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Revealing Mie Resonances with Enhanced and Suppressed Second-Order Nonlinear Optical Responses in a Hexagonal-Prism-Like MoS_2 Nanoparticle

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Supporting Information

Revealing Mie resonances with enhanced and suppressed second-order nonlinear optical responses in a hexagonal-prism-like MoS_2 nanoparticle

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Note 1 Morphology characterization of MoS_2 nanoparticles

We selected MoS_2 nanoparticles with regular morphologies from MoS_2 powder purchased from the supplier (Sixcarbon Tech Shenzhen). In Fig. S1a, we show the SEM image of MoS_2 powder. It contains nanoparticles and nanosheets with different shapes and sizes. In Fig. S1b, we present the magnified images of some nanoparticles and nanosheets. It is noticed that a MoS_2 nanoparticle is generally composed of vertically stacked layers, as shown in Fig. S1c where the side view of a

nanoparticle is presented. In Fig. S1d, we show the SEM image of a hexagonal-prism-like nanoparticle successfully selected from the powder. The AFM image is shown in the inset.

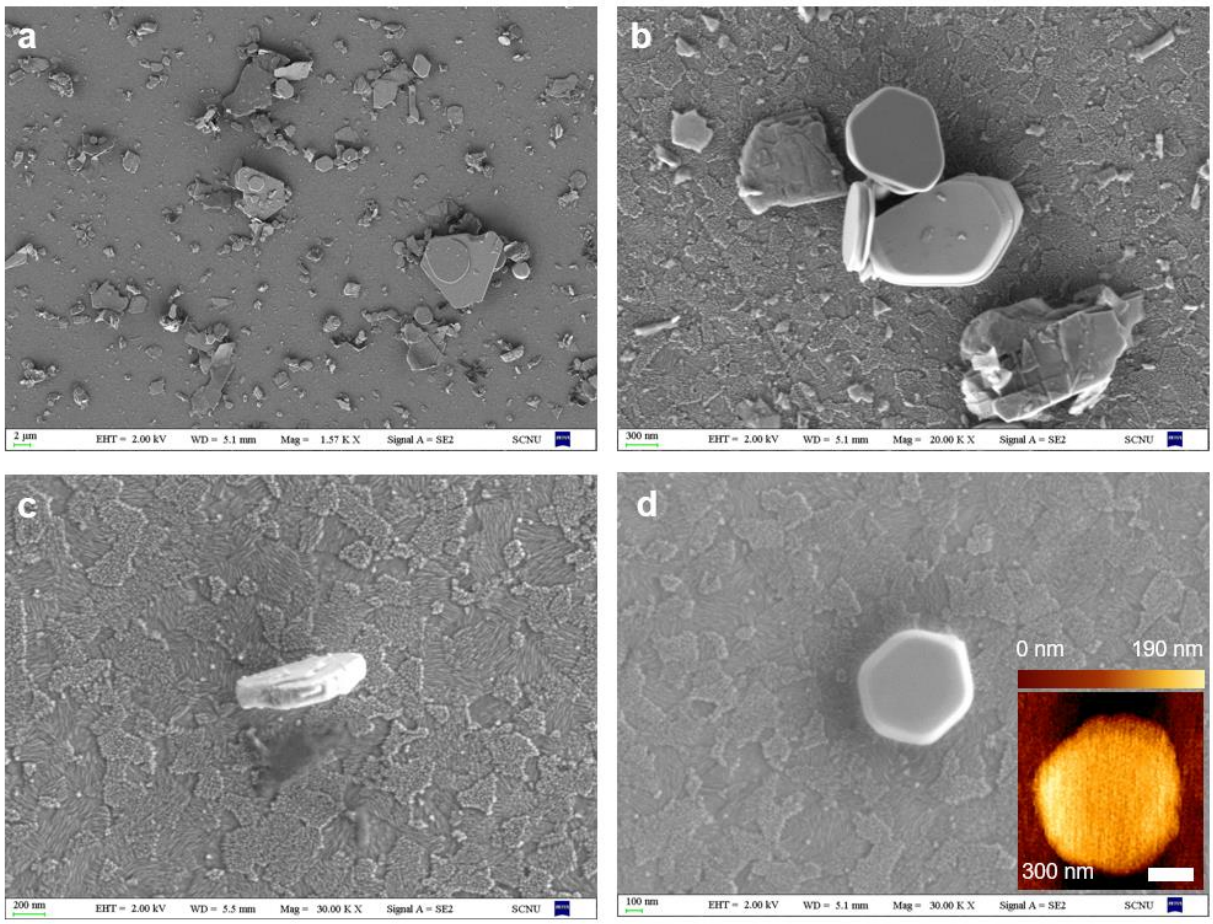

Figure S1 Morphology characterization of MoS_2 nanoparticles. (a) SEM image (top view) of MoS_2 powder distributed on an ITO substrate. (b) Magnified SEM images of MoS_2 nanoparticles and nanosheets. (c) Side view of a MoS_2 nanoparticle. (d) Top view of a hexagonal-prism-like MoS_2 nanoparticle. The AFM image is shown in the inset.

