Supporting information for:

Leveraging nanocavity harmonics for control of optical processes in 2D semiconductors

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Simulation of electric fields in nanocavity

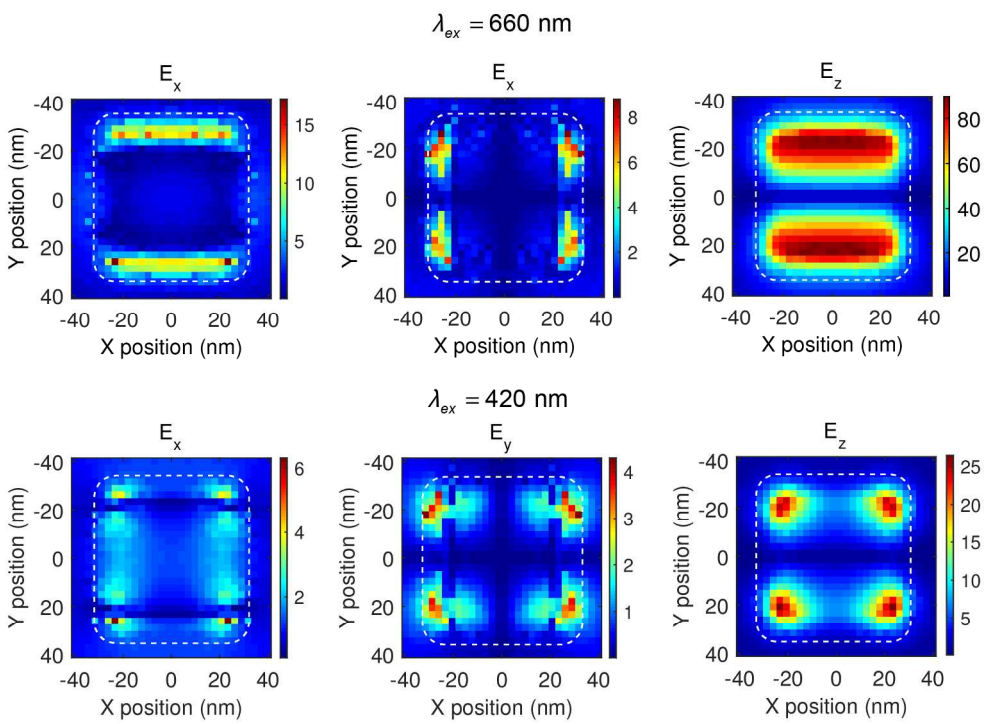

Figure S1. Lateral spatial maps of field enhancement $|E/E_0|$ for each field component at two excitation wavelengths. White outlines indicate the lateral extent of the nanocube. At 660 nm, the fundamental resonance is excited, while at 420 nm the second order resonance is excited.

