



Aalto University
School of Science

Light-matter interactions in quantum plasmonic lattices

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Aalto University

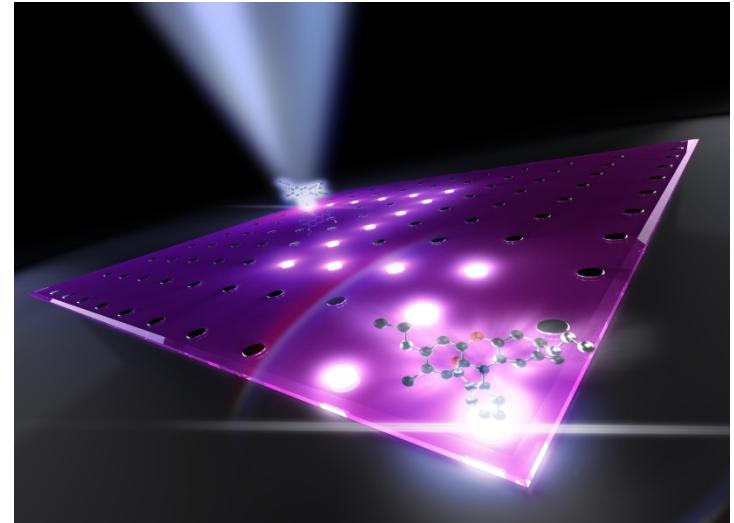
Quantum Plasmonics, Benasque, Spain
9.3.2015



(Periodic) spatial order and quantum physics

By nature:

- Crystal structure in matter
- Quantum objects (electrons) feeling a periodic structure: bands, ...
- (Quantum) objects forming structures: Wigner crystals, ...



By us:

- Semiconductor superlattices, ...
- Photonic crystals, ...
- Optical lattices, ...

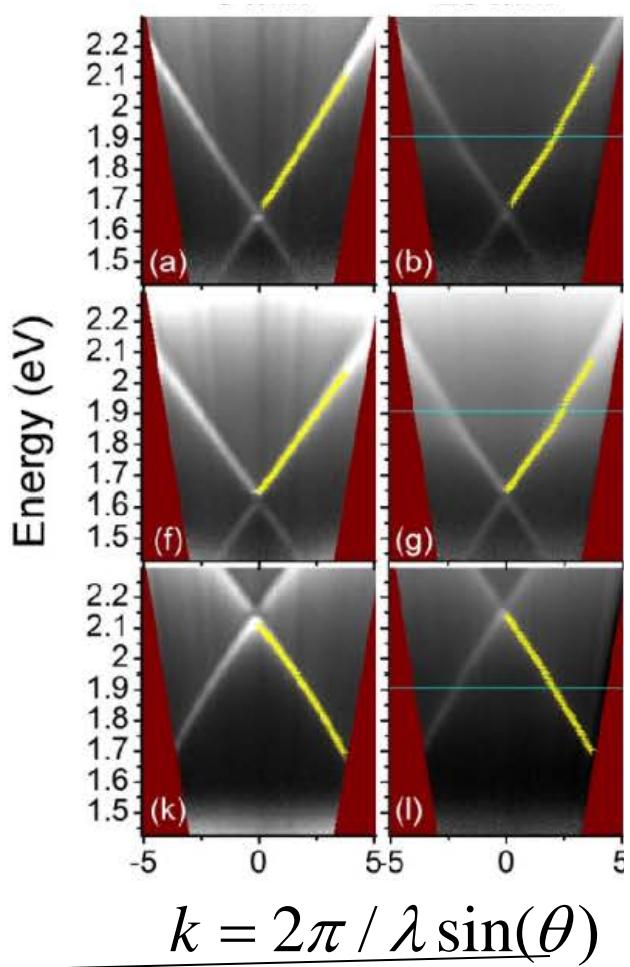
Fundamental influence on
Classical
Quantum
Quantum many-body
phenomena

Also topological properties can be designed

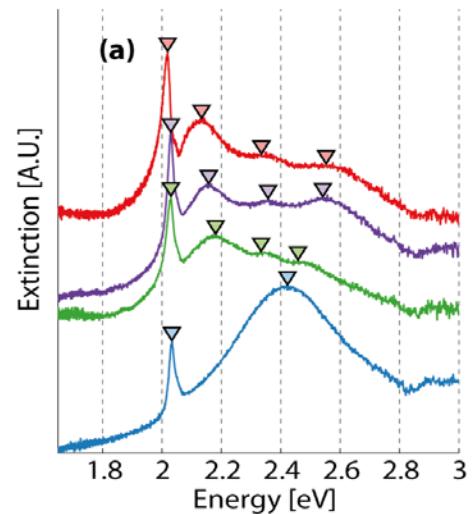
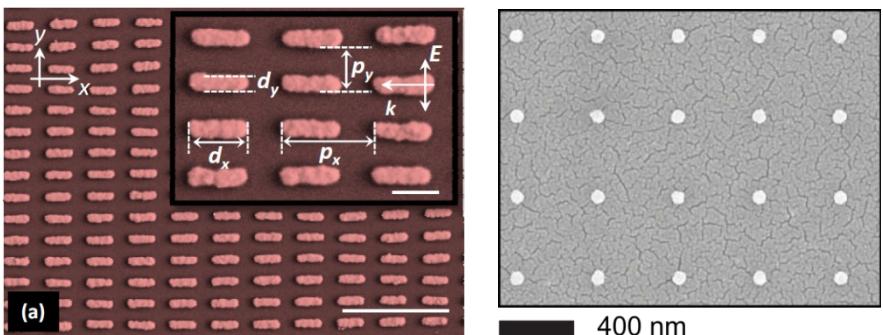
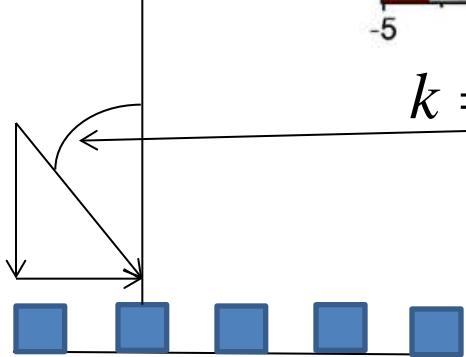


