Excitonic lasers in atomically thin 2D semiconductors

Wen, Wen; Wu, Lishu; Yu, Ting

2020

Wen, W., Wu, L. & Yu, T. (2020). Excitonic lasers in atomically thin 2D semiconductors. ACS Materials Letters, 2(10), 1328-1342. <https://dx.doi.org/10.1021/acsmaterialslett.0c00277>

<https://hdl.handle.net/10356/146650>

<https://doi.org/10.1021/acsmaterialslett.0c00277>

This document is the Accepted Manuscript version of a Published Work that appeared in final form in ACS Materials Letters, copyright © American Chemical Society after peer review and technical editing by the publisher. To access the final edited and published work see <https://doi.org/10.1021/acsmaterialslett.0c00277>

Downloaded on 26 Apr 2023 21:24:47 SGT

Excitonic Lasers in Atomically Thin 2D Semiconductors

Wen Wen[†], Lishu Wu[†], Ting Yu^{†,}*

[†]Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

Abstract

Two-dimensional (2D) atomically thin transition-metal dichalcogenides (TMD) and their van der Waals (vdW) heterostructures offer a platform with tightly bound intralayer/interlayer excitons for the on-chip fabrication of ultracompact nanolasers. Excitons in 2D TMD materials present a considerable binding energy of up to hundreds of meV, which permits a high Mott transition density of 10^{14} cm^{-2} and stable excitonic lasing under room-temperature operation and high pump fluences. Here, we review the recent progress on the lasing emission from intralayer excitons in TMD monolayers and interlayer excitons in vdW heterostructures incorporated with various high-quality optical cavities, including photonic-crystal, whispering-gallery-mode, distributed-feedback, distributed-Bragg-reflector cavities. Lasing emissions in TMD monolayers and heterostructures have been demonstrated by narrow emission peaks, a clear threshold for nonlinear amplification, time- and spatial coherence under either continuous-wave or pulsed light pumping. Finally, prospective and frontier research topics, including large-scale on-chip integration of TMD nanolasers, electrically pumped lasers, spin-polarized nanolasers and exciton-polariton Bose-Einstein condensation (BEC) are highlighted.

1. Introduction

Two-dimensional (2D) atomically thin transition-metal dichalcogenides (TMDs), MX_2 (M is Mo or W; X is S, Se, or Te), are intriguing semiconductors with emission bandgaps ranging from the visible to the near infrared region,¹⁻³ contrary to graphene with zero bandgap. In their monolayer limit, they have been reported to own direct bandgaps,⁴⁻⁶ strong excitonic effect with tightly bound excitons,⁷⁻⁹ strong light-matter interaction,¹⁰ spin-valley locking¹¹⁻²⁰ and considerable charge carrier mobility,²¹⁻²³ which envisage unique electronic, photonic and optoelectronic applications, such as field-effect transistors,²⁴⁻²⁶ light-emitting diodes,²⁷⁻³⁰ optical modulators^{31, 32} and nanolasers.^{33, 34} In 2010, the research into optical properties of TMDs began with the discovery of indirect-to-direct bandgap transition from bulk to monolayer crystals, which results in orders of magnitude improvement of photoluminescence (PL) quantum efficiency from bulks to monolayers.^{5, 6} In 2012, the discovery of spin-valley locking originating from broken inversion symmetry and strong spin-orbit coupling offers new opportunities for the accessibility to explore valleytronic applications that make use of the spin and valley degrees of freedom in TMD monolayers.^{13, 14}

Beyond TMD monolayers, van der Waals (vdW) heterostructures constructed by stacking of diverse atomically thin materials, including semi-metallic graphene, insulating hexagonal boron nitride (hBN), TMDs and other layered materials, provide a novel platform to investigate the exciton excited states, carrier dynamics, spin-valley coupling and moiré potentials.³⁵⁻⁴⁶ Meanwhile, vdW heterostructures also offer a new paradigm to design the superior properties of materials and functionalities of devices because the material properties can be tuned by constituent layers, stacking sequence,^{30, 47, 48} crystallographic orientation⁴⁹⁻⁵¹ and external fields.⁵²⁻⁵⁶ In the vdW heterostructures composed of TMDs, interlayer excitons can be created through either ultrafast charge transfer in optical excitation process or direct electrical injection of electrons/holes into two separated layers.^{37, 39, 42, 53, 57-59} Compared to their intralayer counterparts, interlayer excitons feature longer lifetime,⁵⁷ directional dipole⁵² and longer valley polarization lifetime,⁶⁰ which allow for the long-range exciton diffusion, tunable carrier dynamics by electrostatic gating and manipulation of spin-polarized

carriers at different valleys.^{42, 56, 61, 62}

In addition, based on these atomically thin materials with tightly bound excitons, ultracompact on-chip lasers have been developed. In 2015, the first demonstration of lasing emissions was reported in WSe₂ incorporated with a photonic crystal cavity.³³ After then, the lasing emissions of TMD monolayers and heterostructures have been also implemented by using photonic-crystal,^{33, 63, 64} distributed-feedback,⁶⁵ distributed-Bragg-reflector⁶⁶ and whispering-gallery-mode cavities.^{34, 67} By the material engineering and optical cavity optimization, continuous-wave lasing emissions with a relatively low threshold at room temperature have been realized on both monolayers and vdW heterostructures. In this review, we summarize recent progress in atomically thin 2D nanolasers based on TMD monolayers and heterostructures. Firstly, we briefly introduce the properties of intralayer excitons in TMD monolayers and interlayer excitons in vdW heterostructures. Secondly, the atomically thin lasers based on intralayer excitons and various optical cavities are discussed. Thirdly, lasing emission and Bose-Einstein condensation (BEC) from interlayer excitons in vdW heterostructures are discussed. We describe the material fabrication, optical cavity design and lasing performance of lasers based on TMD monolayers and heterostructures. The lasing emissions in TMD materials have been characterized by a narrowed peak, a threshold in the output intensity and emergent spatial coherence. We conclude the review by highlighting the future research directions, including exciton-polariton lasers, spin lasers, electrically pumped lasers and scalable fabrication of lasers based on TMD materials.

2. Excitons in TMD monolayers and heterostructures

The excitons in TMD monolayers and heterostructures have attracted intense attention owing to the following features. First, TMDs undergo a transition from an indirect to a direct bandgap, when the thickness of TMDs is thinned down to monolayers (Figure 1a),⁵ which is reflected by a great enhancement of PL quantum efficiency with a factor of exceeding 10^4 in monolayers.

Second, the stable excitons with a large binding energy of hundreds of meV have

been demonstrated, which is attributed to the quantum confinement effect and significantly reduced dielectric screening in TMD monolayers (Figure 1b-d). Heinz and his colleagues discovered the excitonic Rydberg series ($n = 1-5$) through reflectance spectra in WS₂ monolayers (Figure 1b).⁸ The $n = 3-5$ peaks can be fitted by a 2D hydrogen model. Exciton transition energies can be calculated by bandgap (E_g) and exciton binding energy of n^{th} excitonic state ($E_b^{(n)}$) as $E_g - E^{(n)}$ and $E_b^{(n)}$ can be expressed as,

$$E_b^{(n)} = \frac{\mu e^4}{2\hbar\epsilon(n - 1/2)^2} \quad (1)$$

where μ is the reduced mass of exciton, ϵ is the permittivity. The model is well fitted with $n = 3-5$ excitons and indicates a bandgap of $E_g = 2.41 \pm 0.04$ eV. And the exciton binding energy of 1s exciton state is 0.32 ± 0.04 eV by considering its exciton transition energy of 2.09 eV (Figure 1c). The non-hydrogenic physics of $n = 1, 2$ excitons are caused by the higher effective dielectric constants of excitons with low quantum numbers (Figure 1c inset). Shan and her colleagues demonstrated the measurement of band-to-band transition in WSe₂ monolayers by combining linear absorption and two-photon PL (2PPL) excitation spectra (Figure 1d).⁷ While linear absorption could not identify a clear band-to-band transition, the 2PPL excitation spectrum can be well fitted by an exciton peak related to the superposition of p states and a band-to-band transition with $E_g = 2.02$ eV. The binding energy of 1s excitons was calculated as 0.37 eV based on the 1s exciton transition energy and band-to-band transition energy. The considerable high exciton binding energy in TMD monolayers permits stable excitons under relatively high population and sustains excitonic lasing emissions.

Figure 1. Excitons in TMD monolayers and heterostructures. (a) Calculated electronic band structure of bulk (left) and monolayer (right) MoS₂. Reproduced from Ref ⁵. Copyright 2010 ACS. (b) Derivative reflectance contrast spectrum of the WS₂ monolayer illustrates excitonic Rydberg states. (c) Experimentally measured and theoretically calculated transition energies of exciton states. The solid line shows the fit of $n=3, 4, 5$ states to the 2D hydrogen model for Wannier excitons. Inset presents effective dielectric constants. Panel (b, c) reproduced from Ref ⁸. Copyright 2014 APS. (d) Linear absorption (red line) and 2PPL excitation spectra of monolayer WSe₂ collected at room temperature. Reproduced from Ref ⁷. Copyright 2014 APS. (e) Band edges of representative TMDs calculated by PBE-SOC (filled grey) and GW (narrower olive) methods. Reproduced from Ref ⁷⁵. Copyright 2017 IOP. (f) PL emissions of WSe₂, MoSe₂ and WSe₂-MoSe₂ heterostructure. Emergent PL peak at 1.4 eV indicates the interlayer excitons in WSe₂-MoSe₂ heterostructure. (g) PL emission of WSe₂-MoSe₂ heterobilayer as a function of applied out-of-plane gate voltage. Panel (f, g) Reproduced from Ref ⁵⁷. Copyright 2015 Springer Nature. (h) Dynamics of total hole population ($p_+ + p_-$), valley polarized hole populations ($p_+ + p_-$) and polarization degree (P) show a hole population lifetime exceeding 1 μ s and valley polarization lifetime of more than 40 μ s. Reproduced from Ref ⁶⁰. Copyright 2017 AAAS.

