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Width-dependent continuous growth of atomically thin quantum nanoribbons from nanoalloy seeds in chalcogen vapor

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Nanoribbons (NRs) of atomic layer transition metal dichalcogenides (TMDs) can boost the rapidly emerging field of quantum materials owing to their width-dependent phases and electronic properties. However, the controllable downscaling of width by direct growth and the underlying mechanism remain elusive. Here, we demonstrate the vapor-liquid-solid growth of single crystal of single layer NRs of a series of TMDs (MeX_2: Me = Mo, W; X = S, Se) under chalcogen vapor atmosphere, seeded by pre-deposited and respective transition metal-alloyed nanoparticles that also control the NR width. We find linear dependence of growth rate on supersaturation, known as a criterion for continues growth mechanism, which decreases with decreasing of NR width driven by the Gibbs-Thomson effect. The NRs show width-dependent photoluminescence and strain-induced quantum emission signatures with up to ≈ 90% purity of single photons. We propose the path and underlying mechanism for width-controllable growth of TMD NRs for applications in quantum optoelectronics.

One-dimensional (1D) or quasi-1D electronic systems including whiskers, nanowires, and nanotubes exhibit exotic physical properties[1–5], which continue to fuel fundamental research and advanced applications. Recently, quasi-1D structures such as graphene nanoribbons (NRs) have been synthesized[6–10], showing enriched electronic structures and quantum phase tuning[6–9] compared to graphene sheets. Theoretical calculations also predicted width-dependent crystalline phases, band structure, and spin-orbit coupling in NRs of atomically thin two-dimensional (2D) transition metal dichalcogenides (TMDs)[11–15]. These anticipated peculiarities have attracted broad interest in creating disruptive technologies, especially in the rapidly emerging area of 1D van der Waals (vdW) quantum materials[16]. To date, the dominant approach for obtaining TMD NRs with controllable widths relies on post-synthesis treatments (e.g., lithography or etching)[17–19]. There are also reports on NR growth mediated by liquid droplets of precursors[20,21], or guided growth on specifically engineered

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substrates through aligned steps[22–24] or chemical treatment[25] to realize aligned NRs with even controllable edge structures[24]. However, the results widths were generally >50 nm and still lack of control on the width. Recently, we reported a direct synthesis of bilayer MoS_2 NRs with width down to sub-10 nm after a mild treatment[18]. Nevertheless, interference with intrinsic properties by post-synthesis treatments and lack of effective control on NRs width during growth made it difficult to study their pristine width-confined properties, thereby hindering their potential applications. On the other hand, deeper understanding of NR growth mechanism could not only guide the direct width-controlled growth but also enrich the fundamental science as an example of formation of 1D or quasi-1D nanocrystals.

Among various established methods, chemical vapor deposition (CVD) has been widely used to synthesize various 1D or quasi-1D nanostructures in a broad diameter range[26–28], from which many peculiarities of growth mechanisms have been revealed since its first exploitation in the 1960s in the growth of Si whiskers[29–35]. Indeed, through classical analysis on the experimental finding that narrower whiskers grew more slowly, Givargizov[30] derived an empirical model predicting quadratic growth rate dependence on reciprocal of the diameter of Au-catalyzed Si whiskers with a critical diameter (≤50 nm), below which the whisker cannot grow. This diameter-dependence and limit was explained by Gibbs-Thomson effect, i.e., the supersaturation of silicon in Au−Si droplet decreases with the droplet size. However, although such a model was later confirmed by many studies on the growth of other semiconducting nanowires[29,33], there were reports showing diameter-independent growth rate, explained by irreversible catalytic dissociation of exploited Si precursor[34]. Moreover, there also have been reports showing the opposite growth rate-diameter relation where narrower nanowires grew faster due to adatom diffusion on the nanowire surface[31,32]. However, the growth mechanism of atomically thin 1D NRs has barely been studied.

In this work, we present a CVD growth of single layer (SL) TMD NRs mediated by seed nanoparticles (NPs) in chalcogen vapor atmosphere. The seed NPs are based on nanoalloys of metal precursor and catalyst (i.e., Mo-Ni or W-Mg), which also control the width of the grown NR. Based on the length vs. width analysis of grown SL MoS_2 NRs under different vapor supersaturation, we discover that unlike previously studied 1-D structures, i.e., whiskers and nanowires, the growth rate of NRs is a linear function of supersaturation, decreasing linearly with the reciprocal of width controlled by the Gibbs-Thomson effect, which is characteristic of continues growth. As an example of exploiting the quantum nature of the NRs, we demonstrate deterministic quantum emission (QE) signatures induced by localized strain, making them promising candidates as single photon sources for quantum communications.

Results

Growth of Single Layer MoS_2 NRs

Synthetic fluorophlogopite mica (F-mica) with (001) surface plane was used as the substrate, which has been used for vdW epitaxial growth of 2D flakes before[36]. Unlike typical one-step synthesis[18,20,21,37], we used a two-step growth approach: First formation of partial precursor/seed NPs followed by introduction of corresponding chalcogen vapor. Specifically taking MoS_2 as an example, we first deposited NPs on the substrate by evaporating a mixture of MoO_2, Ni, and NaBr powder under a moisturized Ar flow in a tube furnace system (Fig. 1a, step 1; Fig. 1b). The presence of NaBr enabled co-deposition of Ni and MoO_2. The deposited NPs are composed of Na-Mo-Ni-O (Fig. 2a, b), in which the Mo atomic ratio within (Mo-Ni) varies in a broad range of ≈ 10%-85%. It is important to note that the presence of moisture was essential for the formation of the Na-Mo-Ni-O compound from the precursor powder mixture; without moisture, only NaBr was deposited on the substrate (Supplementary Fig. 1a–c)[18]. The NPs were then exposed in a sulfur vapor atmosphere, generated by heating sulfur powder, in a separate tube reactor (Fig. 1a, step 2), resulting in "tip"-growth of MoS_2 NRs from the NPs, with widths comparable to the tip-NP (inset of Fig. 1c, Supplementary Fig. 2a, b) and lengths from a few to tens of micrometers (Fig. 1c, Supplementary Fig. 1). Despite the reduction of seed NPs' diameter during the NR growth (Supplementary Fig. 3), their diameter distribution correlates with that of NR width (Supplementary Fig. 2c), which allows control of width distribution of NRs through controlling the diameter distribution of pre-deposited seed NPs. It is also noted that in some areas the as-grown NRs show a certain extent of alignment (i.e., Fig. 1c) while in others they do not, which could be related to the specific surface condition and crystallographic orientations of the F-mica substrate.