Note 2 Scattering spectra of MoS_2 nanoparticles

In Fig. S2, we show the scattering spectra measured for three MoS_2 nanoparticles. The scattering dips originating from the absorption of excitons are clearly identified at ~660 and ~610 nm.

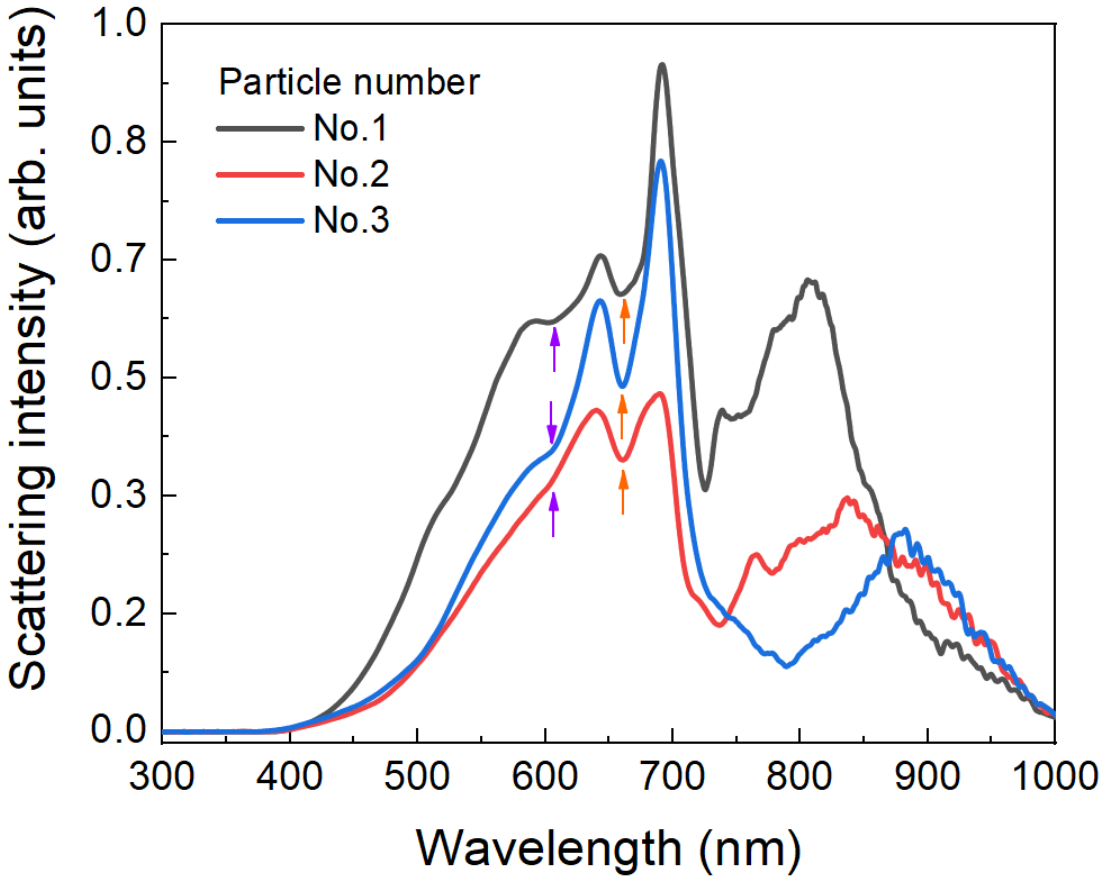

Figure S2 Scattering spectra measured for three MoS_2 nanoparticles with different geometrical parameters. The two exciton resonances appearing as scattering dips are marked by arrows.

Note 3 Dependence of the scattering spectrum on the polarization of the excitation light

We simulated the scattering spectra of a typical MoS_2 nanoparticle with l = 300 nm and h = 170 nm, which is excited by using white light polarized along the arm chair and zigzag directions. As shown in Figs. S3a and S3b, the scattering spectrum of the MoS_2 nanoparticle depends weakly on the polarization of the excitation light.

