Quantum emitters in 2D materials: Emitter engineering, photophysics, and integration in photonic nanostructures

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ABSTRACT

Quantum emitters have become a vital tool for both fundamental science and emerging technologies. In recent years, the focus in the field has shifted to exploration and identification of new quantum systems enabled by the emerging library of atomically thin, two dimensional materials. In this review, we highlight the current state of the art in engineering of quantum emitters in 2D systems, with an emphasis on transition metal dichalcogenides (TMDCs) and hexagonal boron nitride. We start by reviewing progress in TMDCs, with focus on emitter engineering, ability to tune their spectral properties, and observation of interlayer excitons. We then discuss emitters in hBN and focus on emitters' origin, engineering, and emerging phenomena—spanning super-resolution imaging and optical spin readout. We summarize by discussing practical advances of integration of emitters in 2D hosts with plasmonic and dielectric photonic cavities, underpinned by quantum light–matter interactions. We conclude by outlining pathways for practical on-chip quantum photonics applications and highlight challenges and opportunities within this field of research.

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I. INTRODUCTION

Quantum technologies and quantum information science are becoming a mature research field with numerous commercial applications.[1,2] The interest in quantum technologies stems from the ever-growing promise of devices that are not possible via traditional miniaturization of microelectronic components and acceleration of performance. The most common example is quantum key distribution as a means to absolute secure communication, protected by the laws of quantum physics.[3] However, more advanced applications, including quantum optical neural networks, remote quantum sensing, and quantum bio-imaging, have also been proposed and are garnering increasing attention. On the fundamental side, photon-mediated interactions at a single photon level are also intriguing and may eventually lead to new quantum computation and quantum simulation protocols.

Development of quantum infrastructure and, more specifically, physical qubits, has been pursued globally for almost half a century. It is therefore not surprising that some systems—such as superconducting qubits and trapped ions—reached maturity and currently underpin a growing quantum industry.[4,5] Looking ahead, scalable connectivity of quantum nodes by photons is now recognized as a promising next step. In this context, integrated quantum photonics is an emerging field that aims to interface stationary qubits, which can store quantum information and have long coherence times, with flying qubits, such as photons.

A key building block of a quantum photonic network is a single photon source—a source that emits only one photon per excitation cycle. Over the last decade, the field of solid-state sources has exploded, and there are presently dozens of systems with their individual advantages and limitations. In the context of this review, most quantum emitters can be broadly classified into two categories: (1) excitonic emitters in which the quantum emission occurs through a near-bandgap transition and (2) defect-based emitters which have deep states and behave as "artificial atoms" in foreign hosts. The former include quantum dots (QDs), emitters in carbon nanotubes (CNTs), and single molecules. On the other hand, the defects are typically impurities in wide bandgap semiconductors. The most common examples include color centers in diamond, silicon carbide, and hexagonal boron nitride.

Until approximately five years ago, this field of research was focused predominantly on quantum emitter in 3D solid state hosts,[6] and it is fair to say that it was approaching saturation—in part due to a lack of novel experiments and in part due to fundamental physical and technological limitations. However, the emergence of the popularity of two dimensional (2D) materials has led to the nearly simultaneous discovery of quantum emitters in a range of 2D hosts, particularly transition metal dichalcogenides and hexagonal boron nitride. This sparked renewed interest in the field of solid-state quantum systems, and as a result, explorations of quantum emitters in 2D hosts[7] and their integration in nanophotonic devices is constantly and rapidly evolving. In this review, we summarize the current state of quantum emitters in 2D materials with a focus on engineering of these systems, explorations of unique photophysical phenomena, and the latest demonstrations of quantum light–matter interaction that have been enabled by these materials.

II. QUANTUM EMITTERS IN TMDCS

The discovery of graphene[8] has sparked research in other groups of two dimensional materials,[9] such as transition metal dichalcogenides (TMDCs) and oxides.[10–13] TMDCs have the form MX_2 (M = transition metal atom and X = chalcogen atom), whereby the M and X atoms are covalently bonded to form an X–M–X sandwich structure. These materials are promising for optoelectronics due to their unique optical properties and have been incorporated in transistors, photodetectors, light emitting devices, and solar cells.

The discovery of single photon emitters (SPEs) in TMDCs was reported concurrently in four papers in 2015.[14–18] Specifically, photoluminescence (PL) emissions at energies 20–100 meV below the intrinsic excitonic emission were observed from monolayer WSe_2 at cryogenic temperature. The emissions exhibit antibunching, linear polarization, narrow linewidths (~0.1 to 0.7 meV), and lifetimes on the order of 1 ns. Subsequent research has been focused predominantly on two TMDCs—WSe_2[14–17] and MoS_2[19]—although emitters in WS_2,[20] GaSe,[21] and $MoSe_2$[22] have also been reported. The structural origins of these QD-like emitters are a topic of debate. Two theories have been put forward: (a) excitons localized by strain or (b) a defect-to-bound transition, with the defect being a vacancy. The role of strain has been investigated primarily using WSe_2[18,23–27] [Fig. 1(a)], a vacancy alternative has been discussed mainly in the context of MoS_2,[19,28] and an interplay between strain and defects has been proposed in both experimental and theoretical studies.[27,29]

A. Engineering of quantum emitters in TMDCs

The role of strain was originally proposed by Kumar *et al.* and Tonndorf *et al.*[18,23,27,30] They observed SPEs at edges of WSe_2 flakes and at the locations of pillars located underneath WSe_2. They also generated SPEs using an AFM tip that was used to scratch WSe_2 monolayers.[18]

Periodic strain fields can be used to create SPE arrays, which are appealing for scalability and applications. Two approaches have been demonstrated with nanopatterned substrates. In the first, an array of nanopillars is used to induce strain and generate an ordered array of SPEs in a TMDC overlayer.[20] Alternatively, the substrate contains an array of indents that generates the strain field, as is illustrated in Fig. 1(a).[22] Recent results also suggested that electron irradiation of WSe_2 monolayers on top of predefined pillar sites also aids in deterministic emitter creation. Interestingly, these experiments also suggested that

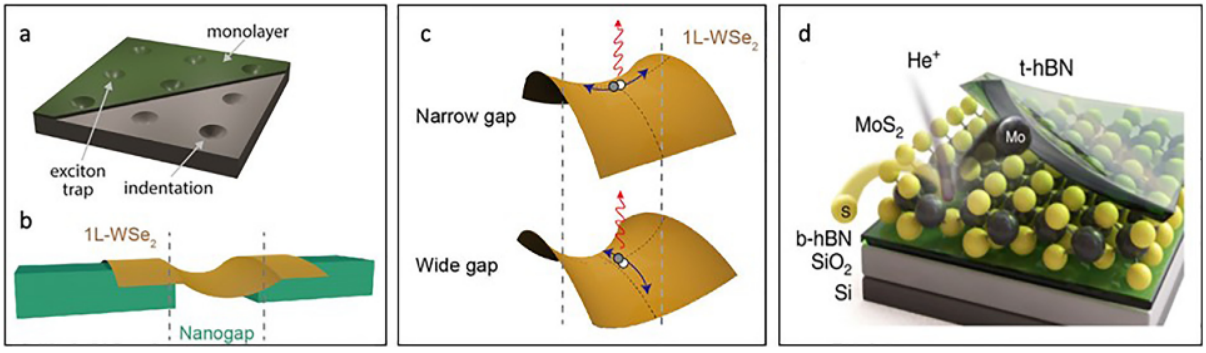

FIG. 1. Engineering of SPEs in TMDCs by strain and ion irradiation. (a) Array of indents in a substrate used to confine excitons and thus generate an array of SPEs in monolayer $MoSe_2$. Reproduced with permission from Yu *et al.*, Nano Lett. **21**, 2376 (2021). Copyright 2021 American Chemical Society. (b) Nanogaps in nanorods used to confine excitons in WSe_2. (c) The nanogap size affects the elongation direction of the WSe_2 and the polarization of non-classic light emitted by the trapped excitons. Reproduced with permission from So *et al.*, Nano Lett. **21**, 1546 (2021). Copyright 2021 American Chemical Society. (d) Defect engineering in MoS_2 by He+ ion irradiation (the MoS_2 was subsequently capped with hBN to stabilize SPEs). Reproduced with permission from Klein *et al.*, Nat. Commun. **10**(1), 2755 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license.

