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Integration of two-dimensional transition metal dichalcogenides with Mie-resonant dielectric nanostructures

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ABSTRACT
Integrating transition metal dichalcogenide (TMD) monolayers with dielectric nanostructures exhibiting Mie-like resonances opens a plethora of opportunities in manipulating their excitonic emission in the near-field and far-field regimes, yielding interactions spanning from weak to strong coupling regimes, and routing valley polarized chiral emission, to name just a few. Moreover, there is a completely new path unraveled recently by realizing dielectric resonators directly from TMDs, thus combining scatterer and emitter into a single entity. This hybrid configuration is promising to realize integrated active meta-optical devices in a facile manner. In this review article, we provide an overview of the state of the art on integrating monolayers of TMDs with Mie-resonant photonic nanostructures.

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1. Introduction

Active meta-optics [1] is gaining new direction and momentum with the emergence of a novel class of nanophotonic systems consisting of Mie-resonant dielectric nanostructures integrated with single or few layers of transition metal dichalcogenides (2D-TMDs). For decades, TMDs have been in use as lubricant material in technical applications due to their low friction, originating from the weak Van der Waals forces between their individual layers. Something very remarkable happens when these bulk crystals are thinned down to a few or single layers. Contrary to their bulk form, TMDs display highly fascinating properties in their monolayer form when their bandgap transitions from indirect to direct [2]. These materials, which have only mono-molecular thickness, display several intriguing optical, mechanical, and electronic properties, thus offering a huge potential for future applications. Some authors even see them as contenders to replace existing silicon-based nano-electronics in the future [3].

Nanostructures make it possible to realize resonant interactions with light at the nanoscale. In particular, optical metasurfaces composed of designed nanoscale resonators arranged in a periodic, aperiodic, or disordered fashion in a plane, offer many opportunities for manipulating light fields and engineering light–matter interactions. Resonant plasmonic nanostructures composed of metals such as gold, silver, and aluminum have been studied extensively to tailor and enhance interaction with light in a broad spectral range from mid-IR to UV wavelengths. In particular, plasmonic nanostructures have been widely employed to achieve a range of functionalities [4], including but not limited to wavefront shaping [5], hot-electron harvesting [6], energy harvesting [7], and drug-delivery [8]. However, the drawback of plasmonic nanostructures is their strong absorption in the optical wavelength range, which is detrimental for many of their potential applications. Dielectric nanostructures, in contrast, realized from high-index materials such as silicon, germanium, gallium arsenide, gallium phosphite, or titanium dioxide, display very low absorption losses compared to their plasmonic counterparts, thus making them ideal for high-efficiency applications down to visible wavelengths [9,10]. Furthermore, dielectric nanostructures can exhibit both electric and magnetic multipolar modes, which further diversifies the engineering options allowing to tailor their response towards specific effects or applications. As such, Mie-resonant dielectric nanostructures have gained immense popularity in recent years, and many potential applications including enhancing light–matter interactions [11], energy harvesting [12], and efficient wavefront shaping devices [13] were suggested. Integrating 2D-TMDs with Mie-resonant dielectric nanostructures
yields hybrid entities which may find applications as novel active nanophotonic platforms and provides interesting routes to harness the unique properties of these molecularly thin materials at the nanoscale.

This review starts with a brief introduction to 2D-TMDs and Mie-resonant dielectric nanostructures in the first section, in particular focusing on those properties which can be mutually useful for hybrid structures. In the second section, we briefly discuss different methods to integrate TMD monolayers with dielectric nanostructures. In the third section, we present a review of selected recent works performed on hybrid nanosystems consisting of 2D-TMDs integrated with Mie-resonant dielectric nanostructures. Specifically, we will discuss concepts like far-field manipulation, weak and strong coupling, and chiral emission in such hybrid architectures. Finally, we will present an outlook of this novel research line, and discuss potential challenges and possible applications.

2. Transition metal dichalcogenides (TMDs)

TMD crystals are stacked arrangements of two-dimensional layers weakly adhered by van der Waals interactions. Each individual crystal layer consists of three atomic layers, X-M-X, which in their semiconducting phase are covalently bound in a trigonal prismatic ($D_{3h}$) configuration, generally represented as MX$_2$. Here M denotes a transition metal atom and X denotes a chalcogen atom. A schematic of such a stacked crystal structure for the example of MoS$_2$ is shown in Figure 1(a). Similar to graphene, the layered structure allows us to obtain even individual layers by direct mechanical exfoliation from a bulk crystal as shown in Figure 1(b). Recently, significant efforts went into the optimization of synthesis methods, including chemical vapor deposition (CVD) [14] and molecular beam epitaxy (MBE) [15] to achieve isolated single-crystal monolayers with high yield, high quality, and large size distribution [16]. For instance, single-crystal MoS$_2$ monolayers grown by CVD are shown in Figure 1(c), typically forming a triangular shape dictated by the crystal symmetry.

While bulk TMDs as shown in Figure 1(b) are known for decades, remarkable optoelectronic properties were revealed in recent years for 2D-TMDs (see Figure 1(c)) consisting of only a single or few molecular layers, which are not only strongly deviating from their bulk counterparts but also changing significantly depending on the number of layers. Most prominently, group-VI TMDs ($M = Mo, W$ and $X = S, Se, Te$) in the semiconducting 2H stacking order exhibit an indirect-to-direct bandgap transition at the $K$-point when thinned down to a single layer crystal as shown in Figure 1(d). As a direct consequence, drastically enhanced photoluminescence (PL) can be observed for single-layer TMDs [17]. With bandgap energies lying in the visible and near-infrared ranges TMDs are
a promising 2D material platform for optoelectronic applications. In this section, we give a brief overview of the optical properties of 2D-TMDs from the viewpoint of their importance for the integration with resonant dielectric nanostructures.

Naturally, 2D-TMDs display strong in-plane confinement of charge carriers accompanied with a reduced dielectric screening. As a result, long-range Coulomb interactions give rise to a variety of many-body phenomena.