Atomic force microscopy (AFM) analysis (Fig. 1d, Supplementary Fig. 2e) and Raman spectroscopy (Fig. 1e, Supplementary Fig. 4) confirm the SL MoS_2 NRs. Scanning transmission electron microscopy (STEM) with fast Fourier transform (FFT) analysis indicates that the NR is single crystal, with the axial growth along the [010] direction (Fig. 1f). Occasionally single crystal NR grown along the [110] direction was also observed (Supplementary Fig. 5). Atomic-resolution high-angle annular dark-field STEM (HAADF-STEM) shows clean, well-defined honeycomb lattice with a lattice parameter $a \approx 0.308$ nm of MoS_2 NRs, which is terminated by zigzag edge structures (Fig. 1g, Supplementary Fig. 5d, e). Detailed Auger electron spectroscopy (AES) analysis (Fig. 2a–c, Supplementary Fig. 6) and HAADF-STEM combined with FFT analysis (Fig. 2d–g) reveal that during NR growth the composition of seed NP evolved from Na-Mo-Ni-O (Fig. 2a, b) to a pure Ni NP with a face-centered cubic (fcc) structure (Fig. 2e, f), as a result of consumption of Mo atoms in the seed NP by MoS_2 NR through precipitation under S vapor atmosphere. The precipitation in turn drives the motion of NP along the axial growth direction of NR (see Supplementary Fig. 7 showing no motion of NP without precipitation). As the growth temperature in step 2 is above the liquefaction temperature of the seed NP (≈725 °C)[18], the growth follows a VLS mechanism. It should also be noted that Na and Ni were not detected from the NR in AES measurement (Fig. 2c, Supplementary Fig. 6), indicating that the as-grown NRs are purely MoS_2. The electrical properties of the SL MoS_2 NR at room temperature show typical n-type behavior with an on/off ratio of ≈ 10^6 (Supplementary Fig. 8) and a current density comparable to the best CVD-grown MoS_2 reported with various contacts[38,39].

Growth mechanism of MoS_2 NRs

To understand the S-driven precipitation of MoS_2 on the atomic level and the role of multiple constituents in the seed NP, we performed molecular dynamics (MD) simulation (Fig. 2h). For an initially equilibrated Na-Mo-Ni-O NP at 650 K with S atoms, shows formation of triangular Mo-S local structural motifs bonded by Ni atoms, which could be the basic building blocks of MoS_2 NRs (Fig. 2i). Ni atoms also play a role in connecting different triangular units together (Fig. 2j). Those Mo-S motifs form even larger two-dimensional Mo-S structures as S continues to react with the system (Fig. 2k). It is noted that the oxygen in the system also plays an important role for the separation of Ni-MoS_2. In contrast to the equilibrated Na-Mo-Ni-O system with a relatively uniform distribution of all constituting elements (Supplementary Fig. 9b), the system without O (Na-Mo-Ni) evolves to a core-shell structure with Na atoms on the surface and a mixture of Ni and Mo in the interior of the nanoparticle (Supplementary Fig. 9c). Such a system keeps a core-shell structure after equilibrated with S atoms, only with slight mixing of Ni atoms with surface Na atoms (Supplementary Fig. 9a).

Next, we studied seed NP-mediated growth mechanism of SL MoS_2 NRs. As mentioned above, investigation on the relation of length (L) vs. diameter (d) of catalytic NP could provide specific insights on VLS growth mechanism of various 1D nanostructures. Therefore, we carefully analyzed our NR L vs. width (W) relationship grown at the same temperature ($T = 730$ °C) for a fixed growth duration $t = 180$ s.

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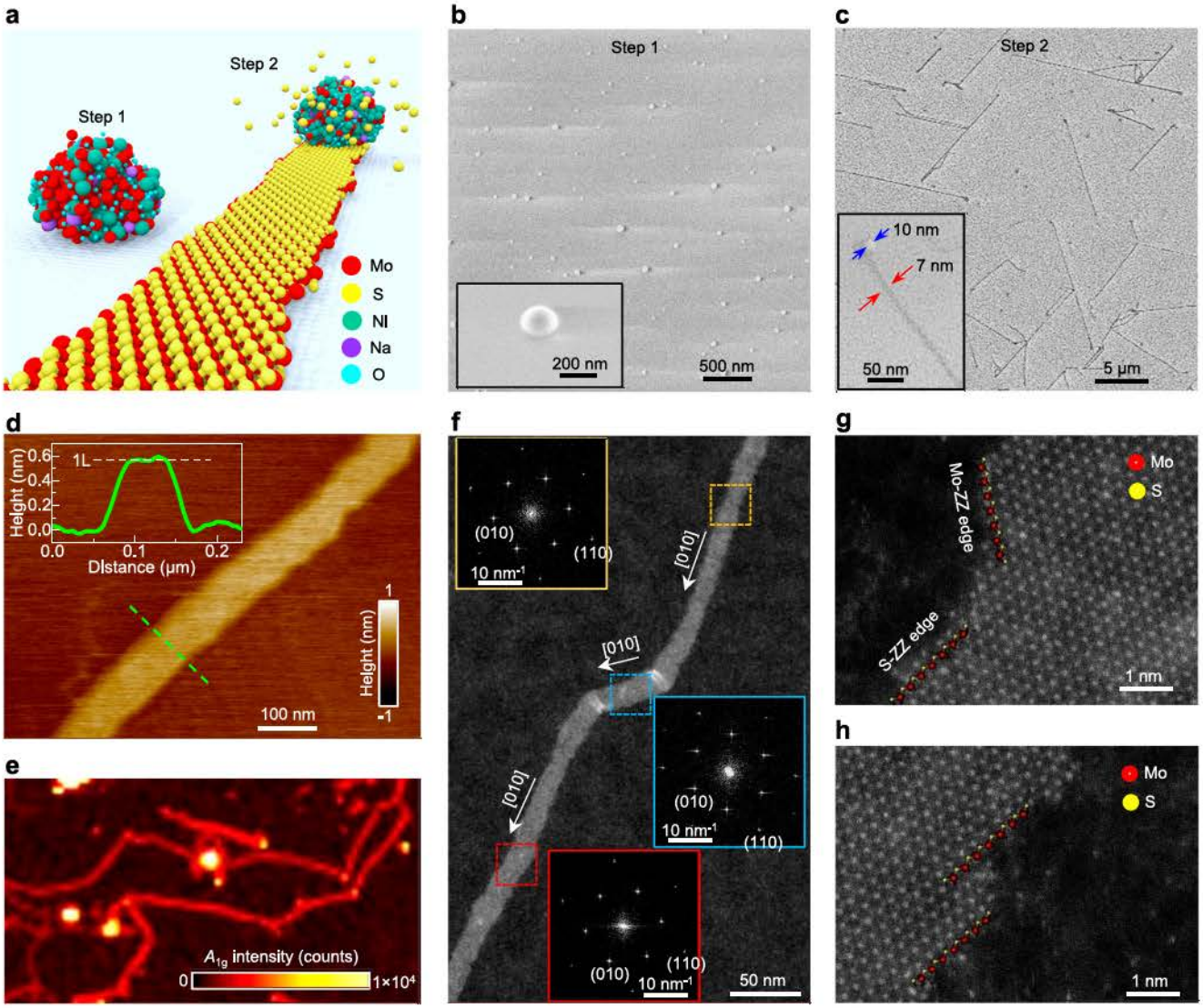