the emitters are optically active up to 150 K, a promising route to operation at liquid nitrogen temperature.[31]

Strain gradients can be used to control not only the emitter locations, but also the emission polarization.[32] This is illustrated in Figs. 1(b) and 1(c) where a gap in a nanorod generates a strain gradient that localizes excitons in a WSe_2 overlayer, and the gap size determines the shape of the strain field and the polarization orientation of the linearly polarized SPE.[26] Similarly, the emission energy can also be tuned by strain, as has been demonstrated using a piezoelectric substrate, which is appealing for on-chip applications of SPEs.[32] Alternatively, stark tuning was also used to tune the emission linewidths of quantum emitters in TMDCs.[33]

A notable alternative to strain for engineering of SPEs in TMDCs is ion irradiation using a focused helium ion beam. This approach has been used to particularly good effect instead of MoS_2 [Fig. 1(d)],[19] where post-irradiation capping of the MoS_2 with hBN flakes has been shown to stabilize the fabricated SPEs and produce arrays.[34] Nevertheless, spectral diffusion of these SPEs is significant, likely due to localized charging of defects generated by the ion beam. Indeed, ion irradiation generates a range of defect species, some of which are optically active, whilst others do not luminesce, but modify the local dielectric environment and can modulate emissions from SPEs. The level of complexity associated with restructuring of materials by ion beams is substantial, and the atomic structure of SPEs generated in MoS_2 is a topic of debate. Several defects have been suggested or observed in post-irradiated MoS_2—including Mo vacancies, S vacancies and defect complexes that include O impurities—but a conclusive correlation between SPE emissions and atomic structure remains to be done.

The optical properties of quantum emitters in TMDCs are still being investigated. Recently, infrared emissions (in the telecom O and C bands, ∼1300 and ∼1550 nm, respectively) have been reported from strained $MoTe_2$.[35] The purity and brightness of the emitters are not optimized, but evidence of telecom emissions from TMDCs is promising for future integration with silicon photonics. Similarly, a number of studies have reported cascade emission[31,36] from localized emitters in WSe_2. These results are important not only to unambiguously prove the presence of bi-excitons, but also for a demonstration of entangled photon pairs from these materials.

The advantages of quantum emitters in TMDCs are exemplified by studies of emission modulation and electrical excitaiton.[37] In a recent work, strain engineering and electrical excitation were combined. Emitters were created deterministically in a graphene–WSe_2–hBN heterostructure using an atomic force microscope (AFM), and the strain-activated emitters were excited electrically [Fig. 2(a)].[38] This device geometry is unique to 2D systems and different from conventional lateral junction-based devices. In a similar device, gate-switchable quantum emitters were also realized with defects in MoS_2.[39]

To controllably modulate emitters in TMDCs, piezo elements can be employed. Figure 2(b) shows an example of a piezoelectric

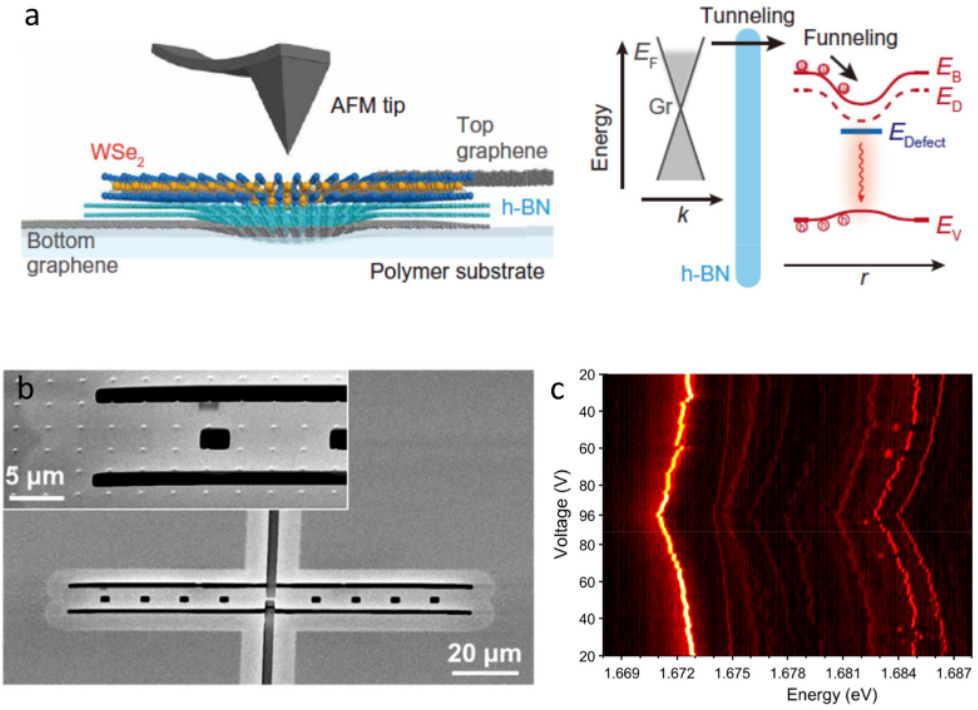

FIG. 2. Combined strain-and-electrical control of quantum emitters in TMDCs. (a). Deterministic generation of quantum emitters in WSe_2 by AFM indentation and subsequent electrical excitation of the device. Graphene serves as a top electrode, as is illustrated in the top-right panel. Reproduced with permission from So *et al.*, Sci. Adv. **7**(43), eabj3176 (2021). Copyright 2021 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (b) SEM image of a piezoelectric microcantilever. The inset shows a close-up of nanopyramid arrays that generate strain-induced quantum emitters in WSe_2. (c) PL spectra vs bias from WSe_2 positioned on top of the microactuator. Reproduced with permission from Kim *et al.*, Nano Lett. **19**, 7534 (2019). Copyright 2019 American Chemical Society.

microactuator with integrated nanopyramid arrays.[40] The actuator consists of two cantilevers connected by a thin tether, and the nanopyramid arrays are used to induce strain in WSe_2. Under voltage, the microcantilever modulates the amount of strain and hence the emission energy of strain-localized emitters in WSe_2. The emission energy varies in the range of 1 to 3 meV, which is in principle sufficient to bring two distinct emitters into resonance.

B. Interlayer excitons (IEs)

Layered 2D heterostructures[41] have emerged as a promising platform for engineering of quantum emitters[12,42] based on interlayer excitons (IEs)[43–45] that form at interfaces between 2D materials. IEs consist of electron–hole pairs donated by adjacent monolayers.

Different combinations of 2D materials and layer stacking angles provide new degrees of freedom and a plethora of new systems for studies of quantum emitters. This is exemplified by monolayer TMDCs which emit in the visible range and can be combined to generate IEs that emit in the infrared spectral range. Karni *et al.* reported a PL emission at ∼1 eV from MoS_2/WSe_2 heterobilayers, as shown in Fig. 3(a).[46] The recombination energy of these IEs can be tuned by up to 80 meV using an out-of-plane electric field. The tunability together with the temperature-dependence of the PL intensity indicate the IE origin of the emission.

IEs can be confined using strain,[47] electric fields,[48] and Moiré[49] potentials. Montblanch *et al.* localized IEs in WS_2/WSe_2 heterobilayers using a pillar array, as is shown in Fig. 3(b). Emissions from the confined IEs are more intense, vary with applied magnetic fields, saturate with excitation power and have lifetimes substantially shorter than the delocalized IE counterparts—all due to confinement by the strain field.[47] IEs can be manipulated further by pressure engineering, which can be used to alter TMDC band structures, as has been demonstrated for WSe_2–$MoSe_2$ heterostructures.[50]

Site-selective trapping of IEs has also been achieved using electric fields. Figure 3(c) shows a potential well generated in an $MoSe_2$–WSe_2 heterostructure by a bias applied to a top-electrode that is made from few-layer graphene and contains a 30 nm hole. The resulting highly

