optical scattering, molecular interactions, aggregation kinetics*

References:

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Strong coupling and symmetry breaking in plasmonic structures

Bjorn Maes

*Micro- and Nanophotonic Materials Group, Research Institute for Materials Science and Engineering,
University of Mons, 20 Place du Parc, Mons B-7000, Belgium
E-mail: bjorn.maes@umons.ac.be*

We employ various types of symmetries, and breaking thereof, for nanophotonic devices with novel functionalities. As a first design we employ a multilayer with hyperbolic dispersion, and with dye molecules integrated for strong absorptive and emissive features. Depending on the dye properties, we obtain a rich variety of strong coupling (such as avoided crossings) and parity-time symmetry- related effects (such as exceptional points) [1]. As a second approach we use time-domain modulations of the refractive index to generate unusual optical behaviors. For example, using a sudden change, or shock, of the Fermi level in graphene, we can create tailored plasmonic reflections at a different frequency. Furthermore, using modulated ribbons of graphene we can efficiently generate a frequency comb, and by stacking multiple gratings we can optimize these combs to stress a particular, desired frequency component [2].

Keywords: *strong coupling, symmetry breaking, plasmonics*

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Quantum Weak Measurements: from Quantum Foundations to Sensitive Optical Measurement

M. Cormann^{1,2}, M. Remy^{1,2}, J. Bouhy² and Yves Caudano^{1,2}

*Department of Physics,
1Namur Institute for Complex Systems (naXys),
2Namur Institute for Structured Matter (NISM),
University of Namur,
Rue de Bruxelles 61, B-5000 Namur, Belgium
E-mail: yves.caudano@unamur.be*

Quantum weak measurements have generated a lot of interest for both their relevance to foundational questions about quantum mechanics and their ability to evidence tiny physical phenomena that would be unobservable otherwise. In a weak measurement, a quantum system is first prepared in an initial state $|\psi\rangle$ (pre-selection). Then, it interacts weakly with a meter acquiring partial information on the weakly perturbed system (observable \hat{A}). In the end, the quantum system is projected on a final state $|\phi\rangle$ (post-selection), for which the meter is measured. The outcome depends on the weak value, an unbounded complex number $A_w = \langle\phi|\hat{A}|\psi\rangle\langle\phi|\psi\rangle^{-1}$ whose interpretation is still debated. We described qubit weak values theoretically in their polar form and showed that the modulus is related to the visibility of an interferometric experiment using entangled photon pairs, while the argument is associated to a geometric phase depicted on the Bloch sphere [1]. Using the Majorana representation of n -level quantum systems, we extended our geometric description on the Bloch sphere to n -level systems in terms of $n-1$ qubits [2]. We applied this formalism to revisit the quantum three-box paradox and also its connection to the quantum Cheshire cat. We designed an experiment to investigate the paradox with entangled photon pairs in a Hong-Ou-Mandel interferometer, which is currently being build. We also investigated the Goos-Hänchen beam shift at optical interfaces using weak measurements.

Keywords: *quantum weak measurements, entangled photons, three box paradox*

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Revealing the Classical Nature of Gravity in a Stern-Gerlach Humpty-Dumpty Experiment

Thomas Durt

*Aix Marseille Univ CNRS
Centrale Marseille, Institut Fresnel
F-13013 Marseille, France
E-mail: thomas.durt@centrale-marseille.fr*

There is no consensus among today's physicists about how to describe properly the gravitational interaction in a quantum framework. We propose to bring this question to the realm of experiment.

To do so we propose to send a freely falling mesoscopic spin $1/2$ microsphere in a Humpty-Dumpty Stern-Gerlach interferometer. As we will show, self-gravity induces a measurable phase shift between the up and down spin components of the microsphere which makes it possible to reveal the existence of a non-linear self-interaction à la Schrodinger-Newton. If our proposal gets realized it would be the first experiment in which the interaction of a mesoscopic object with itself induces measurable effects, which would deliver crucial information about the nature of gravity at the quantum scale.

Keywords: *quantum gravity, interferometry, Schrodinger-Newton equation*

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Fluorescence Control by Photonic Structures in Biological Organisms

Sébastien R. Mouchet

*School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom
Department of Physics, University of Namur, Rue de Bruxelles 61, 5000 Namur, Belgium
E-mail: s.mouchet@exeter.ac.uk / sebastien.mouchet@unamur.be*

Many biological systems including arthropods (e.g., butterflies, beetles, and scorpions), fishes, invertebrates (from corals to worms) or plants are known to give rise to fluorescence emission when they are illuminated by UV light [1,2]. The origin of such phenomenon is the presence of fluorophores, such as bipterin and papiliochrom II, embedded within the teguments of the biological organisms. The confinement of fluorescent sources in natural photonic structures enables the so-called controlled fluorescence [2-4], which is known to lead to modifications of fluorescence properties such as the intensity (enhancement and inhibition) and the spatial distribution of emission.

