Supporting Information

$NiPS_3$ Nanoflakes: A Nonlinear Optical Material for Ultrafast Photonics

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Section 1

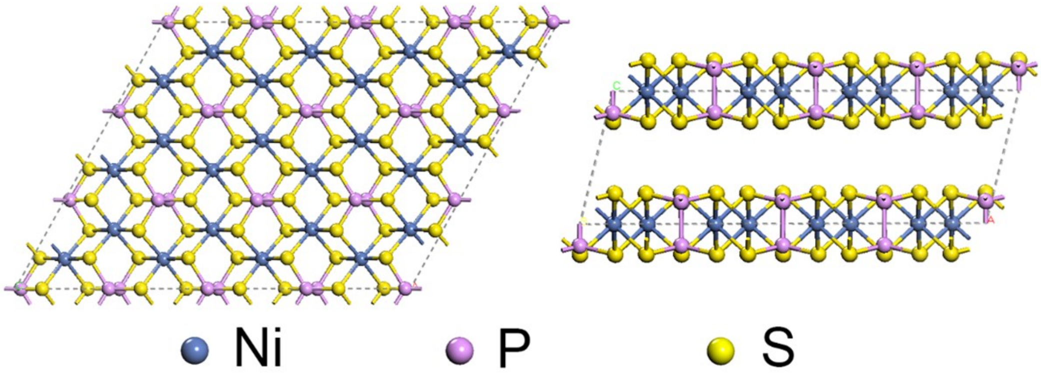

Fig. S1. Crystal structure model of $NiPS_3$.

Table S1. Detailed phase data of $NiPS_3$.

Phase data	
Formula sum	$NiPS_3$
Formula weight	185.87
Crystal system	triclinic
Cell parameters	a=5.812 Å, b=10.070 Å, c=6.632 Å, α=90.00°, β=106.98°, γ=90.00°
Space-group	P 1(1)
Cell ratio	a/b=0.577, b/c=1.518, c/a=1.141
Z	2
Cell volume	371.2 $Å^3$
Formation energy (eV)	-0.601
Density (gm/cc)	3.300

Fig. S2. The optical image of bulk $NiPS_3$ crystals.

Section 2

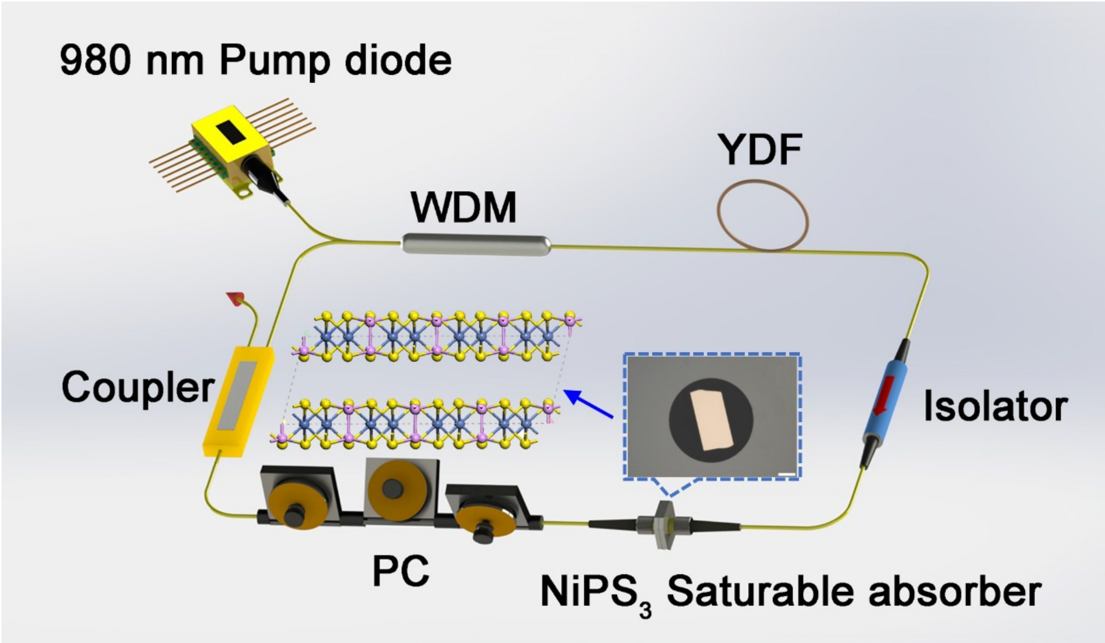

Fig. S3. Configuration of DSR laser cavity. There are 980 nm pump diode, wavelength division multiplexer (WDM), ytterbium-doped fiber (YDF), polarization-independent isolator, $NiPS_3$ saturable absorber, polarization controller (PC) and coupler. Inset: optical image of layered $NiPS_3$ covering on fiber core. The scale bar is 10 μm.

The general cavity configuration basically remained constant apart from the $NiPS_3$ saturable absorber, which was devised into the layered $NiPS_3$ covering on fiber core of the fiber end-facet. Additionally, the total length of cavity was optimized to be 33.4 m.

