Defect and strain engineering of monolayer WSe$_2$ enables site-controlled single-photon emission up to 150 K

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In recent years, quantum-dot-like single-photon emitters in atomically thin van der Waals materials have become a promising platform for future on-chip scalable quantum light sources with unique advantages over existing technologies, notably the potential for site-specific engineering. However, the required cryogenic temperatures for the functionality of these sources has been an inhibitor of their full potential. Existing methods to create emitters in 2D materials face fundamental challenges in extending the working temperature while maintaining the emitter’s fabrication yield and purity. In this work, we demonstrate a method of creating site-controlled single-photon emitters in atomically thin WSe$_2$ with high yield utilizing independent and simultaneous strain engineering via nanoscale stressors and defect engineering via electron-beam irradiation. Many of the emitters exhibit biexciton cascaded emission, single-photon purities above 95%, and working temperatures up to 150 K. This methodology, coupled with possible plasmonic or optical micro-cavity integration, furthers the realization of scalable, room-temperature, and high-quality 2D single- and entangled-photon sources.

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As the second quantum era emerges in the twenty-first century, what were once considered as tangential quantum effects are instead harnessed in new quantum technology. Single-photon emitters (SPEs) are the heart of many quantum photonic applications—such as quantum communication, cryptography, metrology, and linear optical quantum computing. Current state-of-the-art semiconducting single-photon sources—such as InAs self-assembled quantum dots—encounter challenges in scalability and integration with mature photonic technologies due to difficulties in deterministic positioning and growth. In recent years, a new SPE platform has emerged in two-dimensional (2D) materials, including hexagonal boron nitride and transition metal dichalcogenides (TMDs) such as MoSe₂ and WSe₂. Monolayer WSe₂, in comparison to conventional III–V semiconducting quantum dots, offers several advantages, including a high-photon extraction efficiency due to its atomic thickness, the potential for scalable, site-controlled and deterministic manufacturability, and ease of integration with mature photonic technologies via simple transfer methods, which has made them an intriguing contender for future technologically relevant quantum light sources.

However, a challenge facing 2D WSe₂ SPEs lies in their low working temperature and thermal instability. The ultra-sharp emission lines associated with SPEs that appear ~50–200 meV below the free exciton of WSe₂ are observable only at cryogenic temperatures and quench above 30 K due to low confinement potential and quantum yield. Considering that the microscopic origins of these emitters are still not fully understood, their further optimization and engineering become challenging. Recently, it has been hypothesized that SPEs in WSe₂ may be attributed to localized intervalley defect-bound excitons that form when the energy of the dark excitonic band of WSe₂ is lowered and hybridizes with a valley symmetry-breaking defect state at strained regions (Fig. 1). This description clarifies key points regarding the strong brightness and magneto-optic properties of these emitters, which cannot be justified within an only-strain or an only-defect description. Furthermore, it reveals that both strain and defects are pivotal in the creation of SPE. Recent experimental studies have also confirmed parts of this model, however, the essential link, which is the correlation between defects and quantum emitters, remains elusive. This is partially because defects in 2D TMDs are ubiquitous and difficult to control. The challenge compounds further since most strain engineering methods are also prone to damage or replace the TMDs in the process.

In this study, by using electron-beam (e-beam) irradiation as a controllable method to induce structural defects in WSe₂, along with engineering strain fields in encapsulated WSe₂ using dielectric nanopillar structures, we decouple the strain and defect engineering processes and prove that there is a direct correlation between defect density and quantum emitters, which provides strong evidence for the intervalley defect excitation model. Moreover, the accurate positioning of defects within the strain field in WSe₂ minimizes the non-radiative recombination and achieves higher confinement potential. These, in addition to the possible distinct morphology of our e-beam engineered defects that appear at deeper energies than any other observed SPEs in WSe₂, enable engineering SPEs with high yield, high single-photon purity, biexciton-like radiative cascade, and possible working temperatures up to 215 K. This sets the precedent for the highest working temperature of SPEs observed in 2D TMDs without Purcell enhancement (benchmark available in Supplementary Note 1).