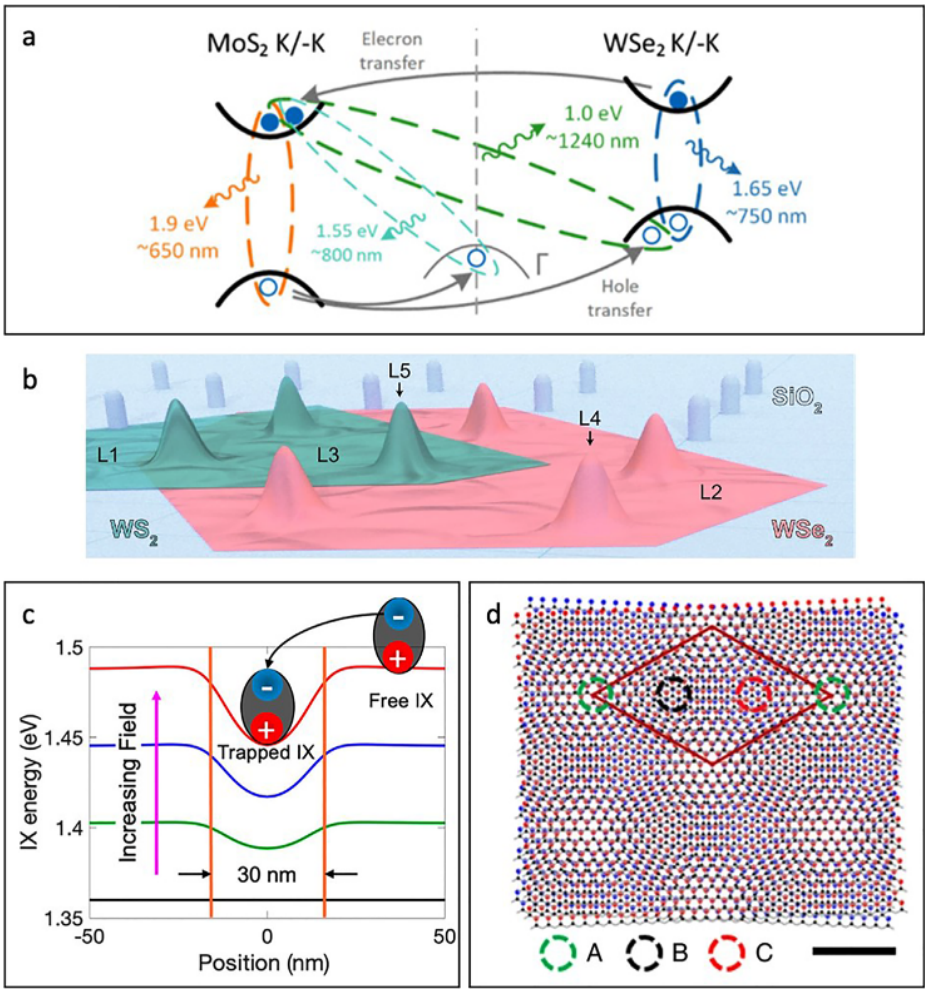

FIG. 3. Interlayer excitons and moiré excitons in TMDCs. (a) Band alignment in a MoS_2/WSe_2 heterostructure. The arrows show charge transfer processes that lead to the formation of interlayer excitons. Reproduced with permission from Karni *et al.*, Phys. Rev. Lett. **123**, 247402 (2019). Copyright 2019 American Physical Society. (b) WS_2/WSe_2 heterostructure atop a SiO_2 pillar array. L1–L5 are representative locations discussed in, and Reproduced with permission from Montblanch *et al.*, Commun. Phys. **4**, 119 (2021). Copyright 2021 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (c) Trapping of interlayer excitons in a potential well generated by a highly localized electric field applied to a $MoSe_2$–WSe_2 heterostructure. Reproduced with permission from Shanks *et al.*, Nano Lett. **21**, 5641 (2021). Copyright 2021 American Chemical Society. (d) Moiré superlattice formed in a $WSe_2/MoSe_2$ heterobilayer with a twist angle of 5°. Scale bar, 2 nm. Reproduced with permission from Brotons-Gisbert *et al.*, Nat. Mater. **19**, 630 (2020). Copyright 2020 Nature Publishing Group.

localized electric field can confine interlayer excitons, whilst IEs under the graphene sheet are suppressed.[48,51] Dipolar interactions between localized IEs were studied using a dynamic electric field by Li *et al.*[52]—these can induce effective photon–photon interactions and repulsive dipolar interactions can potentially lead to exotic many-body states such as self-assembled dipolar crystals with a spin-valley degree of freedom.[53]

More intriguingly, moiré potentials in TMDs can be used to manipulate their electronic and optical properties. Moiré potentials [Fig. 3(d)][54] are generated in layered materials by periodic variations in lattice mismatch and rotation angle between adjacent monolayers.[55] Moiré potentials give rise to moiré-trapped IEs,[54] including valley excitons[56] and quantum emitters. As has been demonstrated by Baek *et al.*, moiré-trapped IEs from a $MoSe_2/WSe_2$ heterobilayer emit at a number of energies, ranging from 1.38 to 1.40 eV; all of these emissions are circularly polarized and split upon application of a magnetic field via the Zeeman effect. These IEs are also tunable by a DC electric field and exhibit photon antibunching with a lifetime of ∼12ns.[49] In short, these recent works illustrate that moiré superlattices provide a new platform for exploring quantum phenomena in 2D heterostructures, with exciting potential applications in nanophotonics and quantum information processing.

The above-mentioned works have made substantial progress in engineering of SPEs in TMDC monolayers and heterostructures, and provide deep insights into the origins of the SPEs and offer pathways to engineer 2D systems for future quantum information applications. There are a few examples of integration of these emitters with plasmonic nanostructures and waveguides.[24,57] As is discussed below, strain localization has been used to develop a number of SPE fabrication methods which provide novel options for integrating SPEs with photonic devices. Focused ion beams provide an approach to create SPEs in 2D materials that is compelling for integration with photonic devices because they retain their layered properties (i.e., flatness). The emerging field of interlayer excitons provides a vast number of options for exploring quantum properties using currently available 2D materials. Nevertheless, there are still challenges for optimizing these SPEs. For example, one concern is stability (i.e., shelf time) of the SPEs—it is essential for long-term applications and needs to be investigated in detail. Other disadvantages, which are also seen in other solid state systems are emission intermittency and spectral wandering. Finally, lifetime-limited linewidths, which are essential for many quantum applications, remain to be demonstrated.

III. QUANTUM EMITTERS IN hBN

A. Emitter engineering

Since the discovery of quantum emitters in hBN,[58] many methods have been developed to create emitters or "activate" defects in hBN,[59–63] whereby "activation" refers to modification of a defect or its enclosing environment so as to make it fluorescent. While the original works[59,61–63] reported methods such as annealing that activate emitters at random locations throughout hBN flakes, deterministic creation of defects both spatially and spectrally has been the focus of recent research. This is indeed a critical challenge in the case of hBN SPEs as the emission energy varies from the UV[64] to the near IR[65] spectral range, and emitters in as-grown material are located primarily at extended defects.[66] Engineering of quantum emitters in a relatively narrow window of the visible spectrum has been achieved by bottom-up growth of hBN[67,68]—the center ZPL emission wavelength can be narrowed down to 580 ± 10 nm by CVD growth of multilayer hBN.[67] Creation of emitters during growth of hBN on copper is grain dependent and emitters usually appear on ⟨100⟩ oriented crystals.[68] It is also possible to tune the center emission wavelength by controlling the growth conditions. For instance, the presence of nickel as a supporting substrate for copper foil has resulted in a 50 nm shift in the ZPL center emission wavelength of emitters in CVD grown hBN.[69] In addition, a rigorous study of hBN films grown by various methods has revealed the role of carbon in quantum emitters in hBN as shown in Fig. 4(a). The result was confirmed using direct implantation of carbon into hBN[70] though control over the spatial position of these emitters during implantation has remained elusive. Density functional theory (DFT) calculations suggest a complex defect, and negatively charged $V_BC_N^-$ could be the source of emission;[70] however, an extensive DFT effort from the community also suggested other carbon-related defects.[71–78]

Positional control of quantum emitters has been overcome by CVD growth onto a substrate of silicon oxide nanopillars [Fig. 4(b)], yielding a near-unity success rate in deterministic fabrication of emitter arrays in hBN.[79] Engineering of quantum emitters in hBN has also been achieved by post-growth approaches such as plasma processing.[61,62,80] Spatial control over the position of quantum emitters was demonstrated using the electron beam of an electron microscope which resulted in activation of single photon emitters with a ZPL emission energy of ∼3 eV (∼435 nm), as is shown in Fig. 4(c).[81]

Some studies have taken advantage of damage sites created by mechanical indentation[82] or ion milling[83] to localize the emitters to the defective regions. Both processes yielded a success rate of ∼30% in creation of quantum emitters. Fabrication of patterned 2D heterostructures provides another avenue for localization of emitters in hBN. Graphene has been shown to suppress emission of quantum emitters in hBN.[84] By placing patterned graphene on top of three-layered hBN, localization of emitters has been demonstrated. Specifically, every opening within graphene resulted in an active emitter in the underlying hBN. A schematic of this structure is shown in Fig. 4(d).[85] An outstanding challenge is to find specific crystallographic structures of defects and develop techniques for engineering of emitters using ion implantation methods and by introducing impurities during growth.

