Remote control of excitonic materials using coupled optical cavities.

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Abstract. Strong coupling with light has emerged as a powerful tool for modifying the properties of optical materials. Typical systems are based on a fluorescent layer embedded in a single optical cavity, whereby the excitonic emission is converted into a polarized, energy-tunable and dispersive polariton emission. There, excitons and photons coexist in the same volume and therefore any change in the emission properties of the excitonic material comes at the expense of simultaneously modifying the photonic environment where excitons reside, i.e., layer thickness and refractive index. Here, we demonstrate remote control over the intensity and total decay rate of the fluorescent layer by adding an extra purely photonic cavity strongly coupled to the first one. By modifying the resonant condition of the extra cavity, we reduce the total decay rate and suppress the fluorescence intensity of the fluorescent layer without explicitly affecting the first cavity. Such modification of the optical properties of the layer is the consequence of a resonant configuration that spatially segregates photons and excitons into different cavities.

Interlayer excitons are unique features that govern the optical properties of stacks of two dimensional materials. The indirect character of these excitations, formed by electrons and holes residing in two different layers, provides interesting properties such as a permanent electric dipole moment, high binding energy and long lifetime, paving the way for the design of room-temperature electrically controlled excitonic devices.

Here, inspired by these fascinating features, we transfer the concept of indirect excitons to optical materials strongly coupled to light. In analogy to the spatial confinement of electrons and holes, we realize intercavity polaritons, whereby photons and excitons forming exciton-polaritons are spatially displaced and segregated in two adjacent optical cavities. This is achieved by modifying the single-cavity set-up typically used in polaritonics. Our system consists of two stacked nanocavities fabricated on a glass substrate using a sequence of multiple sputtering and spin-coating steps. Figure 1 shows a sketch of our structure. The left nanocavity is filled with polymethyl methacrylate (PMMA), whereas the right one embeds a dye-doped polyvinyl alcohol (PVA) layer. The excitonic content is provided by a high concentration of homogeneously dispersed Erythrosin B (ErB) molecules. We demonstrate the ability to remotely control the emission properties of the organic molecules forming the emitting layer solely by acting on the left cavity. [1] This is due to the fact that both cavities are strongly coupled together by means of photon tunneling through a thin intermediate metallic mirror.

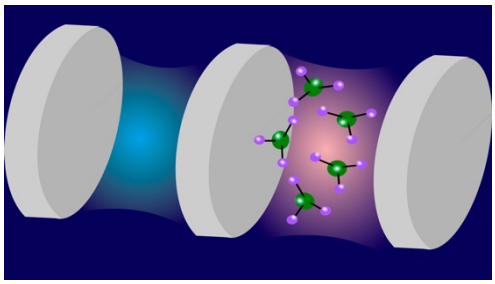

Fig. 1. Representation of the hybrid photonic-polaritonic coupled system.

The combination of the two photon energies and the exciton forms the Lambda-scheme, an energy level configuration well-known in atomic physics. Despite its apparent simplicity, this configuration is used to obtain fascinating phenomena such as dipolaritons, trion-polaritons, slow-light and in general, allows for quantum control experiments. Slow-light is achieved through the dramatic suppression of light dispersion and has garnered considerable interest in the fields of quantum optics, atomic physics and condensed matter. In the quantum domain, slow-light is typically understood in terms of polaritons.

In our system, optical band engineering is possible by tweaking such energy scheme. On resonance, i.e., when the frequency of the left cavity at normal incidence matches the exciton one, one of the polaritons living in our system become a state solely formed by the superposition of the left cavity photon and the exciton: the right cavity photon does not participate in the formation

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of this polariton branch. Moreover, its group velocity reduces forming a tunable quasi-flatband without the need of introducing an external potential or lattice.

This manipulation has far-reaching consequences on the steady-state and time-resolved emission properties of the ErB molecules. In particular, we observe four major features: (i) it allows us to switch on and off the fluorescence from the middle polariton band; (ii) it provides a direct way to modify the photonic and excitonic composition of the polaritons; (iii) it results in a strong suppression of the band dispersion, which may favour the study of strongly correlated polariton phases, in analogy to the flatband physics observed at the atomic scale; (iv) it allows us to slow down the decay rate of the flatband polariton.

1. Y. Garcia, B. Vargas, D. Ley, C. Ordóñez, H. Lara, A. Camacho, G. Pirruccio, Nat. Comm. **15**, 2915 (2024).