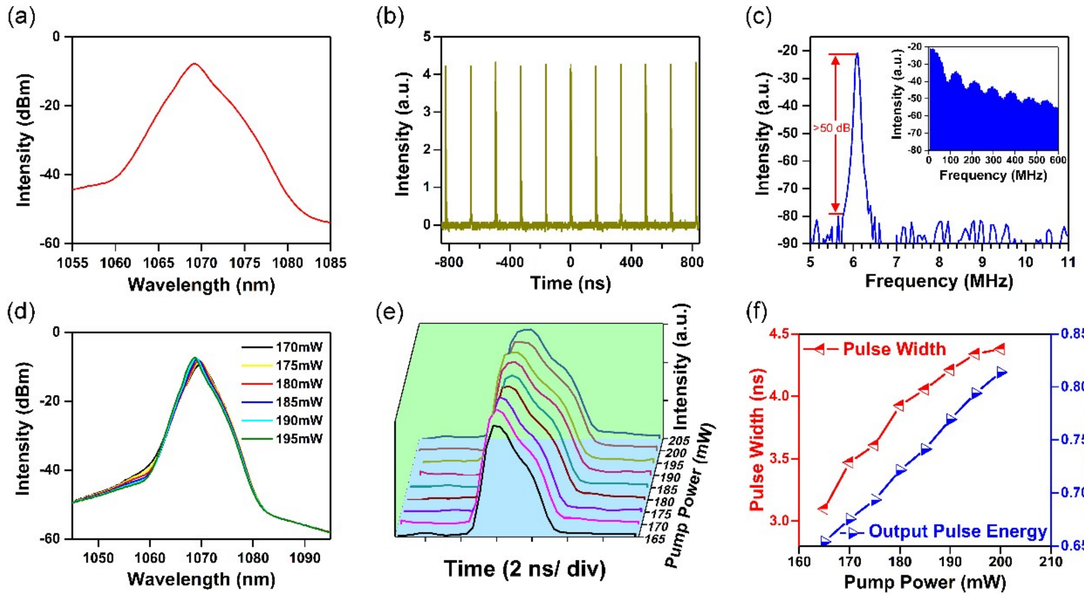

Fig. S4. Characterizations of DSR. (a) spectra. (b) Corresponding pulse train. (c) Radio-frequency trace. (d) The spectra kept nearly unchanged as the pump power increased. (e) Pulse broadening process as the pump power increased. (f) Pulse width and pulse energy under different pump power.

As illustrated in Figure S4, a different self-starting mode-locking pulses regime, dissipative soliton resonance emerged utilizing the lamellar $NiPS_3$. Figure S4a showed the typical smooth optical spectra in DSR regime. The central wavelength of the spectra was 1068.2 nm and the 3-dB spectral bandwidth was 1.85 nm. Figure S4b presented the corresponding pulse-train with the repetition rate of 6.07 MHz. The radio-frequency spectrum was measured to be 55 dB, which shown in the section of Figure S4c. The inset depicted in Figure S4c showed a beatnote envelope modulation, which was a typical feature of the DSR regime.[1]

In addition, the dynamic process of the spectra and pulses profile were carried out to investigate to further confirmed if the obtained pulses were DSR, as shown in Figure S4d-f. It was note that we only changed the pump power once getting the mode-locked state. As it could be seen, when the pump power increased from 165 mW to 205 mW, the central wavelength and the 3-dB bandwidth were almost constant. Correspondingly, the pulse width broadened from 3.1 ns to 4.4 ns. It should be mentioned that the largest output pulse energy we have obtained was limited to be 0.81 nJ. However, according to the previous report,[2] the settings of parameters in the ring cavity laser and the appropriate pump power played a dominant role in obtaining larger pulse energy. Thereby the larger pulse energy can be achieved when appropriately optimizing the parameters in the ring cavity and adjusting the pump power.

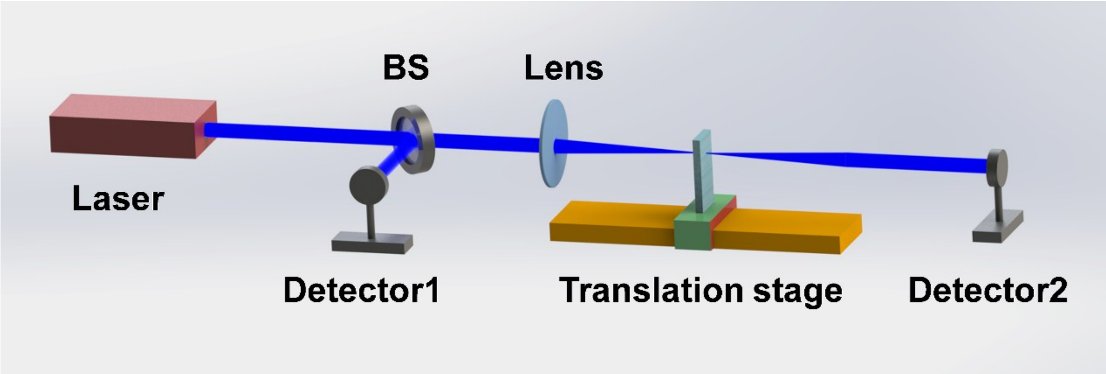

Fig. S5. The setup of open-aperture Z-scan measurement.

Reference

1. K. Krzempek, D. Tomaszewska and K. M. Abramski, *Opt. Express*, 2017, **25**, 24853-24860.
2. E. Ding, P. Grelu and J. N. Kutz, *Opt. Lett.*, 2011, **36**, 1146-1148.