B. Spin defects in hBN

The interface between optical emission and electron spin state underpins a class of defects known as spin defects such as the NV center in diamond[86] and vacancy complexes in silicon carbide.[87] These defects are highly sought after for the realization of scalable quantum information technologies.[88,89] In hBN, a group of spin defects has been reported to exhibit optically detected magnetic resonance (ODMR) at room temperature.[90,91] Negatively charged boron vacancy (V_B^-) centers were the first discovered hBN defect with optically addressable spin. The defect is a spin 1 system with a ground state triplet separated by 3.47 GHz (Dgs) and 50 MHz (Egs) as is shown in Fig. 5(a). Figure 5(b) shows ODMR contrast from V_B^- which is split further in the presence of an external magnetic field due to Zeeman splitting of $M_S = \pm 1$ sublevels in the ground state. The interaction of electron spin in V_B^- with three nitrogen nuclei results in the appearance of hyperfine structure with a splitting of ∼47 MHz, which indicates the presence of three nitrogen nuclei surrounding the defect [Fig. 5(c)]. The creation of these defects via ion implantation independent of the

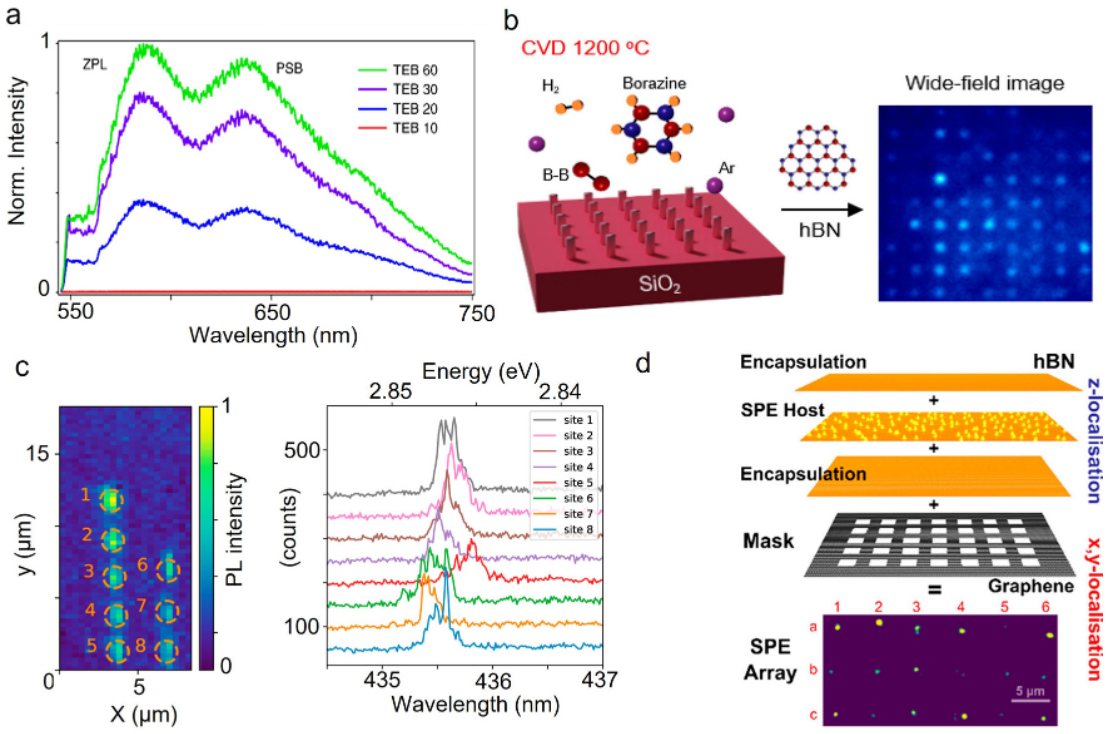

FIG. 4. Deterministic creation of hBN quantum emitters. (a) Photoluminescence emission of carbon-related defects as the carbon content is increased during growth. Reproduced with permission from Mendelson *et al.*, Nat. Mater. **20**, 321 (2021). Copyright 2021 Nature Publishing Group. (b) Localization of emitters during growth to an array of silicon oxide nanopillars. Reproduced with permission from Li *et al.*, Nano Lett. **21**, 3626 (2021). Copyright 2021 American Chemical Society. (c) Deterministic creation of emitters in hBN with an emission wavelength of ∼435 nm using a focused electron beam. Both spatial (left panel) and spectral (right panel) control have been demonstrated. Reproduced with permission from Fournier *et al.*, Nat. Commun. **12**, 3779 (2021). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (d) Localization of emitters in hBN by fabrication of a patterned 2D hetrostructure. Reproduced with permission from Stewart *et al.*, ACS Nano **15**, 13591 (2021). Copyright 2021 American Chemical Society.

ion species together with theoretical calculations confirm the defect structure to be the negatively charged boron vacancy.[92–94] A drawback of these defects is a relatively low quantum efficiency, and hence low brightness, which hinders isolation of a single defect. Consequently, all studies performed to date have been focused on ensembles of V_B^- centers.

Several works have studied spin dephasing of V_B^- centers. The spin lattice relaxation time (T_1) for V_B^- is ∼8 μs, which is independent of external magnetic field due to a strong local field around the V_B^-.[95,96] This value increases to 12.5 ms upon cooling to cryogenic temperature. The spin–spin coherence time (T_2) is ∼2 μs at 300 K and at cryogenic temperatures, while the spin-dephasing time T_2^* is ∼100 ns. It has been shown that the electron spin is mainly affected by the nuclear spins in the far distant coordination shell from the vacancy, but the exact decoherence mechanism has remained elusive to date.[95] V_B^- is a potential candidate for high sensitivity measurements as it can be created near the surface due to the 2D nature of Hbn.[97] The zero field splitting of the V_B^- shifts by more than 300 MHz over the temperature range of 5–600 K.[98] While Dgs is not affected by the magnetic field, the shift with temperature is relatively large compared to known spin systems such as the NV center in diamond. Moreover, the increase in Dgs with temperature is not linear and can be explained by considering the relative change of a and c (η_a and η_c) lattice parameters of hBN with temperature,

$$D_{gs}(T) = D_{gs}(295\,\mathrm{K}) + \theta_a \eta_a(T)h + \theta_c \eta_c(T)h. \quad (1)$$

Considering the possibility of creation of V_B^- in atomically thin hBN and the observation of more than 40% contrast in ODMR,[99] improved sensitivity using V_B^- spin levels is a compelling milestone in this field.

ODMR signatures have also been reported for other defects in hBN, specifically for single photon emitters.[91,100] Ensembles of carbon-related defects show a weak singlet ODMR peak [Fig. 5(d)] which is attributed to the presence of both negative and positive ODMR for single emitters, as is shown in Fig. 5(e).[70,91] ODMR spectra from a single defect consist of an asymmetric peak with a contrast of up to ∼6%. The double resonance measured from these defects is split by ∼35 MHz, and it is unclear whether it is caused by zero field splitting or hyperfine coupling to nuclei. Similarly, paramagnetic single emitters have been reported for groups of emitters with a ZPL in the range of 700–750 nm.[100] The ODMR signature is similar to that of carbon-related defects but hyperfine splitting or quadrupolar coupling