Surface lattice resonances (SLRs)

Markel, J. Mod. Optics 40, 2281–2291 (1993)
 Zou, Janel, Schatz, J. Chem. Phys. 120, 10871–10875 (2004)
 García de Abajo, Rev. Mod. Phys. 79, 1267 (2007)
 Auguié, Barnes, Phys. Rev. Lett. 101, 143902 (2008)
 Kravets, Schedin, Grigorenko, Phys. Rev. Lett. 101, 087403 (2008)
 Rodriguez, Abass, Maes, Janssen, Vecchi, Gómez Rivas, Phys. Rev. X 1, 021019 (2011)



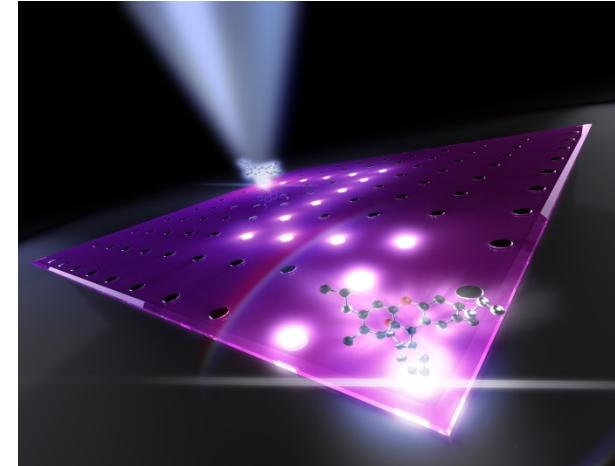
$$d (\sin \theta_i + \sin \theta_m) = m \lambda$$



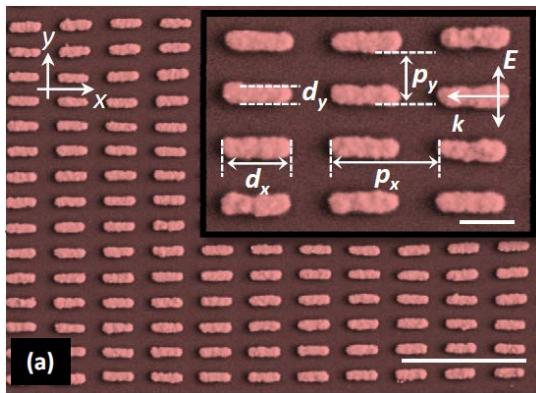
Goal: a new concept - *quantum plasmonic lattices*

J.-P. Martikainen, M.O.J. Heikkilä, PT,
Phys. Rev. A 90, 053604 (2014)

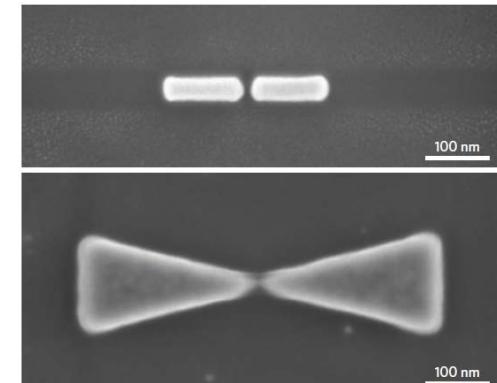
Room T
Nanoscale
On-chip
Ultrafast



$m_{\text{eff}} \sim 10^{-8} \dots 10^{-5} \text{ m}^e \rightarrow \text{BEC at room T (?)}$

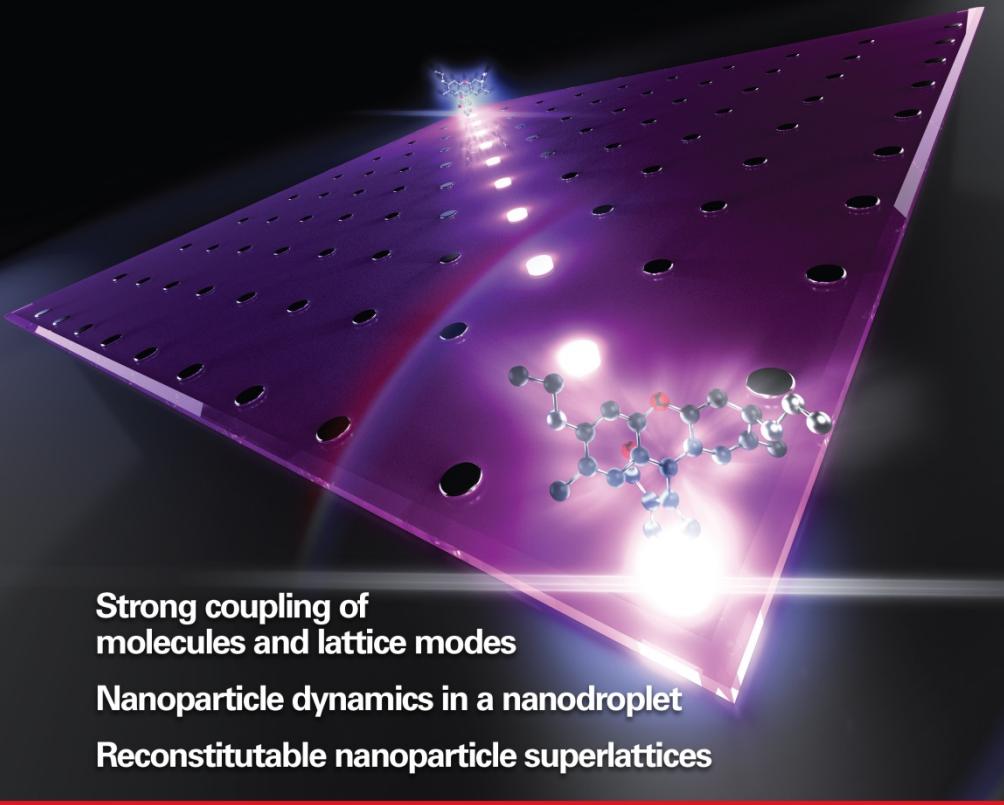


Single-emitter
strong coupling
predicted for hot-spots



Novotny, van Hulst,
Nature Photonics 5, (2011)

Quantum statistics? Interactions between
SLR photons? Quantum fluids?



