Supplementary Information

Substoichiometric Two-Dimensional Molybdenum Oxide Flakes: A Plasmonic Gas Sensing Platform

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S1: Optical gas measurement setup

The chamber consists of two ports for gas inlet and outlet, and two electrical connectors for an electrical heating plate (Fig. S1). Inside the chamber, a holder frame with open window was utilized to mount the molybdenum oxide samples during testing. The chamber was fixed on to a stage with two optical fibre cables and adapters for the connection with the light source and the spectrophotometer, respectively.

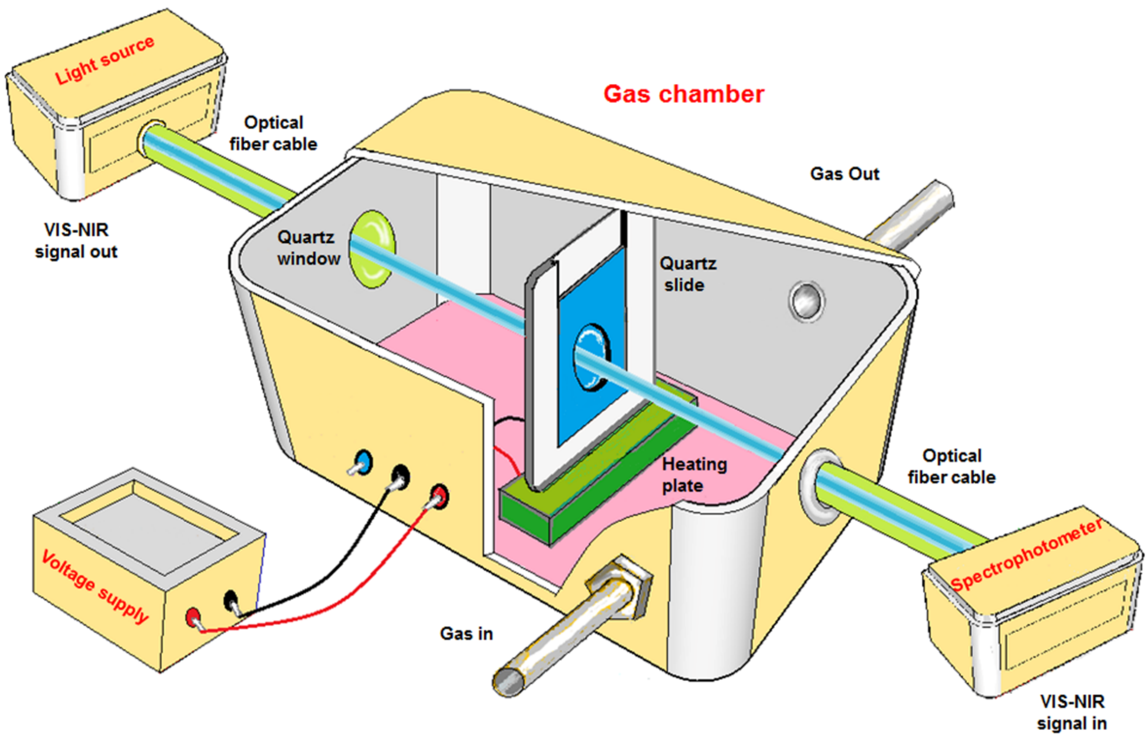

Fig. S1 Illustration of the optical gas testing chamber.

S2: The extended Raman spectra and AFM adhesion mappings of the 2D molybdenum oxide samples