Fig. 1 | SL MoS_2 NRs grown on F-mica substrate. a Illustration of the two-step growth of SL MoS_2 NRs on F-mica substrate. Step 1: Seed NPs deposition. Step 2: Sulfurization of seed NPs resulting in precipitation of MoS_2 NR. Color code of the atoms: Mo (red), S (yellow), Ni (green), Na (purple), O (blue). **b** SEM image of Na-Mo-Ni-O seed NPs deposited for 1 min. Inset is the enlarged view of a NP. **c** SEM image of resulting MoS_2 NRs. Inset is the enlarged view of a NR width of ≈ 7 nm, comparable to the diameter of the tip Ni NP. **d** AFM image and height profile confirming the SL NR. **e** Raman intensity mapping of the A_{1g} peak of the SL MoS_2 NRs grown on F-mica. The color scales in (**d**, **e**) are linear. **f** Low-magnification HAADF-STEM image showing a length of ≈ 500 nm of a SL MoS_2 NR with two kinks. Insets with yellow, blue, and red borderlines are FFT patterns acquired from three different regions indicated by the dashed yellow, blue, and red boxes, respectively, indicating the growth axis of NR along the [010] crystal direction. The good alignment between the NR and diffraction pattern orientation, even in the kinked region (blue box), indicates the NR is single crystal. The kinks are used for strain analyses shown in Supplementary Fig. 19. **g**, **h** Atomic-resolution HAADF-STEM images showing edge structures of the upper side of the NR in the yellow box and lower side in red box, respectively, in (**f**). Although the FFT patterns and atomic resolution lattice show the edge is along the (110) plane, i.e., armchair, they are mostly terminated by zigzag structures resulting in steps. Source data are provided as a Source Data file.

Statistics on measured L and W of individual NRs grown under different sulfur vapor pressures (P_{sulfur}) (Supplementary Fig. 10) based on the SEM images (typical results shown in Supplementary Fig. 11) show a general trend that wider NRs grow longer than shorter ones (Fig. 3a). Since the NRs in general kept growing for $t > 180$ s (Supplementary Fig. 12), we assume the growth rate $V_g = L/t$ (when $t \le 180$ s), which means that wider NRs grow faster than narrower ones. It is also obvious in Fig. 3a that V_g increases with increasing P_{sulfur}, and the overall width distribution shifts to smaller width as P_{sulfur} increases (Fig. 3b), indicating that the smallest width of NR decreases with increasing supersaturation, i.e., shifting from 60–70 nm to 7–10 nm as P_{sulfur} increasing from 7.6×10^{-5} atm to 0.017 atm (see Suppl. Note 1 for more detailed discussion on width distribution).

In crystal growth theory, the growth rate can be described by $V_g = K_g(\Delta\mu/(k_B T))^n$, where K_g is the kinetic factor, $\Delta\mu$ the difference of chemical potentials, k_B the Boltzmann constant, T is the temperature, and n is the order (typically $n = 1$ or 2, corresponding to linear or quadratic growth of nanowires[30]). We consider the following steps for NR growth: i) impingement of S atoms into the liquid Na-Mo-Ni-O droplet from vapor phase due to difference in chemical potentials of S on vapor-liquid boundary, $\Delta\mu_{vl}$, which depends on the droplet diameter, ii) formation of Mo-S motifs and diffusion towards droplet-substrate boundary, and iii) crystallization of the liquid alloy on the substrate under the droplet by precipitation of Mo-S motifs due to chemical potentials difference $\Delta\mu_{ls}$ between the liquid droplet and solid MoS_2 NR, which also depends on droplet diameter. Unlike most

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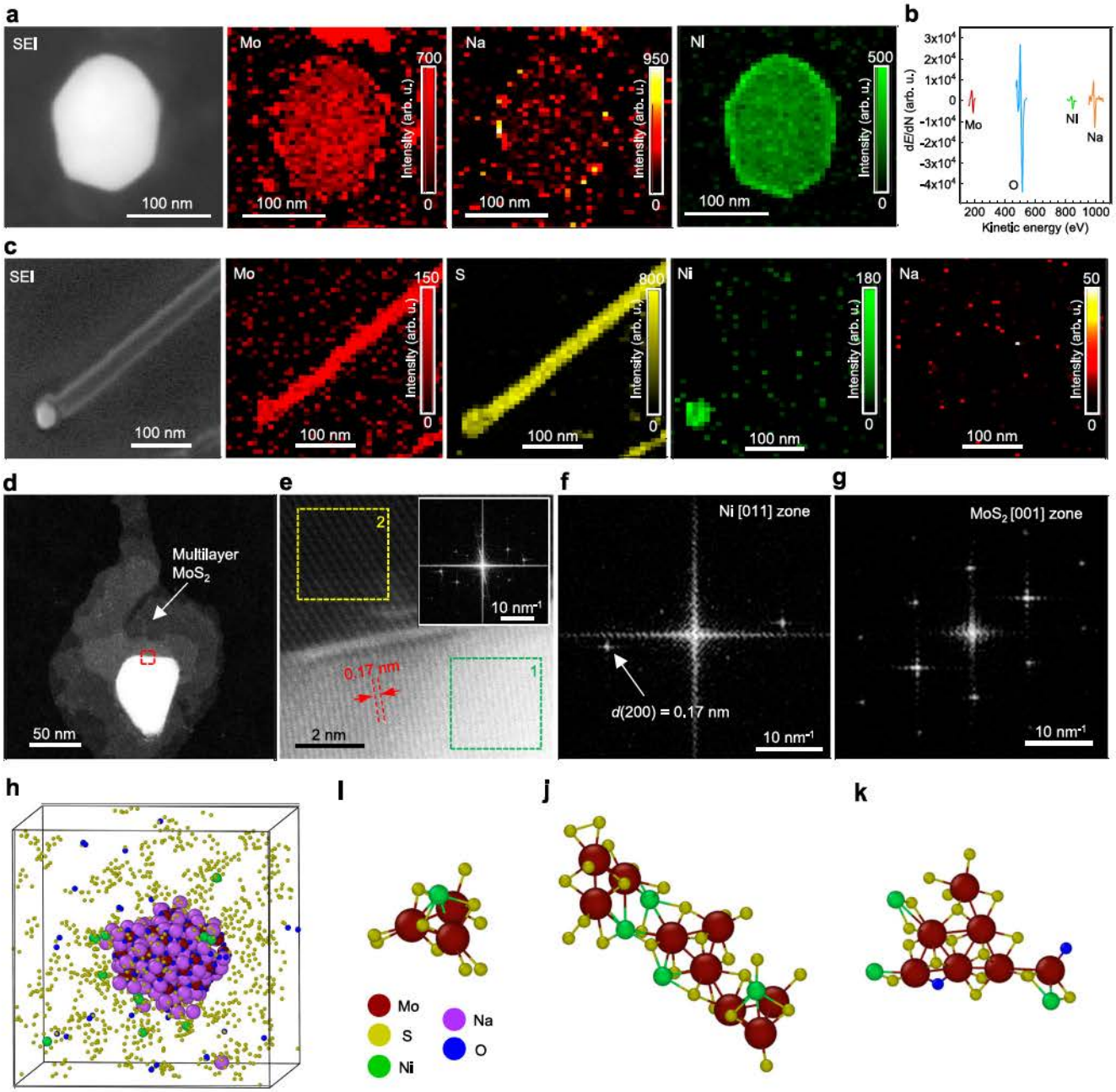


