



# Optical fibres with embedded two-dimensional materials for ultrahigh nonlinearity

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**Nonlinear optical fibres have been employed for a vast number of applications, including optical frequency conversion, ultrafast laser and optical communication<sup>1–4</sup>. In current manufacturing technologies, nonlinearity is realized by the injection of nonlinear materials into fibres<sup>5–7</sup> or the fabrication of microstructured fibres<sup>8–10</sup>. Both strategies, however, suffer from either low optical nonlinearity or poor design flexibility. Here, we report the direct growth of MoS<sub>2</sub>, a highly nonlinear two-dimensional material<sup>11</sup>, onto the internal walls of a SiO<sub>2</sub> optical fibre. This growth is realized via a two-step chemical vapour deposition method, where a solid precursor is pre-deposited to guarantee a homogeneous feedstock before achieving uniform two-dimensional material growth along the entire fibre walls. By using the as-fabricated 25-cm-long fibre, both second- and third-harmonic generation could be enhanced by ~300 times compared with monolayer MoS<sub>2</sub>/silica. Propagation losses remain at ~0.1 dB cm<sup>-1</sup> for a wide frequency range. In addition, we demonstrate an all-fibre mode-locked laser (~6 mW output, ~500 fs pulse width and ~41 MHz repetition rate) by integrating the two-dimensional-material-embedded optical fibre as a saturable absorber. Initial tests show that our fabrication strategy is amenable to other transition metal dichalcogenides, making these embedded fibres versatile for several all-fibre nonlinear optics and optoelectronics applications.**

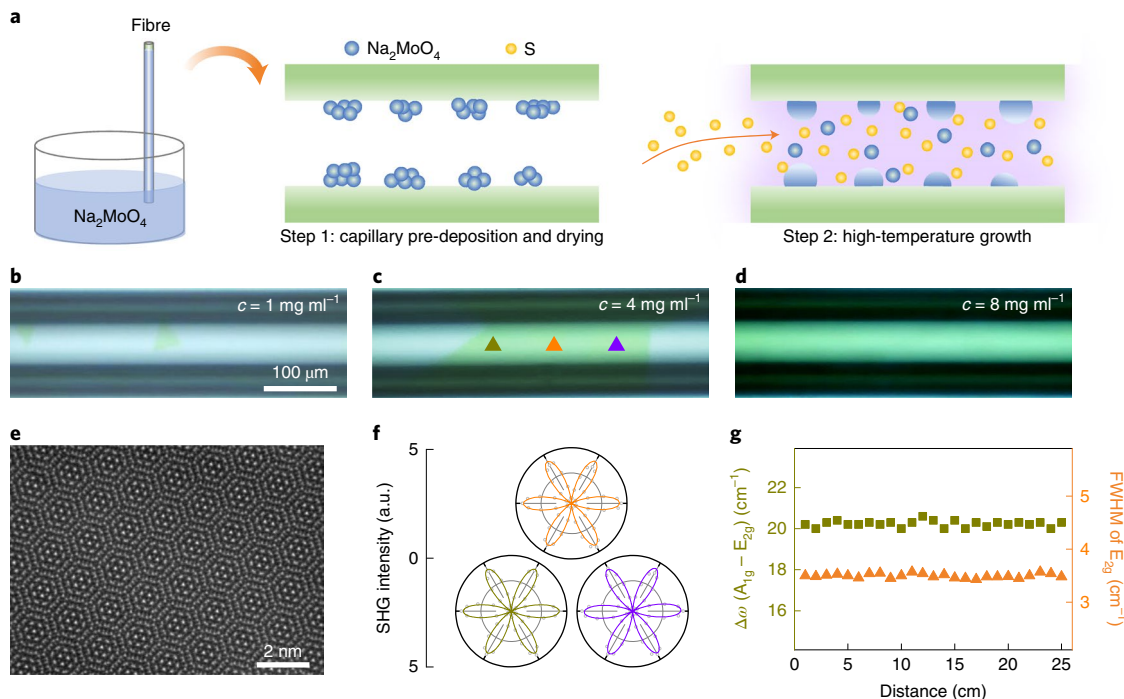
The superiority of two-dimensional (2D) materials used in nonlinear optical fibres is that (1) the atomically thin layer does not destroy the high-quality waveguide modes in the fibres and (2) the enhanced light–2D material interaction within the fibres can induce ultrahigh nonlinear optical responses<sup>12–14</sup>. Previously, 2D materials were mainly attached to optical fibres by transfer techniques, unfortunately suffering from distortions in propagation capacity, shorter light–material interaction length and difficulty in batch manufacturing<sup>15–17</sup>. Owing to the great progress made in 2D material growth, it is now possible to directly fabricate graphene optical fibres, wherein the feedstock is in the gas phase and therefore relatively easy to diffuse into narrow holes for homogeneous growth<sup>18</sup>. However, in transition metal dichalcogenide (TMD)-embedded optical fibre growth, the typical feedstocks are solid precursors and thus very difficult to transfer into the fibre holes effectively and homogeneously<sup>19–22</sup>.