Figure 1. (a) Illustration of a MoS$_2$ crystal composed of a stack of monolayers. (b) Photograph of a bulk crystal of MoS$_2$. (c) CVD-grown MoS$_2$ monolayers on SiO$_2$/Si substrate. (d) Band structure of 1L-MoS$_2$ calculated with density functional theory. The black arrow represents the direct band gap of 1.9 eV at the K-point of the Brillouin zone. (e) Normalized PL spectra (red, green, blue, and purple colored curves) and differential reflectance spectra (gray curves) of monolayer MoS$_2$, MoSe$_2$, WS$_2$, and WSe$_2$ on quartz substrates, for excitation at the C A' for WSe$_2$) resonance. The scale bar indicates a 20% absorption corresponding to differential reflectance spectra. The PL spectra are normalized to the A-exciton differential reflectance from the respective monolayers. (f) Schematic diagram of the band structure at K and K' points in the hexagonal Brillouin zone, giving rise to valley contrasting optical selection rules: Right – σ$^-$ (left – σ$^+$) handed polarized light couples only to K(K') valleys. g) Raman spectra of MoS$_2$ (bulk, 1L to 6L), displaying two peaks corresponding to the first order in-plane ($E_{2g}$) and out-of-plane ($A_{1g}$) phonon modes. The corresponding lattice vibrations of the atomic trilayer (S-Mo-S) are illustrated adjacent to the peaks ($E_{2g}$ and $A_{1g}$). a), Reprinted by permission from [Springer Nature]: [Nature] [Nature Nanotechnology] [30] (Single-layer MoS$_2$ transistors, Radisavljevic et al.), [COPYRIGHT] (2011). (b), Reprinted by permission from [Springer Nature]: [Nature] [Nature Nanotechnology] [31] (Electronics and optoelectronics of two-dimensional transition metal dichalcogenides, Wang et al.), [COPYRIGHT] (2012). (c), Reproduced from [16], https://creativecommons.org/licenses/by/3.0/CC BY 3.0. (d), Reprinted with permission from [17], Copyright (2010) American Chemical Society. e), Reprinted by permission from [Springer Nature]: [Nature] [Nature Communications] [27] (Photocarrier relaxation pathway in two-dimensional semiconducting transition metal dichalcogenides, Daichi Kozawa et al.), [COPYRIGHT] (2014). (f), Reprinted by permission from [Springer Nature]: [Nature] [Nature Nanotechnology] [46] (Valley polarization in MoS$_2$ monolayers by optical pumping, Zeng et al.), [COPYRIGHT] (2012). (g), Reprinted with permission from [53], Copyright (2010) American Chemical Society.
such as the formation of tightly bound electron-hole configurations upon photo-excitation, so-called excitons. In 2D-TMDs, exciton binding energies in the range from 0.2 eV (for MoS$_2$ [18]) to 0.55 eV (for MoSe$_2$ [19]) can be observed exceeding typical values for semiconductor quantum wells by one order of magnitude [20,21]. Such high values of the exciton binding energy are a direct consequence of the mentioned strong in-plane confinement and reduced dielectric screening and are further supported by the large effective masses of electrons and holes at the $K$-points [22,23]. In contrast to conventional semiconductors, excitonic effects hence dominate the optical properties of 2D-TMDs even at room temperature, which may allow for the realization of excitonic devices working at ambient conditions. In Figure 1(e) experimental PL and differential reflection spectra at room temperature for single-layer MoS$_2$, MoSe$_2$, WS$_2$, and WSe$_2$ are shown. The two resonances labelled A and B occurring on the low-energy side of the differential reflection spectra (gray curves) can be attributed to exciton formation at the $K$-point. These two resonances are observed due to strong spin–orbit interaction in 2D-TMDs, resulting in a respective spin-splitting of their valence and conduction bands. In particular, a large splitting can be observed for the valence band mediated by the participation of the transition metal $d$-orbital electrons while the splitting of the conduction band is smaller by one order of magnitude [24]. Hence, the different exciton binding energies leading to the distinct A and B resonances in the differential reflection spectra originate from the contribution of holes in the upper and lower branch of the spin-split valence band, respectively. Generally, the spin-orbit coupling is larger for materials composed of atoms of higher atomic number. Consequently, the valence band of Mo-based compounds splits up to 0.20 eV, that of W-based compounds even up to 0.4 eV [25,26], as can be seen from the separation of the A and B excitons in the respective differential reflection spectra. Resonances on the high-energy side of the differential reflection spectra result from band nesting effects (peaks labelled by C) or the combined effects of band nesting and complex atomic orbital contributions (peaks labelled by A’ and B’ for WSe$_2$) [27]. Despite being atomically thin, 2D-TMDs display comparably strong light-matter coupling reaching absorption values for single-layer crystals up to 15% at their A-excitonic resonance [28], a crucial premise for high-brightness and high-efficiency nanoscale integrated optoelectronic devices. The radiative relaxation of excitons leads to PL (compare colored curves in Figure 1(e)) and respective PL spectra are typically dominated by the A-exciton contribution due to ultrafast relaxation of the charge carriers towards the fundamental band edges at the $K$-points [29].

As seen above, excitonic resonances are strongly affecting the optical properties of 2D-TMDs. Hence, from the optoelectronic integration perspective, knowledge of the excitation and relaxation dynamics of excitons is
of fundamental importance. Generally, excitons can be grouped in bright and dark states, depending on whether their decay can be detected in optical experiments. Bright excitons typically decay radiatively on an ultra-fast time scale [32,33]. However, lifetimes of 2D-TMD excitons on the order of tens of picoseconds to nanoseconds are also practically observed, as the interplay of radiative and nonradiative decay channels is strongly influenced by the presence of disorder (e.g. defects, impurities) [34], dielectric environment [35], temperature [36], excitation power density (e.g. Auger-type exciton-exciton annihilation) [37,38], and defect mediated non-radiative recombination [39]. Besides bright excitons, 2D-TMDs support dark excitons, which can be distinguished into spin-forbidden excitons and momentum-forbidden excitons. The spin-forbidden excitons are intra-valley Coulomb bound electrons in the conduction band and holes in the valence band of opposite spins. Momentum-forbidden dark excitons are Coulomb bound electrons in the conduction band and holes in the valence bands of different valleys. Spin-forbidden dark excitons are especially interesting in the context of coupling to photonic nanostructures, as even though their transition is in-plane dipole-forbidden and therefore not usually observed in standard PL measurements, they can radiatively couple to the far-field by an out-of-plane dipole moment with respect to the monolayer plane. Thus, in order to observe them in optical experiments, usually special techniques such as excitation of the 2D-TMD either parallel to its surface or perpendicular to its surface with high-NA objectives [40] have to be employed. An alternative, simple approach for brightening dark excitons is offered by coupling 2D-TMDs to the tailored electromagnetic nearfields of resonant nanostructures or surface plasmon polaritons [41] while the efficiency of such hybrid devices might further be increased by the use of all-dielectric nanostructures in the future. The spectral overlap of dark and bright excitons in 2D-TMDs will crucially influence their overall PL yield. Hence, the brightening of dark excitons using resonant nanostructures provides a potential approach for further improvement of 2D-TMD integrated devices.