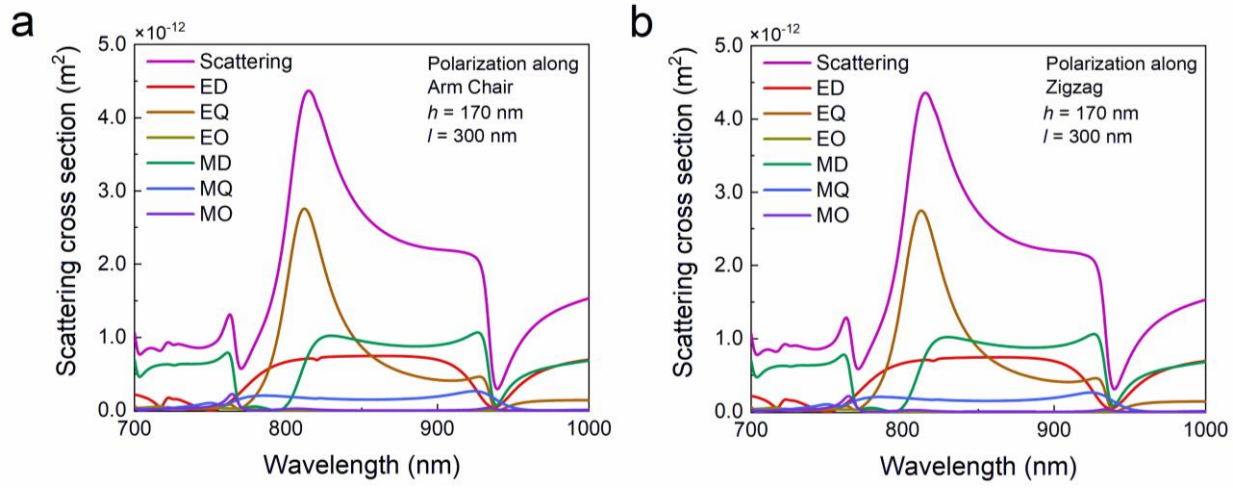

Figure S3 Scattering spectra calculated for a MoS_2 nanoparticles with l = 300 nm and h = 170 nm excited by using white light polarized along the arm chair direction (a) and the zigzag direction (b).

Note 4 Dependences of the optical modes on the geometrical parameters

The thickness of a MoS_2 nanoparticle was characterized by using atomic force microscopy (AFM). The errors in the AFM measurements can be evaluated by the simulation results. In Fig. S4a, we show the scattering spectra of MoS_2 nanoparticles with the same length (l = 300 nm) and different heights (h = 130–210 nm). Thus, the height of a MoS_2 can be estimated from the EQ mode in the measured scattering spectrum. The evolution of the integration of E^4 over the volume of a MoS_2 nanoparticle with increasing height, which refelects the SHG efficiency, is presented in Fig. S4b. Here, we consider only symmetric modes (ED and MD modes). It is found that the the SHG efficiency exhibits a strong dependence on the height of the MoS_2 nanoparticle.

In Fig. S4c, we show the scattering spectra calculated for MoS_2 nanoparticles with the same height (h = 170 nm) and different lengths (l = 130–210 nm). It can bee seen that the scattering cross-sections of all modes increased with increasing lemgth, especially the ED mode. The the SHG efficiencies calculated for the ED and MD modes with increasing height are presented in Fig. S4d. As the length increases, the SHG efficiency also increases rapidly.

