Two-Photon Pumped Exciton-Polariton Condensation

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Two-Photon Absorption (TPA) is widely utilized nowadays for investigation of solid-state quantum-confined structures, revealing various “dark” states of matter inaccessible to conventional spectroscopy but important for quantum information processing. Recently, a condensate of strongly-coupled light-matter exciton-polaritons [1] was proposed to stimulate highly efficient THz emission, in a transition from a two-photon excited 2p “dark” exciton state [2]. This scheme could enable doubly-stimulated THz emission far more efficient than existing sources [3], and introduces new possibilities for nonlinear optics with polariton condensates, so far relying on underlying inter-particle interactions [4]. It further provides a testbed for “dark” state-condensate interactions and can implement coherent control of collective quantum states with individual qubits. So far, several groups have demonstrated TPA-based excitation of non-condensed polaritons [5,6], yet condensation via TPA was not achieved.

Here we demonstrate two-photon pumped polariton condensation, achieved by TPA-based excitation. We show this in a planar GaAs-based microcavity by pumping with ultrafast pulses at half the exciton energies. The resulting angle-resolved photoluminescence (PL) (Fig. 1a-c) exhibits a clear threshold as a function of TPA power, coinciding with an interaction-induced blueshift and a spectral linewidth narrowing (Fig. 1d-f), characteristic of a transition from a polariton thermal distribution to polariton condensation. TPA is evidenced in the quadratic input-output dependence below and above threshold (Fig. 1g), and second-harmonic generation is ruled out by the emission peak energy showing no dependence on pump wavelength.

Fig. 1. a-c. Angle-resolved PL at T=6K and a 1s exciton-cavity detuning energy of Δ = +4.5 meV for three TPA powers. Lines are calculated polariton (white solid), cavity photon C (green dashed-dotted), and 1s (Δ and 2p (Δ) exciton (red dashed) dispersions. A transition from sub-threshold thermally-distributed polaritons to a blueshifting condensate is observed. d-f. PL characteristics vs. TPA power showing clear signatures of condensation. g. Integrated PL intensity vs. TPA peak intensity for two detunings (red-triangles are zero detuning, blue-squares are observed.

References
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