Strong coupling in array

A.I. Väkeväinen, R.J. Moerland,
H.T. Rekola, A.-P. Eskelinan,
J.-P. Martikainen, D.-H. Kim, PT,
Nano Lett. 14, 1721 (2014)

Coherence of the hybrids

L. Shi, T.K. Hakala,
H. T. Rekola, J.-P. Martikainen,
R.J. Moerland, PT,
Phys. Rev. Lett. 112, 153002
(2014)

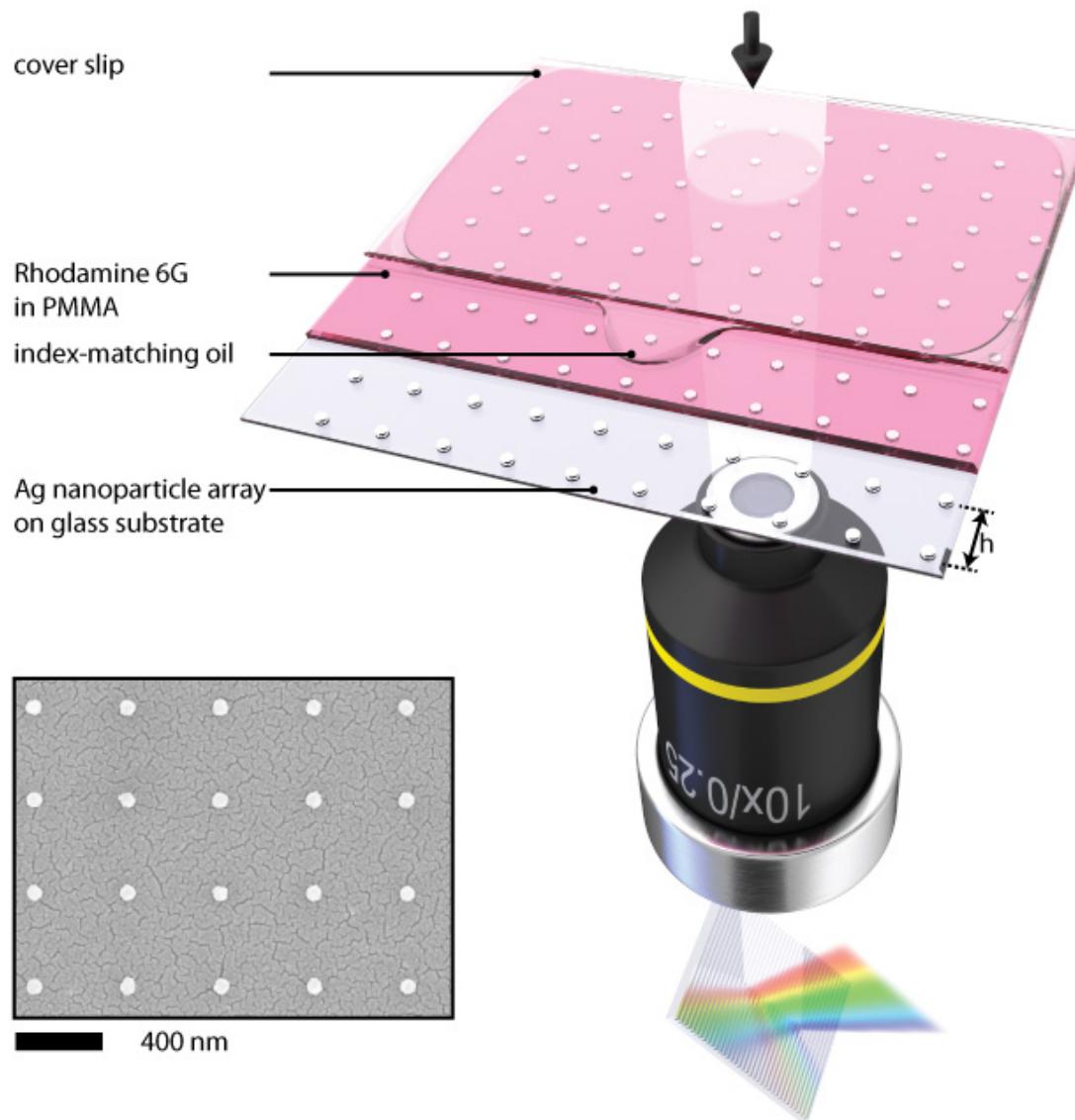
SLRs for magnetic particles

M. Kataja, T.K. Hakala,
A. Julku, M. Huttunen, S. van
Dijken, PT, Nature Comm. in
review (2015)

SLR condensation (theory)

J.-P. Martikainen, M.O.J. Heikkilä,
PT, Phys. Rev. A 90, 053604 (2014)