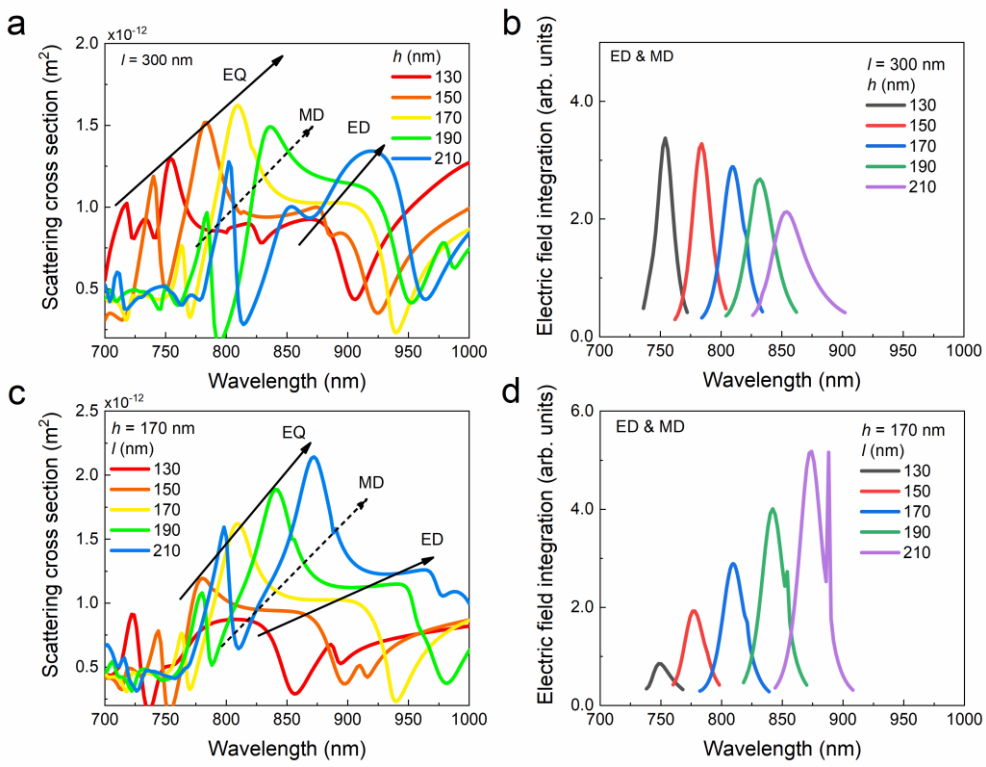

Figure S4 (a) Scattering spectra calculated for MoS_2 nanoparticles with the same length and different heights. (b) The evolution of the SHG efficiency of a MoS_2 nanoparticle with increasing height. (c) Scattering spectra calculated for MoS_2 nanoparticles with the same height and different lengths. (d) Evolution of the SHG efficiency of a MoS_2 nanoparticle with increasing length.

Note 5 CCD images of the SHG

The emission pattern of the SHG from a MoS_2 nanoparticle exhibits a dependence on the excitation wavelength (i.e., the wavelength of the fundamental wave). In Fig. S5, we show the

emission patterns of the SHG observed at different excitation wavelengths, which are recorded by using a charge coupled device (CCD).

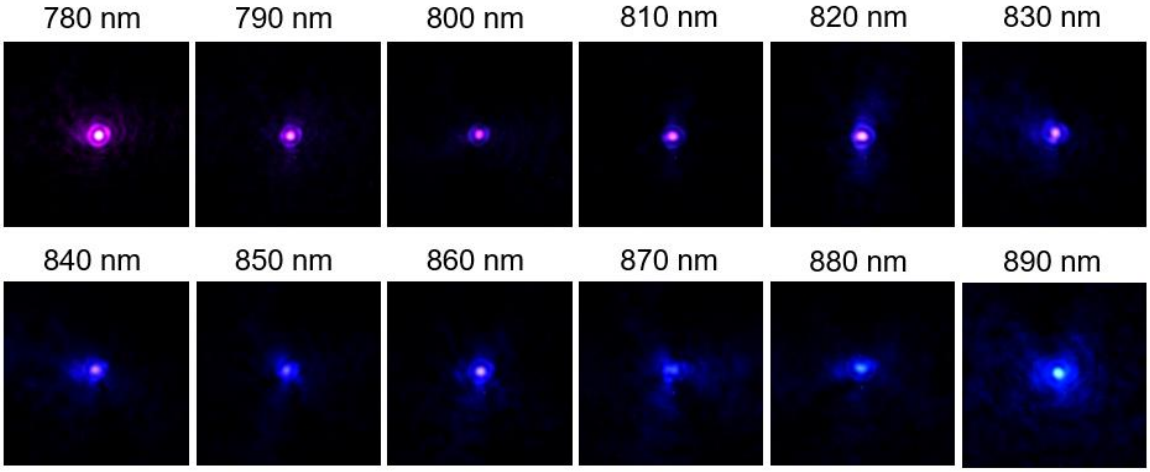

Figure S5 CCD images of the SHG observed at different excitation wavelengths.

Note 6 Optical mode with suppressed SHG

In Fig. S6a, we show the scattering spectrum measured for a MoS_2 nanoparticle. Apparently, a scattering peak appears at ~800 nm. In Fig. S6b, we present the SHG spectra measured at different excitation wavelengths. It is noticed that the minimum SHG is observed at ~800 nm. It implies that the optical mode at ~800 nm is the one with suppressed SHG.