The unconventionally high exciton binding energies in 2D-TMDs allow for various higher-order many-body phenomena, too. Readily observed even at room temperature is the formation of positively (negatively) charged excitons, so-called trions, which are a bound configuration between an exciton with an additional bound hole (electron). Experimentally observed trion binding energies for 2D-TMDs can reach tens of meV [42]. Interestingly, the high stability of charged excitons opens up a huge potential for tunable devices based on 2D-TMDs controlled by electric gating or chemical doping [42–45]. Further members of the exciton family, like bi-excitons and inter-layer excitons in few-layer or heterostructure TMDs, lay out the path for studying a variety of fundamental exciton physics within 2D-TMDs.
For 1L-TMDs (and more generally for TMD crystals with an odd number of layers) the inversion symmetry is naturally broken in their semiconducting trigonal prismatic (2H) phase. Due to time-reversal symmetry further dictating the spin states the combined effect leads to a spin-momentum locking at the K-points, distinguishing between two alternating sets of energetically degenerate but inequivalent states at the $K$ and $K'$ points [46] typically referred to as K/K’ valleys. As K/K’ valleys are well separated in momentum space, this additional valley degree of freedom can be thought of as a pseudo-spin. Interestingly, optical transitions in these valleys occur with opposite helicity opening a route for optically addressing the valley degree of freedom [47]. Light of certain helicity will selectively generate an exciton population in either of the two valley sets. Respectively, the exciton population can only decay radiatively within the same valley by emitting photons of the same helicity hence storing and optically encoding the valley state. Similar to the concept of spintronics the valley degree of freedom can potentially be used for information encoding, storage and readout, opening the field of valleytronics [47,48].

Another important consequence of the broken inversion symmetry in 1L-TMDs is their record high second-order nonlinear susceptibility [49,50]. It allows, e.g. for a strong efficient second harmonic generation (SHG) despite their small thickness, exceeding that of standard transparent nonlinear crystals by about an order of magnitude, if they could be thinned down to the same thickness. As such, 1L-TMDs offer important opportunities for integrated nonlinear-optical devices.

Optical, electronic and vibrational properties of 2D-TMDs are substantially affected by their underlying substrates in terms of surface roughness, chemical composition, and crystallinity [51]. In particular, defects and dangling bonds make the substrate material chemically active toward the supported 2D-TMD crystal potentially resulting in significant doping. Hereby, carriers injected into the 2D-TMD will affect the optical properties by altering the relative population of neutral and charged exciton contributions. For the case of CVD-grown crystals, the choice of the substrate also affects the strain experienced by the 2D-TMD as the strain is built up predominantly in the cooling step of the growth process due to the mismatch of the thermal expansion coefficients of the TMD crystal and the respective growth substrate [52]. Based on the sensitivity of the vibrational properties to these factors, Raman response became a standard method for the determination of crystal quality, chemical and mechanical substrate influences. Typical Raman spectra of exfoliated MoS$_2$ (bulk, 1L to 6L) are shown in Figure 1.(g). The two peaks can be associated with the in-plane ($E_{2g}^1$) and out-of-plane ($A_{1g}$) phonon modes (compare inset in Figure 1.(g)). The separation between the peaks scales with the number of layers of the TMD crystal and hence can be used as a measure for the crystal thickness [53].
The large exciton binding energies, bright & dark excitons, valley degree of freedom, non-blinking character [54], ultrafast lifetimes, tunability of emission with external stimuli (electric, optical, dielectric environment), chiral emission, to name a few, sum up 2D TMDs as light-emitting sources with rich excitonic physics, which can be harnessed for a wide range of applications in quantum photonics, photodetectors [55], excitonic LEDs [56,57], excitonic lasers [58,59], optical modulators, valleytronics, and bio-labelling.

3. **Dielectric nanoresonators**

Dielectric nanoresonators (DNs) made of high refractive index materials such as silicon, germanium, gallium arsenide, or titanium dioxide have received immense research attention during the last several years. Their low absorption makes them ideal candidates to manipulate light at the nanoscale with high efficiency. The current state-of-the-art fabrication technologies make it possible to realize dielectric resonators of diverse shapes, sizes and arrangements. Owing to strong confinement of light inside the high-index dielectric medium, DNs can display electric and magnetic multipolar resonances (see Figure 2(a)), which can be controlled by the DN geometry. To illustrate this, a dark-field true-color image of silicon nanospheres of various dimensions is shown.

![Figure 2](image_url)

**Figure 2.** (a) Schematic of the electric field lines associated to transverse components of the electric (coefficient $a_1$) and magnetic (coefficient $b_1$) dipolar Mie-modes of a spherical particle represented on an imaginary sphere enclosing the particle. (b) Dark-field microscope true-color image of silicon nanospheres of different sizes. (c) Experimentally measured reflectance spectrum of an individual silicon nanosphere, where the peaks can be identified as a magnetic dipole (md), electric dipole (ed), and magnetic quadrupole (mq) resonance. (d) Mode profiles of the electric and magnetic dipolar modes for a silicon nanocylinder. $|E|$ and $|H|^2$ are the absolute value of the electric field and the square absolute value of the magnetic field, respectively. a), Reproduced from [67], Wiley. (b,c), Reprinted by permission from [Springer Nature]: [Nature Scientific Reports] [68], (Magnetic light, Kuznetsov et al.), [COPYRIGHT] (2012). (d), Reproduced from [69], Wiley.
in Figure 2(b), and an experimentally measured reflectance spectra of one of the nanosphere is shown in Figure 2(c). The observed peaks can be identified to originate from excitation of the electric dipole (ed), magnetic dipole (md), and magnetic quadrupole (mq) modes. The mode profiles of the electric dipolar mode and the magnetic dipolar mode of a nanocylinder are illustrated in Figure 2(d). Furthermore, selective interference of these resonances gives rise to several interesting phenomena, which provide additional opportunities to control light at the nanoscale. For example, two important interference phenomena are the Kerker effect [60] and the occurrence of anapole modes [61]. The Kerker effect enables the suppression of backward scattering as well as near-zero forward scattering via the interference of electric dipole and magnetic dipolar scattering contributions. Anapole modes allow for strong field enhancement inside the nanoparticle based on the cancellation of scattering contributions with matching far-field characteristics by destructive interference.

A unique attribute of DNs is their ability to support electric and magnetic type of resonances, which can render them electromagnetically dual symmetric [62,63]. Duality of DNRs enables them to interact with chiral light fields and preserving the chirality after the interaction. In contrast, this is not the case for plasmonic nanoparticles having basic shapes as they can support only electric type resonances [64,65]. This particular characteristic makes DNs suitable candidates to deal with chiral emitters such as TMDs which are strongly chiral at low temperatures [46,66].