To alleviate this dilemma, the key design aspect of our method lies in the pre-deposition step, as shown in Fig. 1a. Mo sources were filled into the fibre holes by the capillarity of the Na<sub>2</sub>MoO<sub>4</sub> aqueous solution. After that, the fibre was loaded into a chemical vapour deposition (CVD) furnace to bake for 30 min at a low temperature of 110 °C for dewetting, and the Na<sub>2</sub>MoO<sub>4</sub> precursors gather into clusters and are randomly coated onto the inner fibre walls, serving as a storage site for the Mo sources (Fig. 1a, middle). Subsequently, at a high growth temperature of ~820 °C, Na<sub>2</sub>MoO<sub>4</sub> clusters on the hole walls transform into liquid droplets and continuously release Mo precursors under vapourization in the confined narrow space of the fibre holes<sup>23</sup>. Sulfur vapour was carried by argon gas from the upstream direction via a low-pressure pump and then uniformly mixed with Na<sub>2</sub>MoO<sub>4</sub> vapour for the subsequent homogeneous MoS<sub>2</sub> growth (Fig. 1a, right, and Supplementary Note 1).

In our two-step growth method, the MoS<sub>2</sub> coverage and thickness can be facily tuned by the concentration (*c*) of Na<sub>2</sub>MoO<sub>4</sub> in the solution. When we increased the solute concentration from 1 to 4 to 8 mg ml<sup>-1</sup>, the MoS<sub>2</sub> gradually enlarged from 40 μm triangles (Fig. 1b), 250 μm triangles (Fig. 1c) and a continuous film (Fig. 1d), respectively. A further increase in the Na<sub>2</sub>MoO<sub>4</sub> concentration led to the growth of few-layered MoS<sub>2</sub> (Supplementary Figs. 1 and 2). Under our optimized conditions, we could readily realize homogeneous MoS<sub>2</sub> growth onto the hole walls of a long optical fibre up to 25 cm (Supplementary Fig. 3). Further, we also carried out controlled experiments, in which instead of the pre-deposition of Mo precursor onto the walls of the fibre holes, MoO<sub>3</sub> was directly sent from outside the fibres (as performed in prevailing growth methods<sup>24</sup>); we found that only a few distributed MoS<sub>2</sub> islands could be grown using this strategy (Supplementary Fig. 4). Thus, the pre-deposition of the Mo source in the fibre holes is the key step for the successful growth of large and uniform MoS<sub>2</sub> films onto the fibres.

A high-quality MoS<sub>2</sub> film grown in the confined spaces of the fibre holes is comparable to that grown on flat substrates in free space, which can be fully confirmed by the photoluminescence (PL) mapping (Supplementary Fig. 5) data of the individual triangular domain. These triangles are single-crystal domains, as proven by the parallel polarization second-harmonic-generated (SHG) patterns at different positions of the single domain (Fig. 1f). Further, we etched away the fused silica fibre and transferred the MoS<sub>2</sub> film

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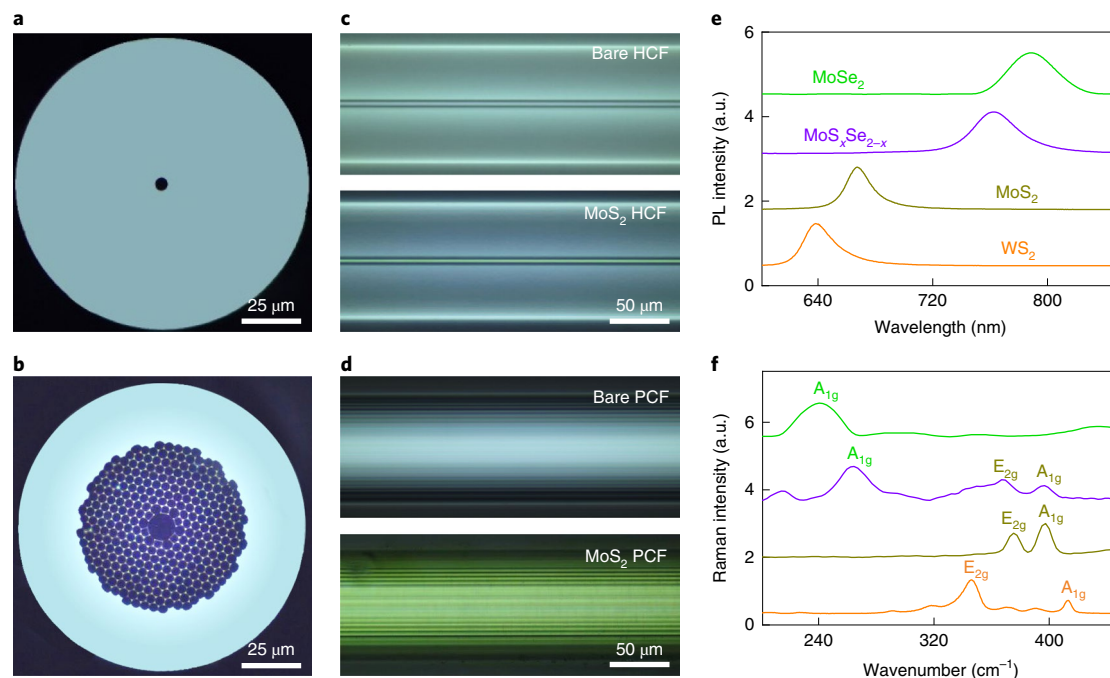
**Fig. 1 | Two-step growth of a high-quality, uniform, monolayer MoS<sub>2</sub>-embedded optical fibre.** **a**, Schematic of the designed two-step growth method, consisting of the pre-deposition of Na<sub>2</sub>MoO<sub>4</sub> onto the walls of the fibre holes by capillary filling and low-temperature drying followed by high-temperature growth. **b–d**, Dependence of MoS<sub>2</sub> coverage on the Na<sub>2</sub>MoO<sub>4</sub> aqueous solution concentrations of 1 mg ml<sup>-1</sup> (**b**), 4 mg ml<sup>-1</sup> (**c**) and 8 mg ml<sup>-1</sup> (**d**), respectively. **e**, STEM image of collapsed tube-like MoS<sub>2</sub> transferred onto a hollow grid, showing the high crystallinity of the as-grown MoS<sub>2</sub> without detectable defects. **f**, Parallely polarized SHG patterns at the different positions labelled in **c**, revealing that the whole MoS<sub>2</sub> domain is a large single crystal. **g**, Statistical  $\Delta\omega$  ( $A_{1g} - E_{2g}$ ) (the frequency difference of Raman shift ( $\omega$ ) between  $A_{1g}$  peak and  $E_{2g}$  peak; left y axis) and FWHM values of the  $E_{2g}$  peak (right y axis) plotted as a function of the numbered sites on the MoS<sub>2</sub>-embedded fibre along the whole length of 25 cm. The slight variations prove that the MoS<sub>2</sub> film is uniformly grown onto the fibre.