Fig. 2 | Evolution of seed NPs. **a** AES elemental mapping of Mo, Na, and Ni in a seed NP. SEI stands for secondary electron image. **b** Corresponding AES spectra of Mo (red), Na (orange), Ni (green), and O (blue) in the NP. Note that AES analysis the NPs were deposited on a SiO_2/Si substrate. **c** AES elemental mapping of Mo, S, Ni, and Na of a SL MoS_2 NR including the tip NP, transferred on a SiO_2/Si substrate after growth. The Mo and S signal at the NP tip area come from the multilayers of MoS_2 precipitated from the tip NP during the cooling down (see **d**). The color scales in (**a**) and (**c**) are linear. **d** Low-magnification STEM image of a MoS_2 NR emanating from the NP tip. **e** High resolution HAADF-STEM image of the region indicated by the dashed red box in (**d**), showing the interface between the tip NP and MoS_2. Inset is the FFT pattern of the whole image. **f**, **g** FFT pattern acquired from the dashed green box 1 and dashed yellow box 2 in (**e**), showing Ni [011] zone and MoS_2 [001] zone, respectively. **h** MD simulation of a Na-Mo-Ni-O NP interactions with S atoms, showing separation of Ni atoms from the other nanoparticles atoms after being equilibrated at 650 K for 6 ns. Color code of the atoms: Mo (dark red), S (yellow), Ni (green), Na (purple), O (blue). **i–k** Extended simulation of the Na-Mo-Ni-O NP in the S environment for 10 ns at 525 K, showing that Mo atoms in the NP start to form triangular local structural motifs with a Ni atom on the top (**i**), inter-connected triangular Mo local motifs (**j**), and embryo of two-dimensional hexagonal structures (**k**) Source data are provided as a Source Data file.

of the typical nanowire and SL TMD growth, there is no growth through the vapor-solid interface by direct incorporation of S atoms from vapor into the solid MoS_2 NR via terrace/edge impingement since there is no Mo vapor in the surrounding atmosphere. Hence, the only driving force for NR growth is the supersaturation of Mo-S motifs concentration in the droplet relative to solid NR, which elevates due to the impinging of S atoms from vapor phase into droplet. Considering S vapor as an ideal gas, the supersaturation of S atoms in the droplet with diameter (d) according to Gibbs-Thomson effect[30] could be expressed as

$$\Delta\mu_{vl}/(k_B T) = \Delta\mu_{\infty}/(k_B T) - 2\sigma_{vl}\Omega/(d k_B T) \quad (1)$$

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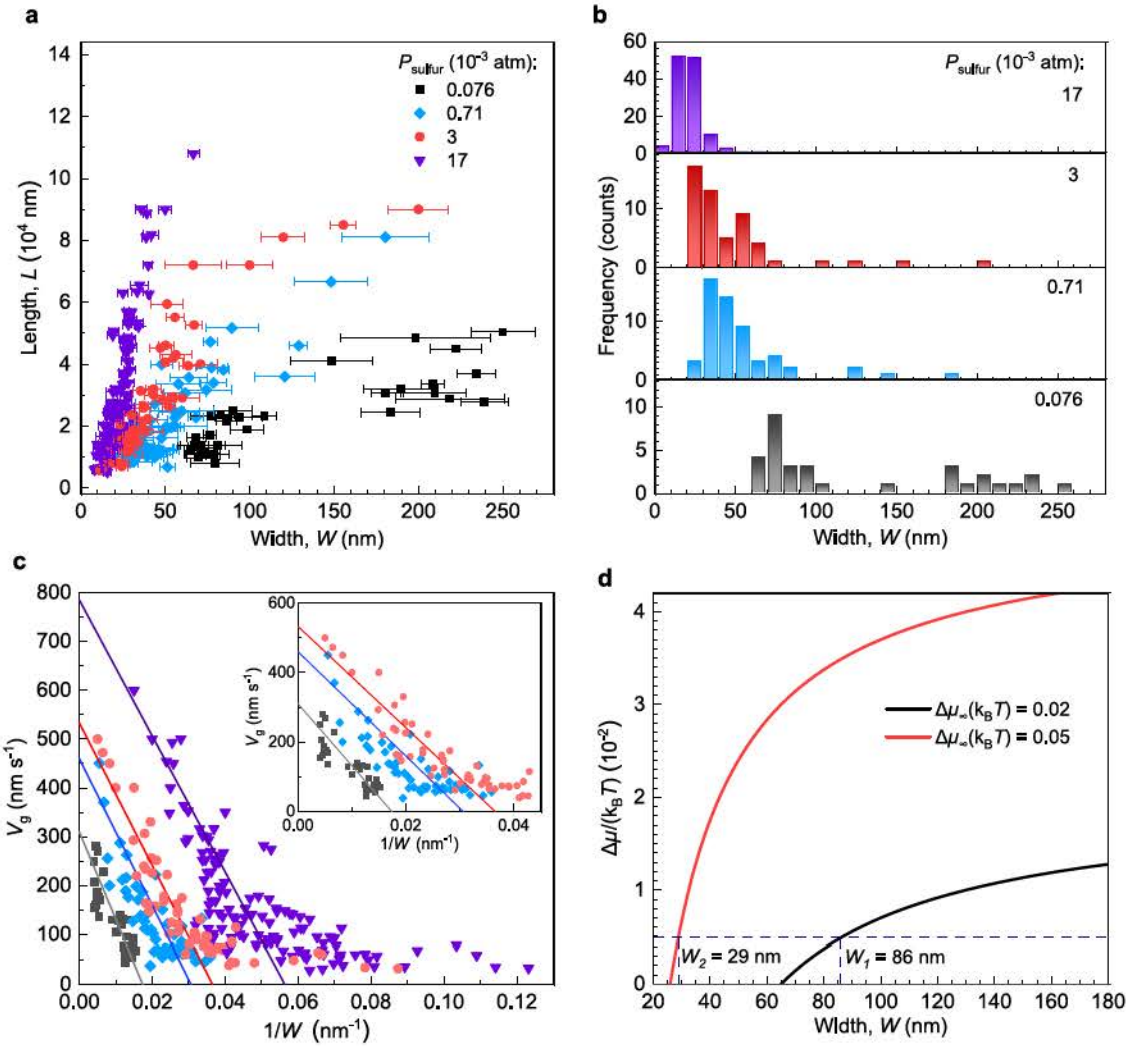


