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Quantum Photonics with layered 2D Materials

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Abstract: Solid-state quantum devices use quantum entanglement for various quantum technologies, such as quantum computation, encryption, communication, and sensing. Solid-state platforms for quantum photonics include single molecules, individual defects in crystals, and semiconductor quantum dots, which have enabled coherent quantum control and read-out of single spins (stationary quantum bits) and generation of indistinguishable single photons (flying quantum bits) and their entanglement. In the past six years, new opportunities have arisen with the emergence of 2D layered van der Waals materials. These materials offer a highly attractive quantum photonic platform that provides maximum versatility, ultra-high light-matter interaction efficiency, and novel opportunities to engineer quantum states. In this Review, we discuss the recent progress in the field of two-dimensional layered materials towards coherent quantum photonic devices. We focus on the current state-of-the-art and summarize the fundamental properties and current challenges. Finally, we provide an outlook for future prospects in this rapidly advancing field.

H1 Introduction

High-performance quantum light sources are required to realize future quantum photonic technologies. For example, stable high-repetition-rate true single-photon sources can enable various quantum communication schemes with unconditional long-term information security[1,2]. Scalable on-chip quantum light sources can facilitate robust photonic quantum computation with the ultimate data processing speed, which is coded into fragile quantum states of individual unperturbed photons[3]. Furthermore, quantum light sources are of great interest for precision optical measurements which can approach the Heisenberg limit[4]. An ideal quantum light source is deterministic, providing on-demand exactly one photon with indistinguishable properties in all degrees of freedom (polarization, spatio-temporal mode, and energy).

Building on concepts from atomic and molecular optical physics, several approaches to generate single-photons and spin-photon interfaces in solid-state materials have been discovered and developed. Solid-state quantum emitters can offer significant advantages over their atomic counterparts, including: the ability to repeatably address a single emitter, tunable energy scales, large oscillator strengths, the capability for added semiconductor or integrated photonic functionality, and (potentially) scalability. Unfortunately, the solid-state environment also contains intrinsic sources of noise and dephasing mechanisms that must be addressed. The foremost solid-state quantum photonics platforms are colour centres in diamond and semiconductor quantum dots[5-8].

Over the past six years, single photon emitters in 2D layered materials were shown to exhibit non-classical quantum light emission[9-14]. Such emitters can be deterministically positioned by local strain fields[15-19], a feature unique to 2D materials. These materials consist of one or few atomically thin layers, inherently providing quantum confinement in one dimension, and possess unique optical and electronic properties[20] and a straightforward means for integration with various optical cavities [21-25] and waveguides [26-30] for integrated photonics. In addition, one can easily stack these materials on top of each other to form devices such as

electrically pumped single photon light-emitting diodes (LEDs)[31], charge-tunable devices for single spin control[32,33], or novel artificial heterostructures[34-36] with completely different chemical and physical properties (electrical, optical, magnetic properties), thus possibly allowing more versatile quantum devices for various applications beyond simulation, communication and computation, such as sensing, imaging, and metrology[37-39].

In this Review, we provide an overview of the basic properties of different 2D layered materials and discuss the recent advances in the field of quantum light sources based on these materials. We conclude by offering a perspective on future directions and challenges in most materials advances and potential applications.

H1 Single photon sources and spin-photon interfaces

A single-photon emitter (SPE) is a few-level quantum system with an optical transition that emits only one photon at a time (see Supplementary Information for an introduction). In the simplest case, it is a two-level system with an optical transition that can be driven coherently or incoherently from the ground to its excited state. Spontaneous emission from the excited state can produce a single photon with a characteristic rate γ_{sp}. In a non-resonant photoluminescence (PL) spectrum, the SPE's spontaneous emission appears as a discrete peak, referred to as the zero-phonon line (ZPL), with a spectral width determined purely by γ_{sp} in the ideal transform-limited case (the case $T_2 = 2T_1$, where T_2 and T_1 are the emitter's coherence time and lifetime, respectively). While incoherent single photons can outperform coherent input states of light for some quantum applications such as quantum random generation [40], it will be difficult for incoherent SPEs to outperform the low-cost and high-data-rates at telecom wavelengths obtained with faint laser pulses and decoy states for other quantum applications such as a practical implementation of intra-city quantum key distribution[41]. However, employing SPEs for quantum technologies which require indistinguishable single photons provides a compelling motivation for their technological development. Photon indistinguishability, defined as the mean wavepacket overlap of two photons, is characterized by the visibility of two-photon interference[42].

One of the more established approaches to a secure quantum network[43] relies on interfacing indistinguishable photons, ideal for sharing quantum states over long distances, with spins, attractive for local storage and quantum processing due to their insensitivity to environmental noise[44-46]. Spin-active optical emitters, such as quantum dots and paramagnetic defects, can provide the required spin-photon interfacing capabilities. A prerequisite for spin-active optical emitters is long electron spin coherence time, which can be achieved by SPEs featuring weak coupling of their states and transitions to phonons and electrical noise in material systems with a low concentration of non-zero nuclear spins (Box 1). Operations such as optical spin initialization and readout, and the creation of spin-photon entanglement, rely on the availability of spin-selective optical transitions. These transitions originate from selection rules set by the symmetry of the system. Efficient spin-photon operations require stable transitions, exhibiting no spectral diffusion **[G]** or blinking **[G]**. In addition, mixing between orbital eigenstates with different spin characters should be minimal to avoid spin-flips induced by optical excitation. If optical excitation generates a detectable amount of PL without causing a spin-flip, single-shot spin readout can be achieved, as shown for nitrogen-vacancy [47,48] and SiV centers[49] in diamond, and for III-V quantum dots[50].

Generation of spin-photon entanglement requires the spin-photon coupling to be coherent [51-53]. Coherence can be achieved in systems with minimal inhomogeneous broadening (approaching the transform limit **[G]**) and with minimal incoherent phonon sidebands, such that a large fraction of emission is in the ZPL. Spin-photon entanglement is a building block for more sophisticated schemes, such as remote entanglement between spins in remote nodes of the quantum network[47,54,55]. These schemes benefit from additional long-lived qubits provided by nuclear spins. Compared to electronic spins, nuclear spins have a reduced coupling to magnetic fields and longer spin coherence time due to a much smaller gyromagnetic ratio. The concentration of accessible nuclear spins in a given material depends on its specific isotopic composition, from the very "dense" spin bath associated with GaAs and hBN (with all elemental isotopes featuring non-zero nuclear spins) to "dilute" baths for Si (95.3% of isotopes have nuclear spin I=0), C (98.8% with I=0), W (85.7% with I=0), Mo (74.5% with I=0), S (99.3% with I=0), and Se (92% with I=0). Control of individual nuclear spins has only been achieved for dilute

baths, where the nuclear spins are sufficiently far apart that their hyperfine interaction with the central electronic spin is resolvable either directly in the electron spin resonance spectrum [56-58] or by tailored pulse sequences [59,60].

[H1] Quantum photonic sources with 2D materials

The simple nature of the lattice construction of 2D materials offers unprecedented opportunities for quantum photonics (see Supplementary Information for an introduction). The lack of dangling bonds (in principle) in the lattice is promising for hosting transform-limited single photon sources, while the atomically thin nature offers potential as a very local electronic probe of the environment or high-efficiency coupling of the radiative emission to optical modes by engineering the dielectric environment. In addition, hybrid approaches to integrated chip technologies benefit from the flexible fabrication processes for integration with optical components (such as cavities, waveguides and others) and light coupling (Box 2). Further, efforts to use nonlinear frequency conversion with 2D materials for quantum light generation and manipulation have been made.