onto a hollow grid for scanning transmission electron microscopy (STEM) characterization with atomic resolution. When the tube-like MoS<sub>2</sub> monolayer collapsed (Supplementary Fig. 6), a clear moiré pattern of the MoS<sub>2</sub> bilayers became evident without observable defect sites (Fig. 1e), reconfirming the formation of high-quality MoS<sub>2</sub> domains. Furthermore, marginal variations in the frequency difference between  $A_{1g}$  and  $E_{2g}$  as well as the narrow distribution of the full-width at half-maximum (FWHM) values of the  $E_{2g}$  peak in the Raman spectra along the entire 25-cm-long fibre indicated that the as-grown monolayer MoS<sub>2</sub> film was highly uniform<sup>25,26</sup> (Fig. 1g).

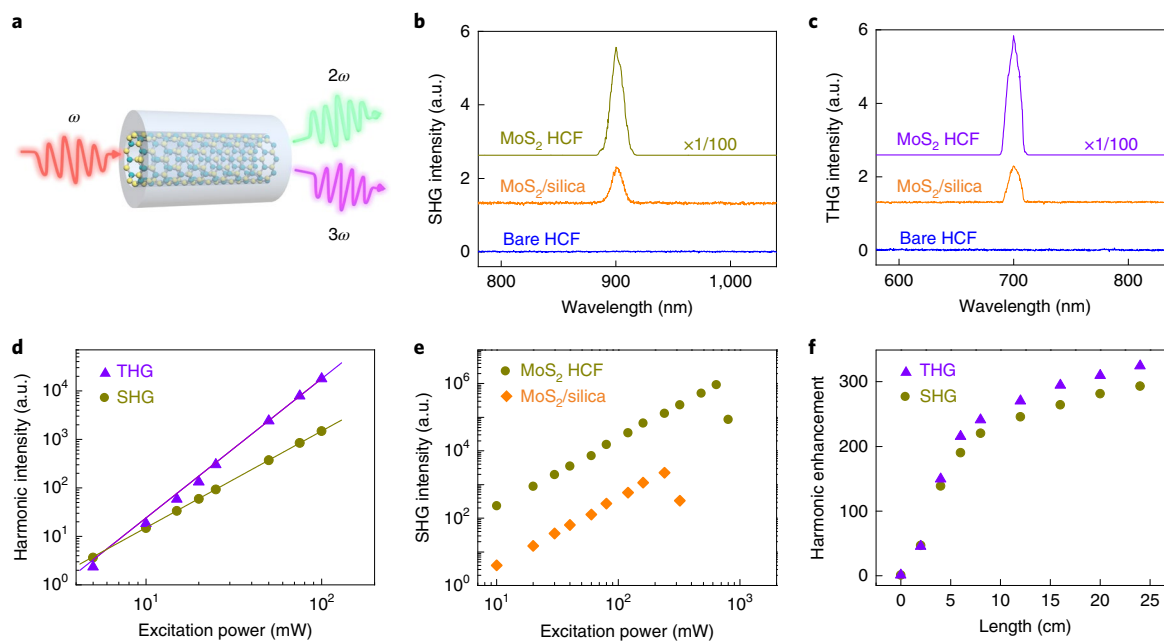
Our growth method for the MoS<sub>2</sub>-embedded optical fibre has been proven to be universal for different fibre structures and 2D materials. Two types of commonly used microstructured optical fibres, namely, hollow capillary fibres (HCFs) with different hole diameters (ranging from 5 to 15 to 50  $\mu\text{m}$  (Fig. 2a and Supplementary Fig. 7)) and photonic crystal fibres (PCFs) with air or solid cores (Fig. 2b and Supplementary Fig. 8), were used to successfully grow uniform monolayer MoS<sub>2</sub> films (Fig. 2c,d). Since the diameter of the hole varies in different fibres, the amount of Mo precursor pre-deposited on the inner hole walls is accordingly adjusted via the solute concentration (that is, the smaller the diameter, the higher the Na<sub>2</sub>MoO<sub>4</sub> concentration). This growth method has also been used to fabricate fibres with different TMDs simply by varying the feedstocks of the transition metals (for example, Na<sub>2</sub>MoO<sub>4</sub> and Na<sub>2</sub>WO<sub>4</sub>) and chalcogen (for example, S and Se) species with adjusted growth temperatures (Methods). The Raman and PL spectra of these obtained homogeneous TMD-embedded fibres demonstrate the construction of optical fibres embedded with MoS<sub>2</sub>, WS<sub>2</sub> and MoSe<sub>2</sub>, as well as alloys such as MoS<sub>x</sub>Se<sub>2-x</sub> (Fig. 2e,f).

