Supplementary Information for "Inkjet-printed tunable 2D metal thiophosphate saturable absorbers for pulsed solid-state laser applications"

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1. AFM and SEM images of $CoPS_3$ and $NiPSe_3$ nanosheets.

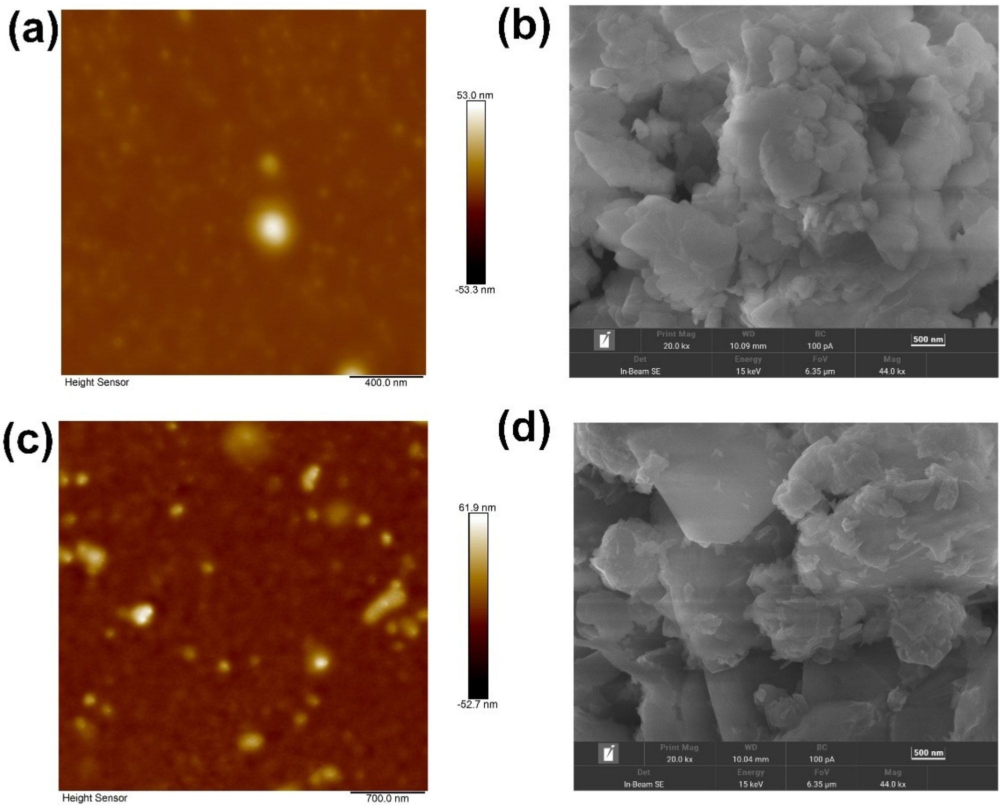

Fig. S1 AFM images of (a) $CoPS_3$ and (c) $NiPSe_3$ nanosheets. SEM images of (b) $CoPS_3$ and (d) $NiPSe_3$ nanosheets.

2. Experimental setup of the OA Z-scan measuring system

The open-aperture Z-scan experimental setup is shown in Fig. S2. The laser pulse source is a commercial mode-locked fiber laser (center wavelength:1030 nm, pulse duration: 500 fs, repetition rate: 25 kHz, FemtoYL-20, YSL photonic Inc., China). The incident laser beam is split into two parts by a 50/50 beam splitter (BS). A power meter (DM1) collects 50% of the portion as a reference light, while the remaining beams are focused by a plano-convex focal lens (FL) with a focal length of 150 mm that produces light and excites the sample, and the resulting signal is collected by another power meter (DM2). Photodetectors (PD1 and PD2) were all connected to an oscilloscope to observe the stability of the signal in real-time. To reduce the error of the measurement, all the light converges to the probe surface of the Power meters and Photodetectors by FL or 50% Partially focal reflector (PFR). The MPT_x nanosheets solution was placed in a cuvette and mounted on a Z-axis translation stage, which was driven by a computer-controlled linear motor.