Third, broken inversion symmetry and strong spin-orbit coupling cause

spin-valley locking, which gives rise to intriguing properties, such as valley Hall effect,^{11, 69, 70} valley polarized excitons,⁴² valley Zeeman⁷¹⁻⁷³ and AC stark effects.¹⁵ Valley polarized lasing emission has yet been realized in TMDs possibly due to the depolarization caused by the electron-hole exchange interaction, which is enhanced under high exciton population. Therefore, in this review, we will not further discuss the spin-valley polarization and some comprehensive reviews on this topic are recommended.^{4, 18, 74}

Fourth, atomically sharp heterostructures can be constructed by stacking different kinds of TMD layers. The library of 2D TMDs allows for constructing type-II heterostructures, which support ultrafast charge transfer and formation of interlayer excitons.⁷⁵ Density functional theory (DFT) calculations have been performed to predict the band edges of TMD monolayers, which has provided guidelines for the construction of type-II heterostructures (Figure 1e).^{68, 75} Before the discovery of interlayer excitons, the ultrafast charge transfer has been demonstrated by Wang and his colleagues in type-II MoS₂/WS₂ heterobilayers.³⁷ PL mapping demonstrated a strongly quenched PL in the heterostructure region, indicating the efficient charge transfer. By using femtosecond pump-probe spectroscopy, ultrafast hole transfer from MoS₂ to WS₂ at a time scale of within 50 fs has been revealed. The ultrafast charge transfer in heterostructures enables the buildup of high population of interlayer excitons before the recombination of intralayer excitons. Xu and his colleagues firstly reported the formation of interlayer excitons between WSe₂-MoSe₂ heterobilayers (Figure 1f).⁵⁷ PL spectrum of WSe₂-MoSe₂ shows an interlayer exciton peak at lower energy of 1.4 eV, while the emissions of neutral (X^0) and charged (X^-) excitons range from 1.55 to 1.75 eV. Interlayer excitons with electrons in MoSe₂ and holes in WSe₂ exhibit directional dipoles from MoSe₂ to WSe₂, which supports tuning of excitonic emission by electric field (Figure 1g). Compared to the rapid exciton recombination with the lifetime of 3-100 ps, the interlayer excitons present a much longer lifetime of over 1 μ s (Figure 1h).⁶⁰ More importantly, exciton-hole exchange interaction for the depolarization process is suppressed owing to the spatial separation of electrons and holes, yielding an ultralong valley polarization lifetime of 40 μ s.⁶⁰

Interlayer excitons with electrons and holes in two separated layers have been demonstrated with longer carrier lifetime, valley polarization lifetime, tunable polarization by electric field and controllable carrier species and dynamics through electrical injection. Therefore, interlayer excitons are promising candidates to be used in spin lasers and electrically pumped lasers based on TMD heterostructures.

3. Lasing emission in TMD monolayers

Lasers comprise gain media for stimulated photon emission and optical cavities for confinement and resonant recirculation of light.⁷⁶ As an ultrathin quantum emitter, monolayer TMD is a category of high-performance gain media with unique advantages of small footprints, low power consumption, efficient coupling with microcavities and feasible on-chip integration.³ TMD monolayers possess tightly-bound excitons with binding energy of exceeding 0.3 eV.⁴ Such a large binding energy sustain stable excitons at the room temperature and high Mott transition density of $\sim 10^{14} \text{ cm}^{-2}$. Therefore, stable excitons can be preserved even at the pump fluence level for achieving population inversion. These excitons also possess high radiative recombination quantum efficiency owing to the intrinsic direct bandgap in TMD monolayers.^{5,6} Strong excitonic resonance and small Bohr radius also benefit efficient and fast radiative recombination, which is important for the buildup of population inversion. For TMD monolayer lasers, the design and optimization of optical cavities are crucial for realizing low-threshold lasing emission. Owing to the ultrathin size, TMD monolayers are not capable of confining emitted photons, which requires additional optical cavities to sustain high optical gain. In this section, we will review lasers constructed by TMD monolayers as gain media and different optical cavities. Before the discussion of specific reports, we would like to recommend the Commentary Article on the criteria of lasing emission, which include narrow linewidth, clear threshold, small beam divergence, polarization, spatial and temporal coherence.⁷⁷

Figure 2. Lasers based on TMD monolayers and photonic-crystal cavities. (a) Schematic illustration of monolayer WSe₂ onto photonic-crystal cavities. (b) SEM image presents a WSe₂ monolayer on photonic crystal. Output intensity of the TMD monolayer laser as a function of incident pump power at (c) 130 K, and (d) 80 K. (e) Representative photoluminescence spectra at pump powers labeled in (d). Panel (a-e) reproduced from ref ³³. Copyright 2015 Springer Nature. (f) Scheme of MoTe₂ monolayer onto a Si photonic crystal suspended in air. (g) Power-dependent photoluminescence spectra of a MoTe₂ laser at room temperature. Inset is a SEM image of a suspended photonic crystal. (h) Output intensity of a MoTe₂ laser as a function of pump power. The solid lines are fitted results by rate equation with different spontaneous emission factors β . Panel (f-h) reproduced from ref ⁶⁴. Copyright 2017 Springer Nature.

3.1 Photonic crystal lasers

Photonic crystal cavities are usually fabricated by etching periodic structures on membranes with high refractive indexes.⁷⁸ These periodic structures show tunable photonic bandgaps, which prevent light propagation at corresponding frequencies. By introducing defects in periodic structures, strong confinement of electromagnetic field can be realized, yielding microcavities with small volumes and large quality factors exceeding 10^5 .⁷⁹ Therefore, photonic-crystal cavities have shown broad applications

in optically/electrically pumped nanolasers, cavity quantum electrodynamics and single-photon sources. The photonic crystal cavities have enabled the fabrication of high-performance nanolasers with low thresholds, high quality factors and ultrafast modulation.⁸⁰⁻⁸³ For instance, quantum dots, a kind of quantum emitters with ultrasmall mode volume, have been incorporated into photonic crystal cavities to create nanolasers.^{84, 85} Photonic crystals also provide a platform for the integrated photonics based on 2D materials. Nanolasers, waveguides, interferometers, modulators and photodetectors can be integrated onto a photonic-crystal chip to build photonic logic devices.^{32, 86, 87}

The first TMD-monolayer laser was reported by Xu and his colleagues in 2015.³³ This nanolaser was constructed by loading a mechanically exfoliated WSe₂ monolayer onto a photonic-crystal cavity (Figure 2a, b). To ensure the high gain and low loss, a L3-type photonic crystal cavity was fabricated on a gallium phosphide (GaP) thin membrane, which is transparent to WSe₂. This photonic crystal cavity presented a quality factor of about 10⁴, which created strong local optical density of states. Power dependent PL measurements unambiguously demonstrated lasing emission of this TMD monolayer laser. At 80 and 130 K, a clear ‘kink’ has been observed in the pump-power-dependent output intensity curves (Figure 2c, d). In contrast, the intensities of spontaneous emission presented no obvious ‘kink’ as a function of pump power. The power-dependent intensity can be fitted by the rate equation as follows,

$$R_{\text{ex}} = \frac{P}{\Gamma t_c (1 + aP)} \left(\frac{1}{\beta} + aP \right) \quad (2)$$

where R_{ex} is the optical pumping rate, P is the cavity photon number, Γ is the cavity confinement factor, a is a coefficient, β factor is defined as the fraction of spontaneous emission into the cavity mode. A β factor of 0.19 is best fitted for the power-dependent output intensity curve. The linewidth of PL peak is also dependent on the pump power (Figure 2e). With the increase of pump power, linewidth of PL peak experienced a narrowing from 0.75 to 0.55 nm below the threshold. Across the threshold regime, a slight broadening of linewidth to 0.65 nm occurred and then the linewidth continuously decreased to 0.55 nm above the threshold. The broadening of

linewidth in threshold regime can be attributed to the coupling between intensity and phase noise during the transition from spontaneous emission to stimulated emission, which has been observed in other photonic-crystal lasers based on quantum dots and quantum wells. This nanolaser also presented localized lasing emission at the defect sites in the photonic crystal. At the defect site, narrow PL peak related to stimulated emission of WSe₂ in cavity can be observed. In contrast, a broad PL peak related to spontaneous emission from WSe₂ can be observed in the off-cavity region. This work demonstrated, for the first time, that lasing emission can be realized on an ultrathin quantum emitter with stable excitons.

The above reported nanolaser based on WSe₂ and GaP photonic crystal cavities can only be operated at cryogenic temperature, as the room-temperature operation of this nanolaser might be limited by its relatively low quality factor. Silicon exhibits a high refractive index of 3.4 and matured fabrication techniques for the construction of photonic crystal structures. However, silicon photonic crystal cavities are not suitable for the fabrication of nanolasers based on MoS₂, WS₂, MoSe₂ and WSe₂, owing to the strong absorption of photons with energy below 1.1 eV for silicon. The self-absorption introduces large losses in cavities, which is detrimental to the lasing emission. Ning and his colleagues circumvented this restriction and reported a room-temperature nanolaser based on silicon photonic cavity by employing monolayer MoTe₂ as gain media.⁶⁴ Monolayer MoTe₂ exhibits an excitonic emission peak at ~1.1 eV, which is 50 meV below the bandgap of silicon. By covering monolayer MoTe₂ onto silicon photonic crystal (Figure 2f), infrared lasing emissions were realized at room temperature.

The photonic crystal cavity was fabricated by electron-beam lithography and etching on a silicon-on-insulator chip, yielding a suspended nanobeam structure (Figure 2g, inset). This nanobeam cavity contained 10 periods of holes at each side as ‘mirror’ sections, while 8 holes with gradually decreased size served as a cavity. This photonic crystal cavity presents three resonant cavity modes at wavelength of 1054, 1132 and 1167 nm with quality factor of 5.2×10^6 , 6.5×10^5 and 1.4×10^3 , respectively. An exfoliated MoTe₂ monolayer was directly transferred onto the nanobeam cavity to

construct a nanolaser. The nanolaser was pumped by a 633 nm continuous-wave laser at room temperature. With the increase of pumped power, a narrow PL peak at 1132 nm can be observed, indicating the lasing emission (Figure 2g). The full width at half maximum (FWHM) is 0.202 nm, corresponding to a quality factor of 5603. This quality factor is higher than that of 2465 observed in the GaP cavity at cryogenic temperature. The greatly enhanced quality factor enables lasing emission at room temperature. The output intensities presented an obvious threshold of 0.097 mW with the increase of pumped power. The output-intensity curve can be fitted by rate equation with a β factor of 0.1. In summary, this work demonstrated a low-threshold infrared laser based on a MoTe₂ monolayer and suspended photonic crystal cavity, which highlights the importance of high-quality factor of cavity to the high-performance nanolasers.

3.2 Whispering gallery mode lasers

Whispering gallery mode (WGM) microcavities,⁷⁶ such as microdisks, microspheres and optical fibers, have been demonstrated with ultrahigh quality factor for the application of small lasers,⁸⁸ chemical/biological sensors,^{89, 90} and frequency combs.^{91, 92} Ultrahigh quality factor of exceeding 10^8 have been demonstrated in microdisks and optical fibers,^{90, 93, 94} permitting the realization of low-threshold nanolasers based on TMD monolayers.

Zhang and his colleagues reported a nanolaser by combining a WS₂ monolayer with a WGM cavity. This nanolaser was fabricated by embedding a WS₂ monolayer between dielectric layers of Si₃N₄ and HSQ to form a WGM cavity (Figure 3a, b).³⁴ By carefully designing microcavity, a microdisk with a diameter of 3.3 μ m was employed, presenting a resonant transverse electric (TE)-polarized WGM at a wavelength of 612 nm (Figure 3c). This resonant mode overlapped with the exciton emission peak and showed a high-quality factor of 2604.

Figure 3. TMD monolayer nanolasers based on whispering gallery mode cavities. (a) Scheme of the crystal structure of WS₂ monolayer. (b) Illustration of microdisk laser based on WS₂ monolayer. (c) Simulated electric field distribution of TE resonance in a microdisk cavity. (d) PL spectra of WS₂ nanolaser under different pump intensities, showing a narrow lasing peak when pump intensity is above the threshold. (e) Output intensity of WS₂ nanolaser as a function of pump intensity. The solid lines indicate the fitting of data by rate equation. (f) FWHM of a monolayer WS₂ laser as a function of pump intensity. Reproduced from ref ³⁴. Copyright 2015 Springer Nature.

