Supporting Information

Light modulation based on enhanced Kerr effect in molybdenum

disulfide nanostructures with curved features

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S.1. The emergence of the spatial self-phase modulation (SSPM) phenomenon

When a laser illuminates on the nonlinear optical medium, the refractive index will change with the intensity distribution. A local refractive-index change leads to a relevant phase-shift $\Delta \varphi$ of laser beam after passing through the MoS₂ NP dispersions of thickness L_{eff} , which can be expressed as

$$\Delta\varphi(r) = \left(\frac{2\pi n_0}{\lambda}\right) \int_0^{L_{eff}} n_2 I(r,z) dz$$
(1)

where I(r,z) stands for the distribution of laser intensity, $r \in [0, +\infty)$ is the transverse coordinate of the beam, and λ is the wavelength of applied laser in vacuum. The effective interaction length can be

$$L_{eff} = \int_{L_1}^{L_2} (1 + z^2/z_0^2)^{-1} dz = z_0 atan^{\frac{1}{100}} (Z/z_0) \Big|_{L_1}^{L_2}$$
, where $z_0 = \pi w_0^2 / \lambda$ defined by the $1/e^2$

intensity diameter of laser beam ω_0 and wavelength of incident laser beam; $L = L_2 - L_1$ equals to the thickness of quartz cuvette. In the experiment, L = 10 mm and $\omega_0 = 800 \mu$ m at the front surface of the cuvette. According to the Gaussian beam, the average laser intensity *I* is half of the central light intensity

I(0,z), and it can be assumed that $\Delta \varphi(r) = \Delta \varphi_0 exp^{[iii]}(-2r^2/w_0^2)$ in the range of $r \in [0, +\infty)$. The fields at the two coordinates r_1 and r_2 can interfere with each other when the satisfies the equation

 $\left(\frac{d\Delta\varphi}{dr}\right)_{r=r_1} = \left(\frac{d\Delta\varphi}{dr}\right)_{r=r_2}$. The bright or dark diffriction ring stripe appears when $\Delta\varphi(r_1) - \Delta\varphi(r_2) = m\pi$, where m equals an even or odd integer. When $\Delta\varphi_0 >> 2\pi$, multiple diffraction

rings appear with the total number of rings N which can be bounded up with $N = [\Delta \varphi(0) - \Delta \varphi(\infty)] \cong \Delta \varphi_0 / 2\pi$. Although the total number of rings N is almost proportional to the applied optical field intensity, N does not increase indefinitely.

S.2. The preparation procedure of three-dimensional (3D) featured MoS₂ Nanoparticles (NPs)

Phosphomolybdic acid (PMA, $H_3PO_4 \bullet 12MoO_3 \bullet nH_2O$, 99.9%) is purchased from Sigma Aldrich. Anhydrous ethanol (C₂H₅OH, \geq 95.0%), hydrofluoric acid (HF, 40%), sodium hydroxide (NaOH, \geq 96.0%) are purchased from Sinopharm Chemical Industry Co., Ltd. All chemicals are directly used without any further purification.

The EP-FDU-12 silica template shows a unique mesostructure consisting of large mesopores and small wormlike mesopores. The structure of unique dual-mesoporous, which have a lot of highly interconnected pore channels, could enhance the mobility of precursors and create nanobridges between particles during nanocasting process. In addition, the morphologies and structures of the obtained products can be controlled by the filling amount of precursor, interior surface feature of silica template and reaction conditions. As illustrated in Fig. S1, the precursor PMA has been filled from the interior of the nanoparticle templates, which leads to a less apparent and uncontrollable mesoporous structure. The filling amount of PMA precursor is the most important factor on the final structures of MoS₂ products. With the increase of PMA concentration from 5 mM to 30 mM, the obtained morphologies of MoS₂ nanostructures could be changed from multilayered MoS₂ half-sphere to MoS₂ sphere, yet, both of the nanostructure have the curved surface.



Fig. S1 The schematic of the fabrication process of 3D MoS_2 NPs *via* template method using EP-FDU-12 mesoporous silica as the hard template.