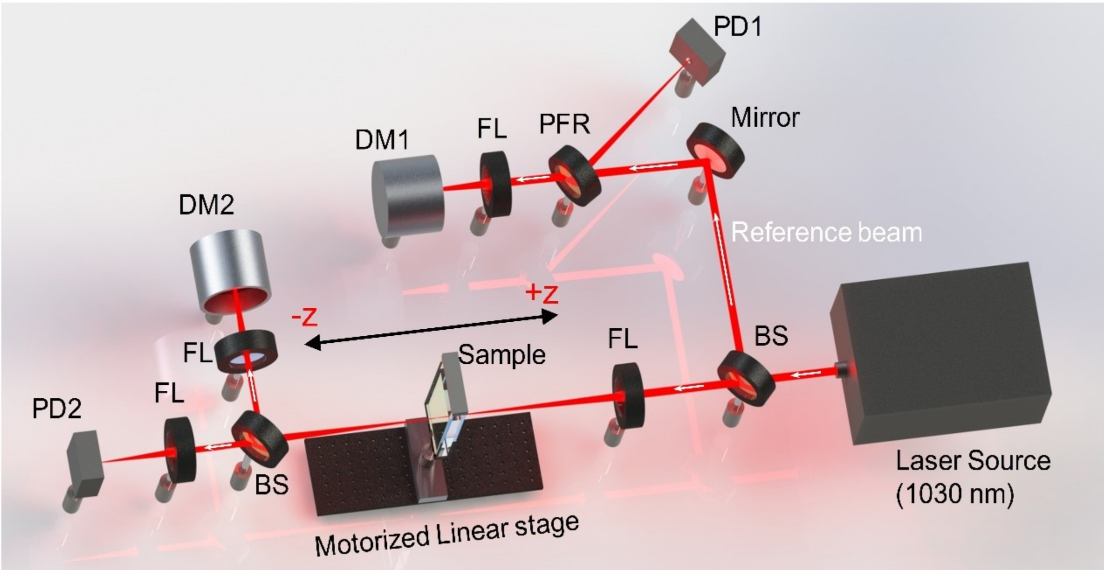

Fig. S2 The schematic diagram of the OA Z-scan system.

3. Equations for the linear and nonlinear parameters

The linear absorption part α_0 has no relation with optical intensity, which can be obtained at low intensity by the following equation:

$$\alpha_0 = \frac{\ln(T_0)}{L} \tag{1}$$

where T_0 is the linear transmittance, and L is the length of the light beam through the solution, which is equal to the length of the cuvette (0.1 cm).

The effective thickness L_{eff} of the sample could be calculated by:

$$L_{eff} = 1 - e^{-\alpha_0 L}/\alpha_0 \tag{2}$$

The Rayleigh length z_0 of one Gaussian beam can be calculated by:

$$z_0 = \pi\omega_0^2/\lambda \tag{3}$$

where ω_0 is the beam waist after the focal length ($45.6\ \mu m$), λ is the wavelength of the laser beam (1030 nm); The calculated $z_0 = 4.89\ mm$, which is longer than the samples.

The imaginary part of the third nonlinear optical susceptibility $lm\chi^{(3)}$ is determined by the β, and the corresponding equation is:[1, 2]

$$\mathrm{Im}\chi^3 = \frac{2\varepsilon_0 c^2 n_0^2}{3\omega}\beta \tag{4}$$

where c is the speed of light in vacuum, ε_0 is the vacuum permittivity, ω is the angular frequency of the light, n_0 is the linear refractive index of the solvent at the wavelength of light, which could calculated by the following analytical expression:[3]

$$n^2 = 1 + \frac{B\cdot\lambda^2}{\lambda^2 - C} + \frac{D\cdot\lambda^2}{\lambda^2 - E} \tag{5}$$

where B, C, D and E is 0.0165, 9.08, 0.8268, and 0.01039 for ethanol solvent.