Results

Strained WSe₂ PL response. A monolayer WSe₂ sample encapsulated by h-BN was prepared using dry transfer techniques (see Methods). h-BN flakes, with thickness of less than 5 nm, were chosen to encapsulate the WSe₂ not only to isolate the WSe₂ from surface states and adsorbates, but to also act as protective layers that shield the WSe₂ from potential damages during the stamping process onto nanopillars. Subsequently, to engineer the necessary strain profile across the 2D flake, the stack was transferred onto a Si/SiO₂ substrate array with a predefined nanopillar dielectric array with height and diameter of 200 and 150 nm, respectively, which was previously identified as an optimal aspect ratio for deterministically introducing SPEs without piercing through the materials. Figure 2a presents the dark-field optical image of the final assembled structure where the brightly illuminated regions correspond to the nanopillar sites. Photoluminescence (PL) spectra of the WSe₂ residing on the nanopillars were taken at T = 5 K (Supplementary Fig. 1). Interestingly, no sharp emission peaks that could be associated with single-photon emitters were observed. In contrast, only slightly redshifted neutral exciton (X) and charged exciton (X⁻) peaks were detected. Given that the strained region is about 2-orders of magnitude smaller than the diffraction-limited spot size, the red-shifted neutral exciton peaks due to strain are dim and cannot be resolved by a far-field diffraction-limited system (see Supplementary Note 2 and Supplementary Figs. 2 and 3), consistent with previous studies. Furthermore, the defect band that usually dominates the PL spectra of 1L-WSe₂ is also absent in the PL spectrum both in the strained and unstrained regions for the typical excitation powers and accumulation times used in our experiments. However, a
very weak broad-defect band, without any sharp peaks, can be observed when PL intensity at low excitation power is integrated over a long time. Interestingly, the correlation between the defect band and quantum emitters has been hinted in previous studies where the spectral histogram of measured emitters reconstruct the shape of the defect band\cite{9,11}. Overall, this provides evidence that in WSe\(_2\) samples, which are devoid of the defect band and are shielded from the substrate with h-BN encapsulation, single-photon emitters cannot form solely due to the strain in WSe\(_2\).

**PL response of defect-induced irradiated WSe\(_2\).** To further test this hypothesis and examine correlations between defects and quantum emitters, electron-beam irradiation, which has been successfully used to induce structural defects\cite{20,21} and phase transformation\cite{22} in 2D TMDs, was utilized to induce defects at desired locations. First, an unstrained region away from the nanopillars was irradiated at five different intensities ranging from \(8 \times 10^4\) electrons/\(\mu\)m\(^2\) to \(10^6\) electrons/\(\mu\)m\(^2\) with an accelerating voltage of 100 keV and an e-beam spot size that is tightly focused (<10 nm) and is held constant to maximize the spatial accuracy. Figure 2b shows the PL spectra of unstrained regions after e-beam irradiation at the energy range corresponding to the WSe\(_2\) defect band. At low irradiation intensity (<\(10^5\) electrons/\(\mu\)m\(^2\)), the band is very dim and difficult to distinguish. However, as the irradiation intensity increases, the defect band intensifies, which indicates that e-beam irradiation generates optically active defects in WSe\(_2\). These observations are consistent with previous studies\cite{21} and it should be emphasized that the broad defect band emission retains its qualitative characteristics irrespective of the power used\cite{21}. It is readily observable that solely with e-beam irradiation, only the defect band can be created, and no sharp quantum emission lines appear in the spectra.