The lasing emissions of WS₂ WGM nanolasers were optically pumped by a 473 nm pulsed laser (190 fs pulse duration, 80 MHz repetition rate) at 10 K. The PL emissions of monolayer WS₂ in cavity shows a clear power dependence: at low pump power intensity below 3.14 MW cm⁻², a broad peak corresponding to spontaneous emission of WS₂ was observed; as the power exceeds 22.4 MW cm⁻², a narrow peak at 612.2 nm appeared, indicating the lasing emission of WS₂ (Figure 3d). The power-dependent emission intensities of a WS₂ nanolaser has been fitted by rate equation, displaying a threshold of ~5-8 MW cm⁻² and a β factor of 0.5 (Figure 3e). The FWHM as a function of pump intensity also shows a clear narrowing of peak

above threshold, demonstrating the transition from spontaneous emission to stimulated lasing emission (Figure 3f). This work represents the first demonstration of a WGM laser based on TMD monolayers. The ultrahigh quality factors of WGM cavities permit the realization of low-threshold lasing emission. Compared to photonic-crystal nanolasers, which restricted by the self-absorption in III-V or Si semiconductor nanocavities, the introduction of dielectric WGM cavities also circumvent the restriction of material selection.

Besides the microdisks, other categories of WGM cavities can also be employed to construct high-performance nanolasers based on TMDs. Salehzadeh et al. reported a room-temperature laser with multiple lasing modes based on four-layer MoS₂ and a free-standing microdisk and microsphere cavity.⁹⁵ The four-layer MoS₂ was employed and treated with O₂ plasma to obtain a direct bandgap. Compared to monolayers, four-layer MoS₂ were selected by considering the suppressed Auger recombination with the increase of thickness. By sandwiching MoS₂ layers between a microdisk and a microsphere, a confined strong optical field can be achieved at the microdisk/microsphere interface. Through 3D finite-difference time-domain (FDTD) simulation, remarkably enhanced electric field at the interface between the SiO₂ microdisk and microsphere were demonstrated, which benefits optical confinement and realization of low-threshold lasing emission. Multi-mode lasing emissions were observed as the pump power exceeds a threshold of 5 μ W. This work demonstrated that lower-threshold and room-temperature lasing emission can be realized by optimizing the microcavity to create a confined strong optical field.

Passivation of TMD monolayers is another significant approach for lowering the threshold and realization of room-temperature operation of lasers.⁹⁶⁻⁹⁹ Recently, Liao et al. demonstrated a WGM mode laser with a threshold of 5 W cm⁻² based on CVD-grown MoS₂.⁶⁷ Micro/nanofibers were employed as WGM cavity and MoS₂ were directly grown onto these cavities by a CVD process. The taper-drawing processing was carried out to fabricate micro/nanofiber cavities of 1 μ m in diameter from standard fibers of 125 μ m in diameter. In this process, the breaking of siloxane bonds occurs to generate oxygen dangling bonds. The authors demonstrated that these

dangling bonds can effectively passivate the defects in MoS₂ monolayers under a wide power dynamic range. At low pump power of less than 1 W cm⁻², the quantum yield of 30% can be achieved. Even under a high pump power of 10⁴ W cm⁻², a relatively high quantum efficiency exceeding 1% can be reached. In contrast, the MoS₂ on planar substrate exhibited quantum yield of less than 0.05%. Well passivated defects in MoS₂ benefits the buildup of population inversion at lower power, therefore effectively lowering the lasing threshold of MoS₂. By pumping the MoS₂ WGM lasers with a 532 nm continuous-wave laser, lasing emission can be observed, which presented a low threshold of 5 W cm⁻² at room temperature. Above the threshold, this WGM laser exhibited a FWHM of ~1.97 nm, corresponding to a quality factor of 350. This result indicates that low-threshold lasing emissions are feasible based on well passivated monolayers. For the practical application of TMD-based nanolasers, this work is illuminating because the CVD grown samples is important to the scalable fabrication and on-chip integration of nanolasers.

3.3 Vertical-cavity surface-emitting lasers

Vertical-cavity surface-emitting lasers (VCSELs) employs distributed Bragg reflectors (DBRs) to achieve a very high reflectivity and strong confinement of photons in a cavity. DBRs are composed of layers with alternating high and low refractive indices and each layer has an optical thickness of $\lambda/4$, where λ is optical wavelength.¹⁰⁰ These alternating layers can cause destructive interference at each layer interface, thus giving rise to a stop band for light transmission. With these high-performance reflectors, long cavity photon lifetime of serval picoseconds and high quality factor exceeding 10⁴ have been realized in VCSELs.^{101, 102} VCSEL geometry has been employed to realize the lasing emission of III-V semiconductors,¹⁰³⁻¹⁰⁵ organic materials,¹⁰⁶ colloidal quantum dots¹⁰⁷ and biological emitters.¹⁰⁸ The optical cavity based on DBRs also provides a platform to study the strong coupling between light and mater.¹⁰⁹⁻¹¹¹ Exciton polaritons (EPs) and EP Bose-Einstein condensation have been demonstrated in DBR cavities based on III-V materials,¹⁰³ organic semiconductors¹¹² and metal-halide perovskites.¹¹³

Figure 4. Vertical-cavity surface-emitting lasers based on WS₂ monolayers. (a) Scheme of WS₂ monolayers sandwiched between top and bottom distributed Bragg reflectors (DBRs). (b) Reflection spectra of WS₂ monolayer embedded in DBRs. (c) PL spectrum of WS₂ in a cavity at the excitation power of 100 nW. (d-f) Integrated intensity, width and position of a WS₂ laser as functions of excitation power. (g) Confocal PL scanning at the position of cavity center (black), surface (red) and 5.1 μ m (blue), respectively. (h) PL intensities of lasing emission (LE) and leaked spontaneous emission (LSE) at different Z positions. Reproduced from ref ⁶⁶. Copyright 2017 Springer Nature.

VCSEL geometry is also important for the realization of low-threshold lasing emission of TMD monolayers due to its sub-wavelength cavity length and ultrahigh quality factor. Additionally, embedding TMD monolayers into DBRs also isolate active materials from ambient environment, which benefits the operating stability of lasers. In 2017, Yu and his colleagues demonstrated, for the first time, the VCSELs based on WS₂ by continuous-wave excitation at room temperature.⁶⁶ The lasers were constructed by embedding WS₂ monolayers into bottom and top DBRs with 12.5 and 8.5 pairs of SiO₂/TiO₂ layers, respectively (Figure 4a). These DBRs creates a stop band for light transmission from 590 nm to 760 nm, which is illustrated in reflection spectrum (Figure 4b). A dip related to a cavity photon mode can be observed at 639.5 nm with a 1.0 ± 0.2 nm, indicating a quality factor of *ca.* 640. Therefore, these DBRs are capable to confine photons for creating a high-quality optical cavity. Based on this device configuration, a sharp PL peak at 636.3 nm can be observed by pumping with a 532-nm continuous-wave laser, suggesting the stimulated lasing emission of WS₂ (Figure 1c). The output intensity as a function of pump power illustrates three obvious regions (Figure 1d). In the first region, PL intensity presents a sublinear relationship with pump power, implying spontaneous exciton emission. The sublinear behavior might be caused by side recombination channels, such as exciton-exciton annihilation under high excitation power. As pump fluence exceeds 4 nW, a superlinear dependence can be observed and a nearly linear dependence can be observed when the pump fluence increases up to 6 nW. This nonlinear response of PL emission strongly evidences the buildup of lasing emission in WS₂.

The lasing emission of WS₂ was also demonstrated by the power-dependent linewidth of PL. With the increase of pump fluence, the linewidth of PL peak decreases from *ca.* 1.2 nm to *ca.* 0.6 nm, while stabilizes at 0.6 nm above the threshold power of 4 nW. The peak position as a function of power illustrates no obvious blueshift with increased power, indicating that this system is a typical photonic laser, instead of exciton-polariton condensation. The lasing behavior of WS₂ VCSELs was also demonstrated by confocal PL measurements along Z-direction. With the focal plane at the cavity centre, both the narrow peak related to lasing

emission and a broad peak corresponding to leaked spontaneous emission can be observed (Figure 1g). The Z-position-dependent PL intensity also indicates that leaked spontaneous emission mainly localized at cavity centre, while the lasing emission is surface emitting. The slower spatial decay of lasing emission indicates its high stability and less diffusion. This work demonstrates low-threshold VCSEL based on TMD monolayers and also creates opportunities to create polariton lasers or electrically pumped lasers based on TMD materials and such a device configuration.

4. Interlayer-exciton lasing based on TMD heterostructures

Ultrafast charge transfer can boost the formation of spatially indirect interlayer excitons with electrons and holes distributed in two different layers in TMD heterostructures. These interlayer excitons possess intriguing optical properties compared to direct excitons in monolayers. Firstly, the interlayer excitons exhibit much longer lifetimes than the intralayer excitons. The longer lifetimes permit the relaxation of excitons into ground states before recombination, thus hopefully leading to the realization of exciton-polariton BEC and lasing emission. Secondly, electrons and holes distributed in different layers result in permanent electric dipole moment of interlayer excitons, which benefits the stronger repulsive nonlinear interactions and electric-field-tunable carrier dynamics. The spatially separated electrons and holes also allows for the electric injection from different sides of heterojunctions, which boosts the development of light-emitting diodes and possible electrically pumped lasers.

4.1 Photonic lasing based on interlayer excitons

For the lasing emission, type-II TMD heterostructures provide a three-level system, which benefits the buildup of population inversion and low-threshold lasing emission. More importantly, such a device configuration is hopeful to realize lasing emission via electric injection, which is crucial for practical applications. Meanwhile, the relatively lower quantum efficiency stemming from the momentum mismatch and reduced exciton oscillation strength also might be the restriction for the realization of

high-performance lasing emissions based on interlayer excitons.

Figure 5. Lasing of interlayer excitons in TMD heterostructures. (a) Scheme of the interlayer-exciton laser constructed by MoS₂/WSe₂ heterostructures onto a photonic crystal cavity. (b) Optical micrograph of MoS₂/WSe₂ heterojunctions onto a photonic crystal. (c) PL spectra of a MoS₂/WSe₂ photonic-crystal laser at room temperature. (d) Output intensity of lasing and spontaneous emission as a function of pump power. (e) Coherent time measurements carried out by a Michelson interferometer present visibility as a function of path decay at pump power of 250 μW. (f) Pump-power-dependent coherence time. Panel (a-f) reproduced from ref ⁶³. Copyright 2019 AAAS. (g) Scheme of a laser based on interlayer excitons constructed by the WSe₂/MoSe₂ heterojunction on a grating cavity. (h) PL spectrum of the

WSe₂/MoSe₂ heterostructure pumped by a 633-nm laser at power of 20 μ W. The shading areas highlights the emission from interlayer excitons, MoSe₂ and WSe₂, respectively. (i) Output intensity and linewidth of lasing emissions as a function of pump power. (j) Scheme of a Michelson interferometer for the measurement of first-order coherence of the interlayer-exciton laser. (k) Representative interference pattern at power of 20 μ W. (l) The coherence length as a function of pump power. The error bars were introduced by the Gaussian fitting. Panel (g-l) reproduced from ref ⁶⁵. Copyright 2019 Springer Nature.