### 3.1. Resonant dielectric metasurfaces

Dielectric metasurfaces can be formed by arranging a large number of designed DNs in a planar fashion. By means of the just mentioned Kerker effect, the interference of electric and magnetic dipolar scattering contributions can be used to obtain uni-directional scattering of light. Typically, this happens at a single wavelength, where the value of the partial scattering cross sections from the electric and magnetic dipolar Mie-type modes coincide. By overlapping electric and magnetic dipolar modes in a DN, the Kerker condition can be approximately fulfilled over the full resonance bandwidth of the two modes. This is harnessed in dielectric Huygens metasurfaces, which consist of DNs whose geometry is carefully tuned to maximize the spectral overlap of their electric and magnetic dipolar Mie-type resonances. In this condition, a near-unity metasurface transmission can be achieved, while the phase of the transmitted electric field undergoes a variation from 0 to $2\pi$ as the wavelength is swept over the spectral width of the resonances [69]. Combining the high transmission and $2\pi$ phase coverage, Huygens’ metasurfaces have been successfully demonstrated for various wavefront-shaping applications [70,71].
Apart from wavefront shaping, Mie-resonant dielectric metasurfaces were recently also extensively employed to enhance and tailor various light–matter interaction processes. By concentrating the electromagnetic near-fields inside and near the individual nanoresonators, strong local field enhancements can be obtained in suitably designed metasurfaces, which was shown to allow for enhancing, e.g., nonlinear optical effects [72] or spontaneous emission processes [1,73]. Further tailoring of the geometry provides additional control of the resonance properties. For example, high-quality-factor resonances can be achieved based on the concepts of Fano resonances [74] or bound states in the continuum [75].

4. Integration approaches

Driven by the fast-paced technological development of modern semiconductor micro- and nanofabrication, also the fabrication of photonic nanos- tructures with top-down and bottom-up approaches has matured well over the past decades, allowing to fabricate nanostructures of diverse shapes over a large scale with high resolution. However, the integration of designed nanostructures with nanoscale emitters remains a challenge, especially when simultaneous requirements of scalability and high sample quality need to be fulfilled. Monolayers of TMDs can be produced either by mechanical exfoliation method or by scalable methods such as CVD and MBE. Though the yield with exfoliation method is low, the quality of crystals obtained is very high and no expensive equipment is required. Correspondingly, crystals produced with this method were used in many research studies. Nowadays, CVD and MBE allow for the fabrication of single-crystal monolayer TMD crystals with high quality, large size and high yield. Efficient integration of 2D-TMDs with designed nanostructures can be expected to represent a crucial step to ultimately harness their optoelectronic properties for scientific and industrial purposes. Significant progress was made in realizing such hybrid systems consisting of nanostructures and 2D-TMDs during the last few years [76,77]. These integration methods generally can be distinguished into dry methods and wet methods.

4.1. Dry transfer method

Dry transfer by viscoelastic stamping has proven to be a viable method for deterministic transfer of layered crystals without the need for any solvents or liquid suspensions. Typically, single- and few-layered crystals are produced by mechanical exfoliation from bulk crystals using scotch tape and polydimethylsiloxane (PDMS) films (Gel-Pak) [78]. The viscoelastic properties of PDMS allow to stick or release material by attaching and detaching in a rapid or slow manner, respectively. The scotch tape carrying multi-layered
crystals will be pressed onto the PDMS film and released fast to form a strong bonding between the crystals and the PDMS. Few- and single-layered crystals that stick to the PDMS film can be identified by visual contrast using a light microscope. After repeating this step until a crystal of desired properties is found, it can be transferred to the target substrate. Using a 3-axis stage, the PDMS film can be laterally aligned with a marker position on and brought in contact with the target substrate. Slow retraction of the PDMS film allows to delaminate the crystal from the PDMS leaving them situated on the target substrate.

### 4.2. Wet transfer methods

All-dry viscoelastic stamping, however, is limited when it comes to the use of synthesized crystals grown by CVD or MBE, as such crystals strongly bond to their growth substrate making the quality-preserving transfer of as-grown crystals to the PDMS film difficult. The most common alternative method is wet-transfer by the help of a sacrificial and a transfer layer, involving solvents or etchants for detachment of the synthesized crystals from their growth substrate.

In refs [79,80], a poly(methyl methacrylate) (PMMA) based process is presented whose steps are sketched in Figure 3. In a first step, TMD

![Figure 3. Wet transfer procedure: 1) CVD grown 2D-TMDs. 2) Spin coating of PMMA over SiO₂/Si substrate with 2D-TMDs. 3) Immersion of PMMA coated substrate into aqueous KOH solution. 4) PMMA film embedded with 2D-TMDs floats on the aqueous KOH solution. 5) Fishing the PMMA film with a target substrate (e.g. substrate with nanostructures). 6) 2D-TMDs integrated with the resonant nanostructure.](image)
monolayers that were synthesized on a SiO$_2$/Si substrate are spin-coated with PMMA. The coated substrate is then immersed into an aqueous KOH solution to separate the PMMA film (with embedded TMD monolayers) from the SiO$_2$ layer. After cleaning in deionized water, the floating PMMA/TMD film can be ‘fished’ using the target substrate. Proper alignment of the PMMA/TMD film can be assured by light microscopy and sliding the PMMA/TMD film atop the water film that is forming between the target substrate and the PMMA/TMD film. After proper alignment, the created stack will be dried on a hot-plate. The remaining PMMA layer can finally be dissolved by acetone. Success rate and quality of the transferred crystals may be further improved by removing the PMMA film using a supercritical dryer instead.

4.3. Direct integration

Wet transfer and dry transfer of crystals synthesized with CVD and MBE can lead to undesirable effects regarding their quality, namely, residue of resist on the crystals or the introduction of strain. Direct integration approaches allow to overcome this issue. In these approaches, the fabrication of hybrid dielectric nanostructures starts directly from a thin film of high refractive index dielectric material covered by or situated on pre-grown TMD crystals. Both the dielectric thin film and the TMD crystals are then nanostructured simultaneously. For example, using such a direct integration approach, germanium metasurfaces were fabricated directly on WS$_2$ crystals (see subsection 4.3).

5. Hybrid nanosystems consisting of 2D-TMDs coupled to resonant dielectric nanostructures

5.1. Shaping directional emission

The realization of industrial applications based on integrated TMD monolayers will crucially depend on their low-cost integrability. Besides large-scale fabrication of high-quality crystals, efficient light management will be important for a sufficient performance without the need for costly equipment such as strong excitation lasers, sensitive detectors, and high-NA collecting objectives. Integrating TMD monolayers with resonant dielectric nanostructures is a potential approach to tailor their PL/SHG emission in the far-field to fulfil the aforementioned requirements. In the following, we discuss some selected recent works on tailoring the far-field emission properties of monolayer TMDs by intergating them with single dielectric resonators or dielectric metasurfaces.