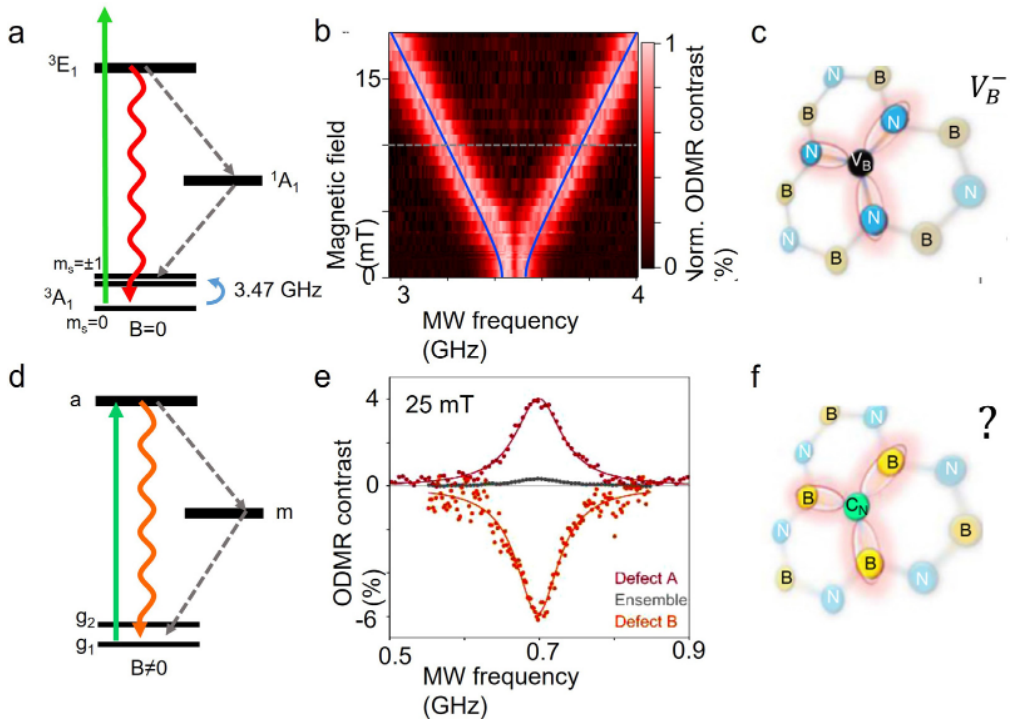

FIG. 5. Spin defects in hBN. (a) Electronic level structure of negatively charged boron vacancies with a zero field splitting of 3.47 GHz at room temperature. (b) ODMR measurement of V_B^- in hBN with Zeeman splitting of its ground state in the presence of an external magnetic field. Reproduced with permission from Gottscholl *et al.*, Nat. Mater. **19**, 540 (2020). Copyright 2020 Nature Publishing Group. (c) Boron vacancy defect in the hBN lattice. (d) Electronic level structure of a quantum emitter in hBN with a doublet ground state. (e) ODMR spectra from these defects can be both positive and negative, resulting in very small contrast when measuring ensembles. Reproduced with permission from Stern *et al.*, arXiv:2103.16494 (2021). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (f) Carbon-related defects have been suggested as possible candidates, but the structural origin for the defect is unknown. Reproduced with permission from Chejanovsky *et al.*, Nat. Mater. **20**, 1079 (2021). Copyright 2021 Nature Publishing Group.

was not observed in this case. The origin of the defect has remained elusive since the experimental data did not match theoretical calculations of possible carbon-related defects [Fig. 5(f)]. Experimental identification of these defects through hyperfine or quadrupolar coupling and the true spin level structure of these defects remain a challenge for future experiments.

C. Photophysics of hBN emitters

Since the discovery of quantum emitters in hBN, many researchers have investigated the optical properties of these defects, including the emission linewidth,[101] as well as stark shift[102–104] and dephasing processes. In general, a strong interaction between emitters in hBN and the underlying substrate or local charge fluctuations within the materials have been reported in multiple studies.[105–108] The first cryogenic study of these emitters under resonant excitation revealed a linewidth of ∼1 GHz, and substantial blinking and spectral diffusion.[109] Subsequent studies reported similar linewidth and confirmed dephasing due to both phonon coupling and spectral diffusion.[110,111] However, Fourier-limited linewidths have, nonetheless, been observed in some hBN emitters,[112,113] which persists even at room temperature.[114] These results have been attributed to complete decoupling of emitters from in-plane phonon modes and the two dimensional nature of hBN.[114,115]

A range of photophysical properties have been reported for various emitters in hBN, indicating that a number of distinct defect species play a role in the reported behaviors. For example, it has been argued that multiple states are involved in the excitation or relaxation of some hBN emitters.[116–119] The role of more than two electronic levels was hypothesized to explain an increase in the autocorrelation function intensity at zero delay time. This was backed further by the presence of two closely spaced phonon sideband lines as well as a double-exponential decay function.[117] Furthermore, cross correlation measurements of two adjacent zero phonon lines confirmed the presence of photochromisms in some hBN emitters, as is shown in Fig. 6(a).[116] Control over the charge switching states was demonstrated by illumination with a higher energy laser,[120] and it was shown [Fig. 6(b)] that co-excitation with a high energy laser can be used to switch controllably quantum emitters in hBN.[118]

Some of the observed photophysical properties have been exploited for super-resolution imaging—a repumping (i.e., co-excitation) method was used to achieve a spatial resolution better than 100 nm in super-resolution imaging of quantum emitters using a ground-state depletion imaging scheme.[120] More recently, the phonon sideband of hBN emitters was mapped out using stimulated emission depletion spectroscopy.[121] Imaging of the emitters using the same technique [Fig. 6(c)] resulted in a spatial resolution of ∼50 nm.[122]

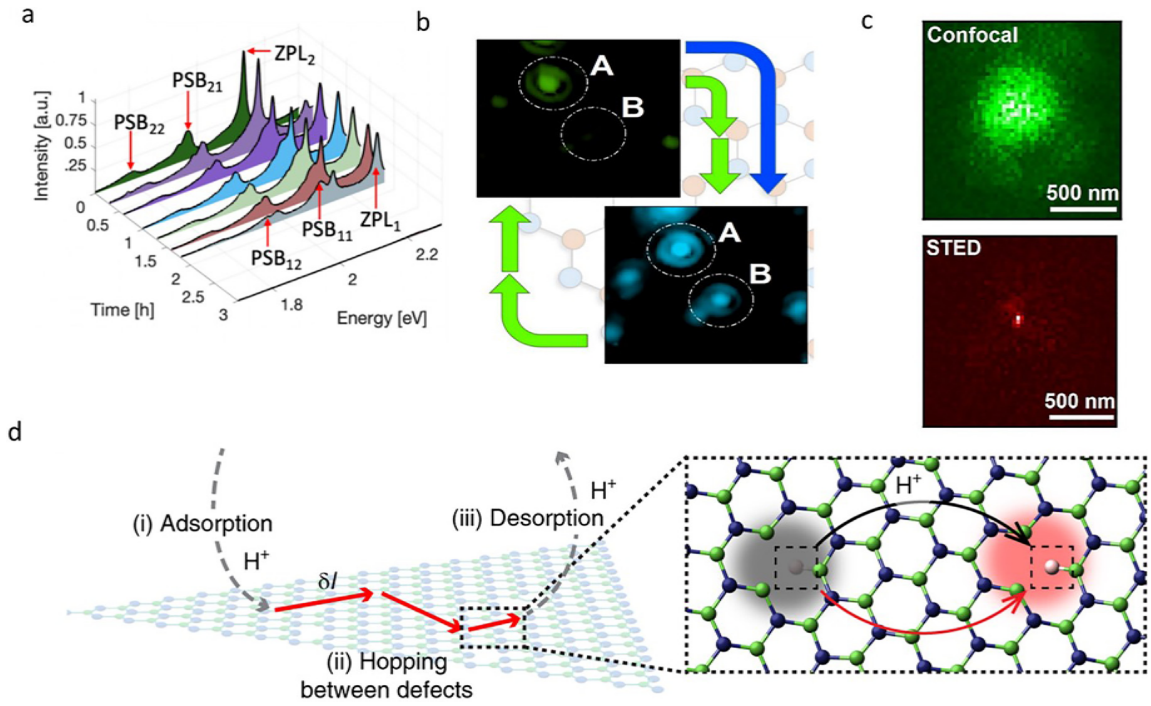

FIG. 6. Photophysical properties and application of hBN quantum emitters. (a) Double ZPL emission from a single emitter in hBN which is switches over time. Reproduced with permission from Feldman *et al.*, Optica **8**, 1 (2021). Copyright 2021 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (b) Optical gating of quantum emitters in hBN using double excitation. Reproduced with permission from Khatri *et al.*, Nano Lett. **20**, 4256 (2020). Copyright 2020 American Chemical Society. (c) Super-resolution microscopy of hBN quantum emitters using the STED method. Reproduced with permission from Khatri *et al.*, ACS Photonics **8**, 2081 (2021). Copyright 2021 American Chemical Society. (d) Schematic illustration of transport of H+ on a hBN monolayer, giving rise to switching of quantum emitters. Reproduced with permission from Comtet *et al.*, Nat. Nanotechnol. **15**, 598 (2020). Copyright 2020 Nature Publishing Group.