Fig. 3 | Precipitation-driven growth mechanism study. a Statistics of dependence of NR L on W with T_{sulfur} = 130, 170, 200, and 250 °C, corresponding to $P_{sulfur} = 7.6 \times 10^{-5}$ (black symbols), 7.1×10^{-4} (blue symbols), 0.003 (red symbols), and 0.017 atm (purple symbols), respectively. The NRs were grown for 180 s. The L vs W data of the NRs have been collected from three independent growth experiments at a given growth temperature (730 °C) for each P_{sulfur} (T_{sulfur}). The error bars represent the variations of width along the length of each NRs. **b** Histograms of width distribution of MoS_2 NR measured in (**a**), with $P_{sulfur} = 7.6 \times 10^{-5}$ (black), 7.1×10^{-4} (blue), 0.003 (red), and 0.017 atm (purple). **c** Growth rate V_g vs. 1/W with different P_{sulfur}. The color code corresponds to (**a**). Solid lines are fitting results using Eq. (2) with $n = 1$. Inset is enlarged view of $P_{sulfur} = 7.6 \times 10^{-5}$, 7.1×10^{-4}, and 0.003 atm. **d** Actual supersaturation $\Delta\mu_{vl}/(k_BT)$ vs. W according to Eq. (1) with $\Delta\mu_\infty/(k_BT) = 0.02$ (black curve) and 0.05 (red curve), corresponding to $P_{sulfur} = 7.6 \times 10^{-5}$ and 0.003 atm. Source data are provided as a Source Data file.

where $\Delta\mu_\infty/(k_BT) = \ln(P_{sulfur}/P_\infty)$ is the supersaturation between vapor and the droplet with infinite interface, P_∞ is the pressure on the infinite boundary of S and droplet, σ_{vl} the surface energy of Ni-Mo alloy (≈1.7 J m^{-2}), Ω is the S atomic volume in the droplet (0.0056 nm^3), $k_B = 1.380649 \times 10^{-23}$ J K^{-1}, and $T = 1003$ K is the synthesis temperature (see Suppl. Note 2 for detailed explanation). Correspondingly, V_g could be expressed as

$$V_g = K_g(\Delta\mu_\infty/(k_BT) - 2\sigma_{vl}\Omega/(dk_BT))^n \quad (2)$$

Furthermore, we found that the width of grown NRs is related to the diameter of the droplets by $d = W/(\sin\alpha) \approx W/0.75$, based on experimental observations of contact angle between NP droplet and the NR (Supplementary Fig. 2b). Hence, we analyzed the plots of V_g vs. 1/W for NRs grown under different P_{sulfur} (Fig. 3c). In the lower supersaturation region (i.e., larger W or d), V_g decreases linearly with increasing 1/W, and each plot could be best fitted using Eq. (2) with $n = 1$, from which nearly constant K_g values for all P_{sulfur} were extracted (Table 1). However, it should be noticed that as supersaturation gets higher, especially for $W < 20$ nm, V_g becomes almost independent of 1/W. Such a behavior is more dominant as P_{sulfur} gets higher (e.g., P_{sulfur} = 0.017 atm), which results in NRs with mostly narrower widths (Fig. 3c). Such a deviation from the Gibbs-Thomson effect could be attributed to the peculiarities of our NR formation. Specifically, in our method the seed droplet is the only source for Mo atoms, whose solubility in the Ni-Mo-O nanoalloy generally depends on the droplet size when $d < 20$ nm, which affects the supersaturation of Mo-S motifs

Table 1 | Kinetic factor (K_g), critical width (W_c) of NRs, and supersaturations at infinite boundary ($\Delta\mu_\infty/(k_BT)$) under different sulfur vapor pressure (P_{sulfur})

P_{sulfur} (atm) / T_{sulfur} (K)	K_g (10^{-5} m s^{-1})	W_c (nm)	$\Delta\mu_\infty/(k_BT)$ (T = 1003 K)
6×10^{-5} / 403	1.15	56	0.02
1×10^{-4} / 443	1.09	33	0.04
3×10^{-3} / 473	1.06	26	0.05
1.7×10^{-2} / 523	1.01	17	0.08

in the droplet and thereby on the growth rate. The reduction of the solubility of Mo in the droplet with decrease of droplet size can drive excess precipitation, resulting in increase of the growth rate[40]. Due to the lack of data for this quaternary nanoalloy phase diagram, further study is needed on this scope.

To further validate the growth mechanism of single-crystal SL MoS_2 NRs, as shown in the structural analysis (Fig. 1f, g), we analyzed the growth rate of NRs with different width but grown under the same supersaturation on the vapor-liquid interface. The interceptions of the fitting lines for V_g vs. $1/W$ with the $1/W$-axis in Fig. 3c, where growth rate is zero, provide critical widths $W_c = 2\sigma_{vl}\Omega/\Delta\mu_\infty$ under different P_{sulfur} according to Gibbs-Thomson model. For given values of W_c, we obtained the corresponding values of supersaturation $\Delta\mu_\infty/(k_BT)$ for different P_{sulfur} from Eq. (2) (Table 1). Using Eq. (1), the width dependence of actual supersaturation $\Delta\mu_{vl}/(k_BT)$ for $\Delta\mu_\infty/(k_BT) = 0.02$ and 0.05 were plotted (Fig. 3d), from which we found two width values $W = 86$ and 27 nm, with a same actual supersaturation, i.e., $\Delta\mu_{vl}/(k_BT) = 0.005$. They correspond to growth rate $V_g = 80$ and 25 nm s^{-1}, respectively, according to V_g vs. $1/W$ experimental graph in Fig. 3c. The results indicate that even though the interface area under the droplet of the wider NR (≈7400 nm^2) is about 10 times larger than that of the narrower one (≈730 nm^2), the V_g is only ≈ 3 times higher, which excludes the monocentric nucleation-mediated growth that could result in observed single crystal NRs. As shown above the growth rate, $V_g = K_g(\Delta\mu/(k_BT))$, depends linearly on $\Delta\mu$ with nearly constant kinetic factor K_g for all P_{sulfur} (Fig. 3c and Table 1), i.e. within the experimental range of applied supersaturations ($\Delta\mu < 0.25k_BT$) K_g can be considered independent of $\Delta\mu$. The linear dependence of V_g on $\Delta\mu$, as an experimental criterion, suggests that the formation of single crystal NR is the result of a continuous growth[41] through the attachment of atoms from the supersaturated droplet alloy to the NR edges with atomic steps/kinks as growth sites of the SL MoS_2 nucleus that grows axially under the droplet.

Growth of other TMD NRs

Beyond MoS_2, we used the two-step method above to grow other SL TMD NRs including $MoSe_2$, WS_2, and WSe_2 on F-mica substrates. $MoSe_2$ NRs were also precipitated from pre-deposited Na-Mo-Ni-O seed NPs, driven by selenium vapor (Supplementary Fig. 13). However, W-based TMD NRs are grown from pre-deposited seed NPs composed of Na-W-Mg-O, in which Mg plays a similar role as Ni (Supplementary Figs. 14–16), which points out the importance of nanoalloy composition.

