Towards active excitonic 2D metasurfaces in monolayer TMDs

Thomas Bauer[1] **and Jorik van de Groep**[1]

1. Van der Waals–Zeeman Institute, Institute of Physics, University of Amsterdam, Amsterdam 1098 XH, the Netherlands

E-mail: t.a.bauer@uva.nl

Monolayer transition metal dichalcogenides (TMDs) like WS_2 exhibit strong exciton resonances in the visible spectral range that dominate their optical response. The excitonic light-matter interaction in these 2D quantum materials is inherently strong and highly tunable, which can be leveraged to realize mutable flat optical elements [1]. To unleash the full potential of exciton-enhanced wavefront shaping and active optical switching in atomically thin metasurfaces, it is essential to gain a detailed understanding of the exciton's quantum mechanical properties and its coupling to hybrid light-matter quasiparticles known as exciton polaritons.

Recently, it was shown theoretically that exciton polaritons can exist as excitations from the continuum of three-dimensional electromagnetic modes even for atomically thin layers of materials [2], with coupling to excitons in a free-standing monolayer membrane demonstrated experimentally [3]. Figure 1(a) shows the calculated dispersion relation of such a TE-polarized exciton polariton in WS_2 on a SiO_2 substrate, capped by PMMA. 2D exciton polaritons (2DEPs) correspond to a strongly coupled light-matter state, enabling novel concepts in photonic sensing and photodetection, as well as probing of fundamental quantum-mechanical properties of light scattering by excitons.

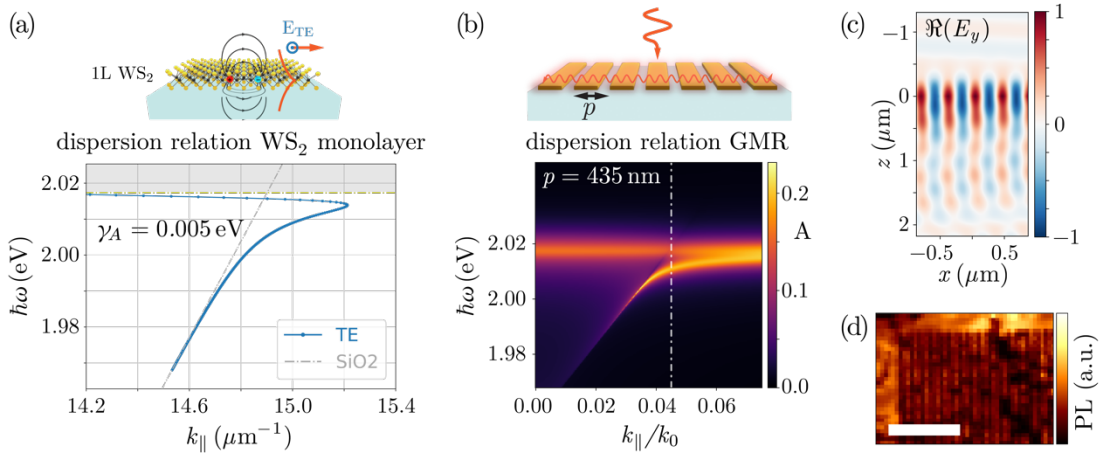

Fig. 1 (a) Dispersion relation of exciton polaritons under TE-polarized excitation in a monolayer WS_2 on top of a SiO_2 substrate (shown as sketch) and covered by 250 nm PMMA, calculated using an eigen-mode solver. A damping constant of $\gamma_A = 5\ meV$ is assumed for the A exciton. The light line of the substrate is shown as dash-dotted gray line, while the A exciton resonance is given by the horizontal dashed ochre line. The high energy side of the exciton is highlighted in gray, where the real part of the susceptibility becomes negative. (b) Dispersion relation of a strongly coupled 2D exciton polariton (2DEP) induced by coupling to a guided mode resonance (GMR) via a grating with period $p = 435\ nm$ structured into the monolayer itself (shown as sketch). The absorption of the 2DEP, calculated via RCWA, shows an enhancement with respect to the bare exciton. (c) Real part of the electric field distribution of the guided mode for an in-plane momentum given by the white dash-dotted line in (b). (d) Photoluminescence (PL) signal of an experimentally fabricated grating structure, demonstrating the negligible influence of the fabrication procedure onto the intrinsic optical response of the grating (scale bar: 2.5 μm).

Here, we experimentally demonstrate active control of coupling to these 2DEPs, allowing for enhanced photonic functionality given directly by the geometry of the monolayer material. Using guided mode resonances in sub-wavelength gratings structured in a WS_2 monolayer (see Fig. 1(b)), we realize polarization-selective dynamic phase control of light scattered off this hybrid light-matter state. The sub-wavelength nature of the grating leads to an electrically tunable phase modulation of the reflected light when exciting close to the 2DEP guided mode resonance. Figure 1(c) visualizes the calculated field distribution of the guided mode, while Figure 1(d) shows the photoluminescence signal of a fabricated grating structure. Further utilization of photonic metasurface concepts allows for active amplitude switching of higher diffraction orders via binary blazed gratings, leading to expected modulation depths exceeding 80%. This opens a path to full active control over the complex optical response in atomically thin metasurfaces via exciton resonance tuning.

References

[1] van de Groep, J., Song, J. H., Celano, U., Li, Q., Kik, P. G. & Brongersma, M. L. (2020). *Nature Photon.*, **14**, 426–430.
[2] Khurgin, J. B. (2015). *Optica*, **2**, 740.
[3] Zhang, X., De-Eknamkul, C., Gu, J., Boehmke, A. L., Menon, V. M., Khurgin, J. & Cubukcu, E. (2019). *Nature Nanotech.,* **14**, 844–850.