Zero-dimensional quantum confinement in bulk semiconductors is typically achieved by one of the following. One option is a point defect in the crystal lattice, such as electrons bound to donor impurities in bulk GaAs[61], diamond[62], or SiC[63]. Another option is a change in material composition at the nanometer-scale, which has a reduced electronic band-gap material compared to the surrounding matrix, often accompanied by local strain. This change in composition can be intentional, for example, in self-assembled quantum dots[64] or can be unintentionally caused by interface disorder, such as at the interface between two epitaxial layers[65]. Similarly, in 2D materials, SPEs can potentially originate from excitons localized due to point defects in the crystal or an electronic perturbation in the crystal environment, for instance, dangling bonds in the substrate material, impurities at a nearby interface, or an electrostatic gate. The former is typically the case with SPEs in wide-band-gap hBN which are mostly based on deep level defects, whereas the precise confinement mechanism in monolayer TMDs is still under debate (a notable exception are SPEs created by He-ion induced defects in MoS_2[66,67]).

Excellent single photon purity is possible for SPEs in 2D materials, which also have the unique capability to be site-controlled and arranged into arrays via localized strain or implantation. For example, moiré-heterostructures, in which periodic electronic interaction between two atomic layers results in the case of a small lattice mismatch or relative twist, present a novel method to create confinement in a semiconductor[34,68]. Moreover, the 2D nature of the layer hosting the SPEs removes the total internal reflection condition typical from bulk-based SPEs, enabling enhanced extraction efficiency of the emitted photons. In general, SPEs based on TMDs have an attractive spin-degree of freedom to realize a spin-photon interface, while only a few defect species in hBN do[69,70]. However, spectral fluctuations, inhomogeneity among emitters, and large phonon sidebands all present significant challenges to realize arrays of indistinguishable single photon sources or coherent spin-photon interfaces. To date, the intrinsic spin coherence and coherence of single photons generated with 2D quantum emitters have yet to be systematically probed. SPEs are usually non-resonantly excited, but steps towards resonance fluorescence and coherent control as well as charge state control and electrical injection using heterostructure devices have been taken.

[H2] Monolayer TMDs

SPEs in TMDs were discovered in mono- and bi-layer WSe_2 in 2015[9-13,15] and monolayer WS_2 soon after[31]. These first reports identified the main features of quantum emitters with W-based TMDs, which exhibited inhomogeneous spectral features at lower energy than the main free-excitons with linewidths down to 0.1 meV (full width at half maximum) (Fig. 1a). The neutral localized excitons consist of a fine-structure split doublet, typically (but not exclusively) with orthogonal linear polarization and a splitting of 0.4 - 0.8 meV. While detailed

theories about the origin of the splitting have been proposed[71], in general, the fine-structure splitting **[G]** (FSS) arises due to electron–hole exchange interaction[72] and the relatively large magnitude of the FSS is due to the strong Coulomb interactions in TMDs. Large gyromagnetic ratios (g-factors) of the excitons were observed for out-of-plane magnetic fields (Faraday geometry), in the range of 8–10[9-13,15]. Beyond single localized excitons, bi-excitons in monolayer WSe_2 SPEs have been observed[73-75], with unambiguous identification based on the distinct polarization dependence and cross-correlation of the exciton and bi-exciton spectral peaks[73] (Fig 1b). At low temperature (T < 10 K), high-purity single photon emission ($g^2(0)$ < 1%, ref.[76]) has been demonstrated from isolated emitters and time-resolved PL measurements reveal lifetimes of a few ns to few tens of ns[9-13,15]. While the fraction of zero-phonon line to total emission is variable in WSe_2 SPEs (likely related to strain conditions), it is typically within the range of 50-80%[76,77].

The spatial correlation of the SPEs with local strain pockets was observed early on[13,15], raising the prospect to spatially define the emitter position and deterministically create arrays of SPEs[16-18]. For instance, a WSe_2 flake can be transferred on top of lithographically defined nanopillars to create point-like strain perturbations that locally modify the electronic band-gap (Fig 1c). Density functional theory results show that a strained WSe_2 flake with a 200 nm tall nanopillar leads to a 50 meV shift in the bandgap[78], which agrees well with the experimental results. Excitons created optically in the vicinity of the localized stressor can diffuse to the potential minimum, creating an efficient exciton funnel[79] for a SPE. By optimizing the nanopillar aspect ratio, a high-yield (> 90%) of deterministic positioning of SPEs with an accuracy up to 30 nm has been achieved[16-18]. Sharp emission lines in PL of $MoSe_2$ arising at local strain sites have also been observed[80-82], which recently have been shown to emit single photons[82].

The precise origin of quantum confinement for TMD based SPEs has been subject to debate. Notably, the local strain features (such as that created by nanopillars or nanobubbles) are typically at the scale of 100 nm[16-18,83,84], which is significantly larger than the nm scale exciton Bohr radius in TMDs[20]. Although local wrinkles around the stressor sites may localize excitons at the 10 nm scale[74,85], the origin for the SPE is unlikely to be solely due to local strain[86], but rather the interplay or combination of local strain with disorder in the environment and / or crystal defects (Box 1, Fig. 1d). There are two important observations to be made here. First, the typical g-factor of the localized excitons is similar to the ground state delocalized dark (free) exciton **[G]** g-factor (~8) in monolayer WSe_2 [20,87]. Second, the SPEs spectrally overlap with a broad band of emission in flat (nominally unstrained) monolayer WSe_2 that is energetically below the free exciton emission. This low-energy, broad bandwidth emission is typically associated with excitons localized at point defects[88], which can be dispersed by local strain pockets[15].

To account for these observations in a microscopic model, one proposal is that the WSe_2 SPEs are intervalley defect excitons arising from hybridization of the dark excitons, modulated in energy by local strain, and defect states associated with intrinsic Se vacancies[71] (Fig. 1e). This theory predicts many physical features (such as the g-factors, FSS, radiative lifetimes, and polarization dependence) observed experimentally for WSe_2 SPEs. Other models based on W-vacancies[89], O interstitial defects[90], and dislocations[91] have also been proposed. Recently, the combination of controlled strain and electron-beam-induced structural defects in WSe_2 has led to the creation of SPEs with a relatively high yield, biexciton formation, and single-photon emission up to 150 K[75].

Recognizing that point defects in WSe_2 likely contribute to the exciton localization responsible for SPEs, efforts to minimize the intrinsic point defect densities (using a flux-growth technique) or deterministic defect creation (using electron-beam irradiation[92]) can be pursued. Recently, He-ion beam lithography has successfully been used to create emitters in other TMDs such as MoS_2 monolayers with a lateral precision of only ~9 nm[66]. Encapsulation of MoS_2 monolayers between two hBN layers after irradiation led to high quality quantum emitters with long excited state lifetimes, showing clear antibunching[93], and the realisation of gate-tuning SPE arrays[94]. The encapsulation shields the 2D material from ambient gases and the SiO_2/Si substrate and reduces the impact by doping and surface roughness[95], as well as minimizes effects of residues from nanofabrication[96].

