Nonlinear Rydberg exciton-polaritons in Cu_2O microcavities

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Rydberg excitons, excited states with high principal numbers and large radii, may enable strong Kerr-like optical nonlinearities due to Rydberg blockade. Rydberg exciton-polaritons [1] are particularly attractive for quantum optical applications, where strong photon-photon interactions are required. Here, for the first time, we investigated Kerr-like nonlinearity of Rydberg exciton-polaritons in Cu_2O-based microcavities for states up to $n = 7$ [2] under resonant excitation using picosecond laser pulses. With increasing pulse power, Rydberg exciton blockade leads to the reduction of the Rabi splitting (see Fig. 1.a). The polariton non-linearity coefficient β scales with principal quantum number n as $n^{4.4}$, which is in agreement with the Rydberg blockade theory (Fig. 1.b).

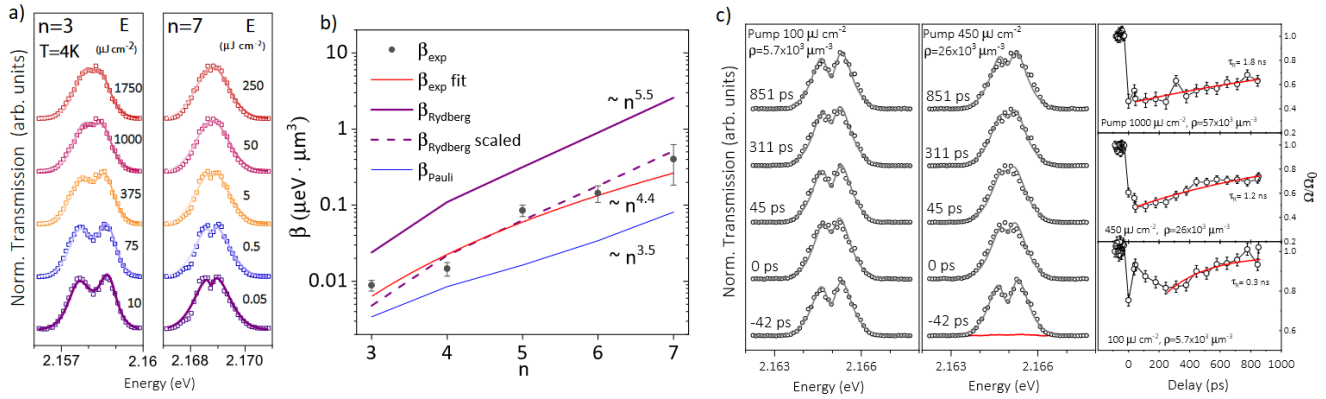

Fig. 1. a) Resonant transmission power dependence for $n = 3$ and 7. b) Nonlinear $\beta = \mathrm{d}\Omega/\mathrm{d}\rho$ coefficient, where Ω is the Rabi splitting, and ρ is the exciton density, as function of quantum number n, with theoretical models (Pauli and Rybderg blockade). c) Left: Normalised transmission as a function of probe delay for different pump fluences. Red line corresponds to pump-only signal. Right: Rabi splitting normalised by Rabi splitting at negative time delay as a function of probe delay.

In a single pulse experiment, the observed nonlinearity must be ultrafast (on the order of the pulse duration, $\sim$ 1ps). In the pump-probe experiments (Fig 1.c), the collapse of the Rabi-splitting is observed on a time scale of a few hundred ps to 1 ns after the pump pulse depending on the pump fluences. Such complex dynamics indicates interplay between the Rydberg exciton-exciton interactions, interactions with long-lived 1s exciton reservoir and plasma arising from Auger recombination. Overall, the ultrafast exciton nonlinear refractive indices are observed to be comparable or higher (for $n = 7$) than in GaAs system, but it is orders of magnitude less than those observed in CW experiments [3], where the role of plasma and the reservoir may be essential. Overall, our work opens up new questions in the field of ultrafast Rydberg exciton-polariton nonlinear optics.

References

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