Thermalization and cooling of photonic/excitonic/plasmonic quasi-particles in a metallic nanoparticle array: towards quantum condensation

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Bose-Einstein Condensation (BEC) – the ground state accumulation of bosons at low temperature and high density – has been reported for atoms [1, 2], photons [3], and solid-state quasi-particles such as exciton-polaritons [4-6]. In contrast, the bosonic excitations at the interface of a metal and a dielectric (surface plasmon polaritons) have never been shown or predicted to condense. The lack of a suitable dispersion relation (e.g. one yielding a light boson mass with a ground state well above k_BT) for thermalization, combined with the strong radiative and Ohmic losses in metals, are likely the reasons for this.

Here we present experimental work towards quantum condensation in a plasmonic system. We investigate a periodic array of metallic nanorods covered by a polymer layer with organic dye molecules at room-temperature. The nanorod array sustains collective resonances arising from the diffractive coupling of localized surface plasmons [7-9]. These are known as surface lattice resonances (SLRs), and we exploit their narrow linewidth and steep dispersion [10] to thermalize low-mass polaritons (~7 orders of magnitude lighter than the electron rest mass) - a precursor for quantum condensation.

First we demonstrate the strong coupling of SLRs to excitons in the solid-state dye layer. This manifests as an anti-crossing in the dispersion diagram of the light extinct by the coupled structure near the molecular resonance energy. Bosonic quasi-particles known as plexcitons emerge from this coupling [11]. Through increased optical pumping we increase the density of plexcitons, while their photonic component leaks out of the open system. At high pumping, the exciton-SLR coupling saturates [12, 13], and a new peak emerges in the emission spectrum. Figure 1 shows the forward-emission spectrum of the sample discussed above, as a function of the critical pump power P_c required for the emergence of the new peak at higher energies. The emerging peak is attributed to the appearance of a plexciton population with weaker coupling, i.e., with a higher (lower) SLR (exciton) fraction. By analyzing the angular spectrum, we find these plexcitons to be the least massive surface polaritons of any kind yet reported. In addition, we observe thermalization of the plexciton population. Therefore, plasmonics provides a suitable platform for room-temperature solid-state studies of macroscopic quantum critical phenomena. We anticipate the possibility to form a plexciton condensate, which may function as a coherent light source.

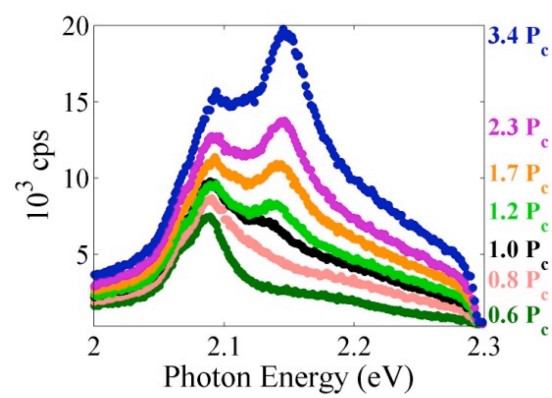

Fig. 1: Measurements of the forward-emission for increasing fractions of the critical pump power P_c required for the emergence of a new band at higher energies.

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