**Strain and defect-engineered quantum emitters in WSe\(_2\).** Next, we irradiated the strained regions at the top of the nanopillars as shown in Fig. 2a with the same nominal intensities. At the lowest intensity, no quantum emitters were observed in our samples. However, at the threshold of \(10^5\) electrons/\(\mu\)m\(^2\) (denoted as \(N_1\)), 12% of sites demonstrated at least one quantum emitter (Fig. 2e). As the electron irradiation intensity increases further, both the yield and number of emitters per site increase. At \(10^6\) electrons/\(\mu\)m\(^2\) (\(N_2\)), 85% of the emitters demonstrated at least one quantum emitter per site (Fig. 2f). It is worth noting that e-beam irradiation has also been utilized to generate SPEs in h-BN, however, the h-BN emitters are not enabled unless a thermal annealing step is performed. Considering our cryogenic temperature and the fact that emitters also appear at lower energies compared to h-BN emitters at room temperature, the emitters studied here are highly unlikely to originate from h-BN. Lastly, a recent study\cite{23} has successfully used helium ion bombardment to create defects exhibiting single-photon emission in 2D MoS\(_2\), however, we must emphasize here that WSe\(_2\) due to its dark ground-state, is a phenomenological interesting case wherein inducing defects through irradiation alone will not result in SPE creation. Integrated PL maps (Fig. 2b) of six irradiated nanopillars (white dashed box region in Fig. 2a) were taken at 5 K for energies below...
1.6 eV. These maps provide evidence for the formation of bright emission lines below the WSe\(_2\) free exciton. The correlation among e-beam intensity, optical emission intensity of the defect band, and the number of engineered quantum emitters in each site strongly supports the hypothesis that both strain and defects have a fundamental role in single-photon emission in WSe\(_2\)\(^{11-16}\).

Figure 2d shows the PL spectrum at the location of three different irradiated nanopillars. Sharp emission lines associated with SPEs in these materials appear at the energy interval of 1515–1580 meV. Note that this energy interval is approximately 100 meV lower than the usual energy range\(^{7,9-12}\) that SPEs appear in WSe\(_2\). This suggests that the morphology of the e-beam-induced defects may be distinct from the previously observed native defects and reside deeper in the bandgap.

Observation of biexciton emission cascade. With a closer look at Fig. 2d, it is readily observable that many sharp emission lines appear to form in pairs with an energy spacing of about 3–5 meV. The features of these peaks resemble those of an exciton-biexciton pair in which the inherent fine-structure splitting results in two doublets, but the sign of the splitting is reversed in each pair as a consequence of the emission cascade (Fig. 3a). The polarization-resolved PL spectrum of the pairs further corroborates the radiative cascade (Fig. 3b). The integrated intensities of the pairs show distinct sub-linear and super-linear characteristics as previously observed for exciton-biexciton emissions in WSe\(_2\)\(^{24}\) (Fig. 3c). Similarly, the time-resolved PL (Supplementary Fig. 4) also shows that the decay time (T\(_1\)) of the exciton (X, T\(_1\) = 6.12 ns) feature is almost 1.5 times higher than the biexciton (XX, T\(_1\) = 4.01 ns), which is comparable with the previously observed dynamics of exciton-biexciton pairs in WSe\(_2\)\(^{24}\). Note that the physical origin of these exciton-biexciton-like features is still ambiguous; previous studies\(^{14}\) have shown that such features can also be attributed to the hybridization of defect states with localized excitons. Spectral diffusion of both exciton and biexciton was measured in a 5-min period, which showed almost appreciable spectral wandering (Supplementary Fig. 5). Finally, second-order correlation measurements were performed using a Hanbury Brown and Twiss setup\(^{25}\) that confirms both emission lines act as single-photon sources with purities as high as 90% (Fig. 3d).