Due to their intact light propagation mode (Supplementary Fig. 9), strong light–matter interaction, various fibre structures and rich TMD species, 2D-material-embedded optical fibres afford ultra-high optical nonlinearity and plentiful function/design flexibility, which intuitively yields different potential applications in the wide scopes of nonlinear optics. Since the optical nonlinear susceptibility contains two parts, namely, the real part and imaginary part, the corresponding applications can be divided into two types: (1) nonlinear wavelength conversion (for example, harmonic generation and four-wave mixing frequency), which is the typical demonstration of the real part, and (2) nonlinear absorption (for example, saturable absorption and multiphoton absorption), which is the typical demonstration of the imaginary part<sup>27</sup>. By selecting proper fibre structures and 2D material species, applications related to the real and imaginary parts have been demonstrated using our TMD-embedded fibres.

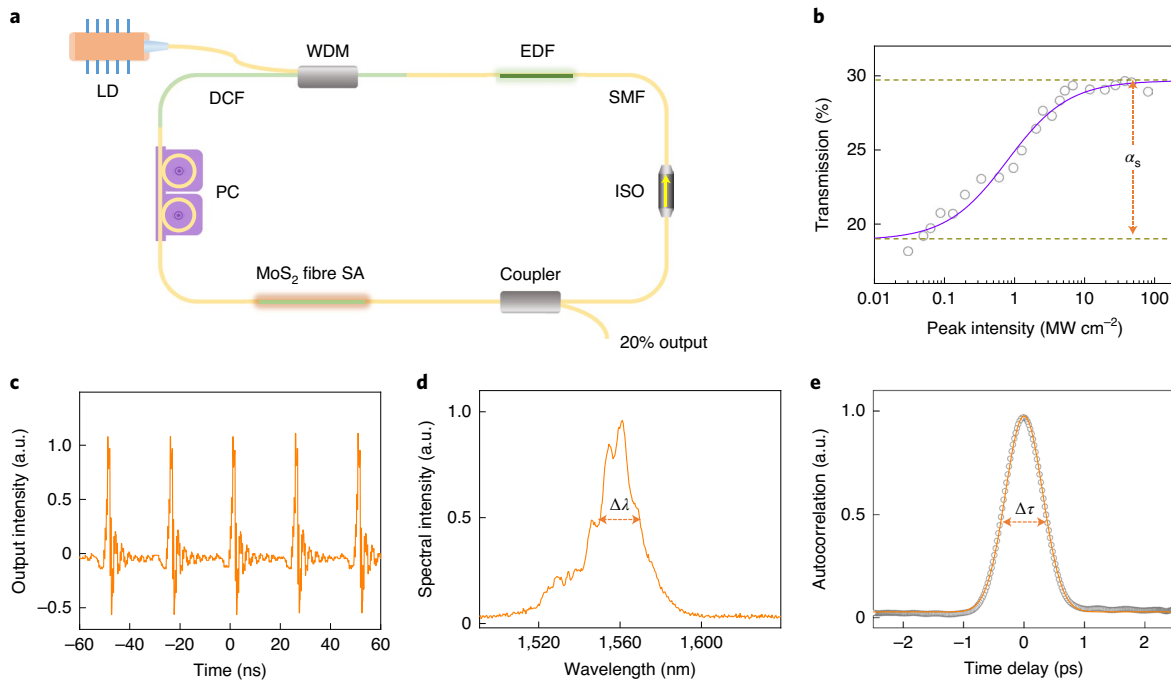
We demonstrated that the nonlinear optical harmonic generation (SHG and third-harmonic generation (THG)), as shown in Fig. 3a) from the TMD-embedded fibres was greatly enhanced compared with that from conventional optical fibres, wherein the SHG and THG were fairly weak and rarely observed before. With regard to practical nonlinear wavelength conversion, the real part of nonlinear susceptibility needs to be utilized, and simultaneously, it should be ensured that the excitation and emission of photons is within the bandgap of the materials. Since the MoS<sub>2</sub> monolayer has a direct bandgap of  $\sim 670$  nm, we chose longer wavelengths (1,800 and 2,100 nm) to generate the SHG and THG (900 and 700 nm, respectively), where the incident and generated photon energies were both smaller than the bandgap to ensure low propagation loss at both the excited and emitted wavelengths (Supplementary



**Fig. 2 | Two-dimensional-material-embedded optical fibres with diverse fibre structures and material species.** **a, b**, Optical images of a HCF with a core diameter of  $\sim 5 \mu\text{m}$  (**a**) and a PCF with a hollow-core honeycomb structure (**b**). **c, d**, The corresponding side views of HCF (**c**) and PCF (**d**) shown in **a** and **b**, respectively. Higher contrast was observed after the growth of  $\text{MoS}_2$  (bottom) compared with the bare ones (top). **e, f**, PL (**e**) and Raman (**f**) spectra of the fibres embedded with as-grown  $\text{MoSe}_2$  (green),  $\text{MoS}_2$  (dark yellow),  $\text{WS}_2$  (orange) and  $\text{MoS}_x\text{Se}_{2-x}$  (violet). The PL and Raman spectra are shifted vertically for clarity.