Gao and his colleagues firstly demonstrated the interlayer-exciton lasers based on MoS₂/WSe₂ heterostructures (Figure 5a).⁶³ The MoS₂ and WSe₂ can forms type-II band alignment. Ultrafast charge transfer has been demonstrated at the time scale within 50 fs, yielding electrons in MoS₂ and holes in WSe₂ to form interlayer excitons. Free-standing photonic crystal cavity comprising 220 nm suspended silicon was selected to confine photons with a high-quality factor and MoS₂/WSe₂ heterostructures were fabricated by dry transfer onto photonic crystals (Figure 5b). The intralayer exciton emissions of MoS₂ and WSe₂ are centred at 668 nm and 750 nm, respectively. In contrast, the interlayer excitons exhibit a PL peak at 1128.6 nm, which is out of absorption band of silicon. With the pump power exceeding the threshold, a sharp cavity mode of stimulated emissions of heterostructures can be observed at room temperature (Figure 5c). The FWHM of cavity mode slightly above the lasing threshold is 2.15 nm. The output power as a function of pump power provides a strong evidence of lasing emission (Figure 5d). For cavity mode, an obvious ‘kink’ can be observed, which can be attributed to the nonlinear behavior of lasing emission. In contrast, there is no obvious ‘kink’ in broad spontaneous emissions.

Coherence is another important criterion of lasing emission. The coherent length of MoS₂/WSe₂ interlayer-exciton lasers were measured by a Michelson interferometer. The visibility is defined as,

$$\mathcal{V} = \frac{I_{\max}(\Delta\tau) - I_{\min}(\Delta\tau)}{I_{\max}(\Delta\tau) + I_{\min}(\Delta\tau)} \quad (3)$$

where $I_{\max(\min)}$ is the maximum (minimum) intensity of envelop function at delay time of $\Delta\tau$. The measured visibility as a function of path delay is shown in Figure 5e. The visibility contains two components: a fast decay corresponding to spontaneous emission and a broad component related to lasing emissions with longer coherent time. By this method, the coherent time of lasing emissions can be extracted at different pump power (Figure 5f). The coherent time of lasing emission achieves ~ 1.7 ps when the pump power exceeds ~ 35 μW , which is consistent with the lasing threshold observed in power-dependent output intensity. With these evidences of spectra, power dependence and coherence, the lasing emission of interlayer excitons in TMD heterostructures was unambiguously demonstrated.

Deng and her colleagues demonstrated an interlayer-exciton laser based on a $\text{WSe}_2/\text{MoSe}_2$ heterobilayer and a grating cavity.⁶⁵ The grating cavity was fabricated onto SiN_x by e-beam lithography and dry etching followed by transferring TMD layers (Figure 5g). To enable the bright interlayer excitons, the axes of WSe_2 and MoSe_2 were strictly aligned with rotation angle of below 1° , which prevents the nonradiative loss caused by momentum mismatch. WSe_2 and MoSe_2 exhibit a type-II band alignment. Ultrafast electron transfer occurs from the conduction band of WSe_2 to that of MoSe_2 on the timescale of 10-100 fs, while the holes are left in WSe_2 . A typical PL spectrum of $\text{WSe}_2/\text{MoSe}_2$ heterobilayer (Figure 5h) illustrates that the intensity of low-energy interlayer exciton peak overwhelms the intralayer excitons of MoSe_2 and WSe_2 at higher energy, indicating the efficient charge transfer and buildup of interlayer-exciton population. The lasing emissions of heterobilayers have been demonstrated by calculating the photon occupancy I_p ($k \approx 0$) at different pump power (Figure 5i). When the I_p reaches one, a transition from linear to superlinear increase with the rise of pump power can be observed, indicating the transition from spontaneous to stimulated emissions above the threshold ($P_{\text{th}} = 0.18$ μW). Above the threshold, the linewidth of lasing emission also experiences a sharp decrease from *ca.* 2.4 meV to *ca.* 1.9 meV.

To rigorously identify the lasing behavior, first-order coherence measurements were carried out. The first-order spatial coherence function $g^{(1)}(\mathbf{r}_1, \mathbf{r}_2)$ is defined as,

$$g^{(1)}(\mathbf{r}_1, \mathbf{r}_2) = \frac{G^{(1)}(\mathbf{r}_1, \mathbf{r}_2)}{\sqrt{G^{(1)}(\mathbf{r}_1, \mathbf{r}_2)G^{(1)}(\mathbf{r}_2, \mathbf{r}_1)}} \quad (4)$$

where $G^{(1)}(\mathbf{r}_1, \mathbf{r}_2)$ is the first order correlation function, defined as,

$$G^{(1)}(\mathbf{r}_1, \mathbf{r}_2) = \text{Tr}\{\rho E^{(-)}(\mathbf{r}_1)E^{(+)}(\mathbf{r}_2)\} \quad (5)$$

where ρ is the density matrix operator, $E^{(+)}$ and $E^{(-)}$ are field creation and annihilation operators, respectively. The modulus of coherence degree $|g^{(1)}(\mathbf{r}_1, \mathbf{r}_2)|$ can be directly characterized by the visibility measured in a Michelson interferometer (Figure 5j). A typical interference pattern indicates coherence of emission, thus demonstrating the lasing emission of interlayer excitons in the WSe₂/MoSe₂ heterobilayer. The coherence length as a function of pump power (Figure 5l) indicates that the buildup of high-coherence lasing emission as the pump power exceeds the threshold. Overall, lasing emission of interlayer excitons can be realized through optical pumping with rational band alignment and high-quality optical cavity.

4.2 Bose-Einstein condensation of interlayer excitons

For dissipative out-of-equilibrium Boson systems, exciton BEC often accompanies with lasing emission, owing to the elementary excitation concentrated into a same ground state. Compared to photonic lasing, which requires population inversion at a relatively high pump density, lasing based on BEC, such as exciton-polariton lasing, often exhibits much lower threshold. Although lasing emission has yet been observed in exciton condensates based on TMD heterostructure system, the realization of exciton BEC suggests the possibility of BEC lasing in the exciton-polariton system after introduction of cavity feedback. Therefore, the development of BEC based on interlayer excitons in TMD heterostructures is feasible to attain electrically pumped lasing based on atomically thin materials with much lower lasing threshold than photonic lasers.

Figure 6. Exciton condensation demonstrated in 2D atomic double layers. (a) Schematic illustration of device configuration. This device is constructed by angle-aligned MoSe₂ and WSe₂ separated by two- to three-layer hBN tunneling barrier. The gate electric field is applied at two sides through fabrication of 20-30 nm hBN gate dielectric and few-layer graphene electrode. (b) Reflection contrast $\delta R/R$ at different photon energies as a function of gate voltage under bias voltage of 5.5 V. The upper panel shows band alignment under this condition. (c) Electroluminescence spectra under different exciton concentrations. (d) Second-order correlation function $g^{(2)}(\tau)$ as a function of exciton concentration. The dash lines highlight the $g^{(2)}(\tau)=1$. (e) Electroluminescence, (f) $g^{(2)}(\tau)$ at zero time delay, and (g) the electroluminescence and tunneling density width as a function of exciton concentration. Reproduced from ref ¹¹⁹. Copyright 2019 Springer Nature.

As a typical Boson, excitons are possible to condensate at a ground state. Compared to ultracold dilute atom gas, excitons exhibit a much lower effective mass, which permits the realization of BEC at higher temperature. However, the short lifetime of photoexcited excitons has been a major restriction to realize such macroscopic quantum phase in the exciton system. In the past decades, indirect excitons with spatially separated electrons and holes, such as bilayer quantum well systems¹¹⁴⁻¹¹⁶ and bilayer graphene isolated by hBN¹¹⁷, have been demonstrated with BEC. However, exciton condensation was observed under high magnetic field and

ultralow temperature in these systems. Recently, a breakthrough of exciton condensation at relatively high temperature of 190 K has been realized in 1T-TiSe₂ semimetals.¹¹⁸ Kogar et al. demonstrated a transition from an electronic, plasmon-like excitation to an exciton condensate by observing falling of the electronic-mode energy to zero at a non-zero momentum using momentum-resolved electron-energy-loss spectroscopy (M-EELS).¹¹⁸ These results can unambiguously demonstrate the BEC phase transition and distinguished the exciton condensate phase from the charge density wave phase originating from Peierls distortion. Although the high-temperature exciton condensation has been realized in this work, the semimetal nature of TiSe₂ restricts its practical implementations toward electroluminescence devices.

2D TMD intralayer/interlayer excitons possess exceptionally high binding energy of several hundreds of millielectronvolts, which supports stable exciton resonance under relatively high concentration ($>10^{12} \text{ cm}^{-2}$) at room temperature. In particular, interlayer excitons with much longer lifetimes than those of intralayer excitons are hopeful to achieve BEC at a ground state. In 2019, Shan, Mak and their colleagues reported BEC of interlayer excitons in MoSe₂-WSe₂ atomic double layers separated by few-layer hBN at a relatively high temperature of 100 K.¹¹⁹ The device was constructed by inserting two- to three-layer hBN tunneling barrier between MoSe₂ and WSe₂ (Figure 6a). Gate voltages can be applied to tune the electron/hole concentration in MoSe₂/WSe₂ by introducing 20-30 nm hBN as gate dielectrics and few-layer graphene as contact. The reflection spectra of MoSe₂-WSe₂ double layers were shown in Figure 6b. At a bias of 5.5 V, a p-n region with excess electrons in MoSe₂ and excess holes WSe₂ was observed, which reflected by reflection signals at lower energy. This indicates the electroluminescence of interlayer excitons. Based on this device, a high interlayer exciton concentration up to 10^{12} cm^{-2} can be achieved. The concentration of interlayer excitons is directly related to the tunneling current of the device.

By tuning the gate and bias voltages, the exciton concentration and electroluminescence of MoSe₂-WSe₂ double layers can be controlled. Figure 6c

illustrates electroluminescence spectra at different exciton concentrations. These spectra show a peak with a FWHM of 10-20 meV originating from the radiative recombination of interlayer excitons. The integrated intensity of electroluminescence spectra and tunneling current as a function of interlayer exciton concentration were plotted in Figure 6e. The electroluminescence intensity presents a clear threshold at $\sim 0.26 \times 10^{12} \text{ cm}^{-2}$. Around the threshold, electroluminescence experiences a rapid increase of two orders of magnitude with the two-fold increase of tunneling current. The second-order correlation function $g^{(2)}(\tau)$ was measured using a Hanbury-Brown-Twiss-type setup to study the coherence of electroluminescence as a function of exciton concentration (Figure 6d). Near the threshold, photon bunching with $g^{(2)}(\tau) > 1$ has been observed, which is absent ($g^{(2)}(\tau)=1$) with the exciton concentration above the threshold. Super-Poissonian photon statistics were observed, indicating the formation of exciton condensation. The threshold behavior together with Poissonian statistics indicates the exciton condensation in MoSe₂-WSe₂ double layers.

