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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 continues-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 continues-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 continue-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 cm^{-2} or J 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 continue-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 MoS2 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 MoS2 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.