

NEWS & VIEWS

Open Access

Optical nonlinearity goes ultrafast in 2D semiconductor-based nanocavities

Armando Genco¹ and Giulio Cerullo¹✉

Abstract

Hybrid systems of silver nanodisks strongly coupled to monolayer tungsten-disulfide (WS_2) show giant room-temperature nonlinearity due to their deeply sub-wavelength localized nature, resulting in ultrafast modifications of nonlinear absorption in a solid-state system.

Decades of scientific progress in the fields of micro and nano-photonics allowed to harness strong interactions between light and matter in solid-state systems, leading to fascinating phenomena and ground-breaking technological applications. Light-matter interactions in semiconductors can be precisely controlled by tuning the coupling between excitons, bosonic quasi-particles made of bound electron-hole pairs, and photons confined in an optical resonator. At high coupling strengths, resonant photons coherently exchange energy with excitons at a rate (Rabi frequency, Ω_R) higher than that of the dephasing processes (i.e., the rate at which photons escape from the cavity or excitons dephase), entering the so-called strong coupling (SC) regime. From this process, new hybrid quantum states are formed, called polaritons. Reaching the SC regime opened new avenues for observing highly nonlinear optical phenomena in the solid state, such as Bose-Einstein condensation^{1,2}, polariton lasing^{3,4} and optical parametric amplification⁵. These discoveries have been recently exploited for the creation of all-optical logic gates⁶ and polariton-based neural networks⁷.

Among the different types of semiconductor systems, monolayers of transition metal dichalcogenides (TMDs) stand out as a highly promising platform for exploring the nonlinear effects produced in the SC regime. Charge carriers in such materials experience an out-of-plane quantum confinement generated by their intrinsic 2D

structure which, together with the reduced Coulomb screening, leads to excitons with huge binding energies (several hundreds of meV) and prominent oscillator strengths, persisting up to room temperature^{8,9}. Moreover, the broken spatial inversion symmetry of the 2D lattice, together with a large spin-orbit coupling, provides an uncommon pseudo-spin degree of freedom for excitons in the K and K' valleys of the Brillouin zone. Valley excitons thus become selectively addressable by exciting them with oppositely circularly polarized light¹⁰, whose valley coherence can be even enhanced by the strong coupling with long-living cavity photons¹¹. Excitons in TMDs uniquely combine high temperature stability with large nonlinearities, which makes them an ideal solution for room-temperature polaritonics.

In order to reach the SC regime, TMD monolayers can be coupled to different optical resonators. Seminal studies of TMD polaritons exploited atomically thin flakes embedded in vertical microcavities (Fig. 1a). Such devices consist of two highly reflective planar or concave mirrors facing each other and distanced hundreds of nanometers, confining photons at discrete energy levels, the “cavity modes”, whose frequencies are determined by the distance between the mirrors. The cavity mirrors can be made of metallic films or of Distributed Bragg Reflector (DBR), a sequence of pairs of dielectric layers with high/low refractive index. In such devices, SC was demonstrated for neutral excitons^{12,13}, trions^{14,15} and excited Rydberg excitons¹⁶, using monolayers either mechanically exfoliated or grown by chemical vapor

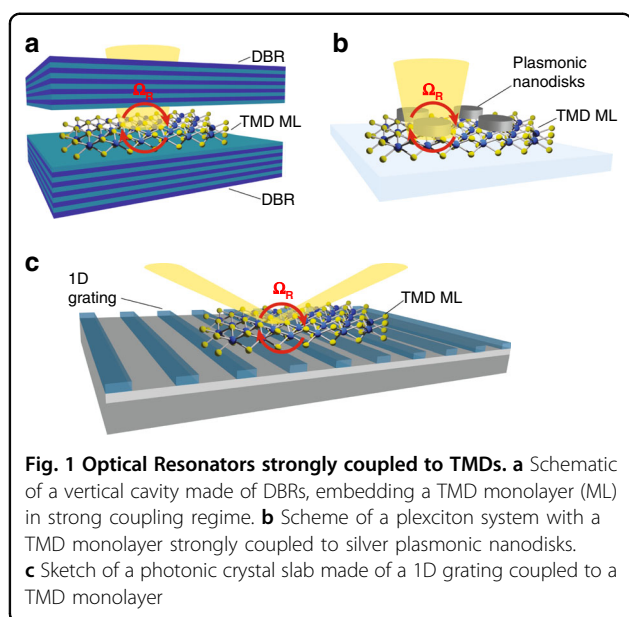
Correspondence: Giulio Cerullo (giulio.cerullo@polimi.it)