Statistics of the defect and strain engineered sites. Engineering high purity and reproducible single-photon emitters with high yield is a requirement for any scalable photonic technology. By decoupling the strain and defect engineering in our nanopillar irradiation process, we were able to achieve a success rate of over 85% in engineering single-photon emitters per site. Note that, since e-beam irradiation is a repeatable process, subsequent irradiation and dosage optimization processes can be leveraged to achieve near-unity-yield at each site. Figure 4 represents the statistical histograms of the single-photon emitters fabricated using our method. The spectral purity of many of the emitters was measured to be above 95% (with an average purity of 92%, Fig. 4a) with average linewidths of 75 \(\mu\)eV and zero-field splitting of 760 \(\mu\)eV, and the linewidth and purities will likely improve with resonance excitation.

Furthermore, the energy interval of the observed SPE emission (1515–1580 meV) is smaller than other approaches. This is likely a feature of our decoupled approach—first a broad-defect band is engineered with e-beam irradiation (~1.5–1.7 eV in Fig. 2). The SPEs then appear when a localized stressor red-shifts the neutral exciton energy into resonance with a defect state in the band. Given the defect range (1.5–1.7 eV) is larger than the spectral range of the emitters (1515–1580 meV), it can be deduced that the variance in the engineered strain profiles sets the limit for the spectral spread of our method. In future experiments, using more precise strain engineering methods may enable engineering identical vdW SPEs.

Temperature evolution of engineered WSe\(_2\) emitters. As discussed previously, one of the main disadvantages of native WSe\(_2\) single-photon emitters is that their PL intensity and single-photon purity quench at temperatures above 30 K. One of the main contributors to this effect is the defect-bound exciton–phonon interaction, which leads to carrier escape from the confined potential, an effect which is more dominant in shallow quantum emitters. As e-beam-induced defects create deeper states within the bandgap, as evidenced by the lower emission energies compared to native defect-based emitters, a higher energy barrier and hence, a higher thermal activation energy is to be expected\(^{26}\). In addition to this, the non-radiative exciton–phonon interaction can become the dominant quenching factor at high-trap densities\(^{27}\). In our process the weights of non-radiative recombination pathways are minimized by using exfoliated 2D WSe\(_2\) with low-defect density (evident from the absence of the defect band). Conversely, the defects are induced in the sample via a controllable process targeting only the desired
decouples the exciton energy from the temperature dependence of strain through more accurate positioning of the defect site within the 2D material. This decoupling can persist up to 150 K, which indicates that deep conduction band states are dedicated to the quantitative analysis of single-emitter properties. It is readily apparent that the emission line is homogeneous within the 2D flake, with a ~5 meV redshift distinguishable up to 150 K. This temperature dependence, this enhancement is limited. This is further corroborated by the previous demonstrations of h-BN encapsulated TMD defect-bound excitons where a general enhancement of working temperatures are observed.

Figure 5e, f show the temperature dependence of the second-order correlation function and demonstrates that the emitters exhibit antibunching up to 150 K with $g^{(2)}(0) = 0.27 \pm 0.05$. Given the high average purity of the array of emitters at 150 K (see Supplementary Fig. 7), it can be expected that the single-photon nature of the SPEs can persist up to 215 K. It can be predicted that our SPE engineering method, in tandem with designs leveraging Purcell enhancement, could enable room temperature single-photon emission.

Figure 6 quantitatively summarizes one plausible mechanism behind the single-photon emission as depicted in Fig. 1. The dielectric nanopillar creates a strain profile akin to Fig. 6a in the 2D flake where the strain is at its maximum around the edges of the nanopillar. The applied strain results in the reduction of the bandgap (Fig. 6b), which creates a potential landscape analogous to Fig. 6a. This potential leads to the localization of free exciton states. From e-beam irradiation, defect states appear within the bandgap of the semiconductor in the strained regions. Provided the energy of these defect states at the strained regions is sufficiently close to the localized exciton states, the defects and excitons hybridize and results in the bright single-photon emission peak. While the engineering methods outlined in this work demonstrate the interplay between defects and strain, as predicted by the above-mentioned picture, the physical morphology of the defects responsible for single-photon emission is elusive. There is a lack of consensus of the defects responsible for single-photon emission, but they have been previously attributed to structures, including selenium vacancies, tungsten centered vacancies, oxygen interstitials, and anti-site defects. It is also plausible that multiple types of defects may be responsible for single-photon emission as long as they break the valley symmetry, and upon application of strain, introduce defect states with favorable energies close to the conduction band.

