Generating persistent sub-radiant states with plasmonic nanocavities

Kalun Bedingfield¹, Ben Yuen¹, <u>Angela Demetriadou¹</u>

1. School of Physics and Astronomy, University of Birmingham, Birmingham, B15 2TT, UK

E-mail: a.demetriadou@bham.ac.uk

Plasmonic nanocavities have gaps between two metallic nano-structures of just 1-2nm, and have been used in the past few years to reach the light-matter strong coupling regime between molecular emitters and plasmons at room temperature [1,2]. This has opened the fascinating prospect of controlling quantum matter with light matter, and ultimately generating quantum states at room temperature, without the complex and cumbersome experimental methods required at cryogenic temperatures.

Since then, a lot of work has been done on understanding and quantifying light-matter interactions at very small plasmoni gaps [3,4]. Here [5], we demonstrate persistent sub-radiant states formed between 2 or more quantum emitters residing within a plasmonic nanocavity. We first develop a quantum electrodynamics description for an open cavity, to obtain both its quantum dynamics of the system and the quantum extinction cross section:

$$\langle \sigma_{ext} \rangle = -\sqrt{\kappa_{in}} \frac{\alpha \langle a^{\dagger} \rangle + \alpha^* \langle a \rangle}{c |\alpha|^2}$$

where $\sqrt{\kappa_{in}}$ is the rate that energy couples into the system by a monochromatic source, α is the amplitude of the coherent state defined by the incident source's photon flux $c|\alpha|^2$ and $\{a, a^{\dagger}\}$ are the bosonic creation and annihilation operators.



Fig. 1 The nanoparticle on mirror (NPoM) with two emitters within the cavity (left) and the quantum dynamics of the system with the symmetric and anti-symmetric states shown in black (right).

The quantum dynamics of two or more quantum emitters placed in a plasmonic nanocavity, and reveal persistent sub-radiant states formed between them (shown in figure 1). Although the Rabi oscillations between the plasmon and emitters decay very fast (~within few tens of fsec), these sub-radiant states persist for up to 100fsec, depending on the inherent non-radiative losses of the molecular emitters chosen. Additionally, the quantum extinction cross section allows us to theoretically predict experimental observables, and in fact in the absence of quantum emitters reduces to the classical behavior of the plasmonic nanocavity.

References

[1] Chikkaraddy, R., de Nijs, B., Benz, F., Barrow, S.J., Scherman, OA, Rosta, E., Demetriadou, A., Fox, P., Hess, O., Baumberg, JJ., 2016, *Nature*, 535, 127-130.

- [2] Zengin, G., Wersäll, M., Nilsson, S., Antosiewicz, T.J., Käll, M., Shegai T., 2015, Physical Review Letters, 114, 157401.
- [3] Kongsuwan, N., Demetriadou, A., Chikkaraddy, R., Benz, F., Turek, VA, Keyser, UF, Baumberg, JJ., Hess O., 2018, ACS Photonics, 5, 186
- [4] Demetriadou, A., Hamm, JM., Luo, Y., Pendry, JB., Baumberg, JJ., Hess O., 2017, ACS Photonics, 4, 2410
- [5] Bedingfield, K, Yuen, B., Demetriadou, A., 2023, submitted