¹Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy

© The Author(s) 2022



Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0/>.



deposition on a large scale¹⁷. TMD polaritons have been also observed in planar architectures, towards on-chip optical circuits, exploiting the sub-diffraction spatial light confinement of plasmonic nanoresonators (Fig. 1b)^{18,19}, or involving propagating modes, as for waveguides²⁰, Bloch Surface Waves (BSW)²¹, surface lattice resonances^{22,23}, photonic crystal slabs (1D gratings)²⁴ (Fig. 1c) and Bound states In the Continuum (BIC)²⁵.

Studies of polariton nonlinearities are of key importance for the understanding of the nonlinear optical processes which form the basis of coherent emission, for ultrafast optical switches and for optical parametric scattering, the latter being observed very recently in a TMD vertical cavity²⁶. However, investigations of such effects on the ultrafast timescale and in deep-subwavelength optical structures have been very limited so far.

Tang et al.¹⁸ explore the ultrafast dynamics of the nonlinear optical response for WS₂ monolayers strongly coupled to plasmonic silver nanodisks, unveiling a giant nonlinearity and studying the underlying physical processes. The non-equilibrium dynamics of the samples were analysed by using femtosecond pump-probe spectroscopy, revealing different exciton-plasmon nonlinear processes over distinct timescales. The main mechanisms which govern the optical nonlinearity in TMD-based microcavity polariton devices²⁷ are identified as (i) the repulsive Coulombic interaction between excitons, which usually reflects in a blueshift of the resonance energy, and (ii) phase space filling (or Pauli blocking), inducing excitonic saturation and reducing the exciton-photon coupling strength. In addition, excitation-induced dephasing in TMDs has been recently found to play a non-negligible role at high fluences²⁸, typically giving rise to a

broadening of excitonic linewidths. The prominent role of the latter mechanism was demonstrated in this work, studying the nonlinear behaviour of plasmon-exciton polaritons (also known as plexcitons) as a function of the excitation fluence. Very interestingly, the authors show a direct application of the strong nonlinear interactions that they found, exploiting the flexibility of the system to easily modify the light-matter coupling strength. This resulted in the observation of direct and reverse absorption saturation at ultrafast timescales, which showcase the potential of TMD polaritons for future uses in sub-wavelength nonlinear optical devices.

There is ample room for further ultrafast studies of the nonlinear optical response of TMD polaritons, from the demonstration of ultrafast all-optical switches working in a low excitation density regime to the holy grail of time-domain observation of Rabi oscillations at the frequency Ω_R ²⁹. Recently, it has been shown that greatly enhanced polariton nonlinearities can be achieved using moiré-lattice excitons in twisted TMD hetero-bilayers³⁰ and hybridized dipolar excitons in homo-bilayers³¹; it will be interesting to study the out-of-equilibrium optical response for those systems. An additional enticing perspective is to extend the investigation of ultrafast exciton-polariton nonlinearities to other systems featuring strongly bound excitons, such as three- and two-dimensional perovskites³².

Acknowledgements

We acknowledge funding from the European Union Horizon 2020 Programme under Grant Agreement No. 881603 Graphene Core 3 and from the Marie Skłodowska-Curie Actions (project ENOSIS) H2020-MSCA-IF-2020-101029644.

Published online: 07 May 2022

References

- Deng, H., Haug, H. & Yamamoto, Y. Exciton-polariton Bose-Einstein condensation. *Rev. Mod. Phys.* **82**, 1489–1537 (2010).
- Kasprzak, J. et al. Bose-Einstein condensation of exciton polaritons. *Nature* **443**, 409–414 (2006).
- Christopoulos, S. et al. Room-temperature polariton lasing in semiconductor microcavities. *Phys. Rev. Lett.* **98**, 126405 (2007).
- Bhattacharya, P. et al. Room temperature electrically injected polariton laser. *Phys. Rev. Lett.* **112**, 236802 (2014).
- Amo, A. et al. Collective fluid dynamics of a polariton condensate in a semiconductor microcavity. *Nature* **457**, 291–295 (2009).
- Ballarini, D. et al. All-optical polariton transistor. *Nat. Commun.* **4**, 1778 (2013).
- Berloff, N. G. et al. Realizing the classical XY hamiltonian in polariton simulators. *Nat. Mater.* **16**, 1120–1126 (2017).
- Wang, G. et al. *Colloquium: excitons in atomically thin transition metal dichalcogenides*. *Rev. Mod. Phys.* **90**, 021001 (2018).
- Mak, K. F. & Shan, J. Photonics and optoelectronics of 2D semiconductor transition metal dichalcogenides. *Nat. Photonics* **10**, 216–226 (2016).
- Xu, X. D. et al. Spin and pseudospins in layered transition metal dichalcogenides. *Nat. Phys.* **10**, 343–350 (2014).
- Dufferwiel, S. et al. Valley coherent exciton-polaritons in a monolayer semiconductor. *Nat. Commun.* **9**, 4797 (2018).
- Dufferwiel, S. et al. Exciton-polaritons in van der Waals heterostructures embedded in tunable microcavities. *Nat. Commun.* **6**, 8579 (2015).

