Supporting Materials

The nonlinear absorption coefficient (β) was used to further reveal the optical limiting mechanism in TMDs sheets. A model containing saturation and reverse saturation was applied to analyze the experimental results to obtain the β values. Total absorption coefficient $\alpha(I)$ can be described as equation (1)[1] :

$$\alpha(I) = \frac{\alpha_0}{1+\frac{I}{I_s}} + \beta I \quad (1)$$

Therefore, the modified normalized transmittance using equation (1) can be written as:

$$T(z) = \frac{Q(z)}{\sqrt{\pi} q(z)} \int_{-\infty}^{+\infty} \ln[1 + q(z)\exp(-\tau^2)] d\tau \quad (2)$$

Where

$$Q(z) = \exp[\frac{\alpha_0 L I}{I+I_s}] \quad (3)$$

$$q(z) = \beta I_0 L_{eff} / (1 + \frac{z^2}{z_0^2}) \quad (4)$$

Where

$$L_{eff} = [1 - \exp(-\alpha_0 L)]/\alpha_0 \quad (5)$$

$$z_0 = \pi \omega_0^2 \quad (6)$$

Where $\boldsymbol{\alpha_0}$ is the linear absorption coefficient; $\boldsymbol{I}$ and $\boldsymbol{I_s}$ represent the laser intensity and the saturated absorption intensity, respectively; $\boldsymbol{I_0}$ means the peak intensity at focus while $\boldsymbol{L_{eff}}$ refers to the effective thickness of the sample; $\boldsymbol{L}$ is the thickness of the sample; $\boldsymbol{z_0}$ means the Rayleigh range; $\boldsymbol{\omega_0}$ is the beam waist radius. Using equation (1-6) to fit the experimental results shown in Figure 3(a~f) can get the β values. It is worth mentioning that when the incident laser intensity was in the range of 0.32-1.28 GW/cm^2, the linear transmittance of samples was unified to be 65%, and corresponding linear absorption coefficient α_0 was approximately equal to 2.15 cm^{-1}. The obtained β values were further fitted to analyze the linearity between the β values and the incident laser energies to estimate the absorption ways as shown in Figure S1.

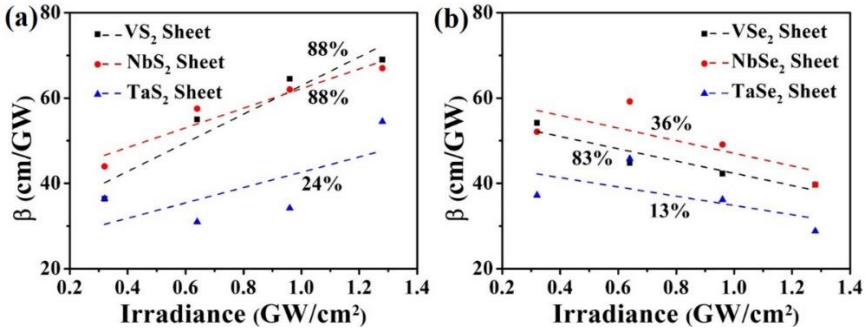

Figure S1. Changes in nonlinear absorption coefficient β of TMDs nanosheets with incident laser energy. a) VS_2, NbS_2 and TaS_2; b) VSe_2, $NbSe_2$ and $TaSe_2$. Dash line: linear fitting data.

The β value would be irrelevant or linearly associated with the incident laser energy in materials with purely TPA or three-photon absorptions[2]. A higher value of linearity indicated a more contribution from TPA or three-photon absorption ways. So, the linearity between the β values and the incident laser energies can be correlated to the absorption ways. The linearity between the β values and irradiance intensity was calculated to be 88% for VS_2, 88% for NbS_2, 24% for TaS_2, 83% for VSe_2, 36% for $NbSe_2$ and 13% for $TaSe_2$, which

indicated that both TPA and RSA existed in these samples. The metal sulfides showed a higher linearity between the β values and irradiance intensity than corresponding metal selenides, especially in NbX_2 (NbS_2 > $NbSe_2$) and TaX_2 (TaS_2 > $TaSe_2$), which may be connected to the different absorption ways.