These results open up avenues for application of hBN emitters in biological imaging, with flexibility in choosing the desired emission energy.

Beyond imaging, the distinctive properties of hBN emitters and the 2D nature of the host material have led to their use in various applications. Bright and stable single photon emitters in hBN have been used for quantum random number generation at room temperature[123] and delayed-choice experiments.[124] Emitters in hBN monolayers have been shown to be affected by protons present in an aqueous environment. This results in emission switching when there is an interaction with He^+ and provides a unique optical method for tracking the transport of He^+ as is shown schematically in Fig. 6(d).[125] Interactions of emitters in hBN with graphene and other two dimensional materials have been also been investigated,[84,126] with an emphasis on charge transfer and energy transfer within the 2D hetrostructures. The ability to fabricate heterostructures from a broad range of 2D materials creates interesting opportunities for investigations of quantum phenomena, and advanced engineering and control of quantum emitters in hBN.

IV. LIGHT–MATTER INTERACTIONS

A. Integration of emitters with photonic structures

On-chip nanophotonics is underpinned by light–matter interactions mediated by SPEs coupled to photonic nanostructures.[127,128] Coupling to cavities and waveguides provides a versatile means to integrate emitters in photonic circuits. Moreover, coupling to cavities in the weak light–matter interaction regime is used to enhance emission rates, reduce emission linewidths, and enhance ODMR contrast,[99,129–143] all of which are needed to improve the competitiveness and appeal of SPEs in layered materials for quantum applications. The strong coupling regime can be used to access cavity quantum electrodynamics; however, while having been demonstrated with ensemble emissions in 2D materials,[144–146] it is yet to be realized using SPEs in layered hosts.

Layered materials are appealing for device and photonic circuit fabrication due to the ergonomic nature of exfoliation and pick-and-place positioning techniques. In addition, the atomic and near-atomic thickness of monolayers and few-layer flakes make them attractive for coupling of SPEs via the evanescent field, and the ability to localize emitters by strain gradients has enabled a unique approach for aligning emitters with edges and asperities at microdisks, nanowires, slot waveguides, nanoparticles, and nanopillars.[57,133,135,137,140–142,147–151] This versatility offered by layered materials has enabled rapid progress in studies of emitter coupling to waveguides,[24,150–153] plasmonic nanostructures,[57,99,131,133,134,138,141,142,147,149,154] dielectric microdisk, bullseye and photonic crystal cavities (PCCs),[26,130,132,137,139,140,155] and open-access Fabry–Pérot microcavities.[129,136,143] We note, however, that most literature on coupling of emissions in TMDCs to PCCs are focused on ensembles,[156–159] while reports on SPEs are relatively sparce,[26,155]

and the reported SPE coupling efficiencies and enhancement factors are low.

The utility of strain field confinement is particularly compelling and illustrated in Fig. 7 by works where SPEs in monolayer WSe_2 were localized to (a) a dielectric waveguide,[151] (b) a plasmonic nanorod waveguide,[147] (c) plasmonic nanocavities,[57] and (d) a circular Bragg grating (bullseye) cavity. In Figs. 7(a)–7(c), waveguide/nanocube edges/ corners were used to generate strain fields in a WSe_2 overlayer/underlayer, while in (d) a conical nanopillar was fabricated in the center of the $Al_{0.31}Ga_{0.69}As$ bullseye in order to localize an SPE at the cavity hotspot.

Coupling to waveguides is highlighted further in Fig. 8 by studies of WSe_2 SPE incorporation in photonic circuits [Figs. 8(a) and 8(b)][150] and coupling of hBN V_B^- SPE ensembles to gold film microwave waveguides [Figs. 8(c) and 8(d)].[99] The gold antennas serve a dual function—(i) plasmonic enhancement of SPE brightness and (ii) enhancement of contrast and signal-to-noise ratio in ODMR spectra of hBN V_B^- spin defects.

B. Purcell enhancement: Plasmonic vs dielectric cavities

In discussing works on coupling of SPEs to cavities, it is useful to delineate approaches used to achieve Purcell enhancement of the spontaneous emission rate which affect the spatial alignment tolerances of the coupled systems. The Purcell enhancement factor is proportional to the cavity quality factor (Q) and inversely proportional to the mode volume (V). Plasmonic cavities suffer from high losses (i.e., low Q factors), but enable ultra-small mode volumes defined by the geometries of plasmonic nanostructures. Plasmonic hot spots are localized at nanostructure edges and asperities which can simultaneously confine electromagnetic fields and achieve spatial matching with strain-localized TMDC SPEs, as has been demonstrated using metallic slot waveguides,[149] nanocubes,[57] nanorods,[141,154] nanocones,[142] and bow-tie antennas. These works are focused predominantly on improving the properties of coupled TMDC SPEs via excitation and emission enhancement. Similarly to TMDCs, plasmonic nanocavities were also used to enhance the optical properties of hBN SPEs,[131,134,138,148] and strain gradients were suggested to play a role in hBN emitter activation in some of these studies.[131,148] The emitters in hBN are, however, deep trap defects—the SPE "activation" mechanism is not well understood and likely distinct from that of exciton localization in TMDCs.

In contrast to plasmonic cavities, dielectric PCCs typically exhibit lower losses and thus enable efficient Purcell enhancement via high Q factors. However, as we noted above, works that demonstrate coupling

