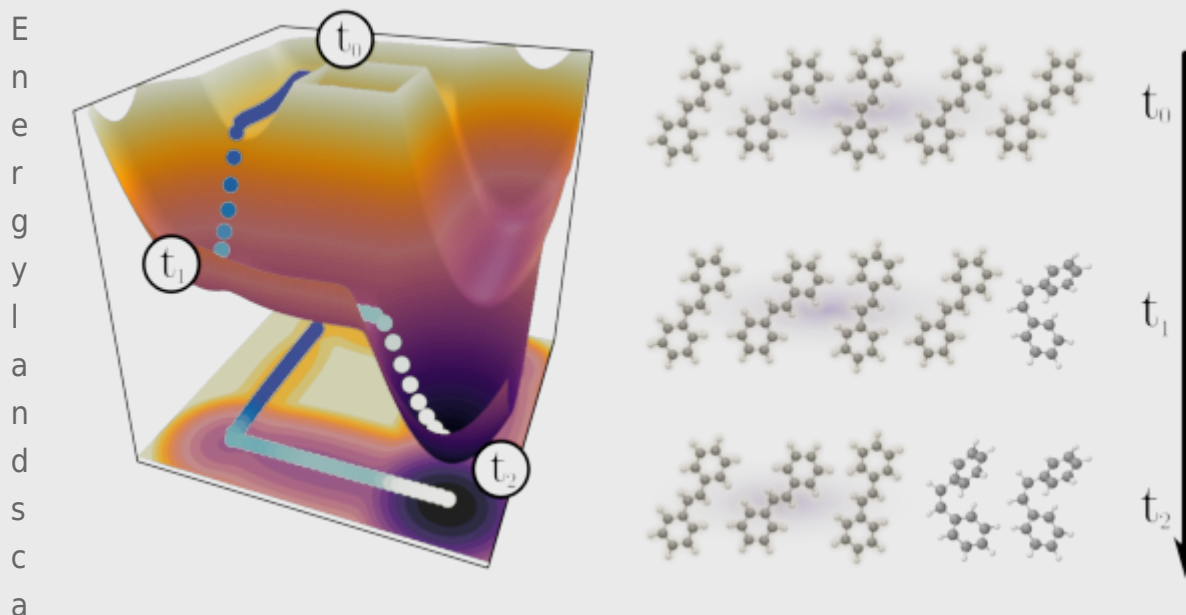


## Triggering Many Molecular Reactions With a Single Photon



pe that governs the chemical reaction of two molecules after the system absorbs one single photon. At the first reaction step  $t_0$  the system consists of five molecules strongly interacting with the electromagnetic vacuum. At the second reaction step  $t_1$  the system has evolved and one molecule has isomerized. In the strong coupling regime this process continues with more steps in which one molecule isomerizes after another.

Article: published in [Physical Review Letters](#) by Javier Galego, [Francisco J. Garcia-Vidal](#), and [Johannes Feist](#), IFIMAC researchers and members of the Department of Theoretical Condensed Matter Physics.

**P**hotoisomerization is a chemical process in which the nuclear structure of an organic molecule is modified after absorption of one photon. It is of great importance to many fundamental processes in nature such as photosynthesis and human vision, and constitutes a great tool for technological applications such as optical switches and solar energy storage. In conventional photochemistry the behaviour of these reactions is ruled by the Stark-Einstein law: only one molecule undergoes a reaction per absorbed photon.

In a recent study published in [Physical Review Letters](#), a theory group from Department of Theoretical Condensed Matter Physics and the [Condensed Matter Physics Center](#) (IFIMAC) at the [Universidad Autónoma de Madrid](#) has proven the possibility of overcoming the Stark-Einstein law in specific situations by taking advantage of the quantum electrodynamics phenomenon of light-matter strong coupling. This realization could have important applications in solar energy storage, where molecules under normal conditions efficiently absorb solar energy and store it for long times. Strong

coupling could allow the release of the stored energy of the whole system on demand by using only one single photon.