The Fig. of merit (FOM) for the third-order optical nonlinearity $lm\chi^{(3)}$ and linear coefficients α_0 can be defined as:

$$FOM = \frac{lm\chi^3}{\alpha_0} \tag{6}$$

4. Experimental setup of the pulsed solid-state lasers

A single mirror system is specially designed to achieve PQS lasers based on $NiPSe_3/CoPS_3$ NSs SAs. The pump source consists of a commercially available fiber-coupled 808 nm diode laser capable of continuous and pulsed output modes. The fiber core diameter is 200 μm and the NA is 0.22. A collimating focus lens set (1:0.8) is used to focus the pump beam into the laser gain medium with a diameter of 180 μm. The laser gain medium is a 3 mm × 3 mm × 4 mm Nd: YAG (1.2 at%, cut in the [111] direction). The Nd: YAG crystal is wrapped by an indium foil and mounted in a copper block cooled by the circulating water with a temperature of 17℃. The S1 film (high transmission for 808 nm and high reflection for 1064 nm) plated on the end surfaces of the crystal acts as an input mirror. The S2 film on the other side (high transmission at 1064 nm and high reflection at 808 nm) plays an important role in ensuring that the pump light can be absorbed twice by the crystal and, also, preventing the pump light from affecting the subsequent SAs. The concave output coupler (OC) M2 with a radius of 50 mm was coated with a partial transmission of 5% at 1064 nm. The film S1 and the OC constitute a simple plane-concave resonator, of which the total length was optimized to be about 25 mm. The $NiPSe_3/CoPS_3$ NSs SAs were positioned in the middle between the OC and the Nd: YAG crystal. According to the ABCD matrix theory, the intracavity spot radius are calculated to be approximately 92 μm at the position of the laser gain medium and 106 μm at the position of $NiPSe_3/CoPS_3$ NSs SAs, as shown in Fig. S3.

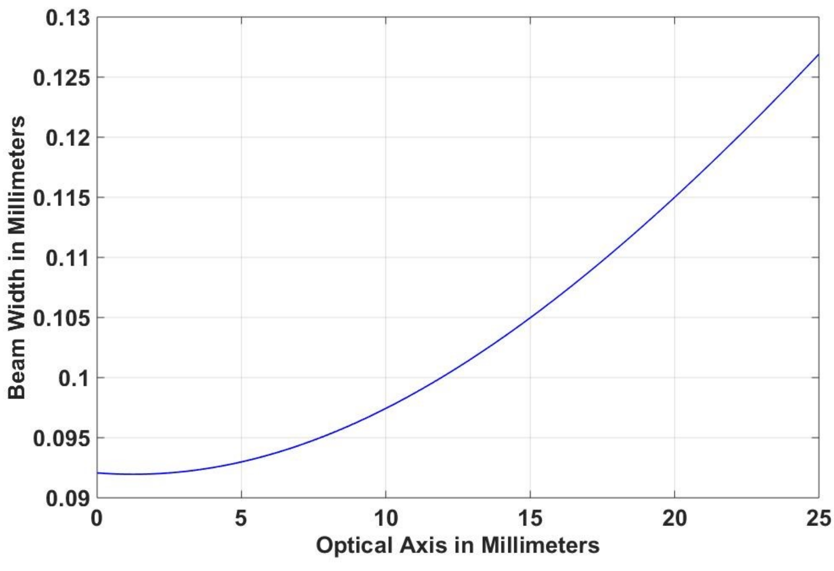

Fig. S3 The intracavity spot size distribution of the PQS single mirror laser resonator