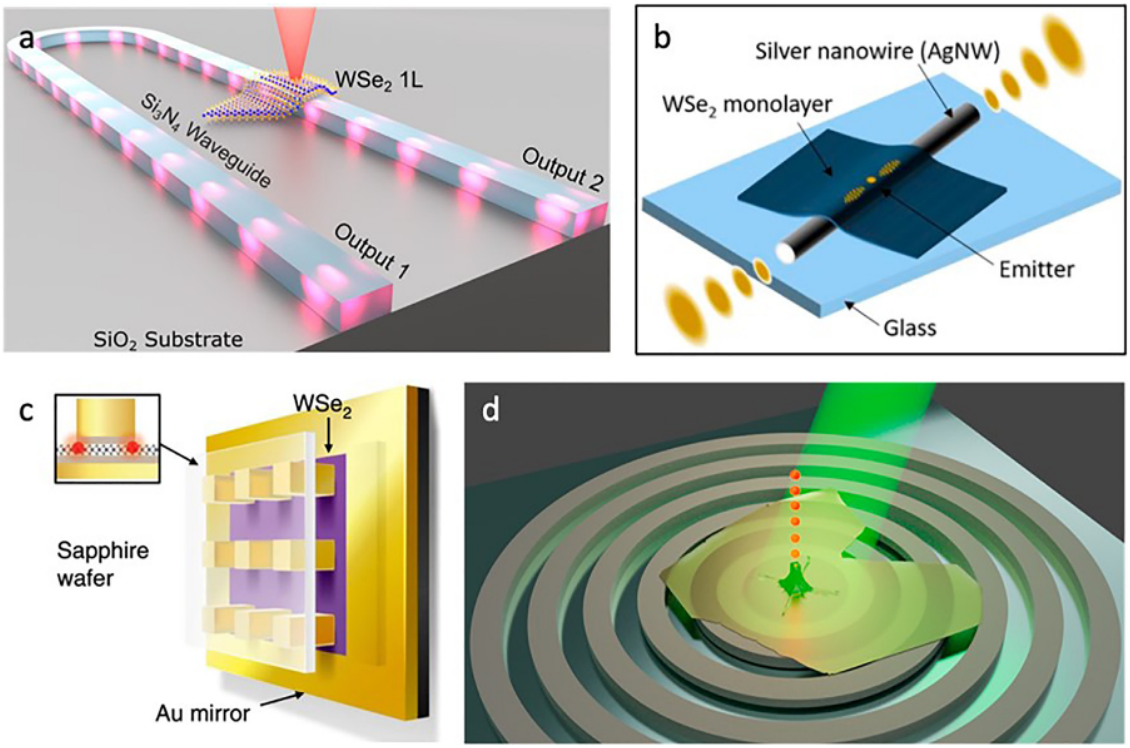

FIG. 7. Spatial alignment of WSe_2 SPEs at photonic nanostructures by strain. Edges of dielectric and metallic nanostructures generate strain gradients in monolayer WSe_2 and confine excitons, enabling coupling of SPEs to photonic circuit components. (a) Emitter coupled to a silicon nitride waveguide in a study of resonant SPE excitation in a photonic circuit. Reproduced with permission from Errando-Herranz *et al.*, ACS Photonics **8**, 1069 (2021). Copyright 2021 American Chemical Society. (b) Emitter coupled to a propagating surface plasmon polariton mode of a silver nanowire. Reproduced with permission from Cai *et al.*, Nano Lett. **17**, 6564 (2017). Copyright 2017 American Chemical Society. (c) Emitters coupled to plasmonic gap nanocavities defined by a gold film and the corners of gold nanocubes in a study that achieved Purcell enhancement factors of up to 551 and SPE emission rates of up to 42 MHz. Reproduced with permission from Lee *et al.*, Nat. Nanotechnol. **13**, 1337 (2018). Copyright 2018 Nature Publishing Group. (d) Emitter coupled to a circular Bragg grating (bullseye) cavity by a strain field generated by a nanopillar incorporated in the center of the cavity. Reproduced with permission from Iff *et al.*, Nano Lett. **21**, 4715 (2021). Copyright 2021 American Chemical Society.

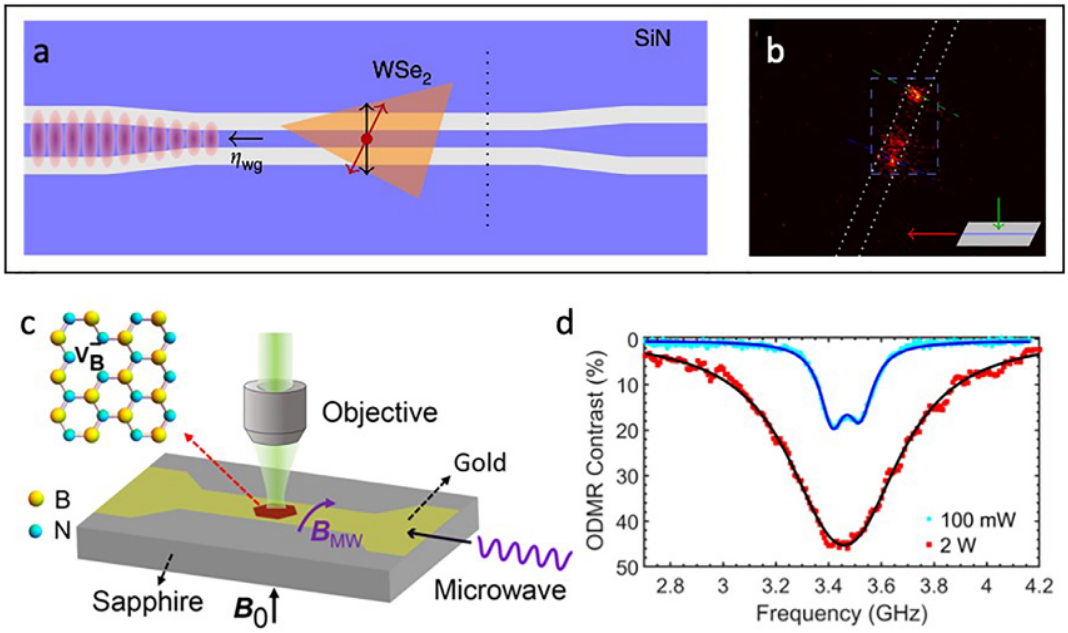

FIG. 8. Coupling of SPEs in layered materials to dielectric and metallic waveguides for photonic circuit integration and ODMR contrast enhancement. (a) and (b) Emitter in monolayer WSe_2 coupled to a silicon nitride circuit in a study of on-chip single photon guiding. Reproduced with permission from Peyskens *et al.*, Nat. Commun. **10**, 4435 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license. The PL map in (b) was generated by scanning an excitation beam across the waveguide (indicated by white lines) from the top and collecting PL through a fiber. The green and red arrows in the inset indicate the excitation and collection paths. (c) and (d) Ensemble of hBN V_B^- spin defects coupled to a gold microwave waveguide used to enhance ODMR contrast and signal-to-noise ratio. The room temperature spectra in (d) show ODMR contrast of approximately 19% and 46% at microwave excitation powers of 100 mW and 2 W, respectively. Reproduced with permission from Gao *et al.*, Nano Lett. **21**, 7708 (2021). Copyright 2021 American Chemical Society.

to TMDC SPEs are limited[26,155]—notably, So *et al.*[26] coupled a WSe_2 SPE to a nanobeam PCC in a configuration where the SPEs were localized to a cavity hot spot by a strain gradient generated by a gap in a dielectric nanorod located below the PCC [Figs. 9(a)–9(c)]. Most works on coupling of SPEs in layered materials to PCCs are, however, focused on hBN, using the so-called hybrid and monolithic integration methods discussed below.

C. Hybrid vs monolithic integration methods

It is instructive to delineate hybrid from monolithic approaches to coupling, particularly in the context of layered materials as their properties can vary dramatically between monolayers and multilayers. In the hybrid integration method, an emitter is placed on a waveguide or the hot spot of a cavity composed of a material that is different from that of the layered SPE host. Conversely, in the monolithic approach, an SPE is embedded within a dielectric nanostructure fabricated from the host material. The monolithic approach can be advantageous in that it avoids losses at interfaces of different materials. It has been applied to hBN[132,139,160,161] since hBN emitters have deep states and are bright despite the indirect nature of the bandgap of hBN. The monolithic approach has, however, not been applied to TMDCs since multilayers are needed to achieve optical confinement, and SPEs in thick TMDCs are very dim due to the indirect nature of the bandgaps of TMDC multilayers.

The monolithic approach has been used to incorporate SPEs in a range of hBN photonic nanostructures such as waveguides, bullseye cavities resonators, and PCCs[132,139,160] [Figs. 9(d)–9(f)]. The hBN nanostructures are typically fabricated by reactive ion etching and/or chemically mediated, direct-write electron beam induced etching [Figs. 9(d) and 9(e)].

Next steps in works on integration of SPEs in photonic nanostructures are foreshadowed by studies of ensemble excitonic emissions in TMDCs. Strong coupling has been achieved in studies of microcavity polaritons and polarons using monolayer MoS_2[146] and $MoSe_2$[144,145] embedded in distributed Bragg reflector (DBR) microcavities [Figs. 10(a) and 10(b)], but is yet to be realized using SPEs in layered materials. Similarly, high refractive index dielectric nanoantennas which can confine optical modes to ultra-small volumes[162,163] have been used to enhance ensemble excitonic emissions in WSe_2 [Figs. 10(c) and 10(d)],[157] and more recently demonstrated also with single photon emitters in WSe_2[164]—interestingly, the coupled emitters exhibited emission enhancement of up to $\times 10^4$, enabling optical coherence measurements at low excitation power. However, the measured coherence times still remains modest (~3 ps).

