Artificial σ-π bonding crossover in ballistic polariton molecules

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The ladder of energy modes in optically trapped exciton-polariton condensates [1, 2] can serve as a low-dimensional out-of-equilibrium analogue of molecular orbitals in an optically accessible setting. When two or more trapped condensates are coupled together, their wavefunctions align such to optimize their mutual interference over an optical-gain landscape thus increasing the condensate amplitude. This high-field seeking behaviour and spontaneous pattern formation has the potential to simulate formation of molecular orbitals and even be implemented as analogue qubits [3]. Interacting artificial atoms have been investigated before in cold-atom systems [4] as well as in microring resonators [5] and in exciton-polariton condensates in open cavities or micropillars [6, 7]. In this study, we investigate the interaction and states between artificial high-order polaritonic atoms in a planar GaAs-based microcavity, using multiple and reconfigurable optical traps.

Our excitation laser is configured into an annular-shape beam profile in order to confine polaritons through their excitonic interactions [1, 2], whilst the dissipative nature of the optical traps enables effective interaction with neighbouring condensates. Using a mixture of $|s\rangle$ and $|p\rangle$ modes between two traps, we demonstrate tunable and reprogrammable artificial polariton molecules that effectively simulate diatomic molecule orbitals in both experiment and theory. In particular, we demonstrate an interaction-induced crossover from the σ to π bonding of the p orbitals [8]. We propose an all-optical polariton simulator and present its scalability in practical problems, showing its tunability by controlling the strength of interaction and the relative energy difference of intertrap modes. We show that these results are consistent with a model of the polariton condensate as an order parameter governed by a 2D generalised Gross-Pitaevskii equation (GPE) coupled to a hot exciton reservoir [9]. Our findings offer insights into reprogrammable coupled polariton systems and open the path for polariton simulators for molecular chemistry.

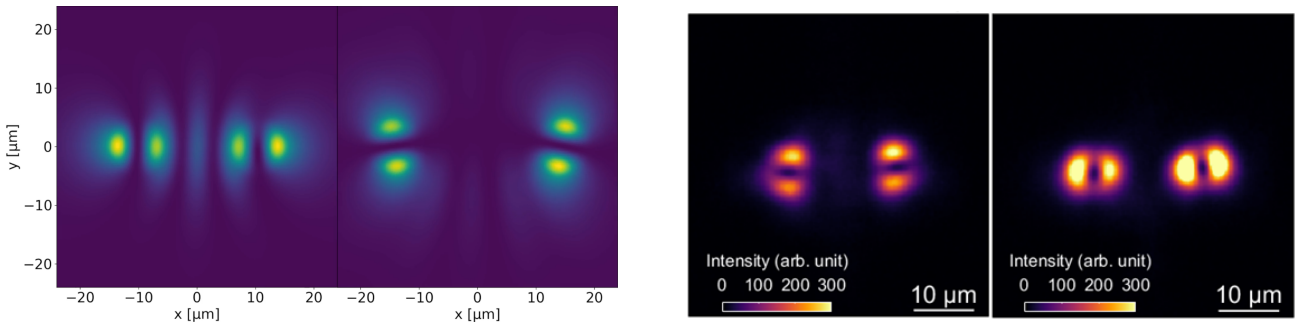

Figure 1: **Left:** Density maps of simulated polariton condensates demonstrating σ - π crossover. **Right:** Experimental PL maps demonstrating σ - π crossover.

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