13. Liu, X. Z. et al. Strong light–matter coupling in two-dimensional atomic crystals. *Nat. Photonics* **9**, 30–34 (2015).
14. Emmanuele, R. P. A. et al. Highly nonlinear trion-polaritons in a monolayer semiconductor. *Nat. Commun.* **11**, 3589 (2020).
15. Sidler, M. et al. Fermi polaron-polaritons in charge-tunable atomically thin semiconductors. *Nat. Phys.* **13**, 255–261 (2017).
16. Gu, J. et al. Enhanced nonlinear interaction of polaritons via excitonic Rydberg states in monolayer WSe₂. *Nat. Commun.* **12**, 2269 (2021).
17. Gillard, D. J. et al. Strong exciton-photon coupling in large area MoSe₂ and WSe₂ heterostructures fabricated from two-dimensional materials grown by chemical vapor deposition. *2D Mater.* **8**, 011002 (2021).
18. Tang, Y. X. et al. Interacting plexcitons for designed ultrafast optical nonlinearity in a monolayer semiconductor. *Light Sci. Appl.* **11**, 94 (2022).
19. Sun, J. W. et al. Strong plasmon–exciton coupling in transition metal dichalcogenides and plasmonic nanostructures. *Nanoscale* **13**, 4408–4419 (2021).
20. Hu, F. et al. Imaging exciton–polariton transport in MoSe₂ waveguides. *Nat. Photonics* **11**, 356–360 (2017).
21. Barachati, F. et al. Interacting polariton fluids in a monolayer of tungsten disulfide. *Nat. Nanotechnol.* **13**, 906–909 (2018).
22. Wang, S. J. et al. Limits to strong coupling of excitons in multilayer WS₂ with collective plasmonic resonances. *ACS Photonics* **6**, 286–293 (2019).
23. Wang, S. J. et al. Coherent coupling of WS₂ monolayers with metallic photonic nanostructures at room temperature. *Nano Lett.* **16**, 4368–4374 (2016).
24. Zhang, L. et al. Photonic-crystal exciton-polaritons in monolayer semiconductors. *Nat. Commun.* **9**, 713 (2018).
25. Kravtsov, V. et al. Nonlinear polaritons in a monolayer semiconductor coupled to optical bound states in the continuum. *Light Sci. Appl.* **9**, 56 (2020).
26. Zhao, J. X. et al. Nonlinear polariton parametric emission in an atomically thin semiconductor based microcavity. *Nat. Nanotechnol.* **17**, 396–402 (2022).
27. Stepanov, P. et al. Exciton-exciton interaction beyond the hydrogenic picture in a MoSe₂ monolayer in the strong light-matter coupling regime. *Phys. Rev. Lett.* **126**, 167401 (2021).
28. Katsch, F., Selig, M. & Knorr, A. Exciton-scattering-induced dephasing in two-dimensional semiconductors. *Phys. Rev. Lett.* **124**, 257402 (2020).
29. Vasa, P. et al. Real-time observation of ultrafast Rabi oscillations between excitons and plasmons in metal nanostructures with J-aggregates. *Nat. Photonics* **7**, 128–132 (2013).
30. Zhang, L. et al. Van der Waals heterostructure polaritons with moiré-induced nonlinearity. *Nature* **591**, 61–65 (2021).
31. Louca, C. et al. Nonlinear interactions of dipolar excitons and polaritons in MoS₂ bilayers. Preprint at <https://doi.org/10.48550/arXiv.2204.00485> (2022).
32. Su, R. et al. Perovskite semiconductors for room-temperature exciton-polaritons. *Nat. Mater.* **20**, 1315–1324 (2021).