TMD SPEs can be incorporated into heterostructure devices with a range of functionality. For example, a single-photon LED can be realized by tunneling carriers from a graphene layer through a hBN barrier to a gated monolayer TMD containing SPEs[31,97] (Fig. 2a-c). Alternatively, a p-i-n diode can be realized with a device that keeps the TMD at intrinsic doping but uses a second graphene gate of opposite polarity to the first[98]. Owing to the Coulomb blockade, deterministic loading of either a single electron or a single hole from graphene into WSe_2

SPEs has also been achieved to realize tuning between negatively charged (X^{1-}), neutral (X^0), or positively charged (X^{1+}), excitons[32] (Fig. 2d-f). The spin-singlet of the charged excitons leads to a clear spectral signature, the lack of FSS. Here, the unique advantages of vdW heterostructures are apparent — thanks to a monolayer hBN tunnel barrier, strong hybridization of the WSe_2 SPE and the Fermi sea (in graphene) is possible. Such devices provide opportunities to explore the potential for spin-valley locking and coherence in TMD SPEs[99], where the ability to optically initialize the resident spin in a specific valley is a valuable first step[33]. In principle, localized excitons are not expected to have permanent electric dipoles out of the plane of the TMD - similar to free excitons in TMDs[100], leading to minimal DC stark tuning[32] **[G]**. However, in some cases, appreciable SPE energy tuning can be realized with applied electric fields[101]. Additionally, an applied field can change the exciton wavefunction symmetry and affect the FSS[102].

Localized excitons in WSe_2 typically have a lifetime of several nanoseconds[9-13]. When WSe_2 is coupled to plasmonic cavities **[G]**, enhanced single-photon radiation is realized. Using plasmonic nanocavities the localized exciton lifetime has been successfully reduced to ~100 ps, increasing the brightness of SPEs without affecting the purity of SPEs[22,23]. This has also been achieved by depositing a WSe_2 monolayer flake on a metallic surface, which causes monolayer WSe_2 to become strained due to nanoscopic metal particles on the substrate, which can also increase the emission rate through plasmonic interactions[21]. In this case, the effect of a plasmonic nanocavity on TMDs based SPEs can be two-fold: SPEs positioning and enhancement of their emission rate. Using gold nanocubes simultaneously as plasmonic cavities and as strain-inducing localization sites, a Purcell enhancement **[G]** was recently measured[22]. Another approach facilitating Purcell enhancement to increase the quantum efficiency and reduce the effects of dephasing on the quality of the emitted single photons are tunable open cavities (Box 2). Tunable open cavities were first used to study polariton effects in 2D materials[103,104] and were later used to realise SPEs with a quantum efficiency of up to 46% ± 3%[25]. Tunable open cavities enable straight forward resonant coupling to the cavity mode[25,103,105]. Otherwise, spectral overlapping of the SPE and the cavity mode requires a post-growth tuning knob, such as electric field tuning[101] or strain tuning[106,107]. Using the latter technique a reversible tuning of 18 meV for localized excitons in WSe_2 has been realized, tailoring the emission properties in the same order of magnitude as the SPE ensemble energy distribution[108].

Another aspiration for the field of 2D material quantum emitters is their integration into photonic circuits (Box 2)[109]. Based on the strain-induced SPE creation using nanopillars, several groups around the world proposed to use waveguide structures as the strain-inducing nanostructure. The waveguide acts not only as the strain-nucleation site but also directly couples the SPE into the photonic circuit, for example, by coupling emitted single-photons from a WSe_2 flake to a plasmonic slot waveguide[27] **[G]**. The peak seen in the PL spectrum indicates coupling of generated photons into the waveguide. Another approach was to transfer a WSe_2 flake on the facet of a titanium-diffused lithium niobate waveguide and directional coupler[26], while PL into a waveguide mode from a WSe_2 monolayer placed on top of a SiN waveguide has also been reported[29]. Recently, antibunching from single-photons coupled to the waveguide has been measured [110] and this geometry also allowed to perform resonance fluorescence. However, in these studies, the coupling efficiency into the waveguide mode was below 10%. To increase the coupling efficiency into the waveguide mode, more complex structures (such as nano-beam cavities[111] or encapsulation of the WSe_2 flake into the waveguide) would be desired. Finally, techniques such as He-ion beam lithography have the potential to simplify the 2D quantum emitter integration into photonic circuits. First, a large 2D material heterostructure could be transferred and integrated into the circuit. In the next step, the quantum emitters would be deterministically created at specific positions on the circuit. This would allow thousands of emitters to be precisely coupled to on-chip cavities with one single transfer step.

[H2] Moiré heterostructures

Vertically stacked monolayer TMDs represent a useful nanoscale system to engineer quantum states and correlations. TMD heterobilayers present atomically sharp interfaces and type-II band alignment[112,113], giving rise to the formation of interlayer excitons **[G]**. A prototypical example is a $WSe_2/MoSe_2$ heterobilayer, in which the resulting interlayer excitons inherit the valley-contrasting physics of the individual monolayers[114-117]. The

spatial separation of the electron and hole results in an increased exciton radiative[114,118] and valley depolarization lifetime[115-117]. The separation of the exciton carriers also results in a large permanent out-of-plane electric dipole moment that enables a large tunability of the exciton transition energy[117] and exciton lifetime[115] by externally applied electric fields. Moreover, similar to monolayer WSe_2 and WS_2[17,18], local strain profiles induced by patterned substrates enable exciton trapping in nanoscale confinement potentials, where the mean number of interlayer excitons can be controlled via the optical excitation level[119,120].

Uniquely, heterobilayers with a lattice mismatch and/or a relative angle twist between the constituent monolayers feature a spatially periodic moiré superlattice[121], with a periodicity that can be precisely controlled by the relative crystallographic alignment of the layers for stacking beyond the theoretically proposed critical angle for lattice reconstruction[122-124]. The moiré superlattice can lead to the hybridization of the intralayer and interlayer excitons[125], which manifests through a pronounced exciton energy shift as a function of the heterobilayer stacking angle. In addition, the moiré superlattice creates a periodic potential landscape for excitons in the heterobilayer[34,68,126], in which three trapping sites (A, B, and C) with specific atomic registries emerge, such as R_h^h(A), R_h^X (B) and R_h^M (C) , where R_h^μ denotes an R-type stacking with the μ site of the electron layer (either h the hexagon centre, X the chalcogen site or M the metal site) vertically aligned with the hexagon centre (h) of the hole layer (Fig. 3a). For moiré periods larger than the exciton Bohr radius, these moiré trapping sites can act as smooth quantum-dot-like confining potentials[34,68].

Experimental evidence of interlayer excitons trapped in a moiré potential has been reported in $MoSe_2/WSe_2$ heterobilayers with twist angles of around 0°, 21.8° and 60° at cryogenic temperatures[35,127,128]. These localized interlayer excitons present linewidths below ~100 μeV and power-dependent emission intensities that can be described by a two-level saturation model[35,127,128], hallmarks of a few-level quantum confined system.