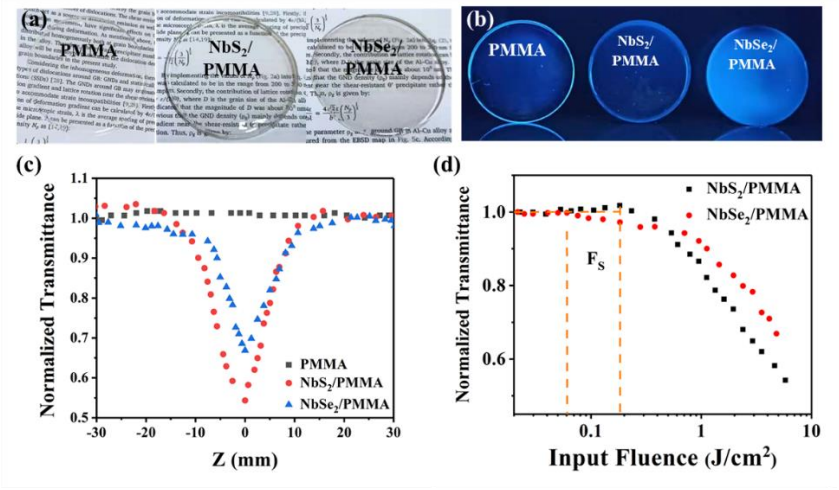

Figure S2. (a) Photos of pristine PMMA (left), NbS_2/PMMA (middle) and $NbSe_2$/PMMA (right). (b) Photos of PMMA (left), NbS_2/PMMA (middle) and $NbSe_2$/PMMA (right) under the UV lamp illumination. (c) Open-aperture Z-scans of pristine PMMA, NbS_2/PMMA and $NbSe_2$/PMMA. (d) The relationship between the normalized transmittance and the input laser fluences of NbS_2/PMMA and $NbSe_2$/PMMA.

In order to explore the application potential of TMDs nanosheets in laser protection equipments and eliminate the influence of nonlinear scattering on optical limiting performance, NbS_2/PMMA and $NbSe_2$/PMMA composites were prepared to evaluate their optical limiting performance in solid state. The photograph of pristine PMMA, NbS_2/PMMA and $NbSe_2$/PMMA were shown in Figure S2(a). The linear transmittance of NbS_2/PMMA (80%) and $NbSe_2$/PMMA (85%) are close to that of pristine PMMA (90%). And NbS_2 and $NbSe_2$ were uniformly dispersed in the solid sample under the UV lamp illumination in Figure S2(b). The normalized transmittances of pristine PMMA, NbS_2/PMMA and $NbSe_2$/PMMA obtained via open-aperture Z-scan were presented in Figure S2(c). The normalized transmittance of NbS_2/PMMA and $NbSe_2$/PMMA was low to 54% and 66% at 1.28 GW/cm^2 while the content of NbS_2 and $NbSe_2$ nanosheets was as low as 0.053 wt‰. While the normalized transmittance of pristine PMMA was about 100% at all scan positions. The relationship between the normalized transmittance and the input laser fluences of NbS_2/PMMA and $NbSe_2$/PMMA was presented in Figure S2(d). The F_S values of NbS_2/PMMA and $NbSe_2$/PMMA were 0.18 J/cm^2 and 0.06 J/cm^2 respectively.

References

[1] Kurian P A, Vijayan C, Sathiyamoorthy K, Sandeep CSS. Excitonic transitions and off-resonant optical limiting in CdS quantum dots stabilized in a synthetic glue matrix. Nanoscale Res Lett 2007, 2, 561-8.

[2] Wang J, Gu B, Ni XW, Wang HT. Z-scan theory with simultaneous two- and three-photon absorption saturation. Opt Laser Technol. 2012, 44, 390-3.