Cihan and coworkers [81] demonstrated enhanced directional scattering of PL from 1L-MoS$_2$ decorated with a silicon nanowire. By considering the
1L-MoS\textsubscript{2} as an electric dipole source, a model was developed based on a modified formulation of Mie-scattering, which allows to describe the excitation of an infinite silicon nanowire by a localized electric dipole. A sketch of the excitation scenario as well as the far-field emission pattern for an electric dipole ($\lambda = 675$ nm) displaced by 1 nm from an infinite silicon nanowire of radius $R = 38$ nm and being oriented along the nanowire axis is shown in the Figure 4(a). Directional scattering in forward direction is observed and explained by the interference of the nanowire’s electric dipolar Mie-resonance, as confirmed by the multipolar contributions shown in Figure 4(b), with the electric field directly emitted by the excitation dipole source. In their work, they further demonstrated experimentally directional scattering from 1L-MoS\textsubscript{2} selectively placed beneath a silicon nanowire (see sketch in Figure 4(c)) by using the nanowire itself as an etch mask. The

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{(a) Electric dipole excitation scheme for an infinite silicon nanowire and respective far-field pattern. (b) Multipolar scattering efficiency spectra for a silicon nanowire as depicted in a). (c) Illustration of spatially truncated monolayer MoS\textsubscript{2} situated below the silicon nanowire. (d) Measured top-to-bottom ratio $T/B$ of the photoluminescence from monolayer MoS\textsubscript{2} beneath a silicon nanowire for a polarization along and perpendicular (inset) to the nanowire axis. (e) Sketch of a CVD-grown monolayer of MoS\textsubscript{2} placed atop a silicon nanocylinder metasurface. (f) Light microscope image of an experimentally obtained assembly of monolayer MoS\textsubscript{2} and a fabricated silicon nanocylinder metasurface. (g) Measured back-focal plane images of the PL emitted by the assembly of MoS\textsubscript{2} monolayers atop various metasurfaces of different nanocylinder diameter. a–d), Reprinted by permission from the [Springer Nature]|[Nature] [Nature Photonics], (Silicon Mie resonators for highly directional light emission from monolayer MoS\textsubscript{2}, Cihan et al.) [81], (Copyright) (2018). (e–g), Reprinted with permission from [82], Copyright (2019) American Chemical Society.}
\end{figure}
spatial truncation of the 1L-MoS\textsubscript{2} to the nanowire’s footprint enhances the front-to-back ratio in the far-field by suppressing background emission. From PL scans of the hybrid system, obtained by confocal laser-scanning microscopy with detection in forward (top) and backward (bottom) direction, a top-to-bottom emission ratio ($T/B$) scan was determined as shown in Figure 4(d). For polarisation along the nanowire axis, ratios of $T/B > 20$ were observed favoring emission into out-of-substrate direction. This directional effect is mostly suppressed for the polarization orthogonal to the nanowire axis (see inset in Figure 4(d)), thus providing polarization control of the forward scattered PL from 1L-MoS\textsubscript{2}.

In a recent work by Bucher and coworkers [82], directional scattering of PL from 1L-MoS\textsubscript{2} was achieved by integration with a resonant dielectric metasurface consisting of a square arrangement of silicon nanocylinders as sketched in Figure 4(e). Various nanocylinder arrays were fabricated with different diameters in order to tune the spectral position of the nanocylinder’s Mie-type resonances across the emission band of 1L-MoS\textsubscript{2}. The hybrid system was then realized by wet-transferring CVD-grown 1L-MoS\textsubscript{2} onto the nanocylinder arrays. A typical fabricated nanocylinder array after the transfer is depicted in Figure 4(f). The authors observed a strong enhancement in the excitonic emission intensity from 1L-MoS\textsubscript{2} when placed atop a nanocylinder array. This effect was attributed to the fact that the nanocylinder arrangement is suspending the 1L-MoS\textsubscript{2} between adjacent nanocylinders. Compared to 1L-MoS\textsubscript{2} situated on bare substrate this results in a reduced substrate-induced doping and reduced local strain manipulation by the roughness of the substrate, the latter effect being known to modulate the crystal’s band structure causing transitioning to an indirect bandgap material. By a systematic study, the authors then showed a directional reshaping of the enhanced emission from freestanding 1L-MoS\textsubscript{2} due to scattering by the Mie-type resonances of the underlying nanocylinders. Respective backfocal plane images of the emission from 1L-MoS\textsubscript{2} atop various nanocylinder arrays are shown in Figure 4(g). A clear redirection of the emission from scattering into large angles (off-resonant case, $D = 240$ nm) to scattering preferentially towards the out-of-plane direction (resonant case, $D = 300$ nm) is observed. In summary, suspending 2D-TMDs to enhance their intrinsic quantum yield while simultaneously redirecting their emission for low-NA collection can be a viable route for the construction of 2D-TMDs-based photonic devices. Their efficiency can further be improved by resonantly enhancing the absorption and emission processes of 2D-TMDs as was demonstrated by Zhang and coworkers [83] using photonic crystal slab geometries.
5.2. Emission enhancement and coupling phenomena

Nanostructures offer an important route to overcome the size mismatch between the relevant wavelength of light and the only molecular thickness of the 2D-TMDs [84–88]. Generally, one distinguishes two regimes of coupling of emitters and resonators, namely the weak and the strong coupling regime. Which coupling regime is established in a particular system depends on several factors. Simply positioning the emitter in the vicinity of a resonator will be sufficient to achieve weak coupling. In order to achieve strong coupling, in contrast, strong light confinement at the spatial position of the emitter and/or a high-quality factor of the resonator will be required.

DNs offer many opportunities for coupling to nanoscale emitters including 2D-TMDs [1,89,90]. In ref [91], Lepeshov and coworkers numerically investigated an arrangement of silicon nanoparticles (Si-NPs) on a glass substrate (see Figure 5(a)). A 1L-TMD was considered to be situated in between a Si-NP and the substrate. Such a particle-on-film configuration is a popular geometry for studying enhancement of the interactions of

