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Strong coupling and catenary field enhancement in the hybrid plasmonic metamaterial cavity and TMDC monolayers

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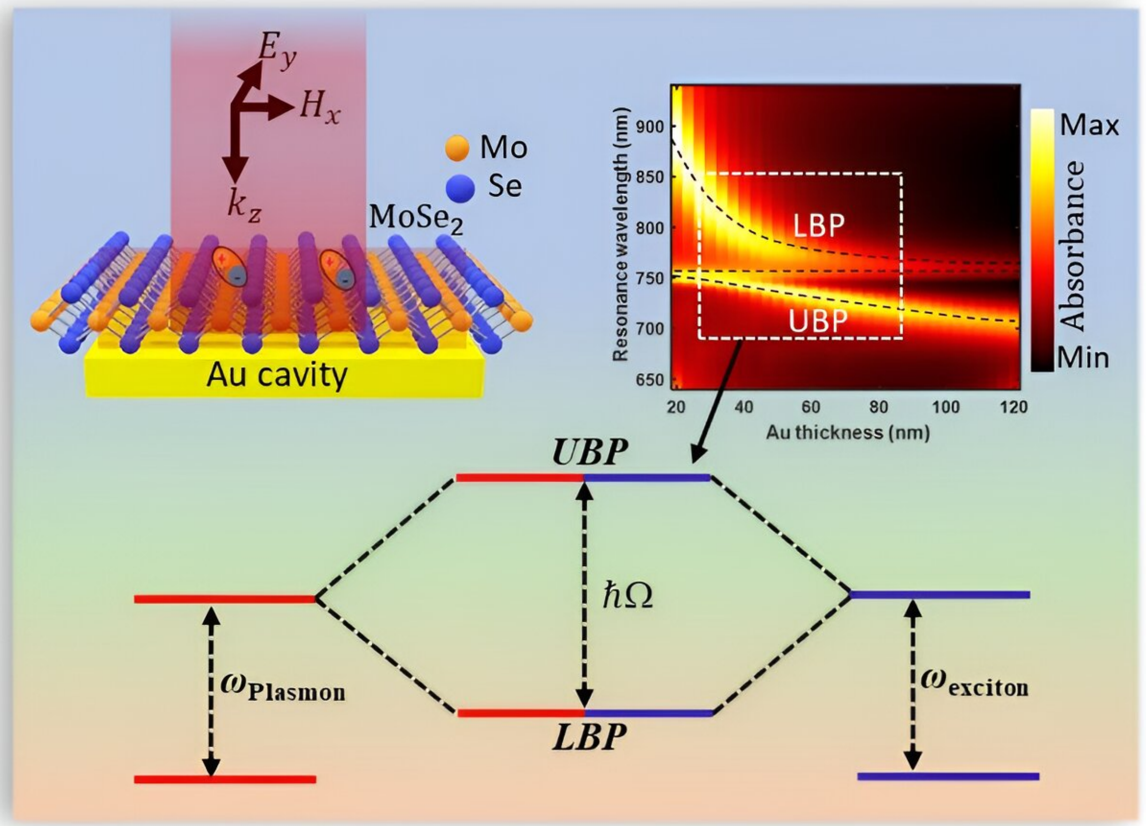

Top: (Left) Schematic illustration of monolayer $MoSe_2$ on top of Au cavity at normal incidence with polarization along the y-axis, and (Right) its corresponding absorption spectrum mapping with varying Au thickness. Insert (bottom): Schematic representation of strong coupling between a plasmon mode and an exciton in a $MoSe_2$ monolayer. Credit: Compuscript Ltd

Researchers in the field of nanophotonics have spent significant time in recent years investigating fascinating concepts known as polaritons and/or plexcitons. These ideas revolve around the strong coupling of light photons and/or plasmons to excitons in semiconductor materials.

Excitons, or bound electron-hole pairs in semiconductors, collectively respond to external light fields. To improve the strong interaction between electromagnetic fields and matter, properly designed cavities such as metasurfaces, metagratings, and metamaterials containing quantum emitters (QEs) are required. For example, their resonance energies should be the same to evaluate the coupling strength between plasmons of metallic nanocavities and excitons in QEs.

As a result, significant coupling between resonantly matched metal surface plasmons and QE excitons results in the development of novel plasmon-exciton hybridized energy states known as excitons. Such significant coupling is possible when the energy exchange rates between these subsystems outpace the decay rates of the plasmon and exciton modes.

Plasmonic nanocavities are essential in plasmon-exciton strong coupling due to their tunability and ability to restrict electromagnetic fields in a compact volume. However, not all plasmonic nanostructures have the same tunability and field confinement properties. For example, single nanoparticles have reduced spatial confinement of electromagnetic fields and restricted tunability to match excitonic resonance. Furthermore, the exciton mode must be stable in order to realize and manage strong coupling for nanophotonic applications.

Researchers now report in *Opto-Electronic Advances* the successful development of strong plasmon-exciton coupling and catenary field enhancement in a hybrid plasmonic metamaterial cavity containing transition metal dichalcogenide (TMDC) monolayers.