The moiré potential minima preserve the three-fold rotational (C_3) symmetry[34,68,129]; excitons trapped in these moiré-defined sites therefore inherit the valley physics properties of the monolayer semiconductors, although with optical selection rules that depend on the spin configuration of the exciton carriers (spin-singlet/spin-triplet) and the atomic registry of the trapping site (Fig. 3b)[130]. Polarization-resolved PL measurements show that the moiré-trapped interlayer excitons retain the strong valley polarisation of the constituent monolayer semiconductors and exhibit strong helical polarization due to the C_3 symmetry, which results in a notable absence of observable fine structure[35,127,128] (Fig. 3c, top). In addition, magneto-optical spectroscopy experiments reveal highly uniform g-factors across several samples, dependent on relative layer twist. The g-factors only take absolute values of ~15.9 and ~7 in samples with relative angle twists of 60° and 0°, respectively, [35,127]clear fingerprints of the spin and valley configurations for excitons composed of band-edge electrons and holes at the ±K points (Fig 3c, bottom). Moreover, in a heterostructure of bilayer 2H-$MoSe_2$ and monolayer WSe_2, the phenomenon of locked electron spin and layer pseudospin can lead to two quantum-confined interlayer exciton (IX) species with distinct spin-layer-valley configurations: the holes, localised in the WSe_2 layer, are strongly Coulomb bound to electrons localised in either the lower or upper $MoSe_2$ layer to form IX^H or IX^R species, respectively[127]. IX^R (IX^H) present carriers with parallel (antiparallel) spin-valley locked contributions, which results in an effective layer-locking of the Landé g-factors of the trapped IX. In addition, the helical polarization of the trapped excitons appears to be determined by the atomic registry[35,127]. Unambiguous evidence of the quantum nature of the moiré-trapped IXs has recently been provided via photon antibunching (Fig. 3d)[128]. Furthermore, by incorporating the moiré-trapped IXs into a device that enables an applied out-of-plane electric field, the large permanent dipole could be exploited to achieve ~40-meV tuning of the SPE emission energy via the DC Stark effect (Fig. 3e)[128]. This capability contrasts significantly with SPEs in monolayers which have very small permanent dipoles in the vertical direction. Last, recent works have shown that controlled electron and hole doping of the $MoSe_2/WSe_2$ heterobilayers results in the formation of negatively- and positively-charged IXs trapped in the moiré potential[131-133] enabling the charge control of the SPEs (Fig. 3f).

These results highlight unique possibilities to engineer highly ordered and uniform array of quantum dots with spin-valley optical selection rules[34,68], where the array of quantum dots (0D) can be tuned into parallel stripes of quantum wires (1D) by strain [134]. Moiré-induced trapping potentials loaded with one or two interlayer excitons[119] can behave as sources of single photons or entangled photon pairs, which can be exploited for applications. Furthermore, the phenomenon of spin-layer locking in TMD heterostructures provides few-level quantum systems in van der Waals heterostructures with a clear spin-photon interface via clean selection rules.

For example, the moiré-trapped IX reported in Ref. [127] create a "vee-type" three-level system. Alternatively, a three-level system analogous to a "spin-lambda" type atom, in which two ground states couple to a common excited state, can be engineered using the layer-pseudospin degree of freedom in a tri-layer heterostructure consisting of bilayer WSe_2/monolayer $MoSe_2$. Finally, the large out-of-plane permanent dipole of these excitons can be exploited to energetically tune the moiré quantum emitters into resonance with a cavity for Purcell enhancement for the generation of indistinguishable photons.

[H2] Hexagonal boron nitride

Bright room-temperature single photon emission in the visible has been reported in bulk crystalline and monolayer hBN[14,135]. SPEs in hBN can be very bright, with count rates exceeding a million counts per second and quantum efficiency as high as 87%[136]. Spectral emission covers a quite large wavelength range (1.6 – 2.5 eV [137,138]) with different classes of defects likely responsible for the emission[139] (Fig. 4a). Tuning of the optical emission wavelength can be achieved by electric fields[140,141] and strain[142]. While the identification of the emitters is not yet conclusive, several proposals have been investigated by ab-initio methods, including the anti-site nitrogen vacancy N_BV_N–[14], the carbon substitutional impurity at nitrogen site (C_N)[143] and the carbon anti-site (C_BV_N) [144] defects, and dangling bonds[145] (Fig. 4b). The latter proposal could possibly explain why these emitters are observed in as-grown material (where the high formation energy of other defects would limit their concentrations) and especially near crystal edges and boundaries, where the crystal lattice might be disrupted.

SPEs with hBN are typically activated with a plasma treatment or by annealing[14,135,146], but other methods, such as irradiation with electron beam[137,146], ultrafast laser pulses[146,147], and focused ion beam [148] have also been demonstrated. Recently, it has been shown that electron beam irradiation leads to the activation of SPEs in hBN with an improved accuracy of the spatial location and control of their emission wavelength [149]. As in the case of TMDs, it is possible to activate emitters by trapping carriers in the deformation potential wells created by inducing a local curvature by a nanopillar[19]. The variety of techniques available to create emitters is a useful resource to couple emitters to photonic structures. Transferring an hBN flake on plasmonic nanocavity arrays has been shown to enhance single-photon emission by a factor of ~ 2 [24,150]. It has also been shown that photonic crystal cavities can be fabricated directly into an hBN flake[151]. Having a SPE inside such a cavity with a spectrally matching mode could provide an emitter-cavity system completely embedded in a single hBN flake. In addition, single photon emission from a hBN flake has been successfully coupled to a tapered optical fibre with 10 % coupling efficiency,[152] which is comparable to the other SPE platforms[153-155]. Finally, another approach has been the integration of SPEs in multi-layer hBN into a tunable plano-concave optical microcavity, which has shown Purcell enhanced single-photon emission by a factor of ~4 at room temperature[156], compared to non-cavity SPEs (Fig. 4c). While hBN emitters are active up to room temperature, quantum interference experiments require a high degree of coherence, only available at cryogenic temperature. PL measurements on individual emitters at cryogenic temperature reveal a large Debye-Waller factor **[G]** (0.82). Low-temperature PL excitation experiments have revealed large inhomogeneous broadening, with emitters subject to blinking and spectral diffusion[157]. Switching between bright and dark states is observed under resonant or quasi-resonant excitation, with luminescence restored by a re-pump laser at a different wavelength. This switching can likely be explained as photo-switching between different charge states[145]. In the weak excitation limit, linewidths of ~0.6 GHz have been observed (lifetime limit ~160 MHz) with spectral diffusion in timescales of a few tens of millisecond[158]. Scans on timescales shorter than the spectral diffusion show transform-limited linewidths down to ~50 MHz[159], which, remarkably, persist as transform-limited up to room temperature[160]. Moreover, Rabi oscillations for a single emitter[158] and the coherent control of spin defects in hBN[70,161,162] have been recently shown, an important step towards coherent optical control of SPEs in 2D materials.

As is the case for other deep-level emitters in diamond and silicon carbide, the availability of optically-detectable electronic spins in hBN would open the way for quantum spintronic applications. The 2D geometry would, for example, enable easy integration in atomic force microscopy-type scanning probes and in heterostructures with increased functionalities. In this direction, a strong dependence of optical emission on the applied magnetic field at room temperature has been reported[69,70] (Fig. 4d). These properties are consistent with the presence of spin-selective intersystem crossing between triplet and singlet states, as for the nitrogen-vacancy

in diamond and for some intrinsic defects in SiC. More recently, a spin-dependent change in PL (optically-detected magnetic resonance) was reported for an ensemble of defects emitting at around 850 nm[70], different from those previously shown as single photon sources, in hBN samples irradiated by neutrons. The optically-detected magnetic resonance signal corresponds to a defect with D_{3h} symmetry, hosting an S=1 electronic spin ground state, with zero-field splitting D~3.5 GHz, and an almost isotropic g-factor g=2. Through electron paramagnetic resonance (EPR) measurements, the defect is identified as an intrinsic defect, the negatively-charged boron vacancy $V_B^{(-)}$ [163]. The zero-field splitting shows a quite strong temperature dependence (changing of ~120 MHz between room temperature and 4 K, as opposed to ~7 MHz for the NV centre in diamond), suggesting possible applications in quantum thermometry. One challenge with quantum spintronics in hBN is that all stable isotopes for both nitrogen and boron only feature non-zero nuclear spins. While the 2D geometry for a suspended monolayer considerably reduces the number of nuclear spins surrounding the central electronic spins, numerical simulations suggest spin coherence times T_2 limited to the microsecond scale[164]. Interestingly, decoherence is dominated by the contribution of ^{11}B nuclear spins, given their high gyromagnetic ratio (13.66 MHz/T, compared to 4.57 MHz/T for ^{10}B and 3.7 MHz/T for ^{14}N). Additionally, ^{11}B dominates the boron nuclear spin bath, with ~80% concentration of ^{11}B and ~20% concentration of ^{10}B. Numerical simulations predict that T_2 would increase from ~40 µs to ~90 µs when replacing all ^{11}B with ^{10}B (ref.[164]).