In the strong light-matter coupling regime the characteristics of each component are mixed and the system develops new states called polaritons, which can show modifications in both the material and chemical properties of the system. In this work the researchers show the possibility of taking advantage of this new “polaritonic chemistry” to open new reaction pathways otherwise forbidden in conventional chemistry. They consider organic molecules proposed for solar energy storage purposes, which have a 50% probability of inducing a useful reaction when absorbing a photon. They show that by putting these molecules in the strong coupling regime, it is possible to entangle the degrees of freedom that describe the reaction of each individual molecule. This way, absorbing a photon causes one molecule to react after another, inducing a stepwise chain reaction for many molecules. Therefore, by using strong coupling it is possible to trigger a reaction of thousands of molecules using only one photon.

This work is another example of the great potential of polaritonic chemistry, of great interest for both fundamental and applied reasons. In particular it is noteworthy how this developing field brings together chemistry and quantum electrodynamics. While in standard chemical reactions light and molecules are separated and have distinct roles, polaritonic chemistry calls for a redefinition of the molecule, and enables novel and exotic chemical processes, even finding ways to defy the well-established laws of conventional photochemistry. [[Full article](#)]

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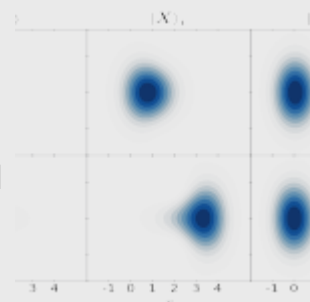
## Modelling Organic Condensates From Weak To Strong Coupling

Title: Modelling Organic Condensates From Weak To Strong Coupling.

When: Wednesday, January 25, (2017), 12:00.

Place: Departamento de Física de la Materia Condensada, Facultad Ciencias, Module 3, Seminar Room (5th Floor).

Speaker: Jonathan Keeling, SUPA – School of Physics and Astronomy, University of St Andrews, St Andrews KY16 9SS, United Kingdom.



**T**he idea of studying strong matter-light coupling using organic molecules has a long history [1], but has recently seen an explosion of experimental interest [2]. In particular exciton-polariton lasing and condensation has now been observed in a variety

of organic media, including anthracene, organic polymers, and fluorenes. Closely related to these strong coupling polariton condensates is the observation, in weak coupling, of Bose-Einstein condensation of photons in a dye-filled microcavity [3]. These experiments pose several questions about the relation of condensation and lasing, and about the role of vibrational modes in the physics of photon and polariton condensation. I will discuss our recent work on these subjects.

In the context of photon condensation, I will discuss the role of vibrational modes in establishing a thermal distribution of photons [4], including the time-evolution toward the thermal state. In the context of polariton condensation I will discuss our recent work exploring the nature of the ground and excited states of a model of such a system [5,6]. In particular, I will focus on the connections to optomechanics in other systems, and changes in the optical properties that can arise from coupling to vibrational modes.

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M. A. Zeb, P. G. Kirton, J. Keeling, arXiv:1608.08929.

[More information on IFIMAC Website](#)

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## Uncoupled Dark States Can Inherit Polaritonic Properties

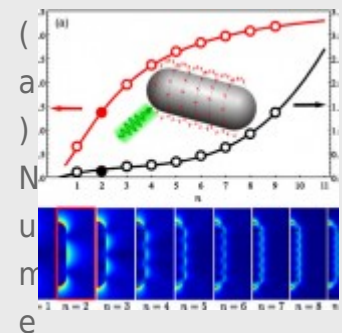


Figure (a) shows numerical results for the real (red) and imaginary parts (black) of the eigenmode frequencies of the metallic nanoparticle.

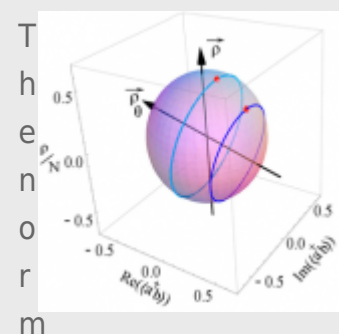
(b) Intensity of the electric field for the corresponding eigenmodes, which are labeled by the mode index  $n$ .