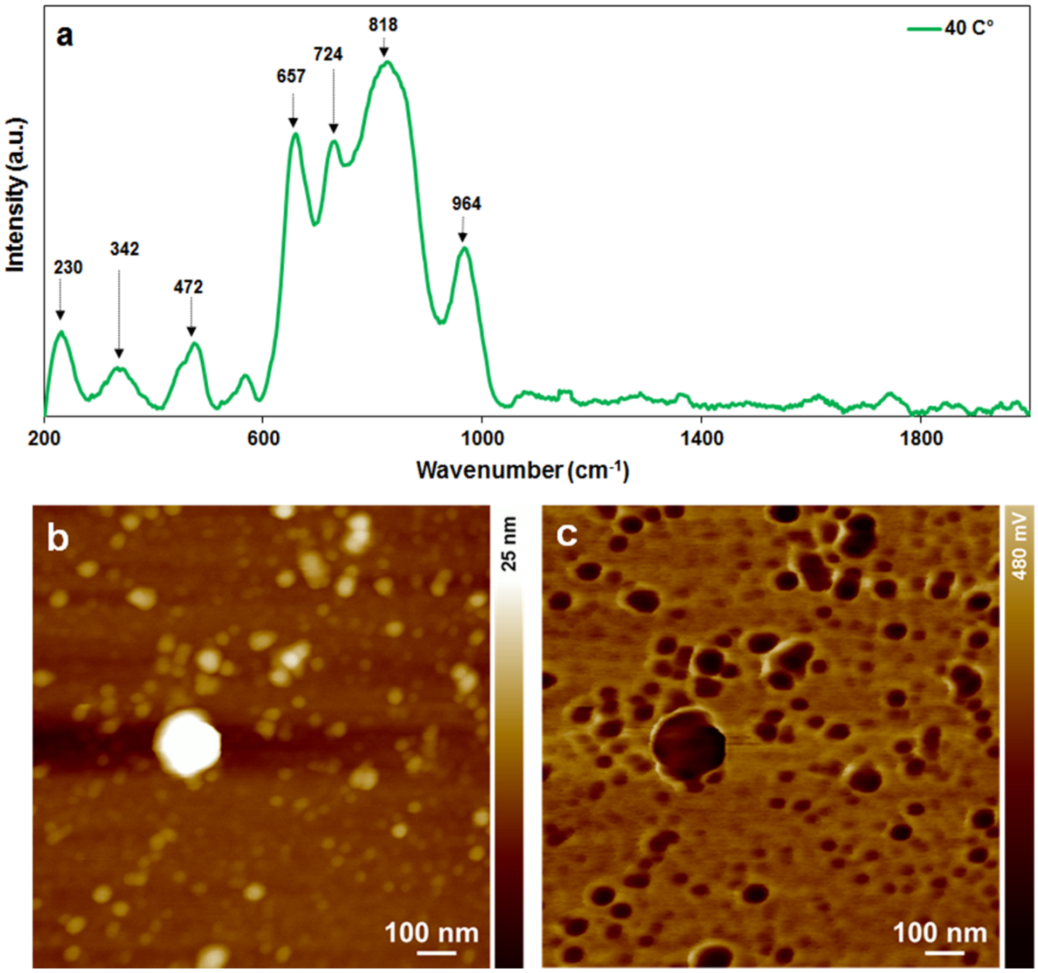

Fig. S2. (a) The Raman spectrum of the drop casted film made of 2D Mo_4O_{11} flakes that were dried at 40 °C. (b) The AFM image of a very low concentration suspension of 2D flakes drop casted onto Si substrate and (c) The corresponding AFM adhesion image of the same area. The adhesion image shows that the surface energy of the flakes are similar (they have similar surface voltages) and hence should demonstrate similar van der Waals forces, assisting them to lie horizontally and homogenous on top of each other. At high concentration of flakes in the suspensions, drop casting results in the formation of stable films with many flakes hold together by these van der Waals forces.

S3: The surface morphology of the 2D molybdenum oxide film

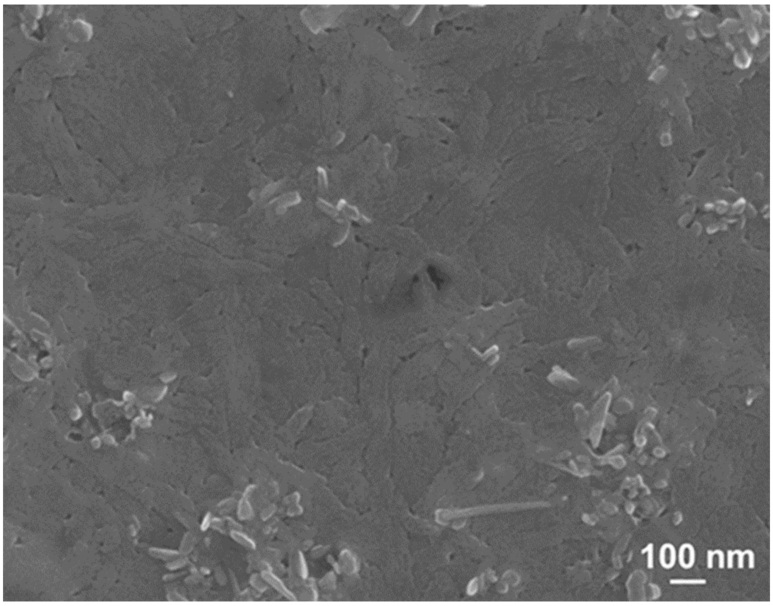

Fig. S3. The SEM image of the film made of high concentration 2D molybdenum oxide flakes after annealing at 40 °C shows the surface morphology and the flakes distribution. It can be observed that these flakes are compactly and horizontally placed onto each other, forming a relatively homogeneous film.