Measurement setup

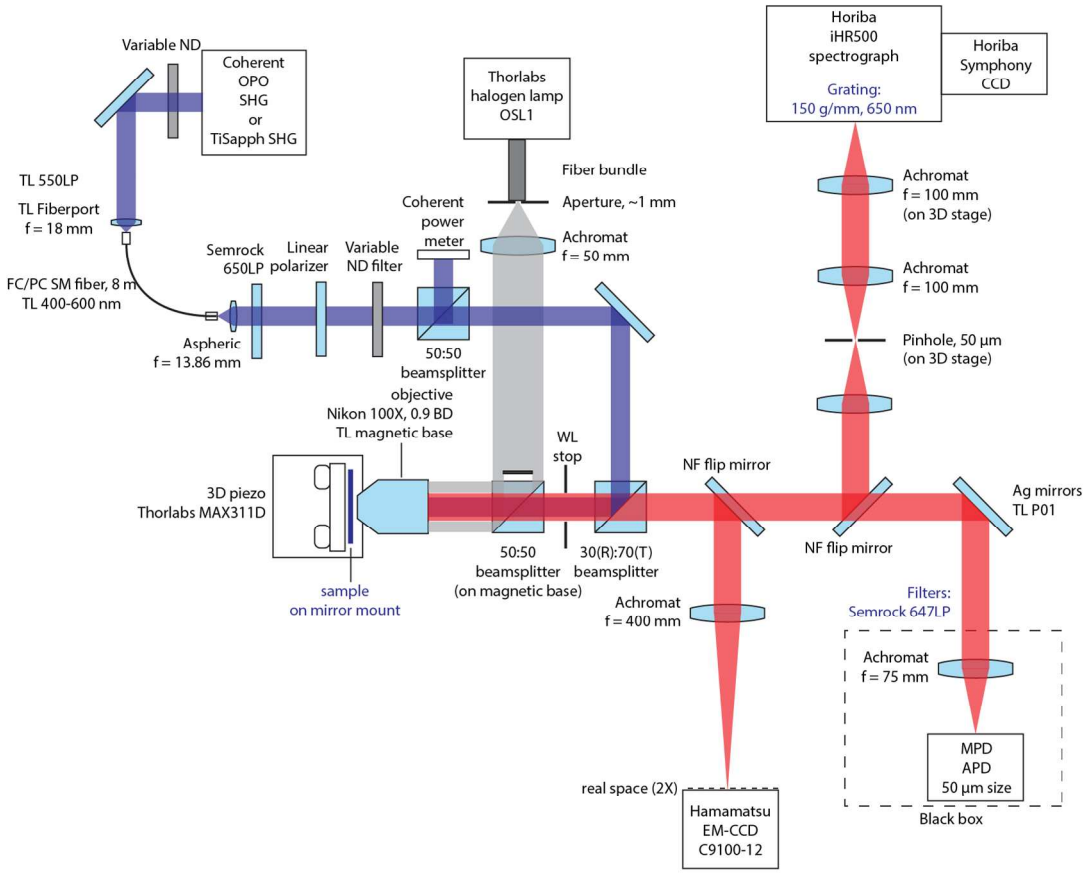

Figure S2. Schematic of home-built optical microscope used for measuring the nanocavities, as described in Methods.

Characterization of MoS_2 crystals

The Raman spectrum indicates that the as grown MoS_2 is a monolayer.[1,2] After transfer, the E_{2g} and A_{1g} peaks slightly shift 1 and 2 wavenumbers towards higher energy, respectively. This tiny blue shift indicates that i) MoS_2 keeps its monolayer nature and ii) is slightly p-doped after transfer.[3]

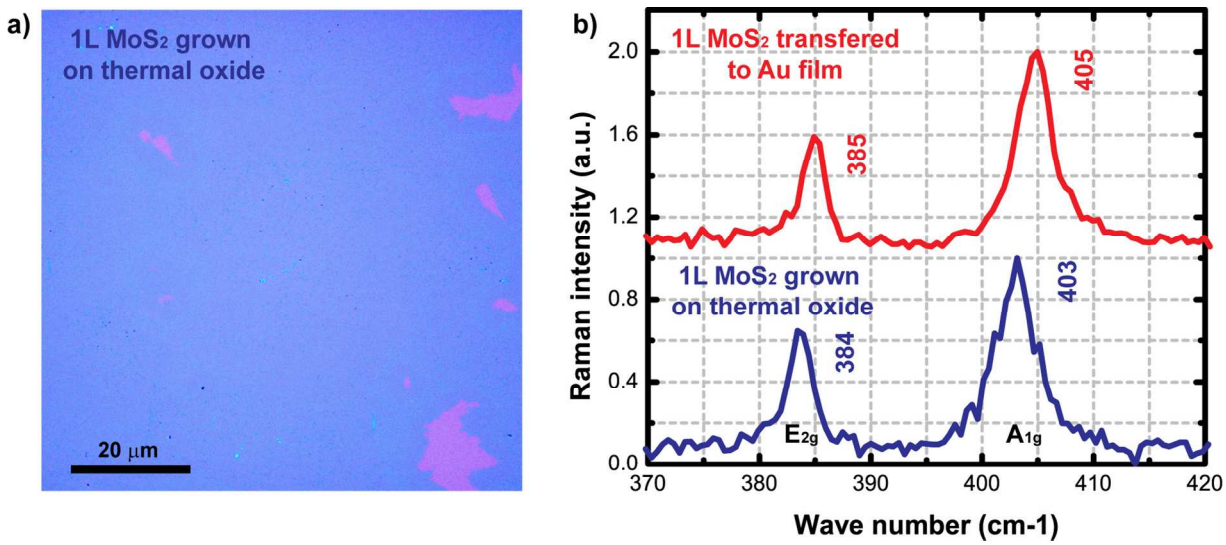

Figure S3. (a) Optical microscopic image of a continuous monolayer MoS_2 film grown on thermal oxide. The reddish areas are holes uncovered by MoS_2. (b) Raman spectra of MoS_2 grown on thermal oxide (blue curve) and transferred on to Au film (red curve), using 532 nm excitation.

References

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(2) Ling, X.; Lee, Y.; Lin, Y.; Fang, W. *Nano Lett.* **2014**, *14*, 464–472.

(3) Shi, Y.; Huang, J.-K.; Jin, L.; Hsu, Y.-T.; Yu, S. F.; Li, L.-J.; Yang, H. Y. *Sci. Rep.* **2013**, *3*, 1839.