

Highly nonlinear Mie-exciton-polaritons in monolayer semiconductors placed on WS₂ nanoantennas on a gold substrate

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Abstract: Subwavelength dielectric photonic structures exhibiting low absorption losses and a wealth of distinct Mie resonances in contrast to their plasmonic counterparts¹. Layered transition metal dichalcogenides (TMDs) have excellent performance with a high refractive index, low losses below the bandgap, and straightforward compatibility with various substrates thanks to the van der Waals forces, thus providing a promising novel nanophotonic platform²⁻⁴. Here, we demonstrate that TMDs can also be used as a versatile platform for observation of the strong light-matter interaction. We observe strong coupling between a Mie resonance in 30 nm tall WS₂ nanoantennas attached to a gold substrate and excitons in a monolayer WSe₂ attached on top of the nanoantennas. Fig.1a shows an SEM image of an individual WS₂ nanoantenna. Fig. 1b shows the optical image of an array of WS₂ nanoantennas covered by a monolayer WSe₂. Fig.1c shows the dark-field scattering spectra before and after transferring monolayer WSe₂ collected for WS₂ nanoantennas with the radius increasing from 84 to 155 nm. Thanks to the high refractive index of WS₂ and additional effect of the gold substrate, Mie resonances with a bandwidth as narrow as ~90 meV are achieved. In Fig.1c and d we observe that a characteristic anti-crossing arises as the Mie mode is tuned in resonance with WSe₂ exciton, which signifies observation of the strong light-matter coupling. Crucially, our results show that the observed Rabi splitting for Mie-exciton-polaritons of ~85 meV is of the same order of magnitude as reported in plasmonic systems (~50 meV⁵), but is accompanied by lower absorption losses. The FDTD simulation of a WS₂ nanoantenna wrapped in a monolayer WSe₂ further supports our interpretation (Fig.1e). Furthermore, we observe strong nonlinearity of Mie-exciton-polariton with increasing optical power in comparison with the exciton in bare monolayer WSe₂. Our results highlight van der Waals nanophotonic structures as a versatile platform for the observation of strong light-matter coupling.

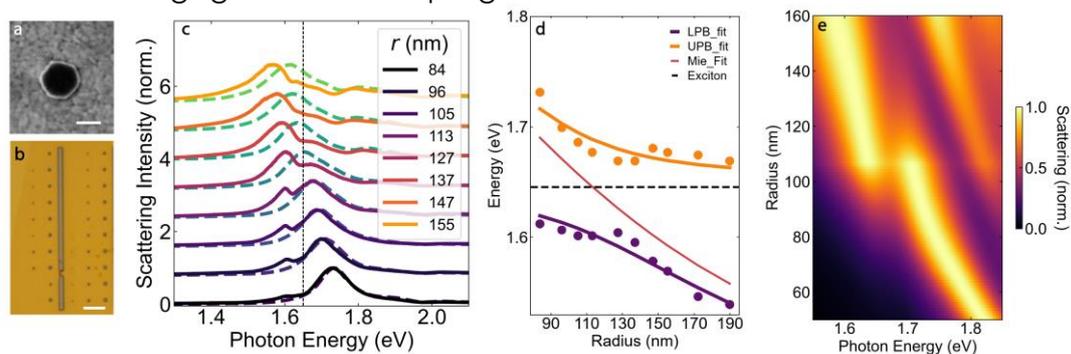


Figure 1. Mie-exciton-polariton achieved by van der Waals materials. a. SEM image of the nanoantenna. The scale bar is 200 nm. b. Optical image of bulk WS₂ nanoantenna covered by monolayer WSe₂ on a gold substrate. The white dashed line indicates the boundary of monolayer region, where the left of line is pure gold. The scale bar is 5 μm. c. Dark-field scattering spectra of WS₂ nanoantenna covered with monolayer WSe₂ (solid curve), with the comparison with nanoantennas before WSe₂ transfer (dashed curve). The vertical dashed line indicates the monolayer exciton position. d. Symbols show peak positions from Fig.c. The solid lines show the fitting obtained using the coupled-oscillator model providing the Rabi splitting of 85 meV. e. Simulated scattering cross section of nanoantenna wrapped with monolayer WSe₂ with the nanoantenna radius.

References

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