Using different microscopy techniques (optical, electron and high-resolution fluorescence microscopies), linear and nonlinear spectrophotometry, spectrofluorimetry, scatterometry and optical simulations, we investigated the cases of fluorescent beetles the porous photonic structures of which contain embedded fluorophores and control their emissions.

Clearly, these biological devices challenge the human imagination which has at his disposal a wide range of materials but is limited so far to the design of comparatively rather simple structures. Understanding these structures and optical effects found in nature and optimized for millions of years of evolution allows in turn the elaboration of new concepts and devices through a bioinspiration approach to develop industrial applications such as LED, VCSEL diodes, solar cells, sensors or biosensors [5].

Keywords: *natural photonics, fluorescence, linear and nonlinear optical characterization*

References:

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Photoelectric Conversion via Local Heating

Wakana Kubo

Division of Advanced Electrical and Electronics Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Naka-cho, Koganei-shi, Tokyo 184-8588, Japan
E-mail: w-kubo@cc.tuat.ac.jp
Website: http://web.tuat.ac.jp/~kubolab/top_En.htm

The abilities of plasmons to generate local heat at the nanoscale are not negligible, and this heating will be valuable as a reliable resource for energy devices. Here, we report a photoelectric conversion triggered only by plasmonic local heat. Periodic silver nanorods covered with an organic thermoelectric thin film were fabricated; the nanorods were then irradiated with monochromatic light. A flow of electric current was detected through the thermoelectric film when plasmons of the silver nanorods were excited. The intensity of the electric current was found to be strongly dependent on the irradiation wavelength and the direction of polarization. The direction of polarization could be used to excite the short-axis plasmon resonance and maximize the electric current intensity, which implies that plasmons are the dominant driving force for generating electric currents. The photoelectric conversion efficiency was observed to be approximately 0.001%. This technology can be applied to photodetectors and thermoelectric devices.[1]

Keywords: *plasmonics, silver nanorods, photoelectric conversion*

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Broadband Energy-Entangled Photons for Quantum Metrology

André Stefanov

*Institute of Applied Physics, University of Bern
Sidlerstrasse 5, 3012 Bern, Switzerland
E-mail: andre.stefanov@iap.unibe.ch*

Energy-time entangled photon pairs can be produced by spontaneous parametric down-conversion [1]. When pumped with a monochromatic laser, they exhibit at the same time narrowband spectral features and short time correlations. Indeed, the energies of both photons sum up to the well-defined pump photon energy, even if each individual photon has a broad spectrum. Respectively in the time domain, the correlation time between the two photons is of the order of few tens of femtoseconds. It is a property of entangled states of light to show strong correlations in two complementary basis, as energy and time here. Such states are a resource to realize measurements beyond the capabilities of classical devices.

I present an experimental setup to manipulate the two-photon energy wave function with the help of a pulse shaper, which combines dispersive elements and a spatial light modulator. At first, the temporal state of the two photons can be characterized by ultrafast optical coincidence in a nonlinear crystal. By controlling with the pulse shaper the added dispersion and time delay, it is possible to proof the entanglement of the state. Then, proof of principle experiments are presented where the ultra-short time properties are used for dispersion measurements and for temporal discrimination of scattered photons. Finally, such quantum light is a promising tool for the implementation of quantum spectroscopy protocols.

Keywords: *quantum metrology, quantum spectroscopy, energy-time entangled photons*

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Merging Intermolecular and Casimir forces with Photonics: Classical and Quantum Approach

Branko Kolaric

*Micro- and Nanophotonic Materials Group, University of Mons
Place du Parc 20, 7000 Mons, Belgium
Photonics Center, Institute of Physics, University of Belgrade
Pregrevica 118, 11080 Belgrade, Serbia
E-mail: branko.kolaric@umons.ac.be*

The interactions between atoms and molecules significantly influence the structural stability and reactivity of molecular and nanoscale systems. While covalent interactions impose the structural integrity of molecules, intermolecular forces govern more subtle phenomena such as self-assembly, surface, and supramolecular interactions, and polymer and biopolymer topology. [1] The understanding of these types of interactions is necessary for the interpretation of many physical and biological phenomena.

Here in this work, we show how the photonic approach, i.e., structuring of materials at the nanoscale, could be used to mold intermolecular forces as well as quantum vacuum fluctuation responsible for the appearance of Casimir interaction [2,3].

Additionally, we also discuss the possibility of light and molecules to bond (within the optical cavity), creating new hybrid light-matter states with far-reaching consequences for these strongly coupled materials. A hybridization model can explain the essence of strong coupling in the language of physicists and physical chemists. The strong interaction between the material entity and light creates new hybrid light-matter states with significantly different energy levels from those of the material entity and the optical system individually [4]. Only recently has it been discovered that strong coupling can affect a host of significant effects at the material and molecular level, which were thought to be independent of the "light" environment: phase transitions, conductivity, chemical reactions, and intermolecular forces.

Keywords: *structuring at nanoscale, strong coupling, intermolecular interactions*

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