Despite the absence of transform-limited emission linewidths at room temperature, the bright and robust single-photon emission of hBN at room temperature makes hBN SPEs as promising candidates for quantum random number generation applications. The intrinsic 2D nature of the material can be harnessed to achieve coupling of the generated quantum random number stream to an on-chip photonic waveguide structure[165].

[H2] III-VI metal chalcogenides

In their bulk form, GaSe and InSe present bandgaps of ~ 2 eV and 1.25 eV, respectively but, by reducing the crystal thickness down to the monolayer limit, these can be increased substantially[166-168]. GaSe and InSe, widely used in the field of nonlinear optics and terahertz generation due to their high nonlinear coefficients, even at the atomic thickness limit[169], are also promising for electro-optical applications[170,171]. GaSe has been used to create SPEs with strain gradients (induced by local deformations created by naturally formed clusters of selenium atoms) which can host localized excitons and biexcitons[172]. Auto-correlation measurements **[G]** confirm the quantum nature of localized excitons, and cross-correlation measurements **[G]** reveal the correlation of the localized excitons and biexcitons. Further, GaSe SPEs have been successfully coupled to on-chip waveguides[28]. Narrow quantum dot-like optical emission has also been observed in InSe at low temperature over a wide range of energy, which was attributed to the localization of excitons around impurities and/or crystal defects[166]. However, the quantum nature of these localized excitons remains to be confirmed.

[H2] Graphene

Since graphene is a semimetal with no bandgap, it is a prominent material for broad-band photodetectors and modulators, but its light emission properties are fairly limited. Nevertheless, the fabrication of graphene quantum dots with atomic level control has been shown possible. These graphene quantum dots have been shown to emit single photons at room temperature with high purity ($g^{(2)}(0) = 0.05$) and brightness (count rate at saturation ~9.7 M counts/s)[173]. The graphene quantum dots consist of 96 carbon atoms with a lateral size of only 2 nm. During fabrication, one can introduce different functional groups to the edges of the graphene quantum dot to tune the emission wavelength [173,174]. The reported PL spectra of a graphene quantum dot and a graphene quantum dot functionalized with chlorine atoms show that the chlorine atoms also induce a redshift to the spectrum. This demonstration indicates the possibility of modifying physical properties (such as brightness, spin structure, and photostability) through the structure, for example, through designing quantum emitters with chemical engineering.

[H1] Materials Advances

Since the initial discoveries of SPEs in 2D materials, the field of 2D materials-based quantum emitters has grown a lot. This is illustrated in a timeline of different SPE material platforms (Fig. 5). However, the research field is still young and fundamental breakthroughs are still expected due to the unique nature of 2D materials (including new materials not yet investigated) and the ability to realize new types of heterostructures with pristine interfaces. For example, the 2D material family is still largely unexplored. The hunt for the new and optimal 2D materials, such as 2D materials with bandgaps close to the fibre telecommunication bands, like $MoTe_2$ and black phosphorus for quantum applications has just begun. In fact, a recent work has reported telecom band quantum emission in $MoTe_2$[175]. Other exciting avenues include new types of layered materials, for example, perovskite quantum dots [176-178].

The increasing theoretical understanding of 2D SPEs provides additional predictive power to aid the search for new SPEs in 2D materials[179]. In addition, the flexibility of the 2D material platform provides novel opportunities to engineer quantum confinement. The 'twist' degree of freedom for heterostructures is an exciting approach unique to this platform. However, while the theoretical description of moiré trapped excitons presented in Ref. [34] accounts for all the experimental signatures reported to date, the experimental results are still subject to debate. In this regard, clear identification of hybridization and moiré effects via resonant spectroscopy of intralayer excitons[125,180,181], the corresponding interlayer PL[35,36,127,128,134] and its interaction with strongly-correlated carriers [182,183], and new optical nanoscopy tools (like near-field scanning microscopy) can provide valuable insights. Further, while there is much excitement regarding the ability to engineer emergent properties of interacting particles in the moiré superlattice, the moiré trapped excitons may provide a means to locally probe or readout single spins in the dilute exciton lattice [132]. In addition to the Fermi-Hubbard model **[G]**, an array of moiré trapped excitons provides a novel platform to investigate superradiance or Bose-Hubbard **[G]** physics and topological collective excitations[184-186]. Other novel approaches to confine particles in 2D semiconductors include electrostatic gating of intralayer[187] or interlayer excitons[188,189]. Although reaching the quantum regime with electrostatic gating will be challenging, the large Coulomb interactions and dipolar repulsion energy for interlayer excitons[119,120] provide a possible route.

From a material perspective, significant advances in crystal growth, including epitaxial techniques, are required to move beyond mechanical exfoliation and manual stacking. Large-scale growth and cost-effective fabrication[190] of TMD monolayers, homo- and hetero-bilayers (with tunable interlayer twist angle[191]) and heterostructures have been realized for various electronic and photonic applications, and it is highly anticipated that the quality and size of 2D materials will be continuously improved at a fast pace. However, in contrast to many other photonic applications (such as photodetectors, modulators, lasers), the intrinsic quantum emission properties and performance of 2D materials based SPEs are more sensitive to structural disorders and environmental perturbations. Therefore, 2D SPEs require substantial more efforts (defect control and/or engineering) for scalable (CMOS compatible growth for on-chip devices) 2D material synthesis and device integration. Exfoliation and stacking are currently still essential for high quality optical devices. Recent advances in large-scale exfoliation[192], flake identification[193] and automated assembly[194] are promising for higher yield and more complex heterostructure devices.

H2 Purcell enhancement and integration.

An important route to successfully bring the performance of 2D SPEs on par with other quantum emitters[5], in particular quantum dots[8,195], is the realization of Purcell enhanced quantum light emission. First, Purcell-enhanced emission would mitigate the spectral diffusion induced inhomogeneous broadening, which is the main limiting factor for the generation of indistinguishable photons. Second, it would increase the collection efficiency and brightness of the 2D material single-photon source and its coupling into a specific fibre mode for long-range communication applications or into a photonic circuit waveguide for on-chip operations. Current approaches to transfer 2D materials on top of photonic waveguides unfortunately only result in coupling efficiencies into the photonic circuits below 10% [29,30,110]. The unique atomically thin nature of 2D materials

comes with advantages and disadvantages for cavity coupling. Compared to single atoms and ions, the 2D SPEs are embedded in a solid-state system; this embedding removes the need for active trapping in cavities, while still allowing for ultra-low cavity mode volumes, which is typically trickier for other platforms. Thanks to the straightforward transfer to other materials, 2D SPEs are easy to integrate into external cavities (Box 2). However, not being incorporated in a matrix also means that nanofabricating a monolithic cavity is more challenging compared to quantum dots.