5 Conclusions and Perspective

In this review, we have summarized the construction of nanolasers based on TMD monolayers and vdW heterostructures incorporated with various optical cavities. Compared to conventional III-V semiconducting quantum wells and dots, TMDs with tightly bound excitons and atomically thin structures are potentially good candidates for the ultracompact nanolasers. Narrowed peaks, clear thresholds and first-order coherence have unambiguously distinguished the lasing emissions from the narrowed emissions caused by other mechanisms.^{33, 34, 63-66} Continuous-wave excitation, room-temperature operation and relatively low threshold have been realized based on TMD monolayers and vdW heterostructures. Integration of TMD monolayers and heterostructures with silicon photonics has created photonic devices, such as modulators³² and photodetectors.¹²⁰ It is expected that ultracompact integrated photonic circuits can be realized after incorporating nanolasers based on TMD materials.

5.1 Scalable fabrication and integration

Despite the rapid progress, it still remains challenging for the practical applications of TMD lasers. At the present, almost all of TMD nanolasers discovered are based on samples exfoliated from bulk crystals. Large-area fabrication of high-quality TMD monolayer and heterostructure thin films onto target substrates is a prerequisite for the on-chip integration of TMD nanolasers. Although the CVD-grown samples on silica fibers have presented the lasing emission,⁶⁷ planar integration of coherent light sources requires growth of TMD materials onto planar substrates, such as silicon, Si_3N_4 and silica plates. For TMD monolayers fabricated by mechanical exfoliation from bulk crystals and chemical vapor deposition (CVD) method, a large defect density gives rise to a relatively low PL quantum yield below 1%.^{121, 122} These defects have caused serious non-radiative recombination, which usually overwhelms the radiative recombination at the room temperature. In particular, TMD monolayers grown by CVD often possess much higher defect density than their mechanically exfoliated counterparts.¹²³ Although mechanically exfoliated monolayers often exhibit higher performance, the CVD method plays a pivotal role in the practical device application of TMD monolayers. Therefore, it is significant to develop CVD process to suppress the defect formation and post-treatment techniques for defect passivation. To further decrease the threshold of lasers, rational passivation of traps plays an important role. Some of surface chemical passivation methods have been developed, but high PL quantum yields are only observed under low exciton population and a significant loss of quantum efficiency occurred at high excitation power, which is not suitable for the high-performance lasing applications.

For the practical application of vdW heterostructures, control of crystallographic alignment is an additional requirement. The efficiency of charge transfer and generation of interlayer excitons related to the momentum need to match with each other between the different layers, which has not been extensively investigated in CVD-grown heterostructures. The properties of interlayer excitons in CVD-grown heterostructures have not been well studied and approaches to improve the PL

quantum efficiency of interlayer excitons in CVD grown structures still elusive.

5.2 Electrically pumped lasers

Electrically pumped lasers are another milestone for the development of integrated lasers based on atomically thin TMD heterostructures. At the present stage, continuous-wave lasing emission and electrically injected light-emitting diodes have been realized based on vdW heterostructures. Development of electrically pumped lasing emission requires further optimization of light-emitting layers, charge carrier transport layers and optical cavities. Firstly, improvement of quantum yield by rational passivation is required to realize high quantum efficiency. Secondly, optimization of transport layers and contact between different layers should be carried out to enable the high-intensity charge injection and buildup of enough exciton population to realize lasing emission. Thirdly, conductive optical cavities can be introduced to achieve charge-carrier transport and optical confinement simultaneously. After the realization of electrically pumped lasers, integration of other photonic components, including waveguides, optical modulators and photodetectors, can accelerate the development of ultracompact integrated photonics based on atomically thin TMD materials.

5.3 Spin-polarized lasers

Introduction of spin-valley polarization into lasing emission offers new opportunities to create functional lasing devices. In III-V semiconducting quantum wells, spin polarized lasers have been achieved by employing anisotropic refractive index, magnetic nanoparticles and ferromagnetic Schottky tunnel contact.^{101, 124, 125} TMD materials with intrinsic spin-valley locking are potential candidates to realize the spin-polarized lasing emission by optical or electrical pumping. Enhanced electron-hole exchange interaction under high population might be the main restriction and the stabilization of spin polarization in TMD materials is still elusive. Fundamental insights into the restriction factors for the valley polarization lifetime are significant for the further development of all valleytronic devices, including the spin

polarized nanolasers.

5.4 Exciton-polariton BEC and lasing emission

Development of exciton-polariton lasers based on TMD materials is another important direction to study the low-threshold lasing emission and many-body physics, such as BEC¹⁰⁰ and superfluidity.¹²⁶⁻¹²⁸ In a semiconducting microcavity, strong coupling of excitons and photons creates a kind of new bosonic quasi-particles termed exciton polariton. With efficient energy relaxation, exciton polaritons are capable to concentrate onto one ground state. The exciton polariton condensates are a typical quasi-equilibrium dissipative system and polaritons present a lifetime of 1-10 ps, which can finally emit coherent photons from the same ground state. Because this process does not require population inversion, polariton lasers often exhibit orders-of-magnitude lower thresholds than photonic lasers. Considerable exciton binding energy of several hundreds of millielectronvolt sustain stable exciton polariton at room temperature, whereas exciton polaritons in GaAs semiconducting quantum wells only stabilize at cryogenic temperature owing to their small exciton binding energy less than 10 meV. Strong coupling and formation of exciton polariton have been demonstrated by incorporating TMD monolayers with DBR microcavities. Based on this device configuration, spin-valley accessibility of exciton polariton has been demonstrated and electrical injection of exciton polaritons has been realized by loading vdW heterostructures in optical microcavities. Observation of polariton condensation and superfluidity of TMD monolayers and vdW heterostructures will be an exciting direction to be explored in the future. Combination of unique physical properties of TMD materials with the microcavity exciton polaritons is an intriguing direction to realize the manipulation of polaritons and polaritonic condensates by employing helical light, external electric and magnetic field.

AUTHOR INFORMATION

Corresponding Authors

Ting Yu — Division of Physics and Applied Physics, School of Physical and

Mathematical Sciences, Nanyang Technological University, Singapore 637371,
Singapore;

Email: yuting@ntu.edu.sg

Authors

Wen Wen — Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

Lishu Wu — Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

Notes

The authors declare no competing financial interests.

ACKNOWLEDGMENT

This work is supported by the Ministry of Education of Singapore (MOE 2019-T2-1-044). This work is supported by the Singapore National Research Foundation (NRF) under the Competitive Research Programs (NRF-CRP-21-2018-0007).

References

1. Mak, K. F.; Shan, J., Photonics and Optoelectronics of 2D Semiconductor Transition Metal Dichalcogenides. *Nat. Photon.* **2016**, *10*, 216-226.
2. Manzeli, S.; Ovchinnikov, D.; Pasquier, D.; Yazyev, O. V.; Kis, A., 2D Transition Metal Dichalcogenides. *Nat. Rev. Mater.* **2017**, *2*, 17033.
3. Xia, F.; Wang, H.; Xiao, D.; Dubey, M.; Ramasubramaniam, A., Two-Dimensional Material Nanophotonics. *Nat. Photon.* **2014**, *8*, 899-907.
4. Wang, G.; Chernikov, A.; Glazov, M. M.; Heinz, T. F.; Marie, X.; Amand, T.; Urbaszek, B., Colloquium: Excitons in Atomically Thin Transition Metal Dichalcogenides. *Rev. Mod. Phys.* **2018**, *90*, 021001.
5. Splendiani, A.; Sun, L.; Zhang, Y.; Li, T.; Kim, J.; Chim, C.-Y.; Galli, G.; Wang, F., Emerging Photoluminescence in Monolayer MoS₂. *Nano Lett.* **2010**, *10*, 1271-1275.
6. Mak, K. F.; Lee, C.; Hone, J.; Shan, J.; Heinz, T. F., Atomically Thin MoS₂: A New Direct-Gap Semiconductor. *Phys. Rev. Lett.* **2010**, *105*, 136805.
7. He, K.; Kumar, N.; Zhao, L.; Wang, Z.; Mak, K. F.; Zhao, H.; Shan, J., Tightly Bound Excitons in Monolayer WSe₂. *Phys. Rev. Lett.* **2014**, *113*, 026803.
8. Chernikov, A.; Berkelbach, T. C.; Hill, H. M.; Rigosi, A.; Li, Y.; Aslan, O. B.; Reichman, D. R.; Hybertsen, M. S.; Heinz, T. F., Exciton Binding Energy and Nonhydrogenic Rydberg Series in Monolayer WS₂. *Phys. Rev. Lett.* **2014**, *113*, 076802.
9. Schuller, J. A.; Karaveli, S.; Schiros, T.; He, K.; Yang, S.; Kyimissis, I.; Shan, J.; Zia, R., Orientation of Luminescent Excitons in Layered Nanomaterials. *Nat. Nanotechnol.* **2013**, *8*, 271-276.
10. Britnell, L.; Ribeiro, R. M.; Eckmann, A.; Jalil, R.; Belle, B. D.; Mishchenko, A.; Kim, Y.-J.; Gorbachev, R. V.; Georgiou, T.; Morozov, S. V.; Grigorenko, A. N.; Geim, A. K.; Casiraghi, C.; Neto, A. H. C.; Novoselov, K. S., Strong Light-Matter Interactions in Heterostructures of Atomically Thin Films. *Science* **2013**, *340*, 1311-1314.
11. Xiao, D.; Liu, G.-B.; Feng, W.; Xu, X.; Yao, W., Coupled Spin and Valley

Physics in Monolayers of MoS₂ and Other Group-VI Dichalcogenides. *Phys. Rev. Lett.* **2012**, *108*, 196802.

12. Cao, T.; Wang, G.; Han, W.; Ye, H.; Zhu, C.; Shi, J.; Niu, Q.; Tan, P.; Wang, E.; Liu, B.; Feng, J., Valley-Selective Circular Dichroism of Monolayer Molybdenum Disulphide. *Nat. Commun.* **2012**, *3*, 887.

13. Mak, K. F.; He, K.; Shan, J.; Heinz, T. F., Control of Valley Polarization in Monolayer MoS₂ by Optical Helicity. *Nat. Nanotechnol.* **2012**, *7*, 494-498.

14. Zeng, H.; Dai, J.; Yao, W.; Xiao, D.; Cui, X., Valley Polarization in MoS₂ Monolayers by Optical Pumping. *Nat. Nanotechnol.* **2012**, *7*, 490-493.

15. Kim, J.; Hong, X.; Jin, C.; Shi, S.-F.; Chang, C.-Y. S.; Chiu, M.-H.; Li, L.-J.; Wang, F., Ultrafast Generation of Pseudo-Magnetic Field for Valley Excitons in WSe₂ Monolayers. *Science* **2014**, *346*, 1205-1208.

16. Jones, A. M.; Yu, H.; Ross, J. S.; Klement, P.; Ghimire, N. J.; Yan, J.; Mandrus, D. G.; Yao, W.; Xu, X., Spin-Layer Locking Effects in Optical Orientation of Exciton Spin in Bilayer WSe₂. *Nat. Phys.* **2014**, *10*, 130-134.

17. Zhang, J.; Du, L.; Feng, S.; Zhang, R.-W.; Cao, B.; Zou, C.; Chen, Y.; Liao, M.; Zhang, B.; Yang, S. A.; Zhang, G.; Yu, T., Enhancing and Controlling Valley Magnetic Response in MoS₂/WS₂ Heterostructures by All-Optical Route. *Nat. Commun.* **2019**, *10*, 4226.