Article: published in [Physical Review Letters](#) by Carlos Gonzalez-Ballester, [Johannes Feist](#), [Esteban Moreno](#) and [F.J. García-Vidal](#), Department of Theoretical Condensed Matter Physics and IFIMAC researchers.

**T**he phenomenon of collective strong coupling emerges when an ensemble of quantum emitters interacts strongly enough with an electromagnetic field. In this regime, hybrid light-matter states, the so-called polaritons, are created. These hybrid modes have the most advantageous properties of their light and matter components and this is why they are usually considered as feasible ingredients in future quantum technologies. Apart from these polaritons, within the Hilbert space there are also many dark states whose origin stems from excitations within the quantum emitters that are not coupled to the light field. Up to date, these modes have been dismissed as it is thought they do not exhibit the promising properties of polaritons. In this work we have demonstrated that this is not the case and that dark states can indeed inherit the delocalized character of the polaritons. We show that these unexpected properties only appear when the electromagnetic field displays a discrete spectrum. When the photonic structure is extended and its electromagnetic spectrum is continuous, the customary picture described above of dark states being localized is fully recovered. We also demonstrate, both numerically and analytically, that dark states can even have superior delocalized properties than polaritons when the main loss mechanism in the system is associated with the light field. [\[Full article\]](#)

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## Polaritonic Rabi and Josephson Oscillations



alized Bloch sphere (Polariton sphere) describes clearly and elegantly the Rabi and Josephson oscillations. Pure Rabi oscillations are circles normal to the polariton axis  $\rho\theta$ . Whether they encircle (light blue) or not (dark blue) the axis of observables  $\rho$  (photon-exciton) makes their relative phase oscillating (as expected) or drifting (usually attributed to Josephson dynamics).

Article: published in [Scientific Reports](#) by [Fabrice P. Laussy](#), Department of Theoretical Condensed Matter Physics and IFIMAC researcher.

**P**olaritons arise from the strong-coupling between light and matter (1). As a particle, a polariton is a quantum superposition of a photon and a quantum of material excitation (let us call the latter an “exciton”, it could be an atomic transition, a plasmon or indeed a semiconductor exciton). As compared to a molecule where both components are present simultaneously to bind together, the polariton constituents are only partially present, being either constantly created and annihilated (the photon becoming the exciton and vice-versa) or in a steady state but with non-unit probability of being there. This is described by an entangled state of the form:

$$|\Psi(t)\rangle = \alpha(t)|1_a, 0_b\rangle + \beta(t)|0_a, 1_b\rangle$$

where  $\alpha$  and  $\beta$  are the amplitudes of probability for the corresponding one-particle states (one photon, 0 exciton and vice-versa). In experiments, however, such genuinely-quantum states are difficult to achieve, and one deals with classical version where the entanglement has vanished. For instance, a coherent superposition of polaritons, that follows a pulsed laser excitation, leads to a product of coherent states of the form (2):

$$|\psi(t)\rangle = |\alpha(t)\rangle|\beta(t)\rangle$$

where  $\alpha$  and  $\beta$  have the same dynamics as before but are now directly the amplitudes of real (classical) fields for coherent states of photons and excitons (3). As such, one can speak of Rabi oscillations of polaritons but in the sense of Leggett (4), not in the sense of Rabi (5). That is to say, polariton Rabi oscillations are the weak-interaction limit of the Josephson dynamics of two coupled condensates. This makes the problem of Josephson oscillation of polaritons more fundamental and central to the polariton physics, and therefore worthy of dedicated attention.