Plasmonic metamaterial cavities were chosen for their capacity to restrict electromagnetic fields in an ultrasmall volume and their ease of integration with intricate structures.

The plasmon resonance of these cavities spans a wide frequency range, which may be adjusted by changing the size or thickness of the cavity gap. This tuning is consistent with the excitons of the WS_2, WSe_2, and $MoSe_2$ monolayers.

TMDC monolayers were chosen for their capacity to facilitate strong light-matter interactions due to their temperature stability, high radiative decay rate, and notable exciton binding energies. By combining these unique properties, a strong coupling regime was realized.

In addition, a concept of catenary-like field enhancement was developed to control coupling strength. It was discovered that the catenary field enhancement's strength decreases as the cavity's gap width rises, resulting in various levels of Rabi splitting.

Consequently, the predicted Rabi splitting in Au-$MoSe_2$ and Au-WSe_2 heterostructures ranged between 77.86 and 320 meV at ambient temperature. Increased cavity gap and thickness reduced the catenary field enhancement's strength and associated Rabi splitting.

Ultimately, the developed plasmonic metamaterial cavities can manipulate excitons in TMDCs and operate active nanodevices at room temperature. The hybrid structure, for example, allows for a single-photon source thanks to cavity-enhanced spontaneous emission, which is critical for developing quantum information technologies.

Furthermore, these developments are critical to creating nanophotonic devices that can outperform semiconductor electronics in terms of speed, addressing the growing need for ultralow-energy data processing.

The authors of this article delve into the interaction between light and a hybrid nanostructure composed of metallic nanocavities and two-dimensional transition metal dichalcogenide (TMDC) monolayers. The study focuses on the exploration of hybrid states known as polaritons and/or plexcitons, which arise from the strong coupling of light photons and/or plasmons with excitons in TMDC semiconductor materials.

Due to this strong coupling effect, the original independent eigenstates are transformed into a mixed state of light and matter. This hybrid state combines the advantages of photons, such as rapid propagation and low effective mass, with the exciton's strong interparticle interactions and non-linearity, providing an ideal platform for exploring a variety of fascinating physical phenomena.

It also has significant implications for the development of nanophotonic devices. For instance, this hybrid state is crucial for developing nanophotonic devices that could surpass the speed of semiconductor electronics, transitioning from the GHz to the THz regime.

Moreover, when the plasmon resonance in a metallic cavity strongly couples with semiconductor excitons, the resulting plexcitons can overcome the size limitations of photonic dielectrics. This advancement makes it feasible to integrate many devices capable of manipulating light signals at energy levels below femtojoule per bit.

Notably, the proposed design has the potential for developing single-photon sources with high purity and indistinguishability by enhancing spontaneous emission in the coupled cavity.

The realization of single-photon sources could significantly impact the development of quantum communication technology. Moreover, the enhanced interaction between plasmon-excitons paves the way to realize compact, low-energy, and high-speed nanolasers, which are crucial for the development of future on-chip interconnects. Additionally, the scalable near-field enhancement in hybrid nanostructures is applicable for enhanced sensors and other optoelectronic devices.

Therefore, to manipulate the strong light-matter interaction for desired applications, the research group designed a hybrid nanostructure containing plasmon–exciton modes to induce large Rabi splitting.

Plasmonic nanocavities play a significant role due to their ability to confine light in an ultrasmall volume to elucidate the presence of energy exchange between plasmon and exciton modes.

Taking advantage of this, several groups have reported strong coupling between plasmons in metallic nanoantennas and excitons in quantum emitters such as J-aggregates, molecules, or quantum dot (QD) semiconductors. However, many organic molecules must be included in metallic nanoantenna-QE interactions to achieve strong coupling in molecular excitons. Moreover, controlling the electric field confinement around the plasmonic cavity is challenging.

Compared to QD semiconductors, two-dimensional transition metal dichalcogenide (TMDC) monolayers are stable at ambient conditions, making them excellent candidates for observing strong coupling. Furthermore, in the strong coupling of plexcitons, the active control of individual metal nanoparticles should be demonstrated.

To address these issues, the researchers investigated the strong coupling of plasmons in metallic metamaterial nanocavities with excitons in TMDC monolayers.

The introduced plasmonic metamaterial cavity exhibits strong catenary-shaped optical fields. These catenary-shaped optical fields in metal-dielectric-metal (MIM) structures can be formed by coupling surface plasmons in the cavity and following a hyperbolic cosine shape.

It was introduced to control the strength of the cavity's electric field confinement and scale the Rabi splitting. Consequently, the article mainly focuses on the gold metamaterial cavity as the plasmon mode and $MoSe_2$ and WSe_2 as the exciton modes.

It is found that large Rabi splitting, ranging from 77.86 to 320 meV, is achieved by Au-$MoSe_2$ and Au-WSe_2 heterostructures based on highly localized field enhancement in the near field of the Au cavity.

More information: Andergachew Mekonnen Berhe et al, Strong coupling and catenary field enhancement in the hybrid plasmonic metamaterial cavity and TMDC monolayers, *Opto-Electronic Advances* (2024). DOI: 10.29026/oea.2024.230181

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