**Figure 5.** (a) Illustration of an assembly consisting of a silicon nanoparticle on top of a 2D-TMD supported by a glass substrate. (b) Tangential electric field intensity map as a function of the radius of the silicon nanoparticle and the wavelength. (c) Electric field distribution of the magnetic quadrupolar mode in a nanoparticle of radius 110 nm at 615 nm wavelength. (d) Experimental photoluminescence spectra from bare WS2 monolayers and assemblies of silicon nanoparticles of different radii on WS2 monolayer. (e) Experimental photoluminescence spectra from bare few layered WS2 (> 10 layers) and assemblies of silicon nanoparticles of different radii on few layered WS2 (10 layers). (a-c) Reproduced with permission from [91], © IOP Publishing. All rights reserved. (d-e), Reproduced from [94], with permission from The Royal Society of Chemistry.
resonant nanoparticles with various emitters placed in the gap between the nanoparticle and the substrate [92,93]. Lepeshov et al. proposed an approach for augmenting the interaction of Si-NPs with 1L-TMDs by simultaneously targeting the excitation rate, Purcell factor, and radiation efficiency. It is stated that the total PL quantum yield from 1L-TMDs coupled to the Si-NPs is proportional to \( d^2 \times N \), where \( d \) is the dipole moment of the exciton and \( N \) is the number of excitons, neglecting interference between excitons due to their incoherent nature. Consequently, a key recommendation was to enhance the number of excitons participating in the interaction by maximizing the tangential electric field component of the excitation field. A map of the tangential electric intensity as a function of the radius of a Si-NP is shown in Figure 5(b). The results show that for a given size of the Si-NP the magnetic quadrupolar mode outperforms the electric and magnetic dipolar modes in localizing the tangential component of the electric field at the 1L-TMD position (see Figure 5(c) for the corresponding electric field distribution). While the considered geometry provides strong confinement of tangential electric field, thus paving the way for a strong interaction, it turns out that this excitation scheme is not sufficient for achieving strong coupling. Nevertheless, the interaction can be strongly enhanced within the weak coupling regime. The authors used the radiative Purcell factor, a quantity accounting both for the Purcell effect and the radiation efficiency, to quantify the enhancement. Varying the geometrical parameters of the nanoparticle allowed to tune the Purcell factor within a chosen emission range of the 2D-TMDs. Large enhancements are predicted for 2D-TMDs such as MoSe\(_2\) and WS\(_2\), which exhibit very small Stokes shifts. This variation of geometrical parameters also allows to couple the emission of the TMDs to different resonance modes, thus allowing to manipulate the emission pattern.

In ref [94], assemblies of Si-NPs with 2D-WS\(_2\) were realized to systematically investigate the influence of Si-NPs on the PL and Raman characteristics of 2D-WS\(_2\). Si-NPs were synthesized using laser ablation from a silicon wafer in deionized water [95]. The obtained nanoparticles were of polycrystalline nature, and displayed electric and magnetic dipolar Mie-type resonances. Interestingly, the PL characteristics of assemblies of Si-NP atop single-layered TMDs and few-layered TMDs show contrasting results. Absorption in the Si-NP creates a local heating effect. Owing to the small thermal conductivity of 2D-WS\(_2\) [96], the heat is slowly dissipated to the substrate, thus causing quenching of the PL and a strong red shift of the PL emission maximum, as shown in Figure 5(d). Furthermore, a red shift of the Raman peaks is observed. In the case of few-layered WS\(_2\), the thermal conductivity is higher as compared to single-layered WS\(_2\) such that the heat can be dissipated more efficiently. Consequently, a smaller red shift of the PL emission maximum is observed. Another difference between the
single-layered and few-layered systems originates from the orientation of the exciton dipole moments. While in the single-layered case the predominant excitons are oriented in-plane, and are thus only weakly coupled with the predominant dipolar field of the Si-NPs, an out-of-plane component arises in the few-layered case, thus facilitating a strong interaction with the out-of-plane dipolar field of the Si-NPs, and ultimately leading to an enhancement of the PL emission (see Figure 5(e)).

The interaction between emitters and the electromagnetic field is a subject of great interest in quantum physics. When this interaction enters the strong coupling regime, the coherent energy exchange between the emitter and the light field becomes a reversible process. The light and the emitter states start to behave as a mixed state, which can be characterized as half light and half matter. The signature of strong coupling is a splitting in the energy spectrum, known as Rabi splitting. The strength of the interaction is described by the coupling constant ($g$), which is inversely proportional to the mode volume $V$. In order to achieve strong coupling, a high-quality factor and/or a small mode volume are required. Typically, these conditions are fulfilled by using light confinement concepts such as photonic bandgap architectures or plasmonic structures with ultra-small gaps. In practice, rigorous design rules and multi-step fabrication procedures are usually required to realize such systems. Strong coupling has, e.g., been achieved in several configurations employing a variety of emitters such as dye molecules, J-aggregates, quantum dots, and 1L-TMDs combined with high-Q cavities, as discussed in detail in three recent review articles [97–99]. 2D-TMDs are excellent candidates for strong coupling because of the strong dipole moments of their excitons [100]. Several studies of strong coupling employing TMDs were performed in plasmonic cavities, which are capable of confining the light up to nanometric volumes. Some relevant examples of strong coupling of 2D-TMDs with plasmonic cavities are discussed in another recent review [89].

Strong coupling of DNs with excitons of 2D-TMDs remains an open quest, owing to the difficulty of achieving sufficiently small mode volumes. A recent work by Lepeshov and co-workers highlights the challenge of achieving strong coupling in such systems [101]. The authors theoretically investigated the possibility to achieve strong coupling in core-shell particles consisting of a Si-NP core and a 1L-WS$_2$ shell. The Si-NP size was chosen such that the spectral position of its magnetic dipolar mode coincided with the A-exciton wavelength of 1L-WS$_2$. In this way, the uniform tangential field of the Si-NP can interact with the bright excitons of the 1L-WS$_2$. They estimated that the described core-shell configuration results in a Rabi splitting energy of up to 116 meV, fulfilling the strong coupling criterion $2g = \hbar \Omega > (\gamma_{ex} + \gamma_{md})/2$, where $\hbar \Omega$, $\gamma_{ex}$ and $\gamma_{md}$ are the Rabi splitting energy and the dissipation rate of the exciton and the magnetic dipolar resonance, respectively. Furthermore,
they demonstrated that by changing the embedding medium of the system from air to water, the Rabi splitting energy increases up to 208 meV unit. They also performed an experimental study of coupling Si-NPs to 1L-WS₂. However, due to the difficulty to realize the described core-shell geometry experimentally, in their experiment they resorted to a particle-on-film configuration as illustrated in Figure 6(a). As mentioned previously, such configurations can provide an ultra-small mode volume while being relatively easy to fabricate.

Si-NPs were hence drop-casted onto the 1L-WS₂. To vary the coupling strength in the system, one possibility is to vary the dipole moment of the excitons. An exciton’s dipole moment can be enhanced by enhancing the dielectric screening, which can be achieved by filling the 1L-WS₂ surrounding medium with a solvent of high static dielectric constant [102,103]. The authors chose water for this purpose considering its high static dielectric constant of ~78 at low frequencies relevant for the screening effect. In a static screening assumption, the enhancement of the effective dipole moment of the A-exciton was estimated to be \( \sqrt{\left(\varepsilon_s + \varepsilon_w\right)/\left(\varepsilon_s + 1\right)} \approx 2.7 \) (where \( \varepsilon \) and \( \varepsilon_w \) are dielectric constants of silicon and water, respectively). Experimental PL spectra of 1L-WS₂ in air and water (see Figure 6(b)) indicate a PL enhancement of 7.5, which corresponds to a dipole moment enhancement of 2.7, thus agreeing well with the static dielectric screening approximation. By filling the space around the Si-NPs with water, a nearly two-fold enhancement of the coupling constant as compared to the Si-NPs in air was achieved. However, even the enhanced coupling constant did not yet meet the criterion of strong coupling due to the participation of fewer excitons in 1L-WS₂ in the particle-on-film configuration as compared to the core-shell geometry.