Width-dependent optical properties of NRs

Next, we attempted to reveal whether the width as an additional degree of freedom enabled by NR geometry could endow tunable intrinsic properties of NRs. We found that the optical properties of NRs are influenced by their width. Specifically, at room temperature, the SL MoS_2 NRs show width-dependent photoluminescence (PL) from the A-exciton (Fig. 4a), which is further corroborated on the nanoscale with nano-PL characterization of individual NRs using tip-enhanced PL (TEPL) measurement. (Supplementary Fig. 17). The PL of the A-exciton blueshifts as the width of the NR decreases, similar to the trend observed in ref. 25. The microscopic origins of such width-dependent PL of the NRs could arise to an interplay between interior states and edge states. The energy blueshift is most pronounced in the range of 50–20 nm, where a linear dependence on $1/W^2$ is shown (inset of Supplementary Fig. 17c), indicating that the energy shift is due to the weak confinement on exciton center-of-mass in an infinite potential well[42–44]; while for NRs with width >50 nm, the energy shift could be dominant by the edge states, where strain relaxation, chemical termination, and or interaction with the substrate result in localized states as previously reported in nearfield studies of 2D crystallites[45,46]. Preliminary measurements show that such edge states may also exists in the MoS_2 NRs (Supplementary Fig. 18), and future measurements will focus on unraveling how these states contribute to the width-dependent effects observed here.

Quantum emission from strain-engineered TMD NRs

Further, strain engineering has been widely used for tailoring excitonic light-matter interactions and deterministically creating localized electronic states in 2D semiconductors[47–51] for potential applications. For our SL NR systems, analysis of atomic resolution HAADF-STEM imaging (Supplementary Fig. 19) reveals that they can accommodate from -14% (compressive) to +14% (tensile) strains. Optically, the SL TMD NRs preserve the strain-tunable intense excitonic light-matter interactions that are expected from SL 2D semiconductors[52]. Exploiting the observed mechanical robustness, we transferred as grown SL MoS_2 and WSe_2 NRs onto arrays of gold nanocones (inset of Fig. 4b)[49,50] to realize strain-induced localized states. The gold nanocones have an average height and base diameter of ≈ 100 nm, and a radius of curvature at the apex of ≈ 5 nm (Supplementary Fig. 20). For a SL MoS_2 NR with a width of 100 nm, the strain induced by being draped over the gold nanocone (Fig. 4b, red curve) generates a pronounced localized exciton state at 1.77 eV that is ≈ 2 × brighter with a narrower linewidth (≈25 meV) than the PL of the free A exciton at 1.97 eV. In contrast, such a strong feature is absent in the PL spectrum acquired from the unstrained NR (Fig. 4b, blue curve). Similar strain-localization of excitons at the same energy (1.77 eV) in SL MoS_2 NRs are observed at room temperature in nanoscale regions directly surrounding a NR draped over a nanocone by nano-PL imaging (Supplementary Fig. 21). In the time evolution of the PL spectra from the strained region (inset of Fig. 4b), the PL intensity and energy of the A exciton 1.97 eV does not show any temporal variation, which is expected for a state that is not localized; in contrast, emission from the strained region exhibits spectral wandering (i.e., random jumps to different energies) and intensity fluctuations on the timescale of seconds, which are both signatures of highly localized quantum dot-like states that can generate single photons[48]. This report introduces purely strain-induced quantum dot-like states in single-layer MoS_2 without the use of ion irradiation[53] (see Suppl. Note 3 for more detailed discussion).

Finally, using the same strain-engineering method on SL WSe_2 NRs, we confirm the ability to fabricate single photon emitters (SPEs) in the more confined geometry of the NRs. Figure 4c shows that bright, narrowband emitters emerge from localized strained regions of a ≈ 100 nm-wide SL WSe_2 NR that is draped over a similar array of gold nanocones (Supplementary Fig. 22a). Nano-PL imaging (inset of Fig. 4c, Supplementary Fig. 22b) reveals a series of bright emissive spots that each correspond to a region where the SL WSe_2 NR is draped over an individual cone. The selected spectrum in Fig. 4c is from the cone site that is marked in the inset. Similar spectra are observed from the other cone-induced strained sites, with emitter energies ranging from 1.6–1.7 eV (Supplementary Fig. 22b, c; 40–50 meV lower than the free exciton in unstrained WSe_2 at 4 K). The linewidths of the strain-induced emitters are >10 × narrower, and their intensities are 100–1000 × brighter than the localized state in the SL MoS_2 NR (Fig. 4b). The much larger intensity of these emitters permits measurement of the second-order autocorrelation function $g^{(2)}(\tau)$ using a Hanbury-Brown-Twiss interferometer (see Methods), which reveals clear photon antibunching with a purity of 80%–90% (Fig. 4d, Supplementary Fig. 23). The demonstration of antibunching in a TMD NR system sets the stage for further research into the width-dependent optical properties of the NRs (i.e., Fig. 4a) and the use of local strain engineering, specific to the NR geometry, to potentially create more homogenous populations of 2D semiconductor-based SPEs for broadscale quantum photonic applications.

In conclusion, we developed a facile method for direct growth of single-crystal SL TMD NRs, which allows controllable downscaling NRs widths down to 7 nm. Deconvolution of seed NP formation and NR

