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Supplementary Materials for

Observation of ultralong valley lifetime in WSe_2/MoS_2 heterostructures

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Supplementary Text

1. Pump fluence dependence of the photo-induced circular dichroism signal in the heterostructure

Figure S1 shows the pump-induced circular dichroism (CD) signal of WSe_2/MoS_2 heterostructure measured through $-\Delta(RC_{\sigma+} - RC_{\sigma-})$ with different pump fluence from 65 $nJ\cdot cm^{-2}$ to 650 $nJ\cdot cm^{-2}$. The pump light energy is at 1.78 eV and probe light at 1.71 eV. All spectra are normalized by pump fluence. The fluence-normalized CD signal shows a constant initial amplitude, indicating that the population of valley-polarized holes generated in WSe_2 is linearly proportional to the pump fluence. This is consistent with the efficient generation of valley-polarized holes due to the ultrafast charge transfer process in the heterostructure. On the other hand, the decay dynamics of the CD signal depends sensitively on pump fluence: the initial fast decay component decreases with lower pump fluence. Such fluence dependence suggests that higher order processes (such as Auger recombination) that involves interactions between photo-generated carriers dominate the initial decay dynamics when the hole density is high. To avoid this complication, we focus on the slow decay component characterizing single-hole behaviour at low excitation density.

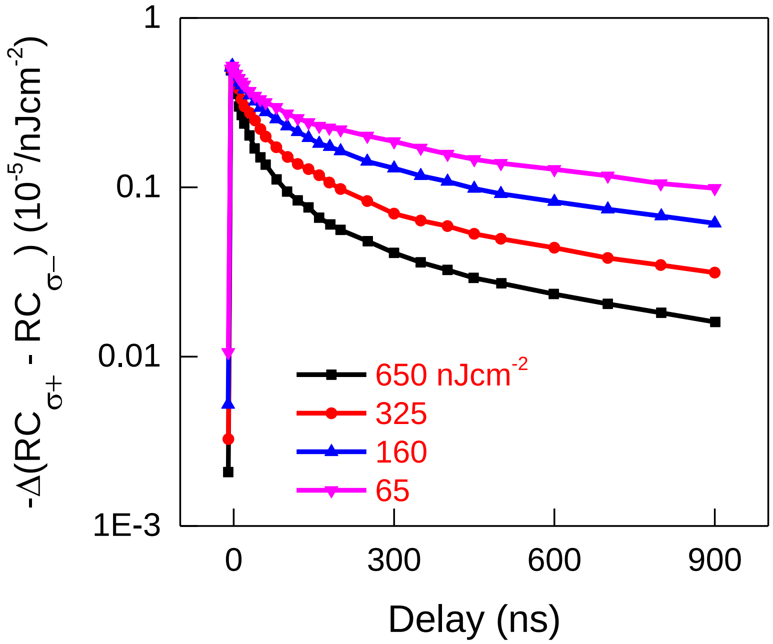

fig. S1. CD signal of the heterostructure at 10 K with different pump fluence from 65 to 650 nJ cm^{-2}. The initial fast decay components depend sensitively on the pump fluence, which is dominated by interactions between photo-excited carriers.

2. Estimation of valley depolarization lifetime at 10 Kelvin

The degree of valley polarization remains almost a constant up to 2.5 μs at 10 Kelvin (Fig. 4c), indicating an ultralong valley depolarization lifetime. We estimate the lower limit of valley depolarization lifetime based on our experimental uncertainty. We determine the experimental uncertainty of valley polarization by calculating the standard deviation σ_P of the measured data between 500ns and 2500ns (see Fig. 4C in the text), and obtain a mean value $\overline{P} = 0.71$ and $\sigma_P = 0.012$. As a conservative estimation, the experimental uncertainty is within $3\sigma_P = 0.036$, which indicates that the valley polarization decay in 2μs is within 5%. This uncertainty gives a lower limit of depolarization lifetime to be 40 μs, and there is no upper bound. The actual value for the valley depolarization lifetime can well be hundreds of μs at 10K.

3. Possible mechanism of valley depolarization

The inter-valley scattering of holes in WSe2 requires a large momentum change from K to K', and a simultaneous spin flipping. One possible mechanism for intervalley scattering is phonon-assisted Elliot-Yafet Mechanism.

In WSe2, the spin at finite momentum is not a perfect quantum number, and in general the wave function of Bloch states have a mixture of the two species of spins. The picture that electrons and holes have perfect up and down spin in the K and K' valley is only a simplified approximation: it is valid for the high symmetry K and K' points, but does not hold strictly for states away from the K and K' points. Therefore, only the scattering between states exactly at K and K' are strictly forbidden, while states close to but not exactly at K and K' can directly scatter between each other. Such spin-flipping process, often called Elliott-Yafet Mechanism, has a characteristic temperature dependence of (*38*)

$$\tau_{EY}^{-1}(T) \sim T^2 \tau_p^{-1}(T)$$

where τ_p is the momentum scattering lifetime that conserves spin. The T^2 dependence originates from the fact that at higher temperature, the thermally excited carriers can be further away from band energy minimum (K or K'), and therefore will have more efficient scattering between opposite spins. However, T^2 is a rather weak temperature dependence that cannot describe the strong temperature dependence we observe, which shows an energy-activated type of behavior that roughly follows $\tau \sim e^{\frac{\Delta}{k_B T}}$, with $\Delta \sim 20\text{meV}$.

The sensitive temperature dependence in our experiment presumably arises from the momentum scattering lifetime τ_p. Indeed, such temperature dependence is expected for phonon-assisted intervalley scattering at low temperature (*35*,*36*). Furthermore, the activation energy of ~20meV does match with WSe_2 phonon energy at K point (to satisfy momentum conservation of intervalley scattering). In this sense, a phonon-assisted intervalley scattering, accompanied by spin-flipping through Elliott-Yafet Mechanism, can potentially account for our observed temperature dependence.