18. Schaibley, J. R.; Yu, H.; Clark, G.; Rivera, P.; Ross, J. S.; Seyler, K. L.; Yao, W.; Xu, X., Valleytronics in 2D Materials. *Nat. Rev. Mater.* **2016**, *1*, 16055.

19. Yang, W.; Shang, J.; Wang, J.; Shen, X.; Cao, B.; Peimyoo, N.; Zou, C.; Chen, Y.; Wang, Y.; Cong, C.; Huang, W.; Yu, T., Electrically Tunable Valley-Light Emitting Diode (vLED) Based on CVD-Grown Monolayer WS₂. *Nano Lett.* **2016**, *16*, 1560-1567.

20. Feng, S.; Cong, C.; Konabe, S.; Zhang, J.; Shang, J.; Chen, Y.; Zou, C.; Cao, B.; Wu, L.; Peimyoo, N.; Zhang, B.; Yu, T., Engineering Valley Polarization of Monolayer WS₂: A Physical Doping Approach. *Small* **2019**, *15*, 1805503.

21. Radisavljevic, B.; Radenovic, A.; Brivio, J.; Giacometti, V.; Kis, A.,

- Single-Layer MoS₂ Transistors. *Nat. Nanotechnol.* **2011**, *6*, 147-150.
22. Cui, X.; Lee, G.-H.; Kim, Y. D.; Arefe, G.; Huang, P. Y.; Lee, C.-H.; Chenet, D. A.; Zhang, X.; Wang, L.; Ye, F.; Pizzocchero, F.; Jessen, B. S.; Watanabe, K.; Taniguchi, T.; Muller, D. A.; Low, T.; Kim, P.; Hone, J., Multi-Terminal Transport Measurements of MoS₂ Using a van der Waals Heterostructure Device Platform. *Nat. Nanotechnol.* **2015**, *10*, 534-540.
23. Kang, K.; Xie, S.; Huang, L.; Han, Y.; Huang, P. Y.; Mak, K. F.; Kim, C.-J.; Muller, D.; Park, J., High-Mobility Three-Atom-Thick Semiconducting Films with Wafer-Scale Homogeneity. *Nature* **2015**, *520*, 656-660.
24. Sarkar, D.; Xie, X.; Liu, W.; Cao, W.; Kang, J.; Gong, Y.; Kraemer, S.; Ajayan, P. M.; Banerjee, K., A Subthermionic Tunnel Field-Effect Transistor with an Atomically Thin Channel. *Nature* **2015**, *526*, 91-95.
25. Fuhrer, M. S.; Hone, J., Measurement of Mobility in Dual-Gated MoS₂ Transistors. *Nat. Nanotechnol.* **2013**, *8*, 146-147.
26. Li, L.; Yu, Y.; Ye, G. J.; Ge, Q.; Ou, X.; Wu, H.; Feng, D.; Chen, X. H.; Zhang, Y., Black Phosphorus Field-Effect Transistors. *Nat. Nanotechnol.* **2014**, *9*, 372-377.
27. Ross, J. S.; Klement, P.; Jones, A. M.; Ghimire, N. J.; Yan, J.; Mandrus, D. G.; Taniguchi, T.; Watanabe, K.; Kitamura, K.; Yao, W.; Cobden, D. H.; Xu, X., Electrically Tunable Excitonic Light-Emitting Diodes Based on Monolayer WSe₂ P-N Junctions. *Nat. Nanotechnol.* **2014**, *9*, 268-272.
28. Baugher, B. W. H.; Churchill, H. O. H.; Yang, Y.; Jarillo-Herrero, P., Optoelectronic Devices based on Electrically Tunable P-N Diodes in a Monolayer Dichalcogenide. *Nat. Nanotechnol.* **2014**, *9*, 262-267.
29. Ye, Y.; Xiao, J.; Wang, H.; Ye, Z.; Zhu, H.; Zhao, M.; Wang, Y.; Zhao, J.; Yin, X.; Zhang, X., Electrical Generation and Control of the Valley Carriers in a Monolayer Transition Metal Dichalcogenide. *Nat. Nanotechnol.* **2016**, *11*, 598-602.
30. Withers, F.; Del Pozo-Zamudio, O.; Mishchenko, A.; Rooney, A. P.; Gholinia, A.; Watanabe, K.; Taniguchi, T.; Haigh, S. J.; Geim, A. K.;

Tartakovskii, A. I.; Novoselov, K. S., Light-emitting diodes by Band-Structure Engineering in van der Waals Heterostructures. *Nat. Mater.* **2015**, *14*, 301-306.

31. Sun, Z.; Martinez, A.; Wang, F., Optical Modulators with 2D Layered Materials. *Nat. Photon.* **2016**, *10*, 227-238.

32. Datta, I.; Chae, S. H.; Bhatt, G. R.; Tadayon, M. A.; Li, B.; Yu, Y.; Park, C.; Park, J.; Cao, L.; Basov, D. N.; Hone, J.; Lipson, M., Low-Loss Composite Photonic Platform Based on 2D Semiconductor Monolayers. *Nat. Photon.* **2020**, *14*, 256-262.

33. Wu, S.; Buckley, S.; Schaibley, J. R.; Feng, L.; Yan, J.; Mandrus, D. G.; Hatami, F.; Yao, W.; Vučković, J.; Majumdar, A.; Xu, X., Monolayer Semiconductor Nanocavity Lasers with Ultralow Thresholds. *Nature* **2015**, *520*, 69-72.

34. Ye, Y.; Wong, Z. J.; Lu, X.; Ni, X.; Zhu, H.; Chen, X.; Wang, Y.; Zhang, X., Monolayer Excitonic Laser. *Nat. Photon.* **2015**, *9*, 733-737.

35. Geim, A. K.; Grigorieva, I. V., Van der Waals Heterostructures. *Nature* **2013**, *499*, 419-425.

36. Novoselov, K. S.; Mishchenko, A.; Carvalho, A.; Castro Neto, A. H., 2D Materials and van der Waals Heterostructures. *Science* **2016**, *353*, aac9439.

37. Hong, X.; Kim, J.; Shi, S.-F.; Zhang, Y.; Jin, C.; Sun, Y.; Tongay, S.; Wu, J.; Zhang, Y.; Wang, F., Ultrafast Charge Transfer in Atomically Thin MoS₂/WS₂ Heterostructures. *Nat. Nanotechnol.* **2014**, *9*, 682-686.

38. Merkl, P.; Mooshammer, F.; Steinleitner, P.; Girnghuber, A.; Lin, K. Q.; Nagler, P.; Holler, J.; Schüller, C.; Lupton, J. M.; Korn, T.; Ovesen, S.; Brem, S.; Malic, E.; Huber, R., Ultrafast Transition between Exciton Phases in van der Waals Heterostructures. *Nat. Mater.* **2019**, *18*, 691-696.

39. Chen, H.; Wen, X.; Zhang, J.; Wu, T.; Gong, Y.; Zhang, X.; Yuan, J.; Yi, C.; Lou, J.; Ajayan, P. M.; Zhuang, W.; Zhang, G.; Zheng, J., Ultrafast Formation of Interlayer Hot Excitons in Atomically Thin MoS₂/WS₂ Heterostructures. *Nat. Commun.* **2016**, *7*, 12512.

40. Liu, Y.; Weiss, N. O.; Duan, X.; Cheng, H.-C.; Huang, Y.; Duan, X., Van

- der Waals Heterostructures and Devices. *Nat. Rev. Mater.* **2016**, *1*, 16042.
41. Tartakovskii, A., Excitons in 2D Heterostructures. *Nat. Rev. Phys.* **2020**, *2*, 8-9.
 42. Rivera, P.; Seyler, K. L.; Yu, H.; Schaibley, J. R.; Yan, J.; Mandrus, D. G.; Yao, W.; Xu, X., Valley-Polarized Exciton Dynamics in a 2D Semiconductor Heterostructure. *Science* **2016**, *351*, 688-691.
 43. Rivera, P.; Yu, H.; Seyler, K. L.; Wilson, N. P.; Yao, W.; Xu, X., Interlayer Valley Excitons in Heterobilayers of Transition Metal Dichalcogenides. *Nat. Nanotechnol.* **2018**, *13*, 1004-1015.
 44. Tong, Q.; Yu, H.; Zhu, Q.; Wang, Y.; Xu, X.; Yao, W., Topological Mosaics in Moiré Superlattices of van der Waals Heterobilayers. *Nat. Phys.* **2017**, *13*, 356-362.
 45. Wu, F.; Lovorn, T.; MacDonald, A. H., Topological Exciton Bands in Moiré Heterojunctions. *Phys. Rev. Lett.* **2017**, *118*, 147401.
 46. Yu, H.; Liu, G.-B.; Tang, J.; Xu, X.; Yao, W., Moiré Excitons: From Programmable Quantum Emitter Arrays to Spin-Orbit-Coupled Artificial Lattices. *Sci. Adv.* **2017**, *3*, e1701696.
 47. Calman, E. V.; Fogler, M. M.; Butov, L. V.; Hu, S.; Mishchenko, A.; Geim, A. K., Indirect Excitons in van der Waals Heterostructures at Room Temperature. *Nat. Commun.* **2018**, *9*, 1895.
 48. Stradi, D.; Papior, N. R.; Hansen, O.; Brandbyge, M., Field Effect in Graphene-Based van der Waals Heterostructures: Stacking Sequence Matters. *Nano Letters* **2017**, *17*, 2660-2666.
 49. Tran, K.; Moody, G.; Wu, F.; Lu, X.; Choi, J.; Kim, K.; Rai, A.; Sanchez, D. A.; Quan, J.; Singh, A.; Embley, J.; Zepeda, A.; Campbell, M.; Autry, T.; Taniguchi, T.; Watanabe, K.; Lu, N.; Banerjee, S. K.; Silverman, K. L.; Kim, S.; Tutuc, E.; Yang, L.; MacDonald, A. H.; Li, X., Evidence for Moiré Excitons in van der Waals Heterostructures. *Nature* **2019**, *567*, 71-75.
 50. Jin, C.; Regan, E. C.; Yan, A.; Iqbal Bakti Utama, M.; Wang, D.; Zhao, S.; Qin, Y.; Yang, S.; Zheng, Z.; Shi, S.; Watanabe, K.; Taniguchi, T.; Tongay, S.; Zettl, A.; Wang, F., Observation of Moiré Excitons in WSe₂/WS₂

Heterostructure Superlattices. *Nature* **2019**, *567*, 76-80.

51. Alexeev, E. M.; Ruiz-Tijerina, D. A.; Danovich, M.; Hamer, M. J.; Terry, D. J.; Nayak, P. K.; Ahn, S.; Pak, S.; Lee, J.; Sohn, J. I.; Molas, M. R.; Koperski, M.; Watanabe, K.; Taniguchi, T.; Novoselov, K. S.; Gorbachev, R. V.; Shin, H. S.; Fal'ko, V. I.; Tartakovskii, A. I., Resonantly Hybridized Excitons in Moiré Superlattices in van der Waals Heterostructures. *Nature* **2019**, *567*, 81-86.

52. Ciarrocchi, A.; Unuchek, D.; Avsar, A.; Watanabe, K.; Taniguchi, T.; Kis, A., Polarization Switching and Electrical Control of Interlayer Excitons in Two-Dimensional van der Waals Heterostructures. *Nat. Photon.* **2019**, *13*, 131-136.