In Scientific Reports (6), we provide the theory for polariton internal Josephson dynamics that generalizes the textbook (atomic) theory for the case of dissipative particles with an arbitrary detuning and possibly different interaction strength. We show how the linear drift of the relative phase is not proper to the Josephson regime and propose an unambiguous criterion to tell apart the Rabi and Josephson regimes based on the stability of the fixed points. The dynamics is greatly simplified on a normalized Bloch sphere (Polar sphere) (7) where the pure Rabi regime corresponds to circles on the sphere, whose relative orientation as compared to the axis of observables account for the possible complex phase dynamics. Complex transitions between the regimes can occur. Several new phenomena are predicted that become in need of their experimental observation. [\[Full article\]](#)

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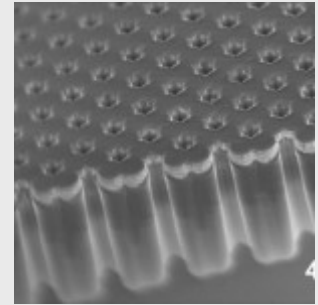
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Title: Polaritons in Lattices: A Nonlinear Photonic Emulator of Graphene.

When: Thursday, 07 July (2016), 16.00-17.00 h.

Place: Departamento de Física Teórica de la Materia Condensada, Facultad Ciencias, Module 5, Seminar Room (5th Floor).

Speaker: Alberto Amo, Centre de Nanosciences et de Nanotechnologies, CNRS, Univ. Paris-Sud, Université Paris-Saclay, France.



**E**xciton polaritons are quasi-particles arising from the strong coupling of quantum well excitons and cavity photons in semiconductor microcavities.

Thanks to their mixed light-matter nature, polaritons present unique nonlinear properties while, simultaneously, allowing the design of the photonic potential landscape. The direct visualisation of polariton eigenfunctions in luminescence experiments, makes microcavities an extraordinary photonic platform to emulate 1D and 2D nonlinear Hamiltonians.

In this way, polaritons allow transposing to the photonic world some of the properties of electrons in solid state systems, and to engineer Hamiltonians with novel transport and nonlinear properties.

In this presentation we will show striking nonlinear effects in two coupled micropillars which can be described by the nonlinear Bose-Hubbard dimer. We will then address the physics of a honeycomb lattice of coupled micropillars [1]. Its band structure emulates for photons the  $\pi$  and  $\pi^*$  bands of graphene [2, 3]. In addition, our system permits exploring orbital degrees of freedom, inaccessible in actual graphene, which give rise to flat bands and novel kind of edge states.

Our system presents interesting perspectives in view of studying nonlinear excitations in engineered photonic Hamiltonians owing to polariton-polariton interactions [4].

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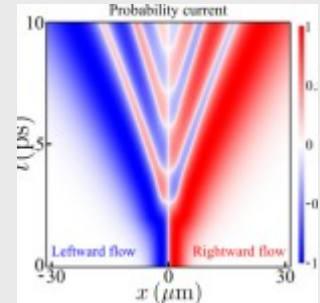
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[More information on IFIMAC Website](#)

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## Self-Interfering Wave Packets

Article: published in [Physical Review Letters](#) by David Colas and [Fabrice P. Laussy](#), Department of Theoretical Condensed Matter Physics and IFIMAC researchers.



**T**he Schrödinger wave packet is a fundamental concept of quantum mechanics that, historically, gives the probability (amplitude) to find a particle at a particular position (or a particular momentum, one excluding the other). Gaussian wave packets are natural solutions of the Schrödinger equation, and their dynamical properties (diffusion, scattering against a potential, etc...) are well known by graduate physics students. Many kinds of wave packets and their connections to particles have since been discovered, such as solitons, that can maintain their shape through propagation if interactions between particles are present, or the Airy beams discovered by Berry and Balazs, that accelerate without any external force or interactions.