Given that the defects in this work were created by e-beam irradiation for which the morphology of resulting defects has been extensively studied, the list of possible suspects can be reduced through a systematic, ab initio approach. Studies have shown that e-beam irradiation processes primarily contribute to the generation of chalcogen and double chalcogen vacancies due to a lower knock-off energy. Furthermore, prolonged exposure of a focused e-beam spot can also lead to the destabilization of the transition metal bond, which causes the transition metal to migrate away from the site, creating more complex vacancy sites such as pore vacancies. In the case of WSe₂, the creation of rotational trefoil defect complexes under e-beam irradiation has also been observed. Therefore, we chose to study five probable defect complexes that might form during the e-beam process, straining the material to further increase the working temperatures. It is worth mentioning that higher confinement potentials can be achieved through more accurate positioning of the defect site within the strain field.

Figure 5a shows the evolution of the single-photon PL emission from 5 to 150 K (Supplementary Note 3 and Supplementary Fig. 6 are dedicated to the quantitative analysis of single-emitter properties). It is readily apparent that the emission line is distinguishable up to 150 K. A ~5 meV redshift is distinguishable up to 150 K (Fig. 5b), which indicates that deep confinement decouples the exciton energy from the temperature dependence of the lattice. Fitting the temperature dependence (Fig. 5b) to a Varshni-form equation (Supplementary Note 3) indicates a temperature coefficient $5.3 \times 10^{-6}$ eV K$^{-1}$, which is smaller than the previously reported values for native defects in CVD grown TMDs. The evolution of the homogenous linewidth broadening with temperature is indicative of a thermally activated pure dephasing from exciton–phonon scattering and can be seen in Fig. 5c. The Arrhenius plot of the integrated intensity of the SPE shows that the integrated intensity quenches to $e^{-1}$ at ~215 K (Fig. 5d). The data are fit with an Arrhenius model $I(T) = I_0/(1 + R \exp(-E_a/kT))$, where $R$ is equal to the ratio of the radiative ($T_r$) and non-radiative ($T_{nr}$) recombination lifetimes and $E_a$ is the thermal activation energy that required to dissociate the defect-bound excitons. The fit to the data results in a quantum yield of 5% and an activation energy of 95 meV, suggesting that the higher activation energies observed in this work is the main factor that contributes to high working temperatures of our emitters (see Supplementary Fig. 6 for a comparative analysis).
namely selenium single vacancy, selenium double vacancy, tungsten vacancy, pore vacancy, and trefoil vacancies (see Supplementary Note 4 and Supplementary Figs. 8–13 for full details).

Density-functional theory (DFT) calculations were performed using Synopsis QuantumATK package by incorporating Perdew–Burke–Ernzerh (PBE) variant of generalized gradient approximation (PBE-GGA) exchange-correlation functional (see Methods for details). Note that given the common bandgap underestimation problem in DFT, along with the uncertainties in the exciton binding energies and the magnitude of the confinement potential, our focus is not on a quantitative comparison. Instead, our main criterion is the relative movement of the conduction band and defect levels with respect to each other upon application of strain, which can be assessed using ab initio simulation.

Figure 6c shows the dynamics of selenium single-vacancy levels with applied biaxial tensile strain. It is readily observable that the defect levels appear at favorable energies close to the conduction band and move to the proximity of the conduction band with strain. However, both selenium vacancies and selenium double vacancies are prone to passivation by oxidation. Given the fact that the SPEs in this work, similar to previous studies, remain active under Arhenious plot of the integrated intensity. The PL peak starts to quench around 150 K and reaches $e^{-1}$ at 215 K. Shaded region in the $T$ = 5 K and $f$ at 150 K exhibiting $g^{(2)}(0) < 0.3$, indicating that the single-photon nature persists up to our maximum measurement temperature of 150 K and likely persists beyond 200 K. Horizontal dashed lines represent the quantum limit of $g^{(2)}(0)$ at 0.5.