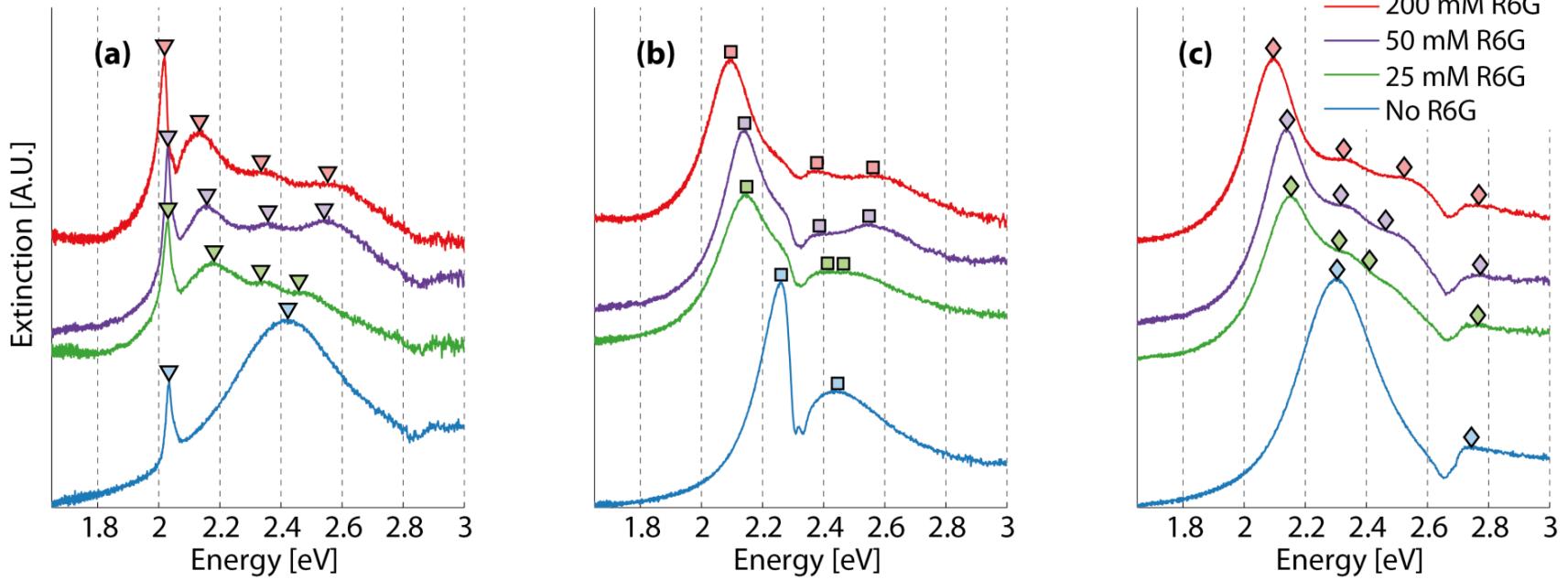
The measurements



Transmission measurements
with inverted optical
microscope

- Symmetrical index environment
- White light incident on from top
- Transmitted light guided to a spectrometer

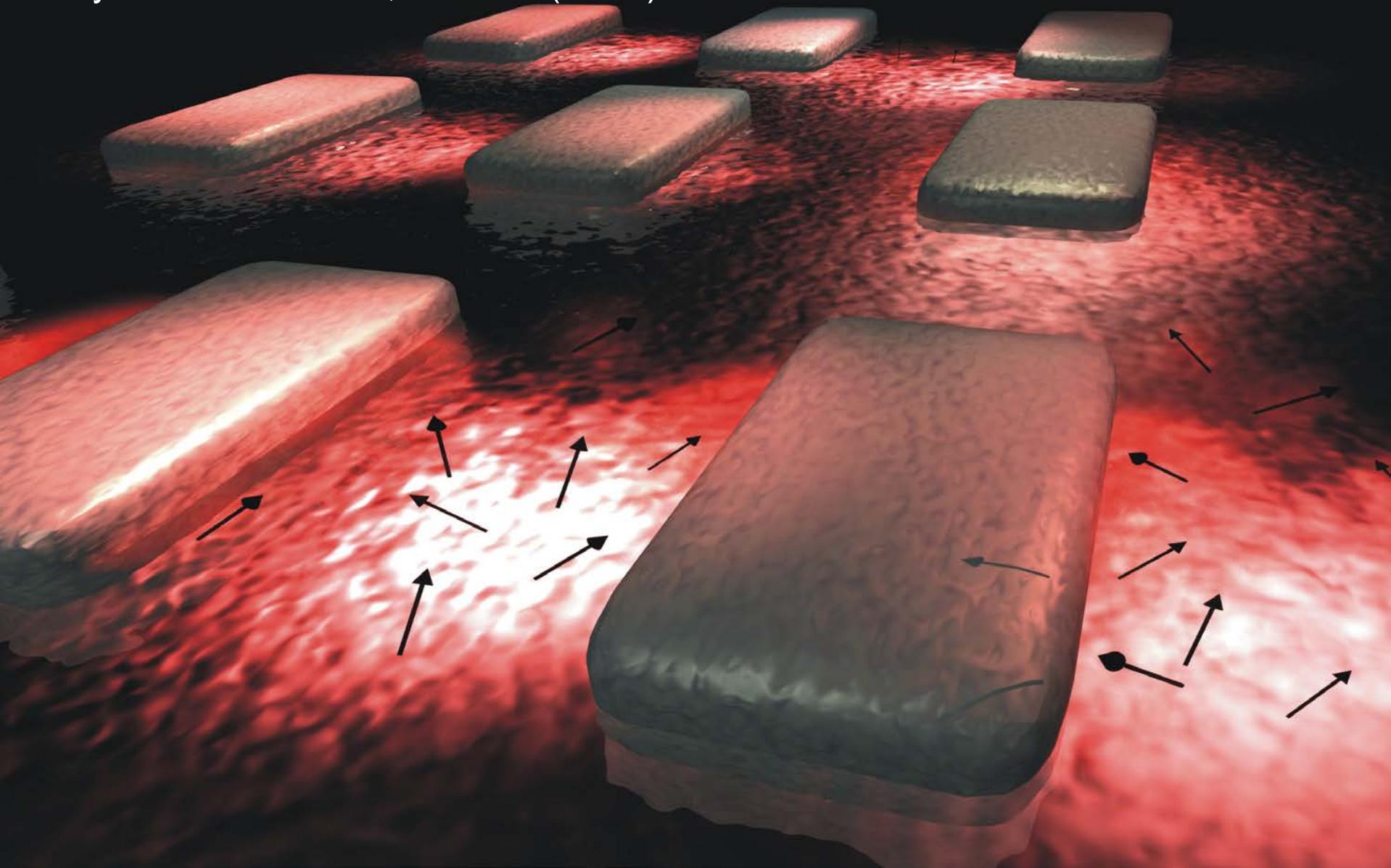
Results (with R6G)



a), b), c): different periods

The splitting grows as $\propto \sqrt{N/Vd}$

L. Shi, T. K. Hakala, H. T. Rekola, J.-P. Martikainen,
R. J. Moerland, and P. Törmä,
Phys. Rev. Lett. 112, 153002 (2014)



Spatial coherence

Localized excitons in molecules/quantum dots:
No long-range spatial coherence

Light-matter hybrids: coherence?