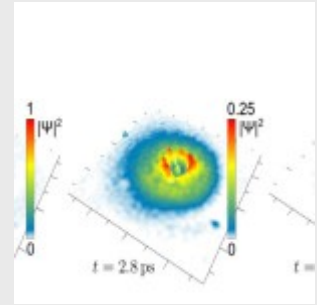
We add another member to the family of noteworthy non-interacting wave packets. Taking advantages of the nonlinear dispersion relation of polaritons—quasiparticles arising in semiconductor microcavities from the coupling of a light field (cavity photons) and a matter fields (excitons of a quantum well)—we demonstrate theoretically that a simple Gaussian pulse can give rise to a Self-Interfering Wave Packet (SIP) that exhibits solitonic properties, although non-interacting. This is powered by the polaritonic dispersion that provides diffusive effective masses of different signs within the same wave packet, leading to self-interferences. A rich phenomenology arises from this concept, such as adding Rabi oscillations that order a spacetime crystal, or by detuning the system which produces ultrafast subpackets. [\[Full article\]](#)

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## Real-space Collapse of a Polariton Condensate



Article: published in [Nature Communications](#) by D. Colas, B. Silva Fernández and [F. P. Laussy](#), Department of Theoretical Condensed Matter Physics and IFIMAC researchers.



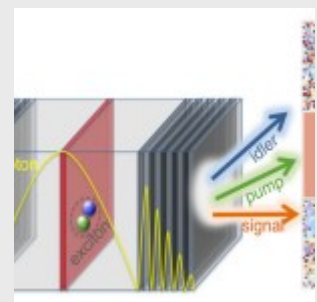
**E**xciton-Polariton are quasiparticles that arise from the coupling of two bosonic fields. Since they are composed by photons and excitons (both trapped in a semiconductor microcavity), they combine both properties of light and matter. A decade ago, the first Bose-Einstein condensate (BEC) of polariton was demonstrated, leading to many experiments on long-range coherence, superfluidity, quantized vorticity etc.

In a recent work published in *Nature Communications* by a collaboration between the groups of Daniele Sanvitto (Lecce, Italy), Alexei Kavokin (Southampton, England) and Fabrice Laussy (Madrid), a unique phenomenology of polariton BEC has been reported. When the polariton vacuum is excited by a light pulse, the quantum fluid suddenly created does not splash but instead coheres into a very bright spot, producing a very sharp, resolution limited, peak. This collapse of the polariton condensate is counter-intuitive since the polariton interactions are known to be repulsive and they have a positive mass. This suggests the intervention of an unconventional mechanism. Our modeling proposes a local heating of the crystal lattice as an explanation for such a self-trapping, described by a collective polaron formed by the polariton condensate. The remarkable phenomenology we report shows once more how polaritons can break new grounds in the physics of quantum fluids, with sui generis physics and phenomena. [\[Full article\]](#)

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## Nonequilibrium Phase Transition in a Two-Dimensional Driven Open Quantum System

Article: published in [Physical Review X](#) by [Francesca M. Marchetti](#), Department of Theoretical Condensed Matter Physics and IFIMAC researchers.



**T**here cannot be absolute order in a two-dimensional fluid because disorder-causing mechanisms are exceptionally strong. Nevertheless, there can be an enormous difference between systems of lesser and greater order (e.g., between an ordinary fluid such as water and a superfluid such as liquid helium). The latter can flow without any friction and even escape up and over its container walls. The transition between superfluid and normal behavior in two dimensions, even for closed systems that are allowed to equilibrate, is particularly dramatic: It is caused by the appearance of a large number of topological defects in the form of vortices—tiny tornadoes—that destroy the more ordered state. An open question is what causes the transition for particles that cannot be perfectly trapped and equilibrated in any container, such as photons. Their inevitable escape has to be counterbalanced by an external influx to keep the situation steady. We find that the transition is still caused by proliferating tornadoes.