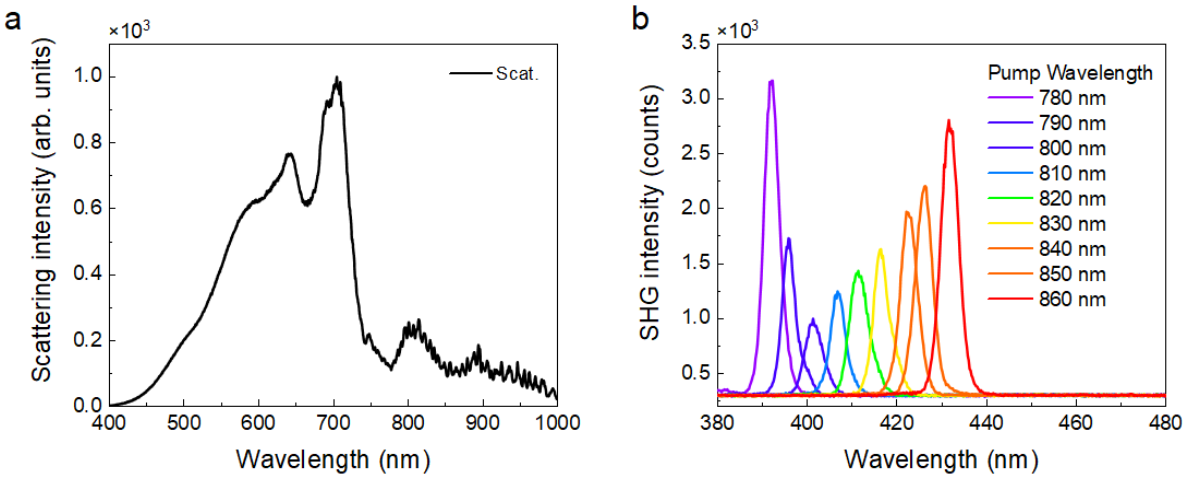

Figure S6 (a) Scattering spectrum measured for a MoS_2 nanoparticle. (b) SHG spectra measured at different excitation wavelengths for the MoS_2 nanoparticle shown in (a).

Note 7 Optical mode with enhanced SHG

In Fig. S7a, we show the scattering spectrum measured for a MoS_2 nanoparticle. Apparently, a scattering peak appears at ~830 nm. In Fig. S7b and S7c, we present the SHG spectra measured at different excitation wavelengths by using a 60× and a 100× objective, respectively. Except the

excitation and collection efficiencies, it is noticed that the maximum SHG is observed at ~830 nm. It implies that the optical mode at ~830 nm is the one with enhanced SHG.

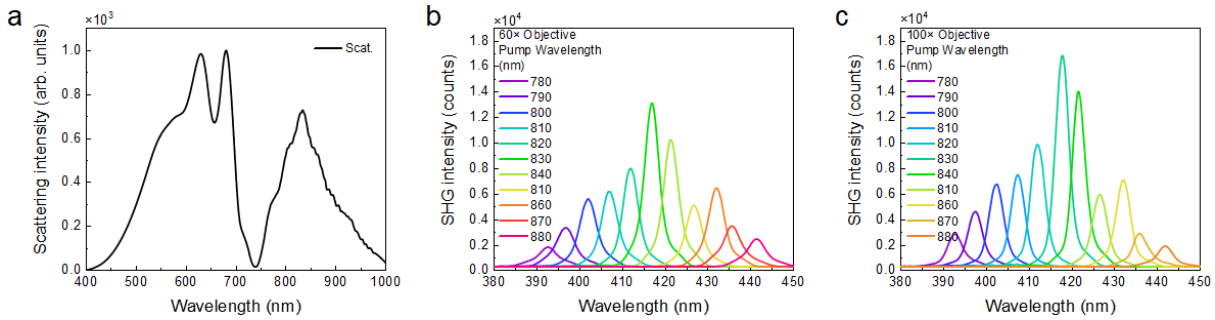

Figure S7 (a) Scattering spectrum measured for a MoS_2 nanoparticle. (b) and (c) SHG spectra measured at different excitation wavelengths for the MoS_2 nanoparticle shown in (a) by using a 60× and a 100× objective, respectively.

Note 8 Emission pattern of the SHG

The three-dimensional (3D) emission pattern of the SHG of a MoS_2 nanoparticle can be obtained by using a laser scanning confocal microscope. In Fig. S8a, we show the 3D emission pattern of the SHG observed for the MoS_2 nanoparticle shown in Figure 3a-c at the optical mode with suppressed SHG. In this case, the suppressed SHG appearing at the center is not revealed due to the existence of the surface nonlinearity. Actually, the nanohole originating from the vanishing of the bulk nonlinearity is hidden inside. In Fig. S8b we show the 3D emission pattern of the SHG for the MoS_2 nanoparticle shown in Figure 3d-f at the optical mode with enhanced SHG.