The ring cavity-type folded resonator for QML operations with $CrPSe_4$ NSs SA can produce focused spots of specific sizes within the cavity, reducing intracavity positive dispersion and the overall size of the laser. The pump light output from the LD is transmitted through a multimode fiber with a core diameter of 105 μm and a numerical aperture (NA) of 0.15, and then collimated and focused onto the Yb: KGW crystal by a 1:1 imaging system composed of lenses. The beam waist radius of the pump light formed on the laser crystal is approximately 52 μm. DM is a dichroic mirror with high transmittance for pump light around 980 nm (T > 95% @ 850 – 985 nm) and high reflectance for the intracavity oscillating laser around 1030 nm (R > 99.5% @ 1020 – 1070 nm). The Yb: KGW crystal, wrapped in indium foil, is clamped to a copper heat sink, which is fixed to a TEC cooling device. The temperature of the crystal is controlled by the TEC to be 18°C (±0.5°C) during the experiment. M1 is a concave mirror with a curvature radius of 300 mm (R > 99.5% @ 1030 nm), and M2 and M3 are concave mirrors with a curvature radius of 200 mm (R > 99.5% @ 1030 nm). The output coupler (OC) is a planar partial reflector with a transmittance of 5% (R = 95% @ 1020 – 1070 nm). GTI1, GTI2, and GTI3 are three gires-tournois interferometer (GTI) mirrors providing negative dispersion to balance the self-phase modulation of the laser crystal and compensate for the positive dispersion of the intracavity elements. They provide single-pass negative dispersion of -300 fs^2, -600 fs^2, and -1000 fs^2 in the spectral range of 1030 nm – 1050 nm, respectively. By controlling the number of reflections of the intracavity laser on GTI1, GTI2, and GTI3, we can achieve a total negative dispersion of -1900 fs^2 to -3800 fs^2, enabling the Yb: KGW oscillator to achieve soliton mode-locking in the femtosecond and picosecond ranges. The $CrPSe_4$ NSs SA is placed near the focus of M3. Similarly, the ABCD matrix method was used to design the resonator so that the cavity mode waist on the laser crystal efficiently matches the pump beam waist in both size and spatial position. The radius of the spot sizes of the cavity mode on the laser crystal and $CrPSe_4$ NSs SA are about 43μm and 54 μm, as shown in Fig. S4.

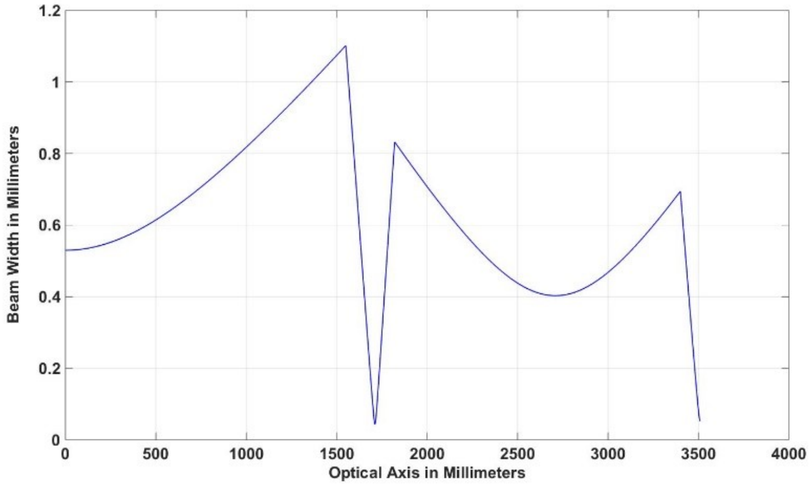

Fig. S4 The intracavity spot size distribution of the QML ring cavity-type folded resonator

Measuring devices and instruments for output parameters: The output power of the pulse lasers was measured using a power meter (30A-P-17, Ophir Optronics Solutions Ltd., Jerusalem, Israel). Output spectra of the lasers were obtained with a spectrometer (USB4000-VIS-NIR, Ocean Optics Inc., Dunedin, FL, USA), covering a range from 200 to 1100 nm. Laser output pulse train signals and RF spectra were collected using an InAsSb photodetector (DET10A/M, Thorlabs, Inc., Newton, NJ, USA), while a high-speed oscilloscope (DPO4104B, Tektronix, Inc., Shawnee Mission, KS, USA) with a bandwidth of 1 GHz and a sampling rate of 5 GHz was used for display measurement.

Reference

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2. N. Xu, H. Li, Y. Gan, H. Chen, W. Li, F. Zhang, X. Jiang, Y. Shi, J. Liu, Q. Wen and H. Zhang, *Advanced Science*, 2020, **7**, 2002209.
3. E. Sani and A. Dell'Oro, *Optical Materials*, 2016, **60**, 137-141.