$$c_l|light\rangle + c_m|matter\rangle$$

***Study the spacial and coherence properties
of the hybrid wavefunction; interference***

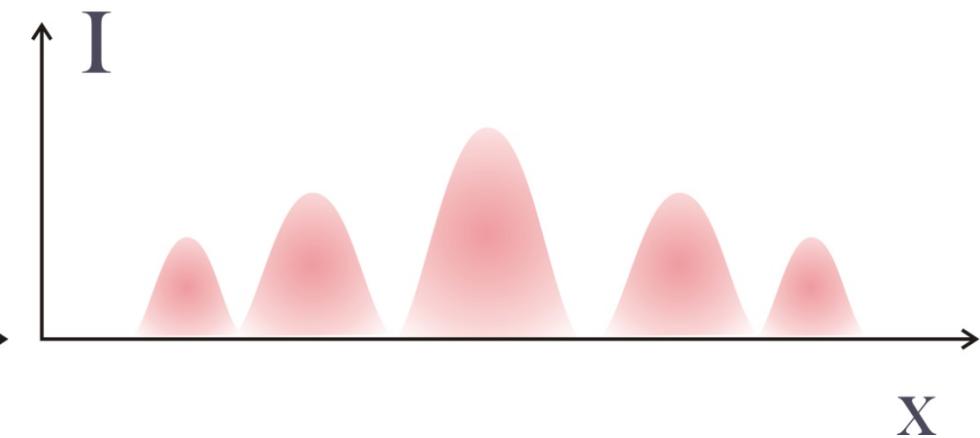
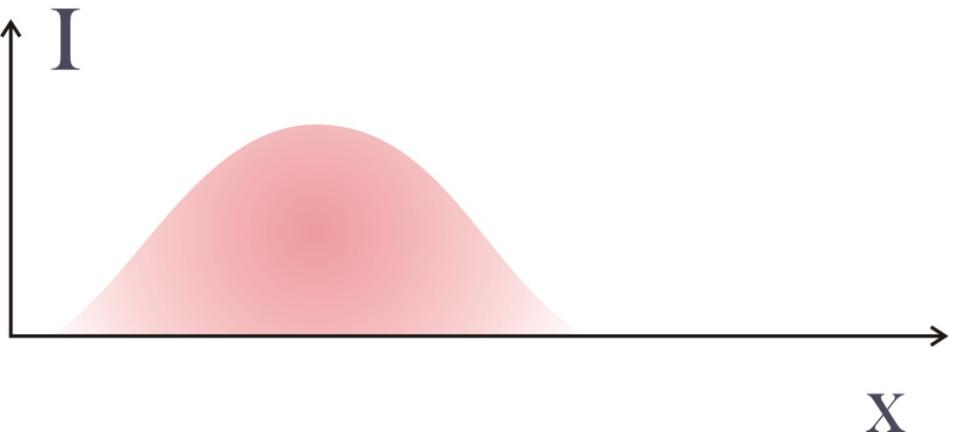
c.f. Aberra Guebrou, Symonds, Homeyer, Plenet,
Gartstein, Agranovich, Bellessa, Phys. Rev. Lett. 108, 066401
(2012); strong coupling regime (weak coupling reference by
a different system), planar metal