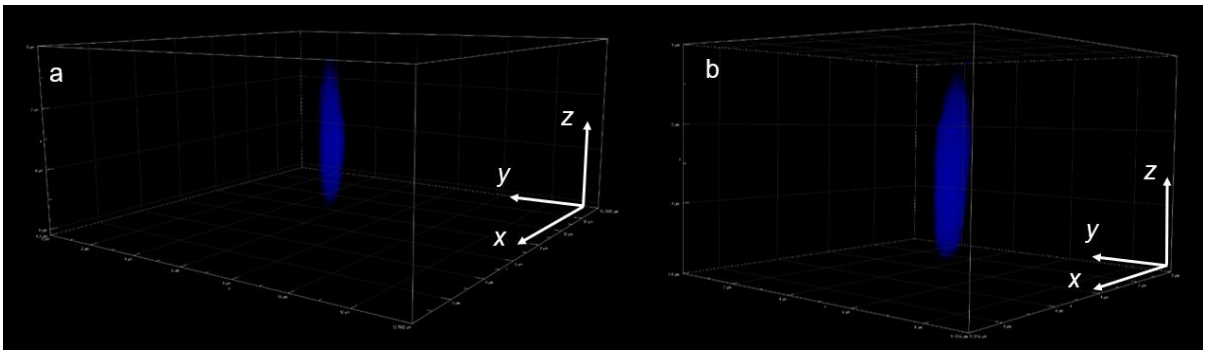

Figure S8 3D emission patterns of the SHG at the optical modes with suppressed (a) and enhanced (b) SHG.

Note 9 Selecting MoS_2 nanoparticles with suppressed SHG by using laser scanning confocal microscopy

In practice, we can identify MoS_2 nanoparticles with suppressed SHG by using a laser scanning confocal microscope. A typical example is shown in Fig. S9 where the SHG emission patterns obtained for MoS_2 nanoparticles distributed on a substrate are presented. The MoS_2 nanoparticle exhibit a suppressed SHG is enclosed by a square. One can observe a nanohole at the center of the emission pattern.

