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Ultrafast dynamics of a polariton gas in organic-semiconductor Fabry-Perot microcavities

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In high-Q microcavities, non-perturbative mixing of the highly confined electromagnetic field and exciton resonances results in new quasi-particles termed exciton-polaritons. These are composite bosons with very low effective mass ($< 10^{-4} m_e$). Above a certain critical density and below a critical temperature, these can undergo Bose-Einstein Condensation (BEC), resulting in macroscopic spontaneous coherence. BEC has been well-documented in inorganic quantum-well microcavities, while this effect has not been convincingly demonstrated for molecular materials. Nevertheless, larger oscillator strengths and higher exciton binding energies should permit polariton condensates at higher temperature than in inorganic devices, even at room temperature, if the coupling is larger than the disorder width. In this oral contribution, I will describe recent efforts to unravel polariton-polariton nonlinear coupling by means of two-dimensional electronic coherence spectroscopy (2D-ECS). It is an ultrafast spectroscopic technique belonging to the family of 2D Fourier Spectroscopies and it allows to measure correlations between quantum transitions induced by the electromagnetic field. In 2D-ECS a sequence of four phase-related ultrafast optical pulses excites quantum states of a material system. Each pulse separately excites a quantum wave packet with spectral phase and amplitude imparted by that pulse, while the effect of the pulse sequence is to collectively excite multiple quantum coherences. Interferences between the various combinations of the pulses wave-packets determine linear and non-linear contributions to the material optical response. With this technique, we address polariton-polariton coupling dynamics towards the formation of quantum condensates.

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