A double slit experiment

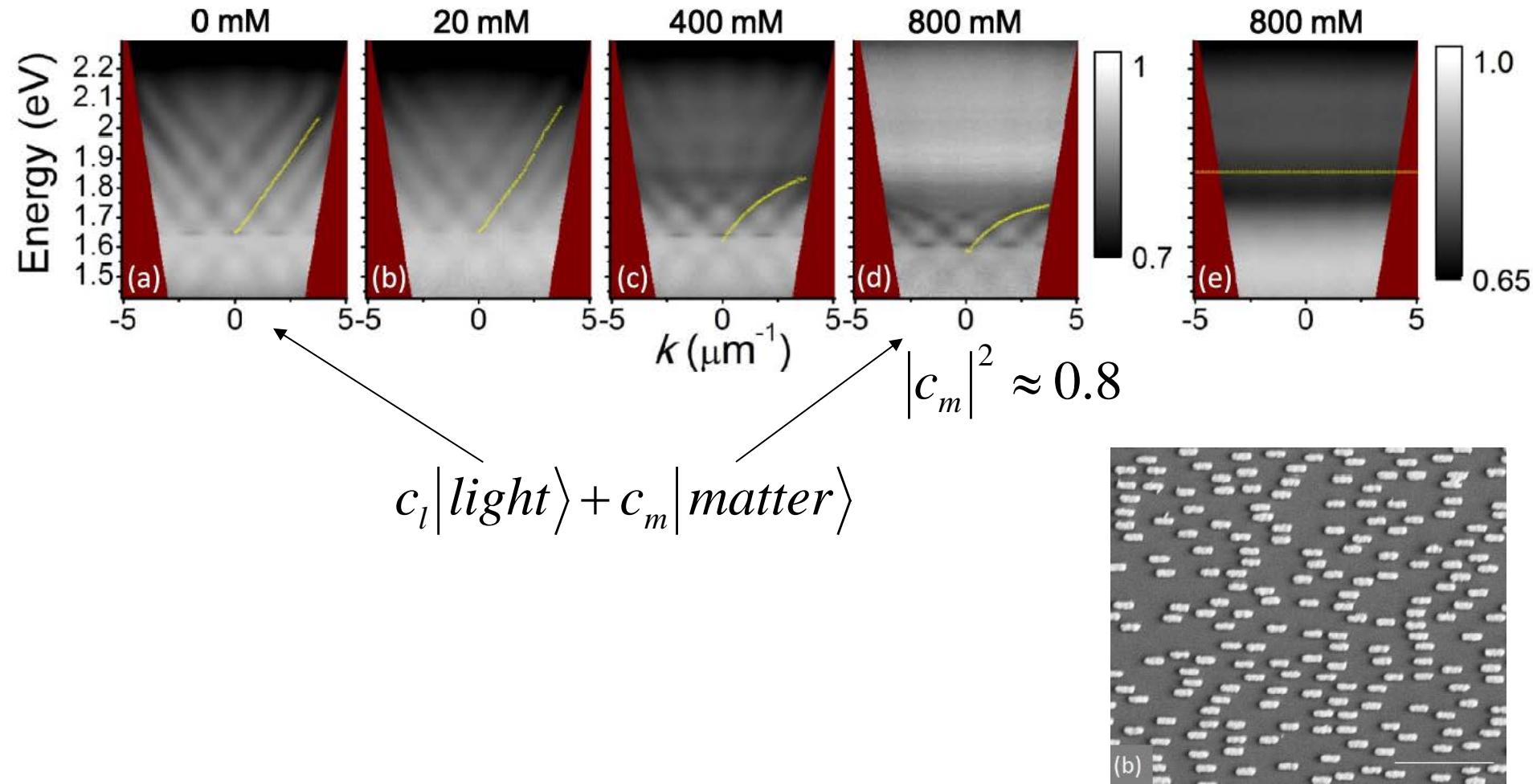
localized mode



delocalized mode



Double slit



Poster on the spatial coherence experiment by Heikki Rekola

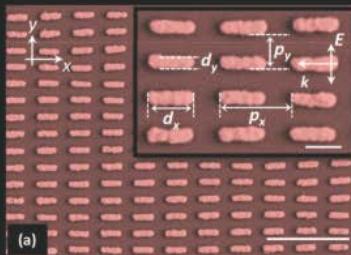


Spatial Coherence Properties of Organic Molecules Coupled to Plasmonic Surface Lattice Resonances in the Weak and Strong Coupling Regime

L. Shi,¹ T. K. Hakala,¹ H. T. Rekola,¹ J.-P. Martikainen,¹ R. J. Moerland,^{2,1} and P. Törmä¹

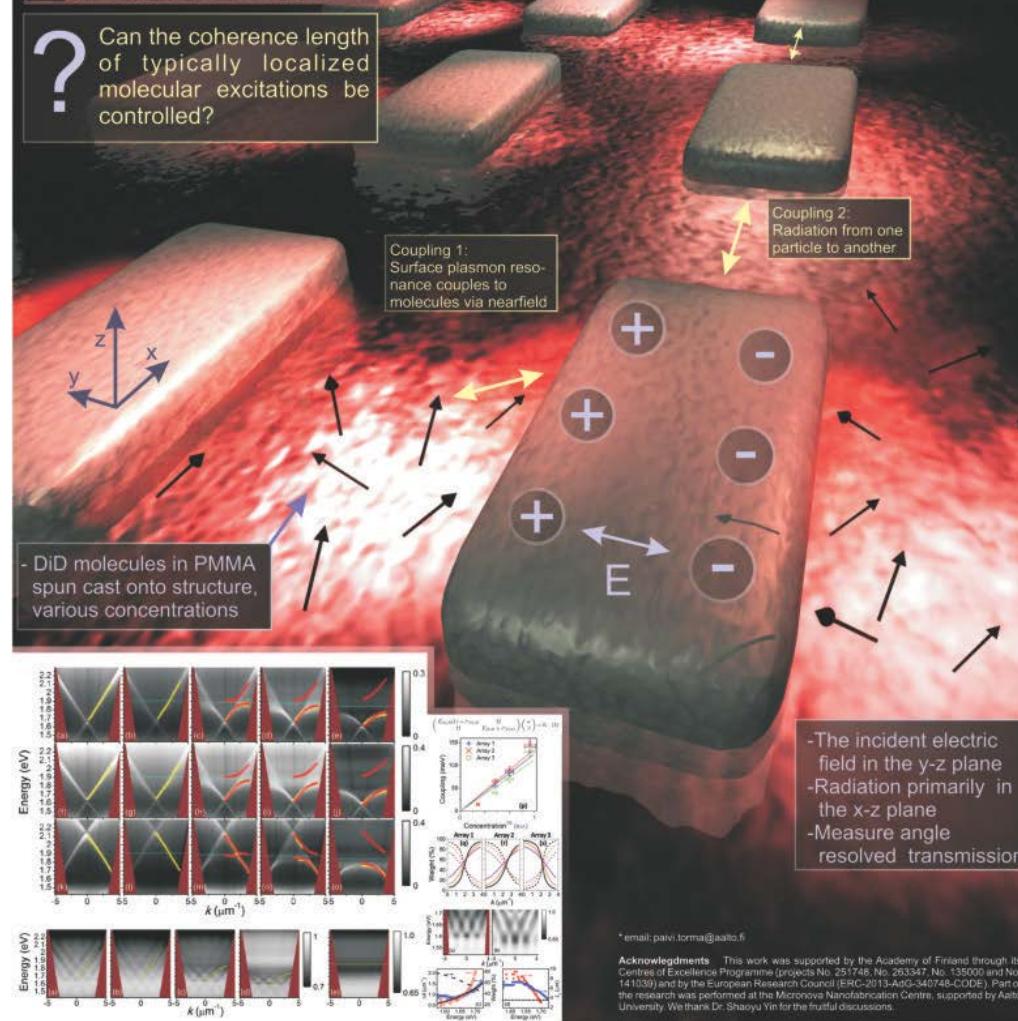
¹ COMP Centre of Excellence, Department of Applied Physics, Aalto University, FI-00076 Aalto, Finland

² Department of Imaging Physics, Faculty of Applied Sciences, Delft University of Technology, Lorentzweg 1, NL-2628 CJ, Delft, The Netherlands



(a)

We study spatial coherence properties of a system composed of periodic silver nanoparticle arrays covered with a fluorescent organic molecule (DiD) film. The evolution of spatial coherence of this composite structure from the weak to the strong coupling regime is investigated by systematically varying the coupling strength between the localized DiD excitons and the collective, delocalized modes of the nanoparticle array known as surface lattice resonances (SLRs). A gradual evolution of coherence from the weak to the strong coupling regime is observed, with the strong coupling features clearly visible in interference fringes. High degree of spatial coherence is demonstrated in the strong coupling regime, even when the mode is very exciton-like (80%). We show that coherence appears in proportion to the weight of the plasmonic component of the mode throughout the weak-to-strong coupling crossover, providing evidence for the hybrid nature of the normal modes.