As 2D SPEs can be simulated as a simple dipole, to a first order approximation, many different cavity coupling designs have been proposed and fabricated (Box 2). One possible approach is the integration of the 2D SPEs in plasmonic cavities[22,24], which in addition to the cavity coupling simultaneously act as the strain-induced nucleation site. Another approach is embedding 2D materials in tunable open cavities[25,103,105] (for example, photonic nanobeam cavities[196], whispering gallery-mode microcavities[197], and possible ring resonators[198] for photonic circuits integration[199]), which aim for high cavity quality factors instead of small mode confinement to realize their Purcell enhancement. One challenge in designing the ideal 2D SPE cavity system is to realize perfect mode overlap between the 2D SPE and the cavity mode. Typically the common transfer method to place the 2D material on top of the cavity[200] or waveguide[201] results in low coupling efficiencies. Additionally, for cavities with small mode volume, a precise lateral position of the 2D SPE is crucial. One possible avenue to success might be 2D SPEs generated by ion-bombardment or other methods with good emitter positioning accuracy in all three axes. Another challenge is the spectral overlap of the SPE and the cavity mode, since typical 2D SPEs have a broad ensemble PL. Therefore, additional tuning knobs such as electric field tuning are required, further complicating the cavity based 2D material device[202]. One approach particularly promising for 2D emitters which can overcome these challenges is the use of a broadband circular Bragg grating cavity[203]. In principle, it ultimately may become possible to integrate all three key quantum technologies (such as photon counters[204-206], linear and nonlinear photonic circuits[207], and single-photon sources) with hybrid 2D materials based platforms, for example with integrated waveguides (Box 2).

H1 Outlook

[H2] Coherence and spin-photon interfaces.

The most obvious applications of the scalable SPEs would be deployment in quantum technologies as either coherent single photon sources or spin-photon interfaces for quantum networking. However, significant challenges must be overcome, such as the generation of indistinguishable single photons from 2D SPEs. To date, Hong-Ou-Mandel interference **[G]** with even limited visibility at short delay times has yet to be reported with any 2D material host, and thus far, time-averaged spectra for all 2D SPEs reveal significant inhomogeneous broadening (typically > 10× the transform limit). In this regard, intrinsic or interface disorder is a formidable obstacle in 2D materials[208] as it can lead to dynamic environmental charging and thus SPE emission energy fluctuations. There are two main strategies to mitigate this: reducing the charge noise in the SPE environment or increasing the transform-limited linewidth by decreasing the emitter lifetime via Purcell enhancement. Both strategies are likely needed. For TMDs, samples encapsulated in hBN have been successfully used to achieve near transform-limited linewidths (meV-scale) for 2D free excitons[95,96,209], but similar efforts have yet to yield transform-limited linewidths for SPEs (μeV-scale), even with resonant excitation. Charge tunable devices, in which the Fermi level is pinned to stabilize the charge or all excess carriers are depleted[210,211], and higher quality intrinsic 2D materials with lower defect densities[212] are attractive options to reduce environmental charge noise in 2D SPE devices. The remaining dephasing mechanisms (like interactions with phonons and nuclear spins; Box 1) are material and emitter dependent. Once the spectral fluctuations are reduced to manageable levels, coherent control techniques developed for trapped atoms, III-V quantum dots, and defects in wide-bandgap materials such as microwave or all-optical spin echo techniques [213]can be applied to the 2D SPEs.

While coherent manipulation of spins associated with emitters in TMDs has not been yet reported, selection rules do provide possible pathways towards spin-photon interfacing. The presence of heavy elements like

transition metals leads to strong spin-orbit coupling. Spin-orbit coupling is typically expected to reduce spin relaxation timescales, by coupling the spin to orbital degrees of freedom (which is then less sensitive to phonons and electric fluctuations). Recent experiments[214] have however shown that this is not necessarily the case, and the full complex interplay between spin-orbit coupling and symmetry needs to be understood. In terms of nuclear spin baths, TMDs are quite promising. The 2D geometry in a free-standing monolayer features a spin bath only in a 2D plane. Additionally, the elements composing popular TMD materials (Mo, W, Se, S) comprise a high fraction of spin-zero isotopes. It has been shown in SiC that, for a multi-isotope bath, decoherence is reduced under an applied magnetic field since the Zeeman splitting associated with different gyromagnetic ratios prevents flip-flops for nuclear pairs in close vicinity[215]. This effect will play a role in reducing decoherence also in 2D materials, most of which feature two or more different isotopes. The combination of 2D geometry and multi-isotope baths with predominantly spin-zero isotopes creates a dilute spin bath, suggesting long dephasing time and possibly the capability to address individual nuclear spins.

Finally, similar to the case of self-assembled quantum dots, the discovery of biexcitons in WSe_2 quantum dots[73] (Fig. 5) offers some prospects towards polarisation-entangled photon pairs via the biexciton cascade.[216,217] However, two significant challenges need to be overcome. First, it is necessary to achieve a better deterministic control of the quantum dot formation. Despite the observation of biexciton formation in WSe_2 quantum dots by different groups[73-75], the majority of reported WSe_2 quantum dots typically do not show biexciton emission. In this sense, further efforts are required to understand the origin of biexciton formation in these quantum dots together with the corresponding improvement in the reproducibility of quantum dot creation. Second, the large FSS characteristic of WSe_2 quantum dots imposes additional limitations towards the creation of entangled-photon pairs. In principle, since the presence of a FSS only leads to a precession of the exciton phase, the degree of entanglement in the biexciton-cascade scheme is only determined by the temporal resolution with which the time delay between the emitted photon pair is measured[218]. Therefore, in theory, it should be possible to produce entangled photon pairs from the biexciton cascade in WSe_2 regardless of the non-vanishing FSS provided that the necessary temporal experimental resolution could be achieved. Unfortunately, the typical FSS values in WSe_2 quantum dots impose an experimental limitation regarding the required temporal resolution of the detectors. Consequently, further efforts are needed to pursue a FSS reduction by different approaches such as controlled electric [102] or strain tuning [107].

[H2] Quantum nonlinear optics in 2D materials.

2D materials exhibit extraordinarily large nonlinearities[169], which shows high promise for photon pair generation[219,220] (see Supplementary Information for an introduction). In principle, using atomically thin nonlinear materials removes the need for phase matching[169,221-224] to manifest several performance advantages compared to traditional bulk crystals. It also has been shown that second-order optical nonlinearity can be permitted in centrosymmetric 2D materials by interlayer twisting to break the symmetry[225-227]. Electrically tuneable second- and third-order nonlinearities have been demonstrated[228-234] indicating the possibility of electrically-tunable quantum sources for future quantum photonics[235]. Nevertheless, the nonlinear optical responses in 2D materials are typically limited by their atomic light-matter interaction length. Therefore, hybrid 2D materials[169,233](for example, with silicon based waveguides or silica based optical fibers[236-238], optical cavities[233,239], antennas[240], or metamaterials[241], exciton-[242] and phonon- polariton[243-245] hybrids) could be potential approaches to enhance the nonlinearity in 2D materials at the quantum level. Further, monolayer TMDs can exhibit near transform-limited 2D excitons. In this limit, an incoming laser resonant with the 2D exciton can be perfectly reflected, and this process can be tunable by gating the TMD to change the exciton resonance[246,247]. With the freedom to freely position the mirror-like atom sheet arbitrarily in front of a dielectric mirror[248-250], the nonlinear optical regime at the quantum level or extreme light-matter interaction may become possible[251]. Despite a lack of successful experimental demonstration[219,220], so far, 2D materials and their heterostructures based hybrid nonlinear optical systems make up an interesting platform for strong photon-photon interactions at

the few-photon level, potentially enabling a number of unique quantum photonic applications, such as generation[219,220,252], manipulation[253-256] and detection of quantum states.