In a similar work, Wang and co-workers also investigated the possibility to achieve strong coupling between Mie-type modes of Si-NPs and 1L-WS₂.
excitons [104], coming to a similar conclusions as Lepeshov et al. [101]. Additionally, they experimentally demonstrated unidirectional scattering and active tuning in the resonantly coupled system using temperature as a control parameter. For an increasing temperature, the 1L-WS$_2$A-excitonic emission was found to red shift. Depending on the radius of the Si-NP, the excitonic emission could be shifted either into or out of spectral overlap with the Si-NP’s magnetic dipolar resonance, thus offering active control over the resonant coupling in the hybrid structure.

A recently demonstrated approach to realize the strong coupling condition in TMDs is to directly fabricate the nanoresonators from the TMD material [105]. A corresponding recent work will be discussed in section 4.4 below.

### 5.3. Chiral effects

One intriguing application scenario of hybrid structures consisting of 2D-TMDs and dielectric nanostructures is to harness the nanostructures for the realization of highly integrated valleytronic devices. However, so far it is not well understood how photonic nanostructures, whether plasmonic or dielectric, influence the chiral emission, which can be used to read out the TMD’s valley polarization state, from 2D-TMDs in their vicinity. A comparative study of emission from chiral dipole sources placed in close proximity to plasmonic or dielectric nanostructures by Zambrana-Puyalto and coworkers has made an important first step to answer this question [64]. They analytically formulated Mie-theory in a helical basis and extracted the scattering cross sections of plasmonic and dielectric nanoparticles of different diameters for the two different helicities. As shown in Figure 7(a), in the case of plasmonic nanoparticles, the emission is scattered into both helicities equally, indicating a conversion of the exciting dipolar emission of given helicity into the opposite helicity channel. Consequently, the net circular dichroism is decreased. While chirality is only perfectly preserved under dual-symmetric condition, dielectric nanoparticles, in contrast, appear to support almost all intermediate values of chirality between $\pm 1$ due to the presence of magnetic resonances. At the electric dipole resonance, however, they display similar depolarizing behavior as their plasmonic counterparts. In the Figure 7(b), the chirality of the scattered field ($C'(\alpha)$) and the scattering efficiency ($Q'$) are plotted for silicon and gold nanoparticles with a radius of 100 nm, indicating that for the Si-NPs $C'(\alpha)$ is maximized around the duality condition (at wavelength 808 nm), while for the gold nanoparticles $C'(\alpha)$ is close to zero inferring achiral behavior.

As another research direction targeting mainly valleytronic applications, enhancing and routing the valley specific chiral emission has attracted intense research interest recently [106–109]. The majority of efforts in this
direction were employing plasmonic nanostructures. However, recently, Guddala and coworkers experimentally demonstrated the possibility to selectively address the specific valleys in 2D-TMDs by making use of a chiral resonant dielectric metasurface [110]. They considered an interesting way of integrating WS$_2$ monolayers with germanium metasurfaces. Specifically, they fabricated the metasurfaces directly on top of the WS$_2$ monolayers, as illustrated in Figure 7(c). The metasurface was designed to generate chiral near-fields for linearly polarized excitation light at 625 nm wavelength. As shown in Figure 7(d), by switching the polarization state of the excitation light between +45° and −45°, the circular polarization state of the emission of the coupled system could be efficiently manipulated. The authors also showed that the valley polarization decreases when the assembly of WS$_2$ and germanium metasurface is excited with wavelengths off-resonant to the excitonic resonance, as expected due to lossy phonon-
assisted intervalley scattering [111] and long-range exciton exchange interaction [112].

5.4. Patterning

Bulk TMDs show an unusually high refractive index in the visible and near infrared range [113], a feature that has not fully been harnessed yet. While their layered structure (strong covalent bonds in-plane and weak Van-der-Waals bonds out-of-plane) makes them intrinsically anisotropic, most studies focus on the in-plane component of the dielectric tensor which was shown to exhibit a significantly higher optical constant for both bulk and 2D-TMDs as compared to the respective out-of-plane components [114,115]. Consequently, high refractive index films can be created from 2D-TMDs. Nanopatterning of such films opens a promising route to ultimately realize atomically thin high-index nanostructures for elastic and inelastic light scattering applications.

Yang and coworkers [116] demonstrated elastic light scattering from patterned multi-layered MoS$_2$ to create the world’s thinnest lens. In their work, they showed that light undergoes multiple reflections inside atomically thin films of layered MoS$_2$ before being transmitted due to the strong impedance mismatch at its interfaces to air or typical substrate materials. Hence, a giant increase in the optical path length (OPL) can be induced by layered MoS$_2$ as shown in Figure 8(a). The measured OPL for 2D-MoS$_2$ on a SiO$_2$/Si substrate greatly exceeds its geometrical OPL as well as the OPL measured for the same number of layers of graphene. This enhanced OPL creates a significant phase difference for light being transmitted through stacks of layered MoS$_2$ with a different number of layers, hence, making them suited for light steering applications based on elastic scattering to the far-field. For experimental demonstration, Yang and coworkers realized an extremely thin concave lens by focused ion beam milling starting from an exfoliated 9L-MoS$_2$ crystal. A line cut through the achieved OPL profile of the patterned stack is displayed in Figure 8(b). The optical performance of the structured stack was then experimentally investigated by scanning optical microscopy. Transverse intensity profiles were acquired in reflection geometry with changing z-position of the stack with respect to the focal plane of a focusing objective. A cut along z-direction through acquired intensity profiles is shown in Figure 8(c) and reveals a clear focusing effect with a focal length of $2f = -480$ μm at a wavelength of 532 nm. In the same work, they further theoretically showed that 1L-MoS$_2$ displays a stronger scattering cross section in comparison to SiO$_2$, graphene, and gold layers of similar thickness. The strong scattering strength of 1L-MoS$_2$ and the capability of discretely tailoring scattering properties by stacking layers on
Inelastic scattering by 2D-TMDs, especially absorption and emission, is strongly dependent on the enhanced excitonic effects commonly observed for this group of 2D materials. Patterning of TMDs provides an opportunity for many possible applications as nanoscale integrated optical devices.