Strong coupling of surface plasmon polaritons and emitters: a review

PT and W.L. Barnes, Rep. Prog. Phys. 78, 013901 (2015)

<u>CLASSICAL</u>	<u>SEMICLASSICAL</u>	<u>QUANTUM</u>
CLASSICAL FIELD + LORENTZIAN OSCILLATOR	CLASSICAL FIELD + QUANTUM 2-LEVEL SYSTEM	QUANTIZED FIELD + QUANTUM 2-LEVEL SYSTEM
<u>SINGLE (N=1) OR MANY (N>1) EMITTERS:</u>	<u>SINGLE EMITTER:</u> RABI OSCILLATIONS RABI FREQUENCY $\Omega = \frac{-dE_0}{\hbar}, \quad \begin{cases} \Omega = 0 \text{ for} \\ E_0 = 0 \end{cases}$ <u>LINEAR POLARIZABILITY</u> (for any N) $\Rightarrow \chi(\omega) \propto \frac{Nd^2}{V\varepsilon_0\hbar} \frac{\omega_0 - \omega + i\gamma}{(\omega - \omega_0)^2 + \Omega_0^2 + \gamma^2}$	<u>SINGLE EMITTER:</u> RABI OSCILLATIONS RABI FREQUENCY $\Omega = g\sqrt{n+1}$ $g \propto d/\sqrt{V}, E_0 \propto \sqrt{n}$ $\Omega = g \text{ for } E_0 = 0$ \rightarrow VACUUM RABI SPLITTING
ELECTRIC SUSCEPTIBILITY $\chi(\omega) = \frac{Ne^2}{V\varepsilon_0 m} \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}$ PERMITTIVITY $\varepsilon(\omega) = 1 + \chi(\omega)$	DISPERSION $\omega = f(k, \chi(\omega))$	MANY EMITTERS: DICKE MODEL LOW EXCITATION LIMIT \rightarrow SPLITTING AND RABI OSCILLATIONS $\Omega \propto g\sqrt{N} \propto d\sqrt{N/V}$

The figure consists of three side-by-side plots. Each plot has the vertical axis labeled ω and the horizontal axis labeled k . In all three plots, there are two curves: a lower solid curve and an upper dashed curve.

- CLASSICAL:** The separation between the curves is constant. A double-headed arrow between them is labeled $\Omega = \frac{e}{\sqrt{\varepsilon_0 m}} \sqrt{\frac{N}{V}}$.
- SEMICLASSICAL:** The separation between the curves increases with k . A double-headed arrow between them is labeled $\Omega \propto \sqrt{\frac{\omega_0}{\hbar \varepsilon_0}} d \sqrt{\frac{N}{V}}$.
- QUANTUM:** The separation between the curves increases with k . A double-headed arrow between them is labeled $\Omega \propto \sqrt{\frac{\omega_0}{\hbar \varepsilon_0}} d \sqrt{\frac{N}{V}}$.

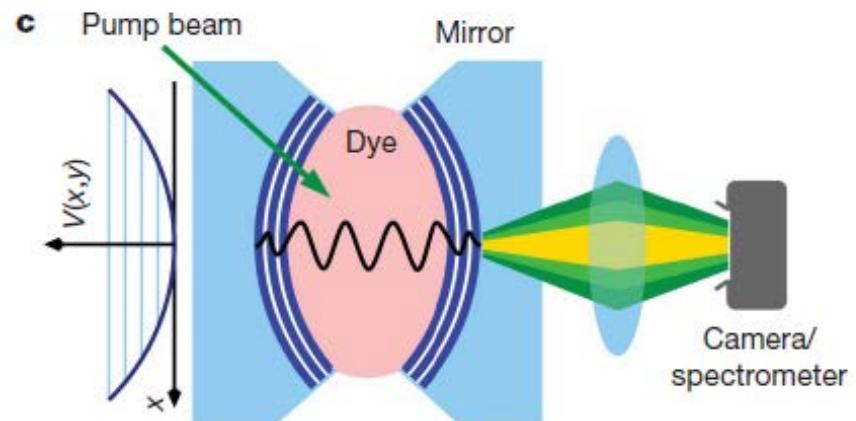
Photon BEC

LETTER

doi:10.1038/nature09567

Bose–Einstein condensation of photons in an optical microcavity

Jan Klaers, Julian Schmitt, Frank Vewinger & Martin Weitz



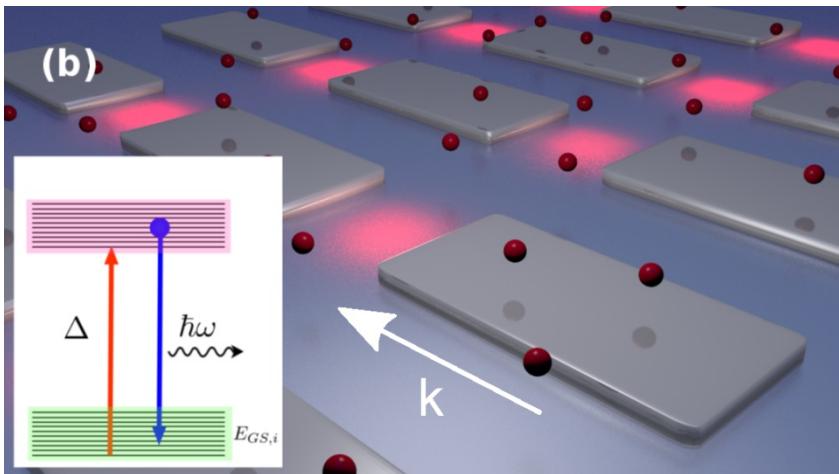
Black body

$$\frac{1}{e^{\hbar\omega/(k_B T)} - 1}$$

BEC

$$\frac{1}{e^{(\hbar\omega-\mu)/(k_B T)} - 1}$$

Question: are similar condensation phenomena possible in plasmonic systems?