S4: Crystal structure of the 2D molybdenum oxide film after light exposure from the UV-Vis-NIR light source

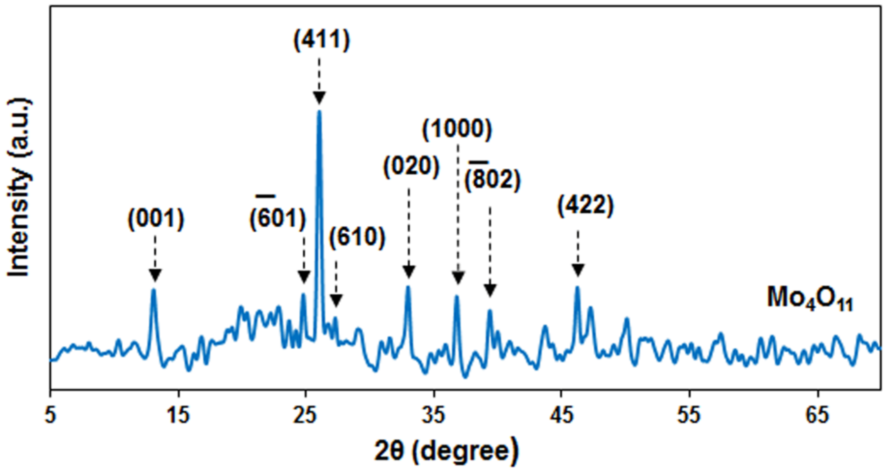

Fig. S4. The XRD pattern of the 2D molybdenum oxide film after light exposure from the UV-Vis-NIR light source.

S5: The DFT calculations

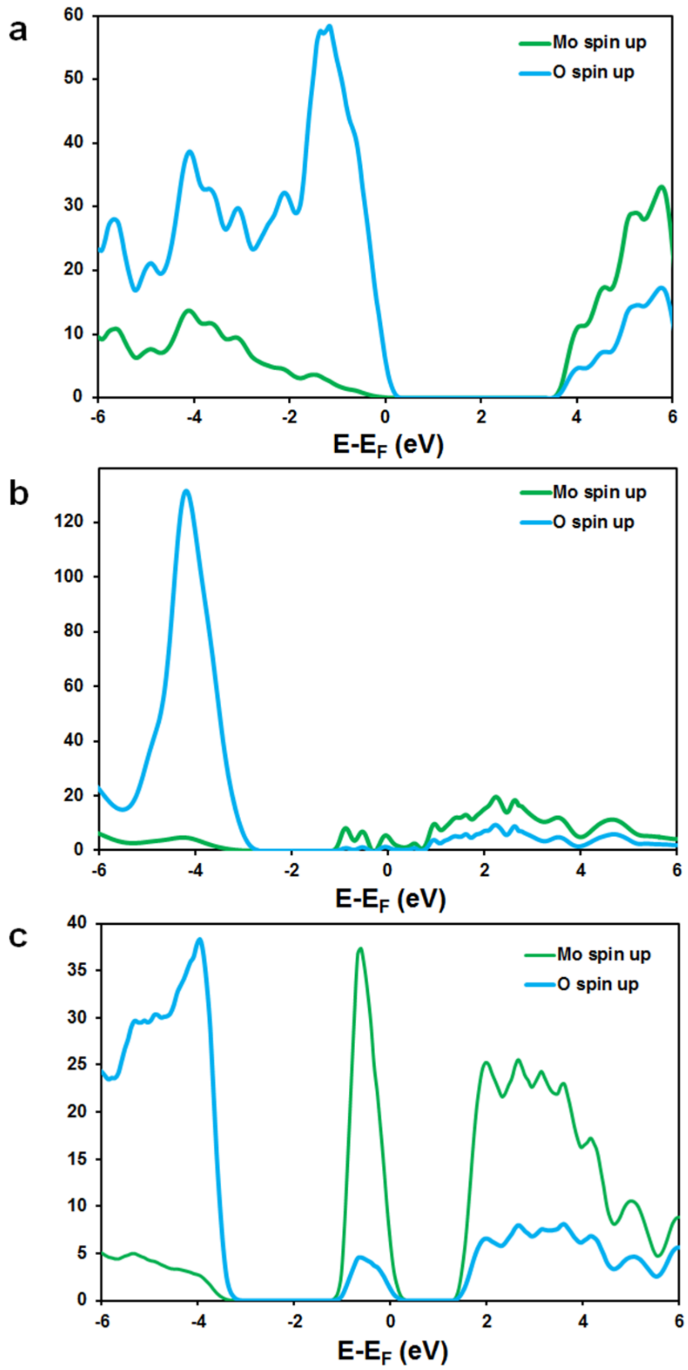

Fig. S5 The density of states calculated by DFT of: (a) α-MoO_3, (b) Monoclinic Mo_4O_{11} and (c) Monoclinic MoO_2.

S6: The Summery of plasmonic H_2 gas sensors reported previously

Table S1 The summary of plasmonic sensors in terms of response factor, response and recovery time for H_2 gas sensing.

Sensitive layer	Operating temperature (°C)	Response factor (%)	Response time (s)	Recovery time (s)	Sensing response range	Ref
Al-ZnO	500	33	N/A	N/A	1500–2500 nm	1
Pt/WO_3	150	0.16	40–55	40–55	200–900 nm	2
Au-YSZ	500	150	N/A	N/A	450–850 nm	3
AuH-rGO	150	0.4	60	120	200–900 nm	4

References

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