Electronic Supplementary Material (ESI) for Nanoscale.
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Supplementary Information for

Ultrafast Auger process in few-layer $PtSe_2$

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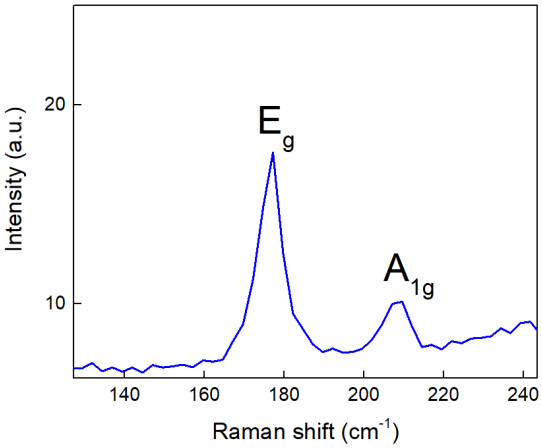

Fig. S1 Raman spectrum of the $PtSe_2$ sample. The E_g mode at $\sim 177\ cm^{-1}$ and the A_{1g} mode at $\sim 288\ cm^{-1}$ are due to the in-plane and out-of-plane vibration of selenium atoms, respectively, in good agreement with previous studies [1,2].

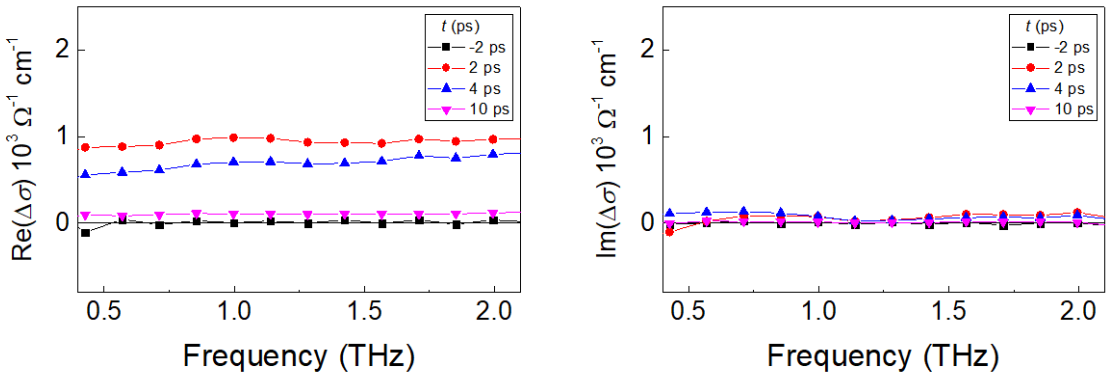

Fig. S2 Complex photoconductivity ($\Delta\sigma$) spectra of the $PtSe_2$ sample for several pump-probe time delays (*t*s). The left and right panels show the real and imaginary parts of $\Delta\sigma$, respectively. The pump fluence is $600\ \mu J\ cm^{-2}$.

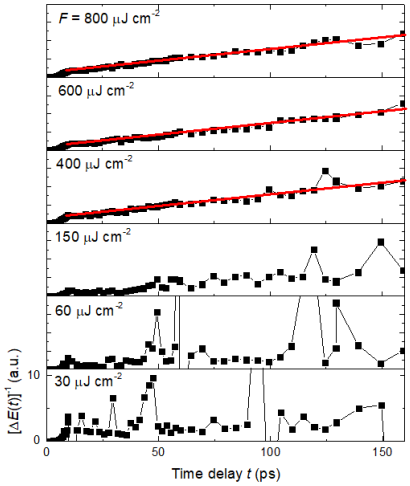

Fig. S3 Pump fluence-dependent $[-\Delta E(t)]^{-1}$ calculated from $-\Delta E(t)/E_0$ traces in Fig. 2a in the main text. As discussed in the main text, $[-\Delta E(t)]^{-1}$ shows linear relationship with t a high pump fluences ($F \geq 400$ μJ cm^{-2}) due to bimolecular Auger dynamics, as indicated by red linear fits. However, it is hard to discern such linearity at lower pump fluences because most excited carriers are trapped by defects before $t \sim 10$ ps and the signal is relatively weak at $t > 10$ ps.

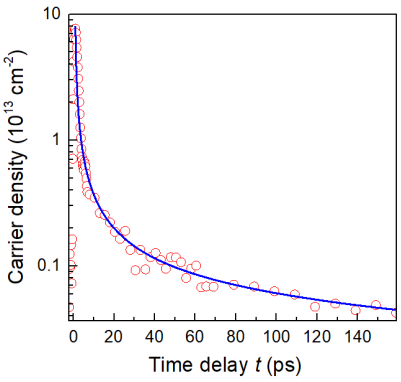

Fig. S4 Red dots are scaled $-\Delta E/E_0$ of the second CVD few-layer $PtSe_2$ sample. The experimental condition is identical to that of Figure 4 in the main text. The blue line indicates simulated temporal evolution of the free carrier density using Eq. (4). The resulting defect density is $n_d \sim 7.0 \times 10^{13}$ cm^{-2}, which is almost similar to that of the main text but slightly higher. The corresponding Auger coefficient is $C_3 \sim 5.3 \times 10^{-17}$ cm^4 s^{-1}. Consequently, the bimolecular Auger recombination rate, $k \sim 3.4 \times 10^{-3}$ cm^2 s^{-1}, is almost the same as that of the first sample in the main text.

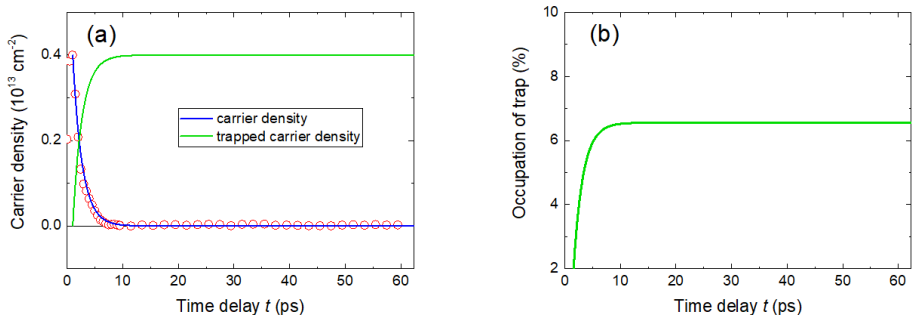

Fig. S5 (a) Red dots are scaled $-\Delta E/E_0$ measured at $F = 30$ μJ cm^{-2}. The blue (green) line are simulated temporal evolution of the free (trapped) carrier density using Eq. (4). (b) Corresponding trap saturation dynamics. It can be seen that only 6.4% of the trap sites are filled and no trap saturation has occurred. This agrees well with the fact that the carrier density after $t \sim 10$ ps in (a) shows an zero-like response.