**Fig. 3 | Greatly enhanced harmonic generation in  $\text{MoS}_2$ -embedded HCF.** **a**, Schematics of SHG ( $2\omega$ ) and THG ( $3\omega$ ) in  $\text{MoS}_2$ -embedded HCF. **b, c**, SHG (**b**) and THG (**c**) spectra of  $\text{MoS}_2$  HCF;  $\text{MoS}_2$  on flat, fused silica substrate ( $\text{MoS}_2/\text{silica}$ ); and bare HCF under 1,800 and 2,100 nm excitation, respectively. With the strong light-matter interaction in 25-cm-long  $\text{MoS}_2$  HCF, both SHG and THG can be enhanced by  $\sim 300$  times compared with monolayer  $\text{MoS}_2/\text{silica}$ . For comparison, no apparent SHG or THG signal can be observed in bare HCF. The SHG and THG spectra are shifted vertically for clarity. **d**, Excitation-power dependence of SHG and THG intensities for  $\text{MoS}_2$  HCF, showing the as-expected quadratic and cubic laws. **e**, Measured SHG intensity of 12-cm-long  $\text{MoS}_2$  HCF and  $\text{MoS}_2/\text{silica}$  as a function of the excitation power under 800 nm excitation. Benefiting from the larger mode area and evanescent light coupling, the damage threshold of  $\text{MoS}_2$  HCF is about three times higher than that of  $\text{MoS}_2/\text{silica}$ . **f**, Fibre-length-dependent SHG and THG enhancements in  $\text{MoS}_2$  HCF with respect to  $\text{MoS}_2/\text{silica}$ , showing a general monotonic increase and saturation behaviour with the fibre length.



**Fig. 4 | Ultrafast laser based on MoS<sub>2</sub>-embedded PCF.** **a**, Schematic of all-fibre mode-locked laser with MoS<sub>2</sub>-embedded fibre as an SA. The optical components consist of a laser diode (LD), wavelength-division multiplexer (WDM), single-mode fibre (SMF), dispersion compensation fibre (DCF), erbium-doped fibre (EDF), isolator (ISO) and polarization controller (PC). **b**, Transmission measurement of the MoS<sub>2</sub> PCF for  $\alpha_s \approx 10\%$  and saturation peak intensity of  $0.8 \text{ MW cm}^{-2}$ . The solid curve is fitted according to the experimental data (circles). **c**, Output pulse train with  $\sim 24 \text{ ns}$  interval ( $\sim 41 \text{ MHz}$  repetition rate). **d**, Spectrum of the output laser with  $\Delta\lambda \approx 19 \text{ nm}$ . **e**, Autocorrelation trace with FWHM ( $\Delta\tau$ ) of  $\sim 720 \text{ fs}$  fitted by a Gaussian function, corresponding to a pulse duration of  $\sim 500 \text{ fs}$ .

Fig. 10). As per this design, we observed notably enhanced harmonic generation in monolayer MoS<sub>2</sub>-based HCF with a length of up to  $\sim 25 \text{ cm}$ , which is more than 300 times higher than that obtainable from monolayer MoS<sub>2</sub> on a flat, fused silica substrate (Fig. 3b,c). The square and cubic dependence of the emission intensity on the excitation power reveals that the observed signals are from the SHG and THG, respectively (Fig. 3d).

Another great advance of 2D-material-embedded optical fibres for nonlinear wavelength conversion is that they yield higher damage threshold power. Unlike the direct focusing of a laser on nonlinear materials in free space, light in the fibre can fill the waveguide core, typically resulting in a larger light-field area. Therefore, fibre optics can withstand much higher power before the nonlinear medium is damaged. Besides, the 2D materials on the fibre walls typically interact with the evanescent light, which is often weaker than light in the fibre core. For the HCF with a hollow core size of  $\sim 50 \mu\text{m}$  and wall thickness of  $\sim 40 \mu\text{m}$ , light can steadily propagate in the side wall (an annular fibre core) and output at the fibre end (Supplementary Fig. 11). The damage threshold power for the MoS<sub>2</sub>-embedded fibre under the 800-nm-excited laser (with high power to reach the damage threshold) is about three times higher than that on a planar substrate with a focal spot of  $\sim 10 \mu\text{m}$  (Fig. 3e). Considering the enhancements in both light-matter interactions and damage threshold power, the output power of harmonic generation in monolayer MoS<sub>2</sub>-embedded optical fibre is three orders of magnitude higher than that of planar 2D MoS<sub>2</sub>; further, energy conversion can reach  $10^{-4}$  to  $10^{-3}$  (see Supplementary Note 2), which is already sufficiently high for some applications such as on-chip new wavelength generation and all-fibre optical parametric oscillator<sup>28,29</sup>. We also observed that the nonlinear enhancement shows a general monotonic increase and saturation behaviour with the fibre length (Fig. 3f). In principle, the SHG or THG signal should have

quadratic dependence on the length<sup>27</sup>, which is indeed the case for fibre lengths less than 5 cm. However, when the fibre becomes longer, the loss effect caused by the absorption tail of monolayer MoS<sub>2</sub> as well as the phase mismatch between the excitation and SHG/THG lights leads to the deviation from the quadratic increase and the saturation shows an upward trend.