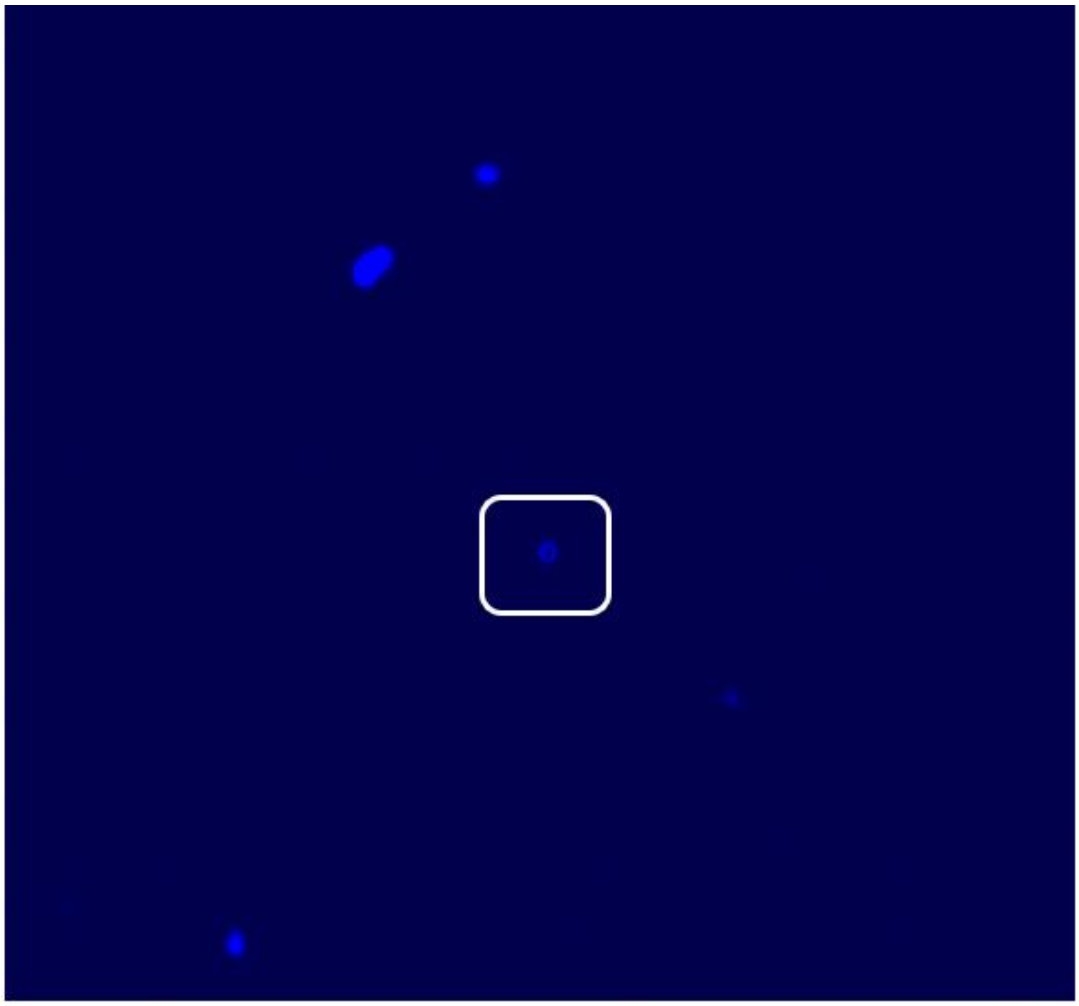

Figure S9 SHG emission patterns obtained for MoS_2 nanoparticles distributed on a substrate by using a laser scanning confocal microscope.

Note 10 MoS_2 monolayer attached on an ITO substrate

In Fig. S10, we show a MoS_2 monolayer attached on an ITO substrate. It was used as a reference sample for the SHG characterization. The MoS_2 monolayer was grown by chemical vapor deposition (CVD) and transferred to the ITO substrate.

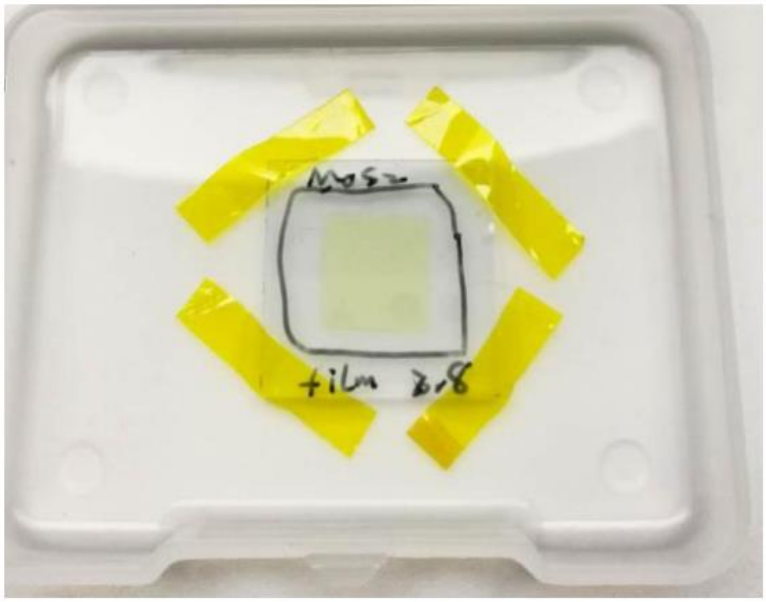

Figure S10 MoS_2 monolayer attached on an ITO substrate used as reference sample.

Note 11 Nonlinear optical responses of MoS_2 nanodisks

The MoS_2 monolayer was grown by chemical vapor deposition (CVD) and transferred to the ITO substrate. In our experiments, we investigated the nonlinear optical responses of hexagonal-prism-like MoS_2 nanoparticles selected from MoS_2 powder. In most cases, their shapes deviate from perfect hexagonal prisms. Thus, it is highly desirable to examine the nonlinear optical response of MoS_2 nanoparticles with regular shapes. For this reason, we fabricated MoS_2 nanodisks from a MoS_2 film grown by chemical vapor deposition on a sapphire substrate with the combination of electron beam lithography and reactive ion etching. The SEM images of MoS_2 nanodisks with different diameters are shown in Figs. S11a and S11b.

We measured the scattering spectra of MoS_2 nanodisks with different diameters, as shown in Fig. S11c. Unfortunately, we did not observed the scattering dips corresponding to the exciton resonances of MoS_2 and any optical resonances in the near infrared spectrum. Also, we did not detect any nonlinear optical signals when such MoS_2 are excited by using femtosecond laser pulses.

In order to find out the physical origin for this abnormal behavior, we measured the X-ray diffraction (XRD) spectrum for the MoS_2 film (red curve) used to fabricate the MoS_2 nanoparticles, as shown in Fig. S11d. The X-ray diffraction spectrum of a hexagonal-prism-like MoS_2 nanoparticle (black curve) is also provided for comparison. It was found that most peaks appearing in the spectrum of the MoS_2 nanoparticle are missing in the spectrum of the MoS_2 film. By comparing the X-ray diffraction spectra reported in literature, it was confirmed that MoS_2 nanoparticles are single crystalline while MoS_2 nanodisks are polycrystalline. This is the reason why both exciton resonances and optical resonances are not observed in MoS_2 nanodisks.