Figure 8. (a) Optical path length measured experimentally with phase-shifting interferometry as a function of layer number for MoS$_2$ and graphene. (b) Optical path length profile across the micro-lens fabricated in 9L-MoS$_2$. (c) Intensity distribution of micro-lens output measured by a scanning optical microscope. (d) Nanoresonators fabricated from a bulk crystal of WS$_2$. (e) Simulated scattering spectrum of the WS$_2$ nanoresonators (height = 200 nm, radius = 150 nm). The field distribution and electric field lines of the electric and magnetic modes are displayed in the insets close to their corresponding resonant peaks. (f) Experimental dark-field scattering spectrum of one of the fabricated WS$_2$ nanoresonators, showing an anticrossing of the two dominant mode signatures in the spectrum. (g) Microscope image of a grating with fork-like dislocation patterned in a monolayer of MoS$_2$. Inset: Zoomed portion of the fork pattern in the center. (h) Back focal plane image of the SHG emission from the patterned 1L-MoS$_2$ showing the zeroth diffraction order in the center, and optical vortices occurring in the 1$^{st}$ diffraction orders on the either side. a–c), Reprinted by permission from [Springer Nature]: [Nature] [Light: Science & Applications] [116] (Atomically thin optical lenses and gratings, Yang et al.), [COPYRIGHT] (2016). d–f), Reprinted by permission from [Springer Nature]: [Nature] [Nature Nanotechnology] [105] (Transition metal dichalcogenide nanodisks as high-index dielectric Mie nanoresonators, Verre et al.), [COPYRIGHT] (2019). g–h), Reprinted with permission from [117], © The Optical Society.
to hybridize inelastic excitonic properties with elastic scattering properties, e.g. by featuring Mie-type resonances supported by their high refractive index, as very recently demonstrated by Verre and coworkers [105]. The authors fabricated nanodisk-shaped Mie-resonators by electron-beam lithography starting from exfoliated multilayer WS$_2$. A false color SEM image of fabricated WS$_2$ nanodisks is shown in Figure 8(d). Mie-type scattering was predicted by numerical simulations (see Figure 8(e)) and experimentally verified by dark-field spectroscopy. Measured dark-field spectra for WS$_2$ nanodisks of varying diameter are shown in Figure 8(f). For a certain range of disk radii the excitonic mode at 630 nm overlaps with a strongly dispersive feature which was attributed to an anapole scattering condition. The observed mode anti-crossing is indicative of the strong coupling regime and respective hybridization of Mie-type and excitonic resonances within a single monolithic nanostructure.

In ref [117], Löchner and coworkers demonstrated manipulation of second harmonic generation (SHG) emission from CVD-grown 1L-MoS$_2$ in the far-field, by patterning the monolayer via focused ion beam milling. For example, 1L-MoS$_2$ gratings with a fork like dislocation were fabricated (see microscope image in Figure 8(g)) to generate optical vortices of topological charge $l = 1$ by the interference of coherent second harmonic (SH) emission from different regions of patterned 1L-MoS$_2$. The angular distribution of the SH emission was imaged by back-focal plane imaging, clearly showing ring-shaped patterns with zero intensity in the center, as characteristic for a vortex beam, in both 1st diffraction orders (see Figure 8(h)). Another recent study [118] also demonstrated complex holographic patterns of the SHG emission by patterning 1L-WS$_2$ by focused ion beam milling.

6. Conclusions and outlook

The topic of coupling monolayers and few layers of TMDs to Mie-resonant dielectric nanostructures is still in its nascent stage. Although only a relatively small collection of research works have been published in this area so far, the high potential of such hybrid systems for optoelectronic or valleytronic devices is already apparent. In particular, Mie-resonant dielectric nanostructures were demonstrated to provide control over the 2D-TMD emission spectrum, direction, polarization, and efficiency. Also, the possibility to optically address the 2D-TMDs’ valley degree of freedom at the nanoscale and to preserve the associated emission chirality using dielectric nanoresonators were discussed.

However, numerous challenges remain to be addressed in order to fully develop this hybrid class of photonic nanosystems for applications.

Regarding the 2D-TMDs as one essential ingredient of these systems, the production of continuous films of these materials having a high quality and
a high quantum yield by scalable methods remains an active research topic. Furthermore, as the methods for the creation of heterostructures of different TMD monolayers are maturing, an entirely new research chapter will likely be opened regarding the hybridization of DNs with van-der-Waals heterostructures [119].

On the side of the DNs, most hybrid systems realized so far used amorphous silicon or germanium as resonator material. These materials display a certain amount of absorption in the visible range, i.e. in the emission bandwith of the 2D-TMDs. The materials such as GaP or TiO$_2$, which also have a high refractive index but only low absorption in the visible range may provide an alternative. As for the integration of 2D-TMDs with dielectric nanostructures, in order to avoid complications due to strain and sample inhomogeneities, there is a need to develop new approaches that do not rely on (dry or wet) transfer procedures. Realizing photonic structures directly on a substrate with CVD-grown monolayers, as demonstrated in ref [110] for a mechanically exfoliated monolayer TMD, could present a possible and scalable solution. Alternatively, monolayers may be grown directly on pre-fabricated nanostructures. Patterning of TMDs is another promising integration approach. Most efforts of patterning TMDs have concentrated on bulk crystals or multilayers of TMDs, aiming to create the photonic structures or nanoresonators directly from the TMD material. As a future direction, integration schemes for patterned monolayers will further enhance the engineering options for tailoring optoelectronic properties of the hybrid systems, e.g. by allowing for a precise placement of the light-emitting, nonlinear or tunable material in the nanostructure.

When considering light–matter interaction enhancement applications, the specifics of Mie-resonant dielectric nanostructures have to be carefully observed. While their low losses enable coupled systems with near-unity radiation efficiency, which is essential for high-efficiency devices, unlike plasmonic nanoparticles, dielectric nanoparticles typically localize the electromagnetic near-fields mostly at their inside. As a result, it can be difficult to create a large overlap of the 2D-TMD and the region of strong field enhancement. Careful engineering of the system geometry, the possible integration of 2D-TMDs inside the DNs, as well as the deployment of special types of modes such as anapoles [61], Fano resonances [74], or bound states in the continuum [75] offer important routes to overcome this challenge.

Apart from their low losses, another useful feature of DNs is their electric and magnetic multipolar response. Especially, their ability to support magnetic dipolar resonances in addition to electric ones was theoretically suggested to play a crucial role in preserving the chirality of valley-polarized emission processes [64]. While more studies, including experimental efforts, are required, based on these initial results we envision that Mie-resonant dielectric nanostructures may play an important future role when it comes
to the preparation and readout of the valley degree of freedom at the nanoscale.

All in all, hybrid nanosystems consisting of Mie-resonant dielectric nanostructures coupled to 2D-TMDs offer many intriguing opportunities for the realization of efficient nanoscale optoelectronic and valleytronic devices. The possibility to choose among several TMD monolayers, their heterostructures, and countless nanostructure architectures opens an enormous design freedom in tailoring the properties or functionalities of the hybrid nanosystems toward particular applications. Finally, electrical gating of the 2D-TMDs, which allows for dynamically controlling the 2D-TMDs PL and SHG properties \[43,44\], is expected to provide a route for the realization of actively tunable hybrid nanoscale devices based on this concept.

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