Further, we utilized the MoS<sub>2</sub>-embedded PCF as a saturable absorber (SA) in a fibre laser for ultrafast pulse generation, which is a typical example of imaginary-part-related applications. The laser structure is similar to the most commonly demonstrated fibre ring cavity, which contains a pump laser diode, an erbium-doped fibre (gain media) and an optical coupler as the output<sup>15</sup> (Fig. 4a). Here the main difference is that we replaced a conventional free-space SA film by the MoS<sub>2</sub>-embedded fibre and therefore realized an all-fibre mode-locked laser. For exploiting the absorptive properties of 2D materials, we intentionally grew 6–8-layered MoS<sub>2</sub>, which had a larger absorption tail at  $\sim 1,550 \text{ nm}$  compared with that of the monolayer<sup>30</sup>. This 3-cm-long few-layered MoS<sub>2</sub> PCF with a hollow-core honeycomb structure shows a relatively low coupling loss of  $\sim 1 \text{ dB}$  (Supplementary Fig. 12) and a nonlinear absorption modulation depth ( $\alpha_s$ ) of 10% with a saturation peak intensity of  $0.8 \text{ MW cm}^{-2}$  (Fig. 4b); these values are already comparable to those of conventional SA semiconductors<sup>15</sup> (for example, modulation depth of  $\sim 5\%$  with a saturation peak intensity of  $\sim 2 \text{ MW cm}^{-2}$  for AlAs/GaAs Bragg mirrors). Then, a stretched-pulse passively mode-locked fibre laser was built by dispersion management. The maximum output power of the pulsed laser is  $\sim 6 \text{ mW}$  (under continuous-wave laser pump power of 600 mW and at room temperature) with  $\sim 41 \text{ MHz}$  repetition frequency (Fig. 4c),  $\sim 19 \text{ nm}$  spectral bandwidth ( $\Delta\lambda$ ), 1,560 nm centre wavelength (Fig. 4d) and  $\sim 500 \text{ fs}$  pulse duration (Fig. 4e and lasing reporting summary for more laser specifics). The subpicosecond pulse train shows a high signal-to-noise ratio



of ~52 dB (Supplementary Fig. 13). Considering the relatively broad spectral bandwidth (~19 nm), the pulse duration could be further compressed to ~200 fs by group-velocity-dispersion compensation outside the laser cavity, if needed, for future applications. The superior performance, massive production ability and environmental adaptability of the MoS<sub>2</sub>-embedded fibre demonstrate its distinct advantages compared with a conventional 2D-materials-integrated fibre on the facets or external surfaces (Supplementary Tables 1 and 2), indicating that it is ready for scientific research and industrial applications in ultrafast lasers.

We predict that this two-step growth method can also be applied to grow other 2D materials (other than TMDs) onto optical fibres. For different applications, there are extremely rich choices of various 2D materials with different nonlinear properties. In addition, by fabricating well-designed fibre structures to control the group velocity dispersion, supercontinuum generation can be realized. Further, if phase matching between the excitation light and nonlinear signal is satisfied, the nonlinear conversion efficiency can even be increased to ~10% (Supplementary Fig. 14 and Supplementary Note 3), which would be comparable to that obtainable from conventional bulk optical crystals. If so, this can open a new regime for the design of nonlinear optical materials, which can inspire versatile nonlinear applications in optical fibres, such as sum-/difference-frequency generation, high-harmonic generation, nonlinear parametric amplification and terahertz sources.

### Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41565-020-0770-x>.

Received: 22 April 2020; Accepted: 26 August 2020;  
Published online: 21 September 2020

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## Methods

**Pre-deposition of transition metal feedstock into optical fibres.** Transition metal sources (Mo or W) were pre-deposited on the fibres by capillarity of their corresponding saline solutions at optimized concentrations. For monolayer film growth, the amount of metal precursor was tuned according to the hole diameters of the different fibres. Typical concentrations are as follows:  $\text{Na}_2\text{MoO}_4$  at 8, 21 and  $45 \text{ mg ml}^{-1}$  or  $\text{Na}_2\text{WO}_4$  at 12, 30 and  $64 \text{ mg ml}^{-1}$  for HCF with core diameters of 50, 15 and  $5 \mu\text{m}$ , respectively;  $\text{Na}_2\text{MoO}_4$  at 32 and  $54 \text{ mg ml}^{-1}$  or  $\text{Na}_2\text{WO}_4$  at 45 and  $72 \text{ mg ml}^{-1}$  for PCF with hollow- and solid-core honeycomb structures, respectively. Then, the fibres were heated to  $110^\circ\text{C}$  and dried in an argon atmosphere for 30 min before CVD growth.

**Syntheses of TMD-embedded optical fibres.** The Mo-precursor-based pre-deposited fibres were placed on a quartz plate in the centre of the CVD furnace, and 1 g sulfur (99%, Sigma Aldrich) powder was placed in the upstream region outside the heating zone. Then, the chamber of the furnace was flushed with argon for creating an inert atmosphere and subsequently ramped up to the optimized growth temperature ( $\sim 820^\circ\text{C}$  for  $\text{MoS}_2$  growth) with 100 s.c.c.m. argon. During the growth, the temperature for sulfur evaporation was  $\sim 120^\circ\text{C}$ . The entire process was carried out under  $\sim 150 \text{ Pa}$ , and the growth duration was set as 15–30 min. After the growth, the system was naturally cooled down to room temperature. Similar growth conditions were used to synthesize other TMD and alloy fibres, such as  $\text{MoSe}_2$ ,  $\text{WS}_2$  and  $\text{MoS}_2\text{Se}_x$ , except for different temperatures for chalcogen sublimation and TMD growth. In particular, the sublimation temperatures were  $\sim 120^\circ\text{C}$  for sulfur and  $\sim 220^\circ\text{C}$  for selenium, and the growth temperatures were  $\sim 800$ ,  $\sim 950$  and  $\sim 820^\circ\text{C}$  for  $\text{MoSe}_2$ ,  $\text{WS}_2$ - and  $\text{MoS}_2\text{Se}_x$ -embedded fibres, respectively.