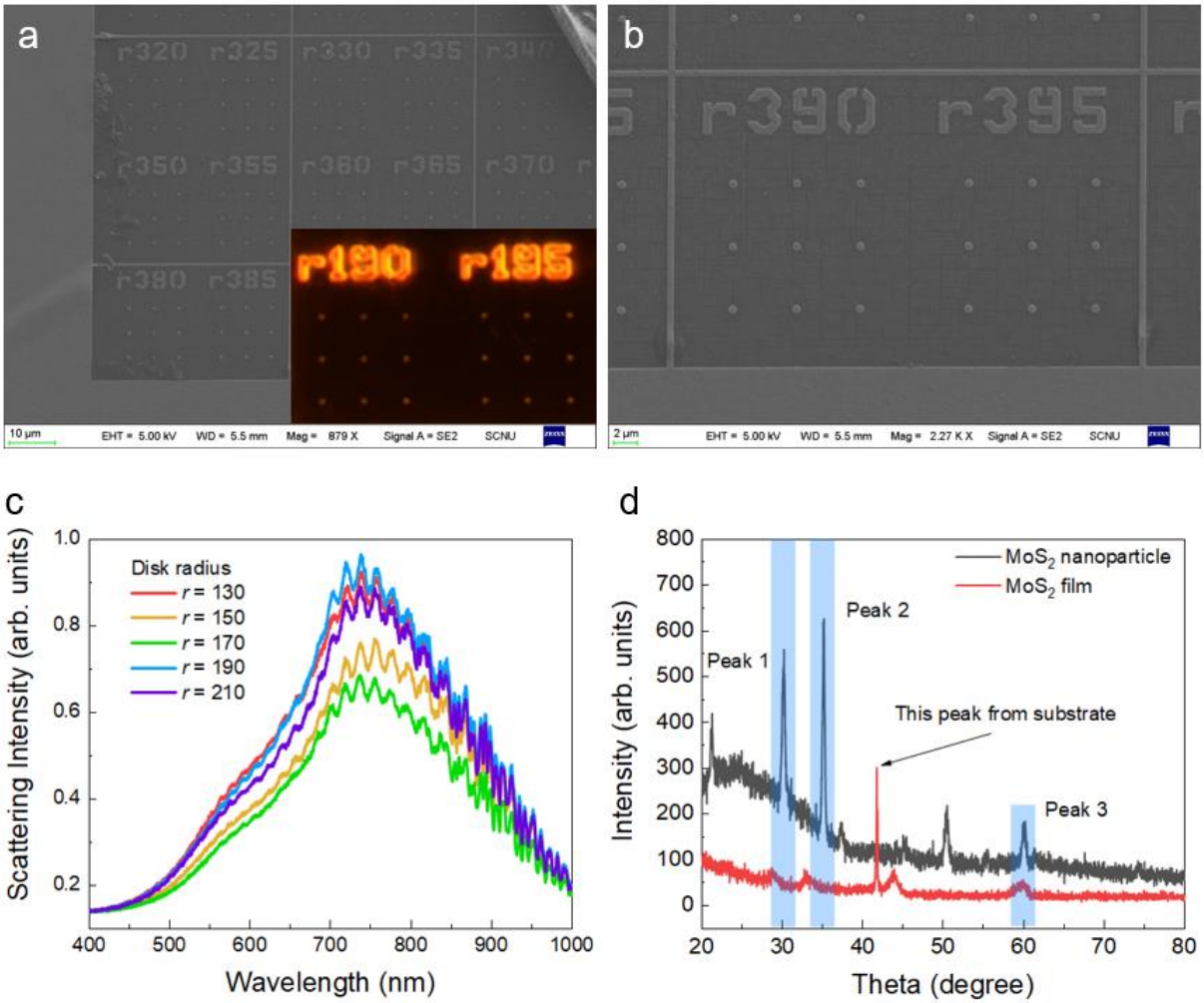

Figure S11 (a) and (b) SEM images of MoS_2 nanodisks with diameters fabricated on a sapphire substrate. (c) Scattering spectra measured for MoS_2 nanodisks with different diameters. (d) X-ray diffraction spectrum measured for the MoS_2 film (red curve) used to fabricate MoS_2 nanodisks. The X-ray diffraction spectrum of a MoS_2 nanoparticle (black curve) is also provided for reference.