Nonlinear Spectroscopy of the Polariton-Polariton Interaction Energy at the Quantum Level

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Exciton-polaritons are hybrid quasi-particles that emerge in the strong-coupling regime of cavity photons and quantum well excitons. They have allowed the observation of many fascinating phenomena such as Bose-Einstein condensation, superfluidity, and nonlinear parametric effects in the solid-state [1]. Very recently, first experiments with exciton-polaritons that go beyond the mean-field regime have been realized and notably signatures of photon anti-bunching, the experimental hallmark of strongly-interacting photons, has been observed [2, 3]. A crucial quantity in these experiments is the polariton-polariton interaction energy U which leads to an anharmonicity in the energy spectrum of the polariton number states and enables photon blockade when exceeding the linewidth of the system. Despite its importance for such kind of experiments, the precise determination of the polariton interaction strength has remained very challenging due to the difficulty of calibrating the mean polariton number in the cavity which is required when extracting U from the mean-field energy shift of the polariton resonance. Additionally, the interactions of polaritons with dark reservoir excitons can overshadow the blue-shift induced by purely polaritonic interactions. These issues have led to strongly scattered measurement results of the polariton interaction strength in the literature [4]. Here, we introduce a radically different measurement approach for U that overcomes these limitations: Using two-color excitation of a semiconductor micropillar (Fig. 1(a)) we spectroscopically resolve the center energy E_2 of the two-polariton state and we compare it with twice the center energy E_1 of the one-photon resonance ((Fig. 1(b),(c)). Based on numerical simulations performed by the direct resolution of the Lindblad master equation as well as the stochastic positive-P method, we predict that $E_2 - 2E_1 = U$. As shown in Fig. 1(d), the experimentally measured energy difference decreases with the intracavity power and stays approximately constant at low power thus giving direct access to U with sub-μeV precision.

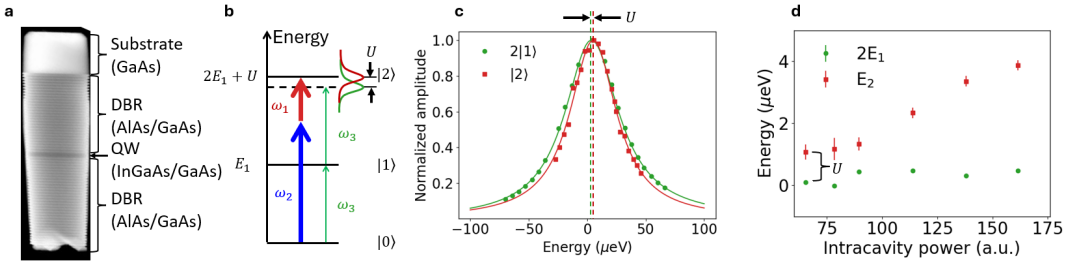

Fig. 1. (a) SEM image of exemplary micropillar as used in the experiment. (b) Nonlinear/linear spectroscopy of the two-/one-polariton transition. (c) Averaged two-polariton (red) and one-polariton (green) spectra with Lorentzian fit at lowest intracavity power. (d) Power-dependence of the two-polariton (red) and twice the one-polariton (green) center energy.

As we perform the determination of the two-polariton and one-polariton energy simultaneously in a three-laser experiment the influence of the reservoir is subtracted when the intracavity power is low enough. We tentatively explain the increased splitting between one- and two-photon transitions at larger powers as a consequence of higher order interaction channels. The introduced method constitutes an invaluable tool for optimizing U for more advanced quantum optics experiments with exciton-polaritons. It is readily applicable to novel material systems such as TMDCs or Cu_2O where a precise characterization method of the interaction strength is highly sought after.

References

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