**Characterization of TMD-embedded optical fibres.** Optical images were taken with an Olympus BX51M microscope by focusing on the holes of the fibre walls. Raman and PL spectra were collected with a homemade optical system excited using a 532 nm laser with power of  $\sim 1 \text{ mW}$ . STEM (FEI Titan Themis G2 300) was conducted to characterize the atomic structure, and a scanning electron microscope (Thermo Fisher, Quattro S) was used to characterize the morphology of the samples.

**Harmonic generation measurements.** An optical parameter oscillator (Coherent, Mira-OPO-X) pumped by a titanium-sapphire oscillator (Coherent, Mira-HP) provided a wavelength-tunable excitation laser (15 mW,  $\sim 150 \text{ fs}$ , 76 MHz) and then it was focused onto the  $\text{MoS}_2$  HCF or  $\text{MoS}_2/\text{silica}$  via an objective (Nikon,  $\times 10$ ,  $\text{NA} = 0.25$ ). The SHG and THG signals were collected by a spectrograph with a silicon-based charge-coupled device (Princeton Instruments, SP2500 and PyLoN 400BRX) via a transmission system.

**Saturable absorption measurements.** A homemade pulsed fibre laser (200 fs, 1,564 nm, 75.4 MHz) was utilized to measure the power-dependence transmission of  $\text{MoS}_2$  PCF. The pulsed laser was equally divided using a fibre-optic coupler to monitor the power. The input/output single-mode fibre (Corning, SMF-28e+) was aligned with a 3-cm-long  $\text{MoS}_2$  PCF (NKT Photonics, HC-1550) by a homemade alignment setup that included two optical microscopes along different directions and multi-axis stages to ensure accurate alignment.

### Implementation and characterization of all-fibre mode-locked laser.

A laser diode (600 mW, 976 nm) was coupled into the fibre ring cavity via a wavelength-division multiplexer as the pump source and 60-cm-long erbium-doped fibre (LIEKKI, Er110) as the gain medium. An isolator guaranteed that the laser in the fibre ring was transferred along a single direction. In this case, we employed a backward pumping configuration for large output power. Further, 3-cm-long  $\text{MoS}_2$  PCF was integrated as the SA with a coupler for 20% output. An additional polarization controller was utilized to optimize the output spectrum and pulse width. The total group velocity dispersion of the fibre ring was  $0.0005 \text{ ps}^2$ ,

comprising a 1 m single-mode fibre, 0.6 m erbium-doped fibre and 3.4 m OSF980 (as the dispersion compensation fibre) with group velocity dispersion of  $-22$ ,  $+12$  and  $+4.5 \text{ fs}^2 \text{ mm}^{-1}$ , respectively. The output spectra and autocorrelation trace were measured by a spectrograph with an infrared charge-coupled device (Princeton Instruments; HRS-300SS with a grating of 150 grooves  $\text{mm}^{-1}$  and NIRvana 640 with spectrometer resolution of  $\sim 1.3 \text{ nm}$ ) and an autocorrelator (APE, pulseCheck USB-15). The pulse train and signal-to-noise ratio were observed by an oscilloscope (Rigol, DS6104) and radio-frequency spectrograph (Rigol, DSA815). A self-starting mode-locked pulse was observed when the pump power was increased to 180 mW.

**Reporting Summary.** Further information on research design is available in the Nature Research Reporting Summary linked to this article.

## Data availability

The authors declare that the data supporting the findings of this study are available within the paper, Supplementary Information and Source Data. Extra data are available from the corresponding authors upon request. Source data are provided with this paper.

## Acknowledgements

This work was supported by the National Natural Science Foundation of China (51991340, 51991342, 51991344 and 51421002); National Key R&D Program of China (2016YFA0300903 and 2016YFA0300804); Beijing Natural Science Foundation (JQ19004); Beijing Excellent Talents Training Support (2017000026833ZK11); Beijing Graphene Innovation Program (Z181100004818003); Beijing Municipal Science & Technology Commission (Z191100007219005); the Key R&D Program of Guangdong Province (2019B010931001, 2020B010189001, 2018B010109009 and 2018B030327001); Guangdong Innovative and Entrepreneurial Research Team Program (2016ZT06D348); Bureau of Industry and Information Technology of Shenzhen (graphene platform 201901161512); the Science, Technology and Innovation Commission of Shenzhen Municipality (KYTDPT20181011104202253); Program of Chinese Academy of Sciences (ZDYZ2015-1 and XDB33030200); National Postdoctoral Program for Innovative Talents (BX20180013 and BX20190016); the Academy of Finland; the ERC (834742); the European Union's Horizon 2020 research and innovation programme (820423, S2QUIP); and China Postdoctoral Science Foundation (2019M660001, 2019M660280 and 2019M660281). We acknowledge the Electron Microscopy Laboratory in Peking University for the use of their electron microscope.