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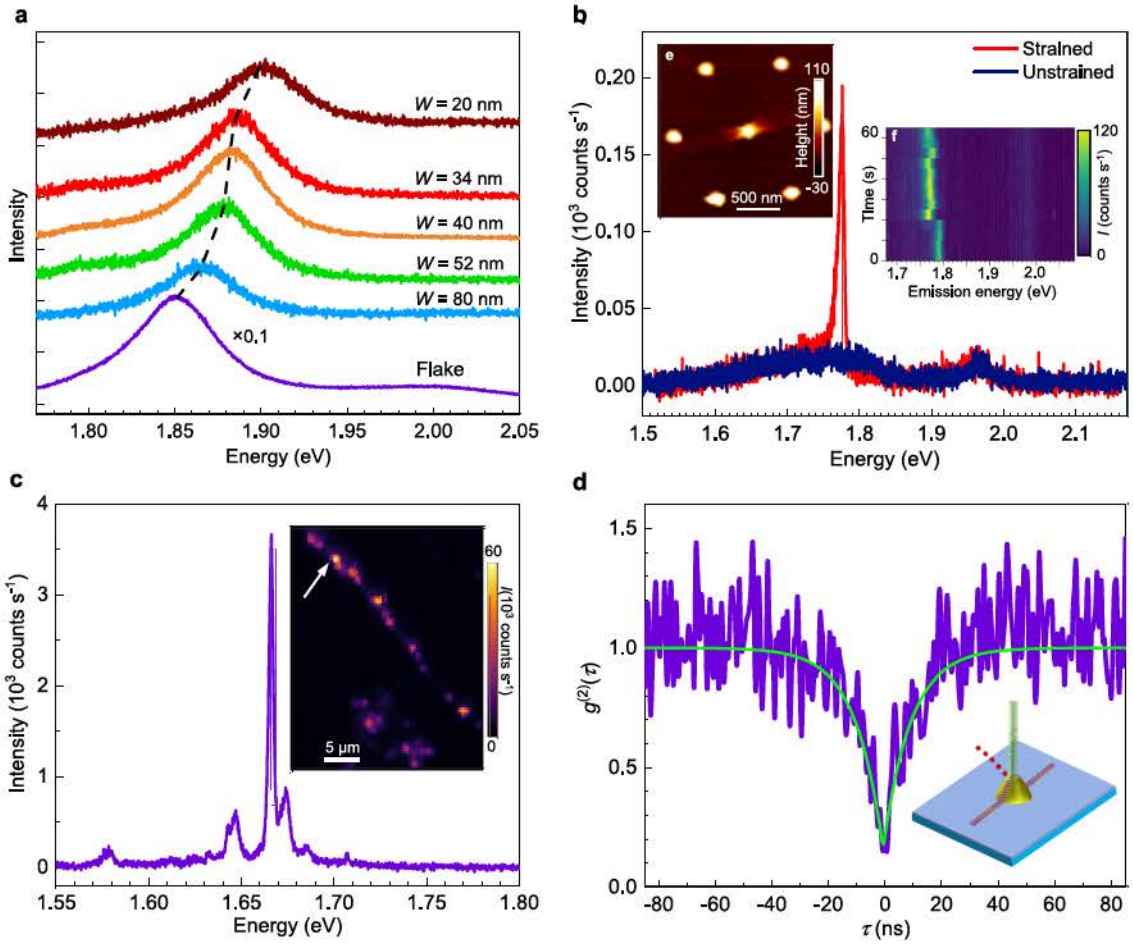


Fig. 4 | Width-dependent PL and quantum light emission from strained SL MoS_2 and WSe_2 NRs. a PL spectra of SL MoS_2 flake (purple curve) and NRs with width of 80 (blue curve), 52 (green curve), 40 (orange curve), 34 (red curve), and 20 nm (dark red curve) at room temperature. (**b**) PL spectra of a MoS_2 NR draped over an Au nanocone, from the region over the cone (strained, red curve) and unstrained region (dark blue curve) at $T = 4$ K. Inset (**e**): AFM topography of the SL MoS_2 NR draped over a cone. Inset (**f**): time-series of the PL spectra from strained region. **c** PL spectrum of one of the strained regions (indicated by the white arrow in the inset) of a SL WSe_2 NR on Au nanocones (see Supplementary Fig. 20) at $T = 4$ K. Inset: Mapping of integrated PL intensity from the WSe_2 NR. The color scales in (**b**) and (**c**) are linear. **d** $g^{(2)}(\tau)$ measurement of the emitter in (**c**) at 1.67 eV. The fitting of the $g^{(2)}(\tau)$ curve (green curve)[60] gives a single photon purity of 81%. Inset is the illustration of quantum light emission generated from strain-engineering NRs by nanocones. Source data are provided as a Source Data file.

growth process allows independent control over the width of the NRs via diameter of seed NPs. Unlike other 1D nanostructures such as whiskers and nanowires, we reveal a continuous growth mechanism for NRs with the growth rate that depends linearly on the reciprocal width of NRs due to the Gibbs-Thomson effect. The NRs show significant mechanical robustness that can accommodate up to ≈14% compression and tensile strain. The strain-induced localized quantum dot like states emit single photons in TMD NRs. The initial results on the SL WSe_2 nanoribbons demonstrate the great potential to realize precisely localized quantum light sources in NRs of TMDs for quantum optoelectronic applications.

Methods

Material synthesis

SL MoS_2 NRs were synthesized in a tube furnace system similar to ref. 18. Specifically, two separate argon gas lines are connected to the tube furnace, in which one directly goes to the reaction chamber (called F_{Ar}) while the other goes through a small bubbler, containing 2 mL of DI water to produce moisturized Ar (called $F_{Ar/H2O}$) before reaching the reaction tube, resulting in a moisturized Ar flow with ≈ 3000 ppm H_2O. In the first step, a mixture of MoO_2, NaBr, and Ni powder at a certain weight ratio was placed in an alumina boat, and a piece of freshly cleaved F-mica was placed ≈ 3 mm above the precursor. The precursor was heated to 730 °C under a gas flow of 40 cm^3 STP min^{-1} of F_{Ar} combined with 40 cm^3 STP min^{-1} of $F_{Ar/H2O}$ to allow seed NPs deposited on the F-mica. In the second step, the F-mica with deposited seed NPs was placed in another clean alumina boat and heated to 730–770 °C at the center of the tube furnace, and meanwhile, sulfur powder, placed at the upper stream, was heated separately by a heating belt to 130–270 °C to provide sulfur vapor, carried by a dry Ar flow of 80 cm^3 STP min^{-1} to sulfurize seed NPs. For the growth of $MoSe_2$ NRs, the same seed NPs were used, while selenium powders were heated to 350–420 °C to provide selenium vapor carried by a combination of 80 cm^3 STP min^{-1} Ar and 6 cm^3 STP min^{-1} H_2. For the growth of WS_2 and WSe_2 NRs, seed NPs were deposited from a mixture of WO_2, NaBr, and Mg precursor.

Characterization

The as-synthesized MoS_2 NRs were characterized using scanning electron microscopy (SEM, FEI QUANTA FEG 650, operating at 5 kV), atomic force microscopy (AFM, Bruker Dimension Icon), Raman spectroscopy (Renishaw inVia Raman microscope, with 1800 grooves mm^{-1} grating and a 532 nm laser, 1 mW laser power, as excitation source). AES was conducted in a PHI 700 scanning Auger nanoprobe

(Physical Electronics) at a working voltage and current of 10 kV and 10 nA. The samples for TEM and STEM characterization were prepared using a wet transfer process. Poly(methyl methacrylate) (PMMA) 495A4 (MicroChem) is first spun onto the F-mica substrate with monolayer crystals at 3500 rounds min^{-1} for 60 s. The PMMA-coated substrate was then floated on deionized (DI) water to detach the PMMA film with the ribbons from the substrate. The floating PMMA film was scooped onto a 3 nm-thick amorphous carbon film supported by a Au TEM grid, which was subsequently soaked in acetone for 12 h to remove PMMA and get a clean sample surface. The transferred samples were finally annealed at 200 °C in vacuum to get rid of the solvent. STEM images were acquired at 60 kV using a Nion UltraSTEM equipped with a probe aberration corrector (the convergence angle was 31 mrad). The inner and outer collection angle of the HAADF detector were 86 and 200 mrad, respectively. To enhance the signal-to-noise ratio, atomic-resolution STEM images were blurred using a 2D Gaussian distribution.