[H2] Quantum sensing

The diversity of 2D materials provides exciting opportunities for investigations and applications exploiting spins and valleys, such as many-body spin physics (with high-fidelity electrical injection of polarized spins[257]), spintronics (by investigating Kondo-phenomena beyond metallic-like Kondo screening[258]), and quantum sensors (by optically polarizing and controlling individual spins associated with quantum emitters to map quantities of physical interest at the nanoscale). For some systems, in particular the nitrogen-vacancy centre in diamond, the electronic structure features an intersystem crossing that results in a difference in PL intensity for different ground-state spin levels (optically-detected magnetic resonance), even at room temperature. This capability has opened up opportunities to study nanoscale magnetism, biological systems, and nanoscale magnetic resonance imaging. The extension of this capability to a 2D geometry, with the discovery of optically-detected magnetic resonance in 2D materials, could open additional exciting opportunities, given the expected ease of integration of such sensors with scanning tips or directly into the surface of the sample to be studied.

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Key points

- 2D materials host quantum emitters with strong light-matter interaction that can be integrated into on-chip devices.
- Some 2D quantum emitters have an intrinsic spin-degree of freedom that can be harnessed for spin-photon entanglement.
- The ease with which 2D materials can be transferred onto photonic circuits to create hybrid devices provides new opportunities for scalable quantum photonic devices.
- While quantum emitters in several 2D materials have been successfully identified, there remain challenges to building functional quantum technologies.

Display Items

Figures

Figure 1: Single photon emitters (SPEs) in WSe_2. (a) Photoluminescence **(**PL) spectrum of SPEs in monolayer WSe_2. Left inset shows the localized emission with fine-structure splitting (FSS) and right inset shows the higher energy free exciton emission. **(b)** The polarization-dependent PL spectra (normalized) from bi-exciton (XX^0) and exciton (X^0) of a single SPE in WSe_2. The FSS and the out-of-phase orthogonal linear polarisations of the XX^0 and X^0 can be observed. **(c)** An optical micrograph (left) of WSe_2 with mono and bi-layer regions placed onto an array of nanopillars. A 2D spatial map of the integrated PL intensity reveals enhanced emission at the location of the nanopillars due to SPEs (right). The SPEs are brightest in the monolayer regions. **(d)** A schematic of atomically thin WSe_2 deformed by a nanopillar to achieve a point-like elastic strain perturbation (top), where the strain locally modulates the exciton energy (bottom). Optically created excitons can efficiently funnel to an individual strain tuned localized exciton trap (due to disorder or point defect) at the nanopillar centre resulting in a single highly efficient quantum emitter. **(e)** A reciprocal space schematic of 'dark' strain-localized exciton states (dashed lines) which hybridize with a point defect (horizontal cyan line), breaking the valley selectivity to lead to efficient photoemission (dark blue and yellow arrows). *K* and *K'* indicate the WSe_2 valleys, $|c_k\rangle$ and $|c_{k'}\rangle$ ($|\upsilon_k\rangle$ and $|\upsilon_{k'}\rangle$) are strain-localised electronic excitations originating from the conduction band (valence band) in each valley, and $|d_{\uparrow,\downarrow}\rangle$ refer to defect states and their spin degree of freedom. Panel a adapted with permission from REF. [10]. Panel b adapted with permission from REF. [73]. Panels c and d adapted with permission from REF. [17]. Panel e adapted with permission from REF. [71].

Figure 2: WSe_2 single photon emitters (SPEs) in heterostructure devices. (a) An optical microscope image of a transition metal dichalcogenide (TMD) based single photon LED. The dotted lines highlight the footprint of the single layer graphene (SLG), hBN and the TMD layers individually. The Cr/Au electrodes contact the graphene and TMD layers to provide an electrical bias. **(b)** Heterostructure band diagram, illustrating the case for zero-applied bias (top) and the case for a finite negative bias applied to the graphene (bottom). Tuning the graphene Fermi level across the TMD conduction band edge (E_C) allows electron tunnelling from the graphene layer to the TMD, resulting in light emission via radiative recombination of the electrons with the holes residing in the p-doped TMD layer. The appearance of valence-band holes below the Fermi level is due to the natural p-doping of WSe_2. **(c)** An example of layered LED emission spectra (PL, photoluminescence; EL, electroluminescence, L identifies the localised exciton emission), for an optically active layer of WSe_2. Top (bottom) spectra correspond to 10 K (room temperature (RT)) operation, where the black and blue spectra are obtained by optical excitation and electrical excitation, respectively. **(d)** Sketch of a charge-tunable WSe_2 SPE device. **(e)** Schematic representation of the electron (filled circles) and hole (open circles) tunnelling through the hBN barrier from the Fermi reservoir in the graphene to the quantum dots in WSe_2. **(f)** Coulomb blockade in a WSe_2 SPE shows voltage control of the charge state. The diagram in the inset represents the energy levels and occupation of the electrons and holes for the pure X^{1-}, X^0 and X^{1+} exciton states of the WSe_2 SPE. CB, conduction band; VB, valence band; $E_{g,}$ energy band gap; U_{ee} (U_{hh}), electron–electron (hole–hole) Coulomb interaction energy; QD, quantum dot. Panels a, b and c adapted with permission from REF. [31]. Panels d, e and f adapted with permission from REF. [32].

Figure 3. Moiré heterostructure based single photon emitters (SPEs). (a) Illustration of the long-period moiré superlattice formed in a transition metal dichalcogenide (TMD) heterobilayer with R-type stacking. The green diamond represents a moiré supercell. Insets are close-ups of three trapping sites (A, B, and C) with the corresponding atomic registries (R_h^h, R_h^X and R_h^M, respectively). **(b)** Schematic illustration of the polarisation selection rules, for K-valley interlayer excitons trapped at A, B and C trapping sites, respectively, for singlet (top

panel) and triplet (bottom panel) spin configurations. **(c)** Magnetic-field-dependent photoluminescence (PL) from interlayer excitons in an $MoSe_2/WSe_2$ heterobilayer with twist angles of 57° (left) and 2° (right). The top panel shows helicity-resolved PL spectra at 3 T. The excitation is linearly polarized, and the σ+ and σ− components of the PL are shown in red and blue, respectively. The bottom panels show plots of the total PL intensity as a function of the magnetic field, showing a linear Zeeman shift of the σ+ and σ− polarized PL. **(d)** Second-order photon correlation statistics of a moiré-trapped IX in a $MoSe_2/WSe_2$ heterobilayer, showing clear antibunching. **(e)** PL spectra of moiré-trapped IXs in a $MoSe_2/WSe_2$ heterobilayer embedded in a dual gate device for different applied electric fields, showing an energy tuning of ~ 40 meV. **(f)** Left: PL spectrum of moiré-trapped IXs in a $MoSe_2/WSe_2$ heterobilayer as a function of the applied gate voltage (V_g). The prominent IX emission peaks are labelled as A-H in each doping region. The dashed lines identify the V_g of the linecuts in the right panel. Right panel: Representative PL spectra for trapped IX^0, IX^-, and IX^+. For direct comparison with IX^0 PL spectrum, the IX^- (IX^+) PL spectra are shifted by 6.5 (6) meV in relative photon energy. Panel a adapted with permission from REF. [34]. Panel b adapted with permission from REF. [130]. Panel c adapted with permission from REF. [35]. Panels d and e adapted with permission from REF. [128]. Panel f adapted with permission from Ref. [132].

Figure 4. hBN based single photon emitters (SPEs). (a). Photoluminescence (PL) spectra of different SPEs in hBN **(b)** Illustration of the N_BV_N and C_BV_N crystalline defects. **(c)** Left: A schematic of a tunable plano-concave optical microcavity. Right: A second-order correlation function, g^2, of cavity-coupled and a free-space SPE in hBN. **(d)** Room temperature optically detected magnetic resonance frequencies from a negatively charged boron vacancy defect in hBN as a function of the magnetic field. Panel a adapted with permission from REF. [138]. Panel b adapted with permission from REFs. [14,144]. Panel c adapted with permission from REF [156]. Panel d adapted with permission from REF [70].