J.-P. Martikainen, M.O.J. Heikkilä
and PT, Condensation phenomena
in plasmonics,
Phys. Rev. A 90, 053604 (2014)

Main conceptual question: the role of losses

Rate equations, steady state: BEC-like distribution at certain conditions

$$n(k) = \frac{1}{g(\omega)e^{\beta(\hbar\omega - \Delta)} - 1}$$

$$g(\omega(k)) = [MB(\omega)R_{abs} + \Gamma(\omega)]/(MR_{spon}B(\omega))$$

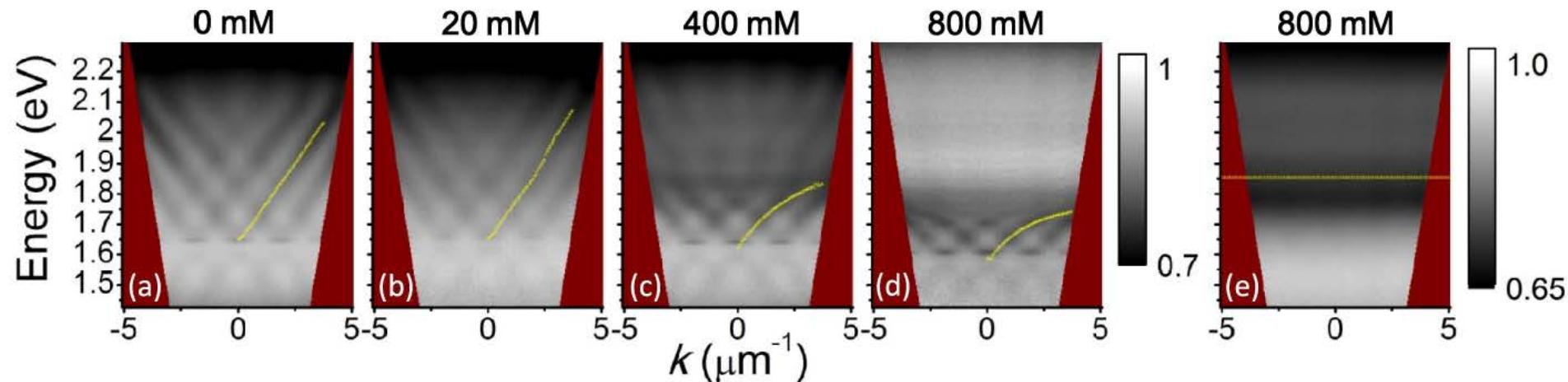
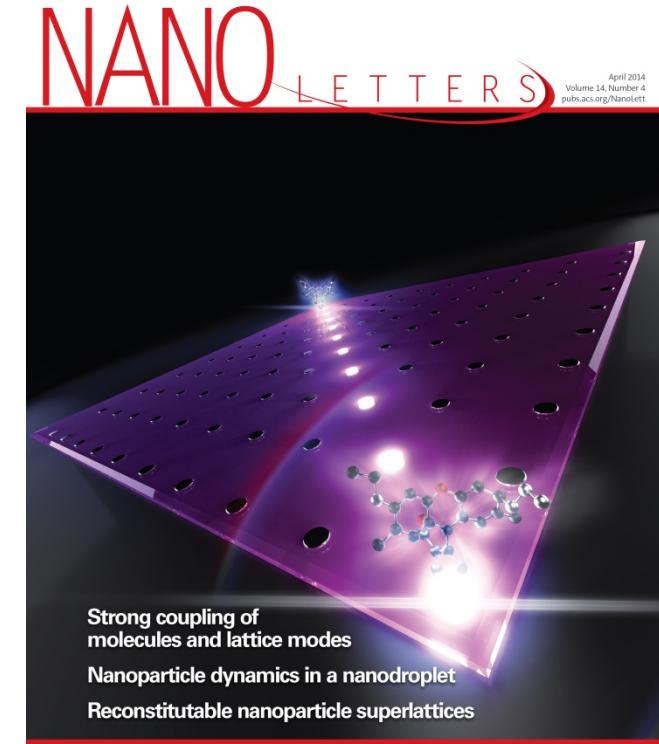
$$\mu(\omega) = \Delta - \frac{1}{\beta} \ln g(\omega)$$
 Effective chemical potential

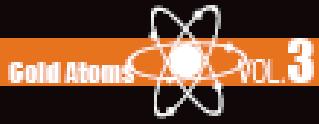
Condition for diverging population

$$\frac{N_e \Phi}{N_g} \geq \frac{1}{1 - \Gamma(\omega) \tau / (MFN_e B(\omega))}$$

Summary

- Goal: quantum plasmonic lattices
- Strong coupling of SLRs and molecules
- Coherence of light-matter hybrids throughout the weak to strong coupling crossover
- Lattice resonances for magnetic particles
- Towards BEC physics





Quantum Gas Experiments

Exploring Many-Body States

This book thoroughly describes the most important experimental techniques in the growing field of ultracold gases, with emphasis on methods especially relevant for studies of many-body quantum physics. As a background for the rest of the book, brief reviews on the many-body physics of optical lattices, and of the BCS-BEC crossover in ultracold Fermi gases are provided. The book then gives in-depth and detailed descriptions of basic experimental techniques for manipulating fermionic atoms, and for the creation of complex optical lattice structures both for bosonic and fermionic gases. Various spectroscopies (Feshbach and lattice modulation) are thoroughly covered. The basic physics of emerging research topics such as dipolar gases and ion-atom gas systems is presented. The book covers advanced techniques especially important for probing many-body aspects of the systems, such as correlation measurements and the newly developed single-site imaging techniques. Most topics are covered both from the theoretical and experimental viewpoint, with interrelated chapters written by theorists and experimentalists.

VOL.3

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Exploring Many-Body States

Quantum Gas Experiments

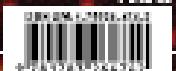
Exploring Many-Body States

editors

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Klaus Sengstock

Tomas
Sengstock

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Quantum Dynamics group



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