Device fabrication

The FET devices were fabricated using electron-beam (e-beam) lithography patterning, performed by Nanobeam n64 Electron Beam Writer system. The electron beam was operated at an accerlating voltage of 80 kV. Metal electrodes of Bi/Au (20 nm/40 nm) were deposited on top of MoS_2 NRs in Angstrom EvoVac Deposition system, the deposition vacuum level is $<5 \times 10^{-7}$ Torr. Transport measurements were performed in a CTI-Cryogenics Model 22 Refrigerator using a Keysight B1500A Semiconductor Device Parameter Analyzer.

MD simulations

MD simulations were carried out using LAMMPS[54], with a recent universal neural network interatomic potential called PFP as implemented on the Matlantis™ platform[55]. PFP has been demonstrated with DFT-level accuracy and has been successfully applied in modeling a wide range of materials. Our MD simulations employed the v4.0.0 version of PFP with the "CRYSTAL_U0" mode. The initial structures of nanoparticles were prepared using the packmol softwar[56]. Nanoparticle with oxygen contains 500 atoms, with 143 Na atoms, 57 Mo atoms, 14 Ni atoms and 286 Oxygen atoms. Nanoparticle without oxygen contains 133 Na atoms, 33 Mo atoms, and 13 Ni atoms. The initial configuration was equilibrated at 300 K for 1 ns, using constant temperature, constant volume MD simulations (NVT), with sufficient vacuum regions (at least >24 Å, the effective interaction range from the graph convolution operations) to avoid image interactions. S atoms were randomly inserted into the simulation box to observe their interactions with nanoparticles. All subsequent simulations at different temperatures were carried out under NVT conditions. Structures were visualized using the Ovito software[57].

Strain analysis

First, we performed filtering to improve the signal-to-noise ratio in the atomic resolution STEM images. Fast Fourier transformation (FFT) has been performed on the red boxes in Supplementary Fig. 19c, d. Reciprocal space around the first-order Bragg peaks is chosen to conduct the inverse FFT, which effectively enhances the contrast of atomic columns and facilitates further detection. The second step is to locate the positions of all the atomic columns that are of interest in the filtered HAADF-STEM images, as shown in Supplementary Fig. 19e, f. The peak finding algorithm offered by the ATOMAP Python library[58] is leveraged to identify initial atomic positions, with the relative intensity threshold and minimum peak separation serving as inputs. The initial positions are further refined, first by determining the center-of-mass and then by two-dimensional (2D) Gaussian peak fitting. This allows for a precise determination of the 2D atomic column positions. Third, two sets of non-parallel planes are selected to measure the relative variations in plane distance compared to the reference value obtained away from the MoS_2 kinks at the middle section in Supplementary Fig. 19b. Previously, the TopoTEM Python library[59] has been employed for strain calculation via interplanar distance variation with the reference value in HAADF-STEM images for crystal with orthogonal axes. Building upon this, we perform a detailed basis vector transformation from inter-planar direction to orthogonal basis in MoS_2 strain study. This enables the computation of MoS_2 displacement field on an orthogonal basis, which subsequently leads to the calculation of in-plane hydrostatic strain mapping results, as shown in Supplementary Fig. 19g, h. The in-plane hydrostatic strain variation along the lines depicted in Supplementary Fig. 19e, f has been calculated. The average value is plotted along the diagonal direction, which reveals a higher magnitude of hydrostatic strain closer to the MoS_2 kinks, as shown in Supplementary Fig. 19i.

Nanocone fabrication

Substrates with nanocone stressor arrays were fabricated according to the process outlined by Yanev et al.[50]. A base layer of gold was evaporated onto a silicon wafer, followed by electron-beam lithography and Al_2O_3 deposition. After lift-off, disks of alumina were left behind, which served as a hard mask during the final argon ion milling step. See ref. 44 for more details and discussion of the fabrication process.

NR transfer on nanocones

A piece of polydimethylsiloxane (PDMS) film ($1 \times 1\,cm^2$, Gel-Pek) is first stamped onto the as-grown NRs on F-mica substrate. The NRs are detached from the substrate and picked up by PDMS film assisted by DI water. The PDMS film with NRs is then stamped on a glass slide, which is subsequently mounted on a 2D materials transfer stage (HQ Graphene) with targeting substrate with fabricated Au nanocones. Specific NR is aligned on Au cone arrays through an optical microscope and placed on top of them. Finally, the stage is heated to 65 °C to lift off the PDMS film, leaving the NR on Au nanocones.

TEPL measurement

Near-field optical measurements were carried out using a commercially available AFM system (OmegaScope; Horiba Scientific) coupled to a Raman spectrometer (LabRAM HR Evolution; Horiba Scientific). In this setup, a 594 nm laser was focused onto the tip of a gold coated AFM probe (AppNano Omni™ TERS-NC-Au) after passing through a long working distance microscope objective (100×, 0.7 NA) mounted at an angle (25° from horizontal). Enhanced PL from the sample was collected through the same objective. Spectra were taken with the tip in contact and also slightly retracted from the surface at each location. This allows for a pixel-wise subtraction of the far-field background, which was used to produce the TEPL maps shown in this work.

Low-temperature PL measurement

Low-temperature spectroscopy was conducted using a homebuilt confocal microscope. Light is focused on samples mounted in a Montana Instruments S50 cryostat using a Nikon Plan Fluor 40× long working distance objective. Excitation for MoS_2 was done with a 532 nm (Cobalt Samba™ 100) diode laser. Laser power on the sample was measured at 20 μW. The excitation source used for the WSe_2 spectra and $g^{(2)}(\tau)$ measurement was a 78 MHz pulsed supercontinuum laser (NKT Photonics SuperK EXTREME). An excitation wavelength of 532 nm was selected using an acoustic optical modulator and a tunable short-pass filter. The laser power on the sample was 60 nW. Integrated PL intensity was collected with a Micro Photon Devices (MPD) APD, and spectra were collected with a Horiba iHR320 spectrometer and a cooled Andor iDus CCD camera.

Data availability

The data that support the findings of this study are available from the corresponding authors upon request. Source data are provided with this paper.

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Author contributions

A.R.H. conceptualized and designed research and suggested growth mechanism. A.R.H. and X.L. planned the experiments and studied the growth mechanism. X.L. developed the synthesis method. Sh.W. and L.M.S. contributed to the NR synthesis. X.L. and Sh.W. developed NR transferring method. P.J.S., J.H., and A.R.H. initiated QE research. P.J.S. and N.B. planned the QE research. E.Y. and P.J.S. fabricated the nanocones and performed near-field measurements. S.W., J.S., M.S., and N.B. performed low-temperature QE property study. Q.-J.L. and J.L. performed the MD simulation. R.R.U. performed STEM imaging. Sh.W. performed AFM measurement. Y.S., Y.Z., and Y.Y. performed strain analysis. A.R.H. and X.L. drafted the manuscript. All the authors contributed to the analysis of data and writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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