53. Jin, C.; Ma, E. Y.; Karni, O.; Regan, E. C.; Wang, F.; Heinz, T. F., Ultrafast Dynamics in van der Waals Heterostructures. *Nat. Nanotechnol.* **2018**, *13*, 994-1003.

54. Mak, K. F.; Shan, J., Opportunities and Challenges of Interlayer Exciton Control and Manipulation. *Nat. Nanotechnol.* **2018**, *13*, 974-976.

55. Ross, J. S.; Rivera, P.; Schaibley, J.; Lee-Wong, E.; Yu, H.; Taniguchi, T.; Watanabe, K.; Yan, J.; Mandrus, D.; Cobden, D.; Yao, W.; Xu, X., Interlayer Exciton Optoelectronics in a 2D Heterostructure p-n Junction. *Nano Lett.* **2017**, *17*, 638-643.

56. Jauregui, L. A.; Joe, A. Y.; Pistunova, K.; Wild, D. S.; High, A. A.; Zhou, Y.; Scuri, G.; De Greve, K.; Sushko, A.; Yu, C.-H.; Taniguchi, T.; Watanabe, K.; Needleman, D. J.; Lukin, M. D.; Park, H.; Kim, P., Electrical Control of Interlayer Exciton Dynamics in Atomically Thin Heterostructures. *Science* **2019**, *366*, 870-875.

57. Rivera, P.; Schaibley, J. R.; Jones, A. M.; Ross, J. S.; Wu, S.; Aivazian, G.; Klement, P.; Seyler, K.; Clark, G.; Ghimire, N. J.; Yan, J.; Mandrus, D. G.; Yao, W.; Xu, X., Observation of Long-Lived Interlayer Excitons in Monolayer MoSe₂-WSe₂ Heterostructures. *Nat. Commun.* **2015**, *6*, 6242.

58. Yu, Y.; Hu, S.; Su, L.; Huang, L.; Liu, Y.; Jin, Z.; Purezky, A. A.; Geohegan, D. B.; Kim, K. W.; Zhang, Y.; Cao, L., Equally Efficient Interlayer Exciton Relaxation and Improved Absorption in Epitaxial and Nonepitaxial

MoS₂/WS₂ Heterostructures. *Nano Lett.* **2015**, *15*, 486-491.

59. Yu, H.; Wang, Y.; Tong, Q.; Xu, X.; Yao, W., Anomalous Light Cones and Valley Optical Selection Rules of Interlayer Excitons in Twisted Heterobilayers. *Phys. Rev. Lett.* **2015**, *115*, 187002.

60. Kim, J.; Jin, C.; Chen, B.; Cai, H.; Zhao, T.; Lee, P.; Kahn, S.; Watanabe, K.; Taniguchi, T.; Tongay, S.; Crommie, M. F.; Wang, F., Observation of Ultralong Valley Lifetime in WSe₂/MoS₂ Heterostructures. *Sci. Adv.* **2017**, *3*, e1700518.

61. Unuchek, D.; Ciarrocchi, A.; Avsar, A.; Watanabe, K.; Taniguchi, T.; Kis, A., Room-Temperature Electrical Control of Exciton Flux in a van der Waals Heterostructure. *Nature* **2018**, *560*, 340-344.

62. Wang, Z.; Chiu, Y.-H.; Honz, K.; Mak, K. F.; Shan, J., Electrical Tuning of Interlayer Exciton Gases in WSe₂ Bilayers. *Nano Lett.* **2018**, *18*, 137-143.

63. Liu, Y.; Fang, H.; Rasmita, A.; Zhou, Y.; Li, J.; Yu, T.; Xiong, Q.; Zheludev, N.; Liu, J.; Gao, W., Room Temperature Nanocavity Laser with Interlayer Excitons in 2D Heterostructures. *Sci. Adv.* **2019**, *5*, eaav4506.

64. Li, Y.; Zhang, J.; Huang, D.; Sun, H.; Fan, F.; Feng, J.; Wang, Z.; Ning, C. Z., Room-Temperature Continuous-Wave Lasing from Monolayer Molybdenum Ditelluride Integrated with a Silicon Nanobeam Cavity. *Nat. Nanotechnol.* **2017**, *12*, 987-992.

65. Paik, E. Y.; Zhang, L.; Burg, G. W.; Gogna, R.; Tutuc, E.; Deng, H., Interlayer Exciton Laser of Extended Spatial Coherence in Atomically Thin Heterostructures. *Nature* **2019**, *576*, 80-84.

66. Shang, J.; Cong, C.; Wang, Z.; Peimyoo, N.; Wu, L.; Zou, C.; Chen, Y.; Chin, X. Y.; Wang, J.; Soci, C.; Huang, W.; Yu, T., Room-Temperature 2D Semiconductor Activated Vertical-Cavity Surface-Emitting Lasers. *Nat. Commun.* **2017**, *8*, 543.

67. Liao, F.; Yu, J.; Gu, Z.; Yang, Z.; Hasan, T.; Linghu, S.; Peng, J.; Fang, W.; Zhuang, S.; Gu, M.; Gu, F., Enhancing Monolayer Photoluminescence on Optical Micro/Nanofibers for Low-Threshold Lasing. *Sci. Adv.* **2019**, *5*, eaax7398.

68. Zhang, C.; Gong, C.; Nie, Y.; Min, K.-A.; Liang, C.; Oh, Y. J.; Zhang, H.; Wang, W.; Hong, S.; Colombo, L.; Wallace, R. M.; Cho, K., Systematic Study of Electronic Structure and Band Alignment of Monolayer Transition Metal Dichalcogenides in van der Waals Heterostructures. *2D Mater.* **2016**, *4*, 015026.
69. Mak, K. F.; McGill, K. L.; Park, J.; McEuen, P. L., The Valley Hall Effect in MoS₂ Transistors. *Science* **2014**, *344*, 1489-1492.
70. Lee, J.; Mak, K. F.; Shan, J., Electrical Control of the Valley Hall Effect in Bilayer MoS₂ Transistors. *Nat. Nanotechnol.* **2016**, *11*, 421-425.
71. Srivastava, A.; Sidler, M.; Allain, A. V.; Lembke, D. S.; Kis, A.; Imamoglu, A., Valley Zeeman Effect in Elementary Optical Excitations of Monolayer WSe₂. *Nat. Phys.* **2015**, *11*, 141-147.
72. Wang, Z.; Mak, K. F.; Shan, J., Strongly Interaction-Enhanced Valley Magnetic Response in Monolayer WSe₂. *Phys. Rev. Lett.* **2018**, *120*, 066402.
73. Stier, A. V.; McCreary, K. M.; Jonker, B. T.; Kono, J.; Crooker, S. A., Exciton Diamagnetic Shifts and Valley Zeeman Effects in Monolayer WS₂ and MoS₂ to 65 Tesla. *Nat. Commun.* **2016**, *7*, 10643.
74. Xu, X.; Yao, W.; Xiao, D.; Heinz, T. F., Spin and Pseudospins in Layered Transition Metal Dichalcogenides. *Nat. Phys.* **2014**, *10*, 343-350.
75. Gong, C.; Zhang, H.; Wang, W.; Colombo, L.; Wallace, R. M.; Cho, K. J. A. P. L., Band Alignment of Two-Dimensional Transition Metal Dichalcogenides: Application in Tunnel Field Effect Transistors. *Appl. Phys. Lett.* **2013**, *103*, 053513.
76. Hill, M. T.; Gather, M. C., Advances in Small Lasers. *Nat. Photon.* **2014**, *8*, 908.
77. Samuel, I. D. W.; Nanddas, E. B.; Turnbull, G. A., How to Recognize Lasing. *Nat. Photon.* **2009**, *3*, 546-549.
78. Vahala, K. J., Optical Microcavities. *Nature* **2003**, *424*, 839-846.
79. Lodahl, P.; Mahmoodian, S.; Stobbe, S., Interfacing Single Photons and Single Quantum Dots with Photonic Nanostructures. *Rev. Mod. Phys.* **2015**, *87*, 347-400.
80. Ellis, B.; Mayer, M. A.; Shambat, G.; Sarmiento, T.; Harris, J.; Haller, E. E.; Vučković, J., Ultralow-Threshold Electrically Pumped Quantum-Dot Photonic-Crystal Nanocavity Laser. *Nat. Photon.* **2011**, *5*, 297-300.

81. Strauf, S.; Hennessy, K.; Rakher, M. T.; Choi, Y. S.; Badolato, A.; Andreani, L. C.; Hu, E. L.; Petroff, P. M.; Bouwmeester, D., Self-Tuned Quantum Dot Gain in Photonic Crystal Lasers. *Phys. Rev. Lett.* **2006**, *96*, 127404.
82. Altug, H.; Englund, D.; Vučković, J., Ultrafast Photonic Crystal Nanocavity Laser. *Nat. Phys.* **2006**, *2*, 484-488.
83. Tandraechanurat, A.; Ishida, S.; Guimard, D.; Nomura, M.; Iwamoto, S.; Arakawa, Y., Lasing Oscillation in a Three-Dimensional Photonic Crystal Nanocavity with a Complete Bandgap. *Nat. Photon.* **2011**, *5*, 91-94.
84. Aoki, K.; Guimard, D.; Nishioka, M.; Nomura, M.; Iwamoto, S.; Arakawa, Y., Coupling of Quantum-Dot Light Emission with a Three-Dimensional Photonic-Crystal Nanocavity. *Nat. Photon.* **2008**, *2*, 688-692.
85. Nomura, M.; Kumagai, N.; Iwamoto, S.; Ota, Y.; Arakawa, Y., Laser Oscillation in a Strongly Coupled Single-Quantum-Dot–Nanocavity System. *Nat. Phys.* **2010**, *6*, 279-283.
86. Kuramochi, E.; Nozaki, K.; Shinya, A.; Takeda, K.; Sato, T.; Matsuo, S.; Taniyama, H.; Sumikura, H.; Notomi, M., Large-Scale Integration of Wavelength-Addressable All-Optical Memories on a Photonic Crystal Chip. *Nat. Photon.* **2014**, *8*, 474-481.
87. Molesky, S.; Lin, Z.; Piggott, A. Y.; Jin, W.; Vucković, J.; Rodriguez, A. W., Inverse Design in Nanophotonics. *Nat. Photon.* **2018**, *12*, 659-670.
88. Tamboli, A. C.; Haberer, E. D.; Sharma, R.; Lee, K. H.; Nakamura, S.; Hu, E. L., Room-Temperature Continuous-Wave Lasing in GaN/InGaN Microdisks. *Nat. Photon.* **2007**, *1*, 61-64.
89. He, L.; Özdemir, Ş. K.; Zhu, J.; Kim, W.; Yang, L., Detecting Single Viruses and Nanoparticles Using Whispering Gallery Microlasers. *Nat. Nanotechnol.* **2011**, *6*, 428-432.
90. Zhu, J.; Ozdemir, S. K.; Xiao, Y.-F.; Li, L.; He, L.; Chen, D.-R.; Yang, L., On-Chip Single Nanoparticle Detection and Sizing by Mode Splitting in an Ultrahigh-Q Microresonator. *Nat. Photon.* **2010**, *4*, 46-49.
91. Rueda, A.; Sedlmeir, F.; Kumari, M.; Leuchs, G.; Schwefel, H. G. L.,