Figure 5. A development timeline of quantum photonics platforms including QDs,[51,53,259-269] NV centers in diamond,[270-277] WSe_2,[9-13,17,30,76,101] hBN,[14,30,32,73-75,151,158,161,278] Moiré[119,128,131-133], MoS_2,[93] $MoSe_2$[82] and $MoTe_2$ [175].

Boxes

Box 1: Intrinsic solid-state decoherence mechanisms

Electron-phonon interactions: Localized carriers in solid-state emitters interact with a many-body environment, leading to non-trivial modifications of the emitter's properties. The interaction of the trapped carrier with the surrounding lattice ions causes displacements of the latter from their equilibrium position, which in turn leads to absorption or emission of phonons (panel a). The equilibrium position of the crystal lattice depends on the local charge state due to the deformation potential and electron-phonon interaction. The two main contributions to the electron-phonon interaction are the deformation potential and the piezoelectric coupling to the long-wavelength acoustic phonons. A third type of electron-phonon coupling relevant for semiconductors is the polar coupling to longitudinal optical phonons, which give rise to a dipole polarisation field that interacts with the charge-carrier via the Fröhlich Hamiltonian[279]. These interactions affect several properties of 2D materials, including charge carrier mobility [280], resistivity [281], photon linewidth and lineshape dependence, and a temperature-dependent fraction of the photons emitted incoherently via the phonon sideband[282,283].

Electron-nuclear spin interactions: Despite the atomic thickness of 2D materials, the finite effective width of the exciton wavefunction along the axis perpendicular to the plane results not only in the Fröhlich interaction[284], but also in non-vanishing Fermi contact and dipole-dipole interactions between the trapped charge-carrier and the surrounding nuclei (panel b; red oval represents the wavefunction of trapped charge). A charge can be trapped (wavefunction represented by red oval) at the nanoscale due to intrinsic or extrinsic defects, such as transition metal or chalcogen vacancies (empty circles in the lattice) or dangling bonds in the substrate (represented by loops with positive charges), respectively. The localized wavefunction might extend across several lattice sites and into the encapsulating hBN layers, resulting in hyperfine interactions between the electron and encapsulating and substrate nuclei (depicted by randomly oriented arrows). The interactions of the entire nuclear ensemble with the trapped electron result in a fluctuating effective magnetic field, further limiting the coherence time of the central spin[285]. This fluctuating field can vary considerably from one material to another. Furthermore, adatoms, substitutional vacancies, and defects such as metal or chalcogen vacancies (which are abundant in TMDs[286-289]) provide a rich source of electronic spins that interact with the trapped charge carrier via the hyperfine interaction, resulting in an additional source of decoherence.

Box 2: Cavity integration and hybrid quantum photonic chips

Cavity Integration: Optical microcavities are devices capable of enhancing the light-matter interaction by concentrating the electromagnetic field at optical frequencies into sub-wavelength volumes. The inset in the figure shows a schematic of a single photon emitter (SPE) coupled to a cavity: a two-level emitter embedded inside an optical micro-resonator with optical losses κ couples to the optical mode of the cavity with a coupling strength g. As a result of the interaction, the emitter radiates with an enhanced spontaneous emission rate $\gamma_c = F_P\gamma_{sp}$, where γ_{sp} is the intrinsic spontaneous emission rate of the emitter and F_P is the Purcell factor, which is inversely proportional to the volume of the optical cavity mode, V. Finally, the spontaneous emission coupling factor (β) quantifies the fraction of the light emitted that is coupled into the cavity mode ($\beta = F_P/(F_P + 1)$).

The atomically-thin nature of the host crystals for 2D-based SPEs allows the integration of the SPEs with a large variety of optical cavity architectures with different properties (panel a). From left to right: engineered substrate, distributed-Bragg-reflector cavity, open dielectric cavity, plasmonic cavity, ridge waveguide, and a photonic crystal nanobeam. The colour bars show a qualitative comparison of the mode volume (V), Purcell enhancement factor (F_P), and spontaneous emission coupling factor (β) for each type of cavity. The integration of 2D SPEs in optical cavities offers potential to mitigate two of the main challenges of these emitters: SPE coherence and brightness. The enhanced radiative emission rate via the Purcell effect could simultaneously increase the photon brightness and enhance the spontaneous emission rate above the rate of dephasing processes responsible of the low coherence of these SPEs.

Quantum Photonic Integrated Circuits: For scalability reasons, most applications in quantum photonics require an on-chip solution where all components of the device are integrated on a photonic circuit (panel b). The functionality of a photonic circuit and its performance can be reconfigured using electric contacts (gold) to control active elements (like tunable ring resonators and modulators). After different gate operations and routing, the photons are detected using on-chip single-photon detectors (on the far right). Fundamental building blocks for quantum photonic circuits are quantum light sources, photonic circuits, and single photon detectors. Finding a single material platform where all building blocks are monolithically integrated with state-of-the-art

performance of each component remains challenging. Another route is to take a well-developed circuit platform, such as silicon-based photonics, and deterministically integrate quantum emitters using a transfer method[290-296] Such a heterogeneous approach enables the integration of all required building blocks on a single chip[297,298], and perfectly matches with the pick and place exfoliation technique used for 2D materials. After a monolayer is isolated on a transfer stamp, it can be precisely placed on a photonic circuit, and first steps have already been taken[26-30,110]. Given the hybrid approach, there is no restriction on which material platform can be used. Different platforms offer different advantages and challenges, and an overview can be found in Ref.[299]. Current preferred choices are SiN[300], AlN[301], and $LiNbO_3$[302].

Glossary

Spectral fluctuations: Changes in the energy levels of an emitter due to electrostatic noise in the emitter's environment.

Blinking: Random switching of bright and dark states of the emitter.

Transform limit: The ideal coherence limit: $T_2 = 2T_1$, where T_2 and T_1 are the emitter's coherence time and lifetime, respectively

Fine-structure splitting (FSS): Splitting of exciton energy levels caused by spin interactions and / or wavefunction asymmetry.

Dark exciton: In dark exciton the spins of the electron and hole are parallel and spontaneous emission is forbidden due to spin momentum conservation.

DC stark tuning: Tuning of emission spectra using an external electric field.

Plasmonic cavities: Cavity where the light is enhanced by the interaction of surface plasmons

Purcell enhancement: Environmental enhancement of light emission rate of a quantum system. Typically caused by a resonant cavity.

Slot waveguide: A waveguide, where light is confined between two slabs of high-refractive index materials.

Interlayer excitons: Electron-hole Coulomb bound states between electrons and holes spatially separated in different monolayers

Debye-Waller factor: Describes the magnitude of thermal vibrations in a crystalline lattice and is used as a measure for structural disorder of material.

Auto-correlation measurements: Second-order correlation measurement used to measure the time delay between two successive photons.

Cross-correlation measurements: Correlation measurement of two different signals

Fermi-Hubbard model: Interaction model of fermions in a lattice.

Bose-Hubbard: Interaction model of Bosons on a lattice

Hong-Ou-Mandel interference: This bosonic interference describes the situation where two photons approach a 50/50 beam splitter from different input ports. If the photons are indistinguishable and they enter the beam splitter at the same time, both photons will exit together in a superposition from the output ports of the beam splitter.

Website summary

Quantum photonics offers an integrated and scalable approach to quantum information processing and communication. This article summarises the state-of-the-art and an outlook on future challenges and opportunities for quantum photonics based on 2D materials.