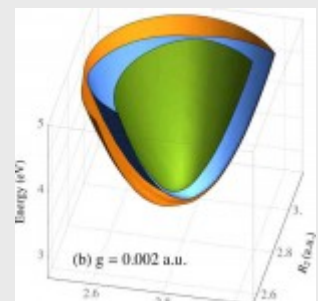
We focus on a quantum fluid of polaritons, and our analysis, based on a stochastic field formalism, accounts for topological defects and fluctuations. Surprisingly, we find that systems that are externally disturbed can remain a superfluid in an overall less-ordered state than their equilibrium counterparts. Whether these systems are more robust to vortex proliferation or simply more disordered by collective fluctuations remains to be determined. This externally overshaken-but-not-stirred quantum fluid clearly constitutes an interesting new laboratory to explore nonequilibrium phases of matter.

We expect that our results will motivate future studies of nonequilibrium phase transitions in driven-dissipative systems, in particular, optical ones. [\[Full article\]](#)

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## Cavity-Induced Modifications of Molecular Structure in the Strong-Coupling Regime

Article: published in [Physical Review X](#) by Javier Galego, Francisco J. Garcia-Vidal, and Johannes Feist, Department of Theoretical Condensed Matter Physics and IFIMAC researchers.



**W**hen organic molecules interact with light modes confined in nanostructures, hybrid light-matter states (polaritons) can be created via so-called “strong coupling.” The participating molecules are affected by the interaction, and this effect can be used

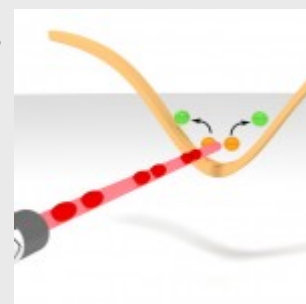
for controlling chemical reactions or modifying a material's properties. Up until now, there has been no consensus on how these modifications occur, and there has accordingly been no way to accurately predict these modifications or design novel structures to exploit them. Now, IFIMAC researchers Javier Galego, Prof. Francisco J. García Vidal, and Johannes Feist demonstrated that it is possible to understand and predict these modifications by adapting well-known techniques from chemical physics. The extent of the modifications turns out to depend sensitively on which observable is interrogated.

Progress up until now had been hindered by the fact that organic molecules possess a large number of internal (rovibrational) degrees of freedom and do not behave like simple two-level emitters. The researchers employed a first-principles approach that takes into account electronic, nuclear, and electromagnetic degrees of freedom on an equal footing. They demonstrated that the Born-Oppenheimer approximation, which underlies most of our understanding of chemical structure and dynamics and assumes that nuclear and electronic motion can be separated, can break down under strong coupling. However, exactly how this happens can be predicted, and in addition, in many important cases it does not break down. By exploiting this approach, the researchers show that molecular structure is modified even for so-called dark states, which are typically thought to be uncoupled from electromagnetic modes. These findings provide the basis for understanding how to manipulate chemical structure and reactions through strong coupling, and could have wide-ranging implications for the design of novel nanostructures. [\[Full article\]](#)

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## Exciting Polaritons with Quantum Light

Article: published in [Physical Review Letters](#) by J. C. López Carreño, C. Sánchez Muñoz, E. del Valle, and F. P. Laussy Department of Theoretical Condensed Matter Physics and IFIMAC researchers.



**F**or about five decades, research in quantum optics has been focused on photon correlations of systems excited by a laser. In this Letter, we propose a change in this paradigm, namely, we replace the laser with the emission of a quantum system. This emission has special features that a laser doesn't have, e.g., it is highly correlated. Turning to quantum light thus allows us to explore new regimes of excitation. As an

illustration of this approach, we consider the quantum light emitted by a two-level system, impinging on a passive cavity or one embedding quantum wells. The later case gives rise to so-called polaritons, particles of light and matter, with wondrous properties including high temperature BEC, superfluidity and a full gamut of nonlinear optical effects. Specifically, we show how quantum light, thanks to its reduced fluctuation and statistical properties, allows us to measure exactly the polariton interaction, even when it is weak as compared to dissipation. This technique that we refer to as “Mollow spectroscopy”, after the Mollow lineshape of resonance fluorescence used as the quantum source, should be of general interest for a wide range of optical targets and open new grounds in the nascent field of quantum spectroscopy. [\[Full article\]](#)

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