## Author contributions

Z.L., X.B. and K.L. conceived the experiments and supervised the project. Y.G.Z. and C.L. contributed to the growth experiments. W.Y. performed the optical experiments and fibre laser setup. X.C. contributed to the theoretical modelling. R.Q., P.G. and X.B. conducted the STEM experiments. J.L., X.Z., J.W., M.W. and Y.Z. conducted the SEM, PL and Raman characterizations. S.W. and Z.S. suggested the optical experiments. All the authors discussed the results and wrote the manuscript.

## Competing interests

The authors declare no competing interests.

## Additional information

**Supplementary information** is available for this paper at <https://doi.org/10.1038/s41565-020-0770-x>.

**Correspondence and requests for materials** should be addressed to K.L., X.B. or Z.L.

**Peer review information** *Nature Nanotechnology* thanks Baohua Jia, Zhiyi Wei and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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## Lasing Reporting Summary

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### ► Experimental design

#### Please check: are the following details reported in the manuscript?

##### 1. Threshold

Plots of device output power versus pump power over a wide range of values indicating a clear threshold

Yes  
 No

The plot of output power versus pump power is necessary for the device for indicating the lasing threshold (i.e., the lowest excitation level at which a laser's output is dominated by stimulated emission rather than by spontaneous emission). But in our mode-locked fibre laser, the continuous-wave (CW) laser output can be observed though at the lowest controllable pump power of 50 mW. Hence, the real key point should be the pump power that at which the pulsed laser's output rather than CW laser, and we have indicated that the mode-locked self-starting pump power is ~180 mW in Methods part.

##### 2. Linewidth narrowing

Plots of spectral power density for the emission at pump powers below, around, and above the lasing threshold, indicating a clear linewidth narrowing at threshold

Yes  
 No

For our ultrafast laser, the linewidth narrowing at the lasing threshold cannot be observed in our experiment. This is because our device always works at laser emission mode, rather than spontaneous emission. Besides, the spectral bandwidth of output ultrafast laser is measured as ~19 nm shown in Fig. 4d, which is much wider than the conventional continuous-wave (CW) laser. The wide spectrum is required for the mode locking and ultrafast laser (~500 fs) generation.

Resolution of the spectrometer used to make spectral measurements

Yes  
 No

The resolution of the spectrometer is ~1.3 nm as mentioned in Methods part.

##### 3. Coherent emission

Measurements of the coherence and/or polarization of the emission

Yes  
 No

For the ultrafast laser, the coherent is not an important parameter. This is because that the large spectral width (for ultrafast laser) determines the short coherent length.

##### 4. Beam spatial profile

Image and/or measurement of the spatial shape and profile of the emission, showing a well-defined beam above threshold

Yes  
 No

Our device always works at the situation of laser emission rather than spontaneous emission, so the beam spatial profile is not relevant to show a well-defined beam above threshold. Besides, in our device, the ultrafast laser exported from the single-mode optical fibre, which ensured the fundamental mode beam output.

##### 5. Operating conditions

Description of the laser and pumping conditions  
*Continuous-wave, pulsed, temperature of operation*

Yes  
 No

We have demonstrated the pumping conditions is continuous-wave laser and under room temperature, and the output laser is pulsed laser in fibre laser part of main text.

Threshold values provided as density values (e.g.  $W\text{ cm}^{-2}$  or  $J\text{ cm}^{-2}$ ) taking into account the area of the device

Yes  
 No

Our device always works at the situation of laser emission rather than spontaneous emission, so the lasing threshold is not observed in our experiment.

##### 6. Alternative explanations

Reasoning as to why alternative explanations have been ruled out as responsible for the emission characteristics  
*e.g. amplified spontaneous, directional scattering; modification of fluorescence spectrum by the cavity*

Yes  
 No

Our device always works at the situation of laser emission and we have used the TMD embedded fibre as saturable absorber to change the continuous-wave laser into ultrafast pulsed laser output. The pulsed laser pulse output can directly rule out the alternative explanations, such as amplified spontaneous, directional scattering and modification of fluorescence spectrum by the cavity.

##### 7. Theoretical analysis

Theoretical analysis that ensures that the experimental values measured are realistic and reasonable  
*e.g. laser threshold, linewidth, cavity gain-loss, efficiency*

- Yes  
 No

The mode-locked fibre lasers based saturable absorbers are widely studied and the theoretical analysis should be similar as some previous articles and books, such as Fiber Lasers (Wiley, 2012). Hence, the additional theoretical analysis is not necessary in our manuscript.

## 8. Statistics

Number of devices fabricated and tested

- Yes  
 No

In our all-fibre mode-locked laser, the main difference here is that we replaced the conventional free-space SA film by the MoS<sub>2</sub> embedded fibre. Benefiting from the controllability and repeatability of directly CVD method, the different fibre SAs with the same fabrication condition have demonstrated the same performances. So the number of devices fabricated and tested is not important in our works.

Statistical analysis of the device performance and lifetime (time to failure)

- Yes  
 No

Benefiting from the controllability and repeatability of directly CVD method for MoS<sub>2</sub> fibre, the SA and mode-locked laser performance should be similar in our experiment. For lifetime, during our experiments (near two weeks), the performance still keep stable, so it's difficult to measure the lifetime.