Investigating strong light-matter interactions in monocrystalline gold nanodisks coupled to tungsten disulfide

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Abstract: Plasmonic nanodisks deposited on mono- and multilayer tungsten disulfide show Rabi splitting of 108 meV and 175 meV, respectively, the highest splitting reported in transition metal dichalcogenides coupled to plasmonic nanostructures.

Tailoring the interaction between excitons in semi-conducting materials and electromagnetic fields focused inside optical cavities is the cornerstone of many applied and fundamental researches in nanophotonics, optoelectronics, and plasmonics. The coupling strength between matter and light can range from the weak to the strong regime, where light and matter can no longer be treated independently but instead as hybridized states sharing both properties of light and matter [1]. In order to reach that regime, the coherent energy exchange between the excitons and the optical field must happen faster than any dissipation processes. However, for many nanophotonics devices, this regime is only reached at the cost of working in a cryogenic environment, where dissipation processes become sufficiently low. With the recent discovery of new low-dimensional materials such as transition metal dichalcogenides (TMDC), hosting excitons with binding-energies above 0.3 eV and exhibiting large transition dipole moments [2], it has been shown experimentally, in combination with localized plasmonic nanostructures, that it is possible to reach the strong-coupling regime at room temperature [3,4].

Here, we report the observation of strong coupling at room temperature between plasmons in ultra-thin (9 nm) monocrystalline gold nanodisks deposited on top of tungsten disulfide (WS_2). The ultra-thin geometry of the disk is advantageous, allowing us to determine both scattering and absorption spectra, thereby enabling unequivocal identification of strong coupling [5]. Dark- and bright-field spectroscopies show a clear Rabi splitting of 108 ± 8 meV at the onset of the strong coupling regime. Inspired by recent results [4] we use multi-layer WS_2 to push this coupling deeper into the strong-coupling regime and reach a splitting as high as 175 ± 9 meV.

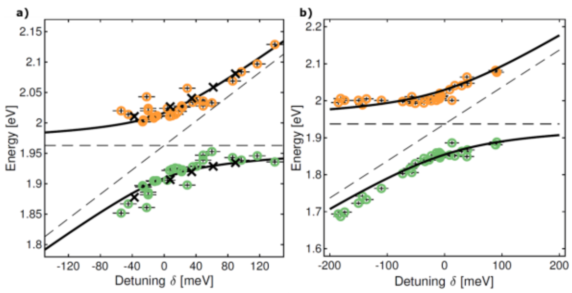

Fig. 1 a) Experimental dispersion of the lower (green) and upper (orange) polariton branch extracted from far-field scattering spectra measured on individual nanodisks deposited on top of monolayer tungsten disulfide (WS_2) and fitted to the coupled oscillator model (black line). The Rabi splitting at zero detuning is 108 ± 8 meV. Numerical calculations are shown with black crosses. **b)** Experimental dispersion exctracted from individual nanodisks deposited on top of multilayer WS_2. The Rabi splitting at zero detuning is 175 ± 9 meV.

This corresponds to a major increase of 62% of the Rabi splitting and is, to our knowledge, the highest reported for TMDC materials coupled to plasmonic nanoparticles [5]. We explain this important increase in the coupling strength by the higher number of exciton participating in the coupling, and the low mode volume of the ultra-thin gold nanodisks. This investigation can lead to the design of ultra-compact and highly efficient light sources down to the few- and single-exciton level, where novel nanophotonic research and applications become possible.

References

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