- Resonant Electro-Optic Frequency Comb. *Nature* **2019**, *568*, 378-381.
92. Zhang, M.; Buscaino, B.; Wang, C.; Shams-Ansari, A.; Reimer, C.; Zhu, R.; Kahn, J. M.; Lončar, M., Broadband Electro-Optic Frequency Comb Generation in a Lithium Niobate Microring Resonator. *Nature* **2019**, *568*, 373-377.
 93. Pöllinger, M.; O'Shea, D.; Warken, F.; Rauschenbeutel, A., Ultrahigh-Q Tunable Whispering-Gallery-Mode Microresonator. *Phys. Rev. Lett.* **2009**, *103*, 053901.
 94. Armani, D. K.; Kippenberg, T. J.; Spillane, S. M.; Vahala, K. J., Ultra-High-Q Toroid Microcavity on a Chip. *Nature* **2003**, *421*, 925-928.
 95. Salehzadeh, O.; Djavid, M.; Tran, N. H.; Shih, I.; Mi, Z., Optically Pumped Two-Dimensional MoS₂ Lasers Operating at Room-Temperature. *Nano Lett.* **2015**, *15*, 5302-5306.
 96. Amani, M.; Lien, D.-H.; Kiriya, D.; Xiao, J.; Azcatl, A.; Noh, J.; Madhupathy, S. R.; Addou, R.; KC, S.; Dubey, M.; Cho, K.; Wallace, R. M.; Lee, S.-C.; He, J.-H.; Ager, J. W.; Zhang, X.; Yablonovitch, E.; Javey, A., Near-Unity Photoluminescence Quantum Yield in MoS₂. *Science* **2015**, *350*, 1065-1068.
 97. Kim, H.; Lien, D.-H.; Amani, M.; Ager, J. W.; Javey, A., Highly Stable Near-Unity Photoluminescence Yield in Monolayer MoS₂ by Fluoropolymer Encapsulation and Superacid Treatment. *ACS Nano* **2017**, *11*, 5179-5185.
 98. Kim, H.; Ahn, G. H.; Cho, J.; Amani, M.; Mastandrea, J. P.; Groschner, C. K.; Lien, D.-H.; Zhao, Y.; Ager, J. W.; Scott, M. C.; Chrzan, D. C.; Javey, A., Synthetic WSe₂ Monolayers with High Photoluminescence Quantum Yield. *Sci. Adv.* **2019**, *5*, eaau4728.
 99. Lien, D.-H.; Uddin, S. Z.; Yeh, M.; Amani, M.; Kim, H.; Ager, J. W.; Yablonovitch, E.; Javey, A., Electrical Suppression of all Nonradiative Recombination Pathways in Monolayer Semiconductors. *Science* **2019**, *364*, 468-471.
 100. Deng, H.; Haug, H.; Yamamoto, Y., Exciton-Polariton Bose-Einstein Condensation. *Rev. Mod. Phys.* **2010**, *82*, 1489-1537.
 101. Holub, M.; Shin, J.; Saha, D.; Bhattacharya, P., Electrical Spin Injection

- and Threshold Reduction in a Semiconductor Laser. *Phys. Rev. Lett.* **2007**, *98*, 146603.
102. Fink, T.; Schade, A.; Höfling, S.; Schneider, C.; Imamoglu, A., Signatures of a Dissipative Phase Transition in Photon Correlation Measurements. *Nat. Phys.* **2018**, *14*, 365-369.
103. Schneider, C.; Rahimi-Iman, A.; Kim, N. Y.; Fischer, J.; Savenko, I. G.; Amthor, M.; Lerner, M.; Wolf, A.; Worschech, L.; Kulakovskii, V. D.; Shelykh, I. A.; Kamp, M.; Reitzenstein, S.; Forchel, A.; Yamamoto, Y.; Höfling, S., An Electrically Pumped Polariton Laser. *Nature* **2013**, *497*, 348-352.
104. Slusher, R. E., Laser Technology. *Rev. Mod. Phys.* **1999**, *71*, S471-S479.
105. Mei, Y.; Weng, G.-E.; Zhang, B.-P.; Liu, J.-P.; Hofmann, W.; Ying, L.-Y.; Zhang, J.-Y.; Li, Z.-C.; Yang, H.; Kuo, H.-C., Quantum Dot Vertical-Cavity Surface-Emitting Lasers Covering the ‘Green Gap’. *Light Sci. Appl.* **2017**, *6*, e16199.
106. Koschorreck, M.; Gehlhaar, R.; Lyssenko, V. G.; Swoboda, M.; Hoffmann, M.; Leo, K., Dynamics of a High-Q Vertical-Cavity Organic Laser. *Appl. Phys. Lett.* **2005**, *87*, 181108.
107. Dang, C.; Lee, J.; Breen, C.; Steckel, J. S.; Coe-Sullivan, S.; Nurmikko, A., Red, Green and Blue Lasing Enabled by Single-Exciton Gain in Colloidal Quantum Dot Films. *Nat. Nanotechnol.* **2012**, *7*, 335-339.
108. Gather, M. C.; Yun, S. H., Bio-optimized Energy Transfer in Densely Packed Fluorescent Protein Enables Near-Maximal Luminescence and Solid-State Lasers. *Nat. Commun.* **2014**, *5*, 5722.
109. Kasprzak, J.; Richard, M.; Kundermann, S.; Baas, A.; Jeambrun, P.; Keeling, J. M. J.; Marchetti, F. M.; Szymańska, M. H.; André, R.; Staehli, J. L.; Savona, V.; Littlewood, P. B.; Deveaud, B.; Dang, L. S., Bose–Einstein Condensation of Exciton Polaritons. *Nature* **2006**, *443*, 409-414.
110. Sanvitto, D.; Kéna-Cohen, S., The Road towards Polaritonic Devices. *Nat. Mater.* **2016**, *15*, 1061-1073.
111. Liu, X.; Galfsky, T.; Sun, Z.; Xia, F.; Lin, E.-c.; Lee, Y.-H.;

- Kéna-Cohen, S.; Menon, V. M., Strong Light–Matter Coupling in Two-Dimensional Atomic Crystals. *Nat. Photon.* **2015**, *9*, 30-34.
- 112.Daskalakis, K. S.; Maier, S. A.; Murray, R.; Kéna-Cohen, S., Nonlinear Interactions in an Organic Polariton Condensate. *Nat. Mater.* **2014**, *13*, 271-278.
- 113.Su, R.; Diederichs, C.; Wang, J.; Liew, T. C. H.; Zhao, J.; Liu, S.; Xu, W.; Chen, Z.; Xiong, Q., Room-Temperature Polariton Lasing in All-Inorganic Perovskite Nanoplatelets. *Nano Lett.* **2017**, *17*, 3982-3988.
- 114.Eisenstein, J.; MacDonald, A. H., Bose–Einstein Condensation of Excitons in Bilayer Electron Systems. *Nature* **2004**, *432*, 691-694.
- 115.Nandi, D.; Finck, A.; Eisenstein, J.; Pfeiffer, L.; West, K., Exciton Condensation and Perfect Coulomb Drag. *Nature* **2012**, *488*, 481-484.
- 116.Eisenstein, J., Exciton Condensation in Bilayer Quantum Hall Systems. *Annu. Rev. Condens. Matter Phys.* **2014**, *5*, 159-181.
- 117.Liu, X.; Watanabe, K.; Taniguchi, T.; Halperin, B. I.; Kim, P., Quantum Hall Drag of Exciton Condensate in Graphene. *Nat. Phys.* **2017**, *13*, 746-750.
- 118.Kogar, A.; Rak, M. S.; Vig, S.; Husain, A. A.; Flicker, F.; Joe, Y. I.; Venema, L.; MacDougall, G. J.; Chiang, T. C.; Fradkin, E., Signatures of Exciton Condensation in a Transition Metal Dichalcogenide. *Science* **2017**, *358*, 1314-1317.
- 119.Wang, Z.; Rhodes, D. A.; Watanabe, K.; Taniguchi, T.; Hone, J. C.; Shan, J.; Mak, K. F., Evidence of High-Temperature Exciton Condensation in Two-Dimensional Atomic Double Layers. *Nature* **2019**, *574*, 76-80.
120. Maiti, R.; Patil, C.; Saadi, M. A. S. R.; Xie, T.; Azadani, J. G.; Uluutku, B.; Amin, R.; Briggs, A. F.; Miscuglio, M.; Van Thourhout, D.; Solares, S. D.; Low, T.; Agarwal, R.; Bank, S. R.; Sorger, V. J., Strain-Engineered High-Responsivity MoTe₂ Photodetector for Silicon Photonic Integrated Circuits. *Nat. Photon.* **2020**, doi: 10.1038/s41566-020-0647-4.
121. Wang, H.; Zhang, C.; Rana, F., Ultrafast Dynamics of Defect-Assisted Electron–Hole Recombination in Monolayer MoS₂. *Nano Lett.* **2015**, *15*, 339-345.
122. Yuan, L.; Huang, L., Exciton Dynamics and Annihilation in WS₂ 2D Semiconductors. *Nanoscale* **2015**, *7*, 7402-7408.

123. Cunningham, P. D.; McCreary, K. M.; Hanbicki, A. T.; Currie, M.; Jonker, B. T.; Hayden, L. M., Charge Trapping and Exciton Dynamics in Large-Area CVD Grown MoS₂. *J. Phys. Chem. C* **2016**, *120*, 5819-5826.
124. Chen, J.-Y.; Wong, T.-M.; Chang, C.-W.; Dong, C.-Y.; Chen, Y.-F., Self-Polarized Spin-Nanolasers. *Nat. Nanotechnol.* **2014**, *9*, 845-850.
125. Lindemann, M.; Xu, G.; Pusch, T.; Michalzik, R.; Hofmann, M. R.; Žutić, I.; Gerhardt, N. C., Ultrafast Spin-Lasers. *Nature* **2019**, *568*, 212-215.
126. Amo, A.; Lefrère, J.; Pigeon, S.; Adrados, C.; Ciuti, C.; Carusotto, I.; Houdré, R.; Giacobino, E.; Bramati, A., Superfluidity of Polaritons in Semiconductor Microcavities. *Nat. Phys.* **2009**, *5*, 805-810.
127. Sanvitto, D.; Marchetti, F. M.; Szymańska, M. H.; Tosi, G.; Baudisch, M.; Laussy, F. P.; Krizhanovskii, D. N.; Skolnick, M. S.; Marrucci, L.; Lemaître, A.; Bloch, J.; Tejedor, C.; Viña, L., Persistent Currents and Quantized Vortices in a Polariton Superfluid. *Nat. Phys.* **2010**, *6*, 527-533.
128. Amo, A.; Pigeon, S.; Sanvitto, D.; Sala, V. G.; Hivet, R.; Carusotto, I.; Pisanello, F.; Leménager, G.; Houdré, R.; Giacobino, E.; Ciuti, C.; Bramati, A., Polariton Superfluids Reveal Quantum Hydrodynamic Solitons. *Science* **2011**, *332*, 1167-1170.

TOC graphic