**Fig. 5 Statistics of single-photon emission up to 150 K.** a Evolution of the PL spectrum as function of the temperature. b Redshift of the SPE line with temperature increase due to the reduction of the bandgap following Varshni’s empirical relationship. c Homogenous linewidth broadening due to the increase of temperature. d Arhenious plot of the integrated intensity. The PL peak starts to quench around 150 K and reaches $e^{-1}$ at 215 K. Shaded region in b, c, and d demonstrate the range of recorded parameters for an ensemble of emitters. Open circles demonstrate measured data for a single emitter in the ensemble and the solid black lines are theoretical fit to the data. e Second-order correlation measurement $g^{(2)}(T) = 0.05±0.04$ exhibiting $g^{(2)}(0) < 0.3$, evidencing that the single-photon nature persists up to our maximum measurement temperature of 150 K and likely persists beyond 200 K. Horizontal dashed lines represent the quantum limit of $g^{(2)}(0)$ at 0.5.

**Methods**

**Sample preparation.** The nanopillar array was fabricated with high-resolution electron-beam lithography following the procedures described in ref. 10. Briefly, a silicon wafer with thermal oxide was cleaved into a 1 cm$^2$ chip followed by a solvent clean. The chip was spin-coated with hydrogen silsesquioxane (HSQ) resist diluted with methyl isobutyl ketone (MiBK). After a 5-min bake at 90 °C, pillar arrays and chip alignment markers were defined via electron-beam lithography and then developed in a 25% tetramethyl ammonium hydroxide (TMAH) solution and rinsed with methanol. A final rapid thermal anneal step at 1000 °C in oxygen converts the HSQ pattern into SiO2. The nanopillars have a nominal diameter of 150 nm and height of 200 nm.

We used a dry viscoelastic transfer technique to create the h-BN-encapsulated WSe2 structure. Intrinsic WSe2 flakes (2D Semiconductors) were exfoliated onto an unpatterned preparation SiO2/silicon substrate for identification and subsequent transfer. Next a thin flake of h-BN is exfoliated onto the transfer stamp, which was used to exfoliate and transfer WSe2 monolayers from thicker flakes on the preparation substrate. Finally, the WSe2/h-BN stack was transferred to an h-BN layer exfoliated on the patterned nanopillar array, which ensures that the WSe2 layer is encapsulated by the h-BN and free of any contaminants or residues during and after the transfer process. Quantum emitters were created using a 100 keV, <10 nm spot size electron-beam to expose the sample at the location of each nanopillar with ~10$^8$ electrons/μm$^2$.

**Optical spectroscopy.** For steady-state micro-photoluminescence (PL) measurements, the sample temperature was held fixed between 5 and 150 K. The emitters were excited non-resonantly with a continuous-wave 633 nm laser focused to a ~1 μm full-width at half-maximum spot size using a long working distance 0.7 numerical apertures microscope objective. The backscattered PL was collected and measured with a spectrometer and TE-cooled charge-coupled device with a spectral resolution of ~30 μeV. For time-resolved PL measurements, a 40 MHz, 532 nm pulsed laser diode was used as the excitation source. The PL was spectrally filtered with ~1 nm bandwidth and detected using a superconducting nanowire single-photon detector and time-tagging electronics with ~40 ps detector temporal
Note that the conduction band movement is larger with respect to the valence band. The inset band diagram denotes the qualitative movements of conduction band (Ec) and valence band (Ev) with strain. After the application of strain, the energy spacing between the conduction band and closest defect-level decreases. The Fermi-level energy position depends on the size of the supercell.

**Data availability**
The data in this manuscript is available upon reasonable request.

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