Works on coherent light emission from two dimensional materials[158,159,165] are also instructive here as they illustrate how coupling to microcavities can be used to overcome limitations imposed by the extremely low thickness of monolayers. Specifically, atomically flat materials are challenging for the design of on-chip excitonic lasers due to the small volume and a lack of light confinement in a 2D gain

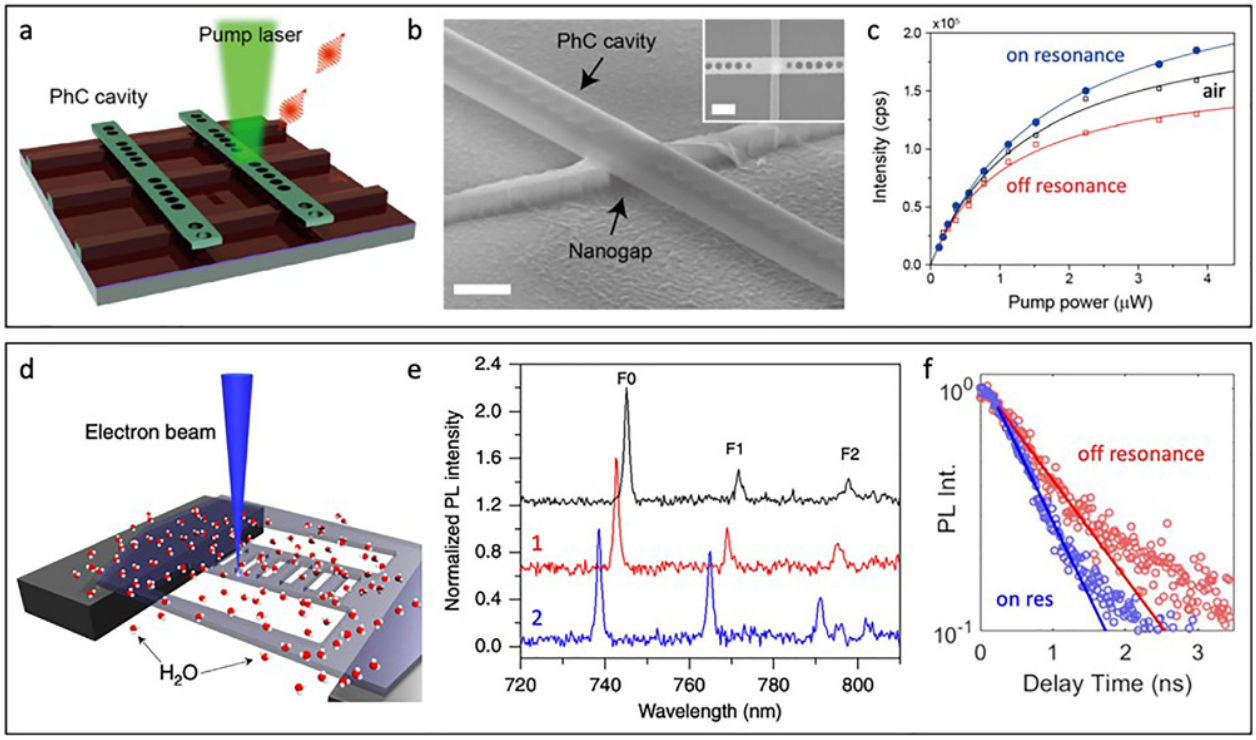

FIG. 9. Hybrid (a)–(c) and monolithic (d)–(f) integration methods used to couple SPEs to nanobeam photonic crystal cavities. (a)–(c) WSe_2 emitter coupled to a silicon nitride cavity. Reproduced with permission from So *et al.*, Nano Lett. **21**, 1546 (2021) Copyright 2021 American Chemical Society. The emitter was localized to the cavity hotspot by strain generated in the WSe_2 monolayer by a gap in a silicon nitride nanorod located below and oriented perpendicular to the nanobeam. The electron micrograph in (b) shows the nanorod–nanobeam structure. The photoluminescence saturation curves in (c) show the brightness of three different emitters located in a region of WSe_2 that was suspended in air, positioned at but not coupled to a cavity, and coupled to a cavity. (d)–(f) Studies of SPEs in monolithically fabricated hBN nanobeam cavities. In (d), a nanobeam cavity was tuned iteratively by direct-write electron beam induced chemical etching. Reproduced with permission from Kim *et al.*, Nat. Commun. **9**, 2623 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution (CC BY) license. The photoluminescence spectra in (e) show the wavelengths of three modes in an as-grown cavity (black), and the same cavity after one and two tuning cycles. The photoluminescence decay time measurements in (f) are from a subsequent study and show the lifetime of an SPE before and after it was brought into resonance with a nanobeam cavity mode by gas condensation. Reproduced with permission from Fröch *et al.*, Small **2021**, 2104805. Copyright 2021 Willey.

medium. To overcome this issue, exitonic TMDC lasers rely on coupling of TMDC excitons to microcavities, e.g., where WSe_2 and WS_2 were coupled to a PCC and microdisk cavity, respectively.[158,165]

V. CONCLUSIONS

The emergence of single defects in 2D materials has spurred several interesting research directions. Fundamentally, new systems that were not envisioned previously, such as moiré trapped excitons, will be vital in understanding the underlying photodynamics of these unique superlattices. Technologically, coupling to waveguides and on-chip guiding of quantum light has now became easier and more accessible since the photonic circuits can be engineered using conventional methods and materials, and layered materials can be incorporated using ergonomic exfoliation and transfer techniques. This is important as it eliminates the need for high-end fabrication steps such as the growth of ultra-pure materials, as was often the case for arsenic and nitride-based quantum dots and diamond. The rapid progress in engineering of emitters in 2D materials on-demand is also noteworthy. Focused ion beam systems are now commonplace, and ion implantation and irradiation techniques are in the spotlight as the most promising approach for introducing localized defects in 2D lattices. Indeed, the rapid rate of progress in the field has, in large part, been enabled by the ease and broad accessibility of the material and device fabrication techniques.

Questions remain, however, about the ultimate performance of quantum emitters in 2D systems compared to the more established platforms. Indeed, key quantum experiments are still missing and a number of performance metrics are inadequate, including the quantum efficiency of TMDC emitters, demonstrations of two photon interference, examination of the spin-photon interface, isolation of single V_B^- defects in hBN (the only defect in a layered material to date with a triplet ground state), and extension of the V_B^- spin coherence time. Moreover, consensus is yet to be reached regarding the atomic structure of most emitters in both hBN and TMDCs. We will undoubtedly see increased research activity aimed at solving these challenges as an increasing number of groups join this fascinating journey into the quantum world in flatland.

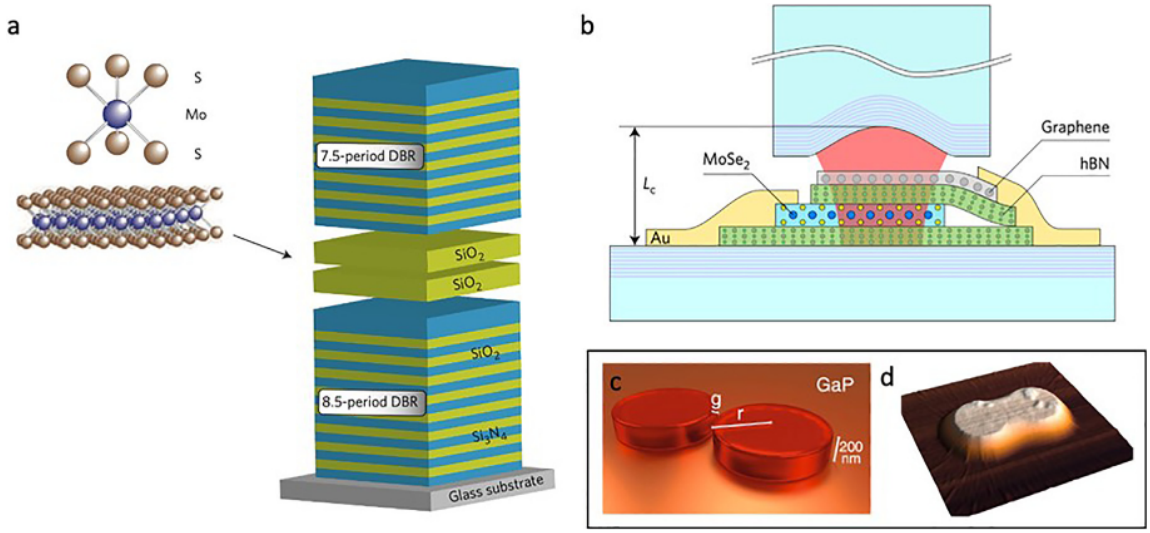

FIG. 10. (a) and (b) Strong light–matter interactions realized by coupling emissions from TMDCs to distributed Bragg reflector (DBR) microcavities in works that serve as stepping stones to polaritonic devices based on 2D materials. Reproduced with permission from Sidler *et al.*, Nat. Phys. **13**, 255 (2017). Copyright 2015 Nature Publishing Group and Liu *et al.*, Nat. Photonics **9**, 30 (2015). Copyright 2017 Nature Publishing Group. (c) and (d) Cylindrical gallium phosphide nanoantennas used to increase excitonic photoluminescence intensities in monolayer and bilayer WSe_2 by enhancement factors as high as 10^4. Reproduced with permission from Sortino *et al.*, Nat. Commun. **10**, 5119 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors declare no conflict of interest.

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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