Supporting Information: Long-lived Hole Spin/Valley Polarization Probed by Kerr Rotation in Monolayer WSe_2

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1. Finding τ_3 from the Kerr rotation overlap measurements

The time evolution of the long-lived Kerr rotation signal can be described by an exponential decay with a characteristic lifetime τ_3:

$$A_0 \exp(-\frac{t}{\tau_3}) \tag{1}$$

where A_0 is the Kerr rotation amplitude. Since τ_3 is longer than the laser repetition period t_{rep} (13.16 ns) at low temperature, the previous pump pulses also contribute to the Kerr rotation signal:

$$\begin{aligned} A(t) &= A_0[\exp(-\frac{t}{\tau_3}) + \exp(-\frac{t+t_{rep}}{\tau_3}) + \exp(-\frac{t+2t_{rep}}{\tau_3}) + \cdots] \\ &= A_0 \exp(-\frac{t}{\tau_3})[1 + \exp(-\frac{t_{rep}}{\tau_3}) + \exp(-\frac{2t_{rep}}{\tau_3}) + \cdots] \\ &= A_0 \exp(-\frac{t}{\tau_3}) \frac{1}{1-\exp(-\frac{t_{rep}}{\tau_3})} \end{aligned} \tag{2}$$

In overlap Kerr rotation measurements, the delay time t is set at 1 ns and -200 ps (12.96 ns) and the Kerr rotation amplitude $A(1)$ and $A(12.96)$ are obtained from the Gaussian heights. Therefore:

$$A(1) = A_0 \exp(-\frac{1}{\tau_3}) \frac{1}{1-\exp(-\frac{t_{rep}}{\tau_3})} \tag{3}$$

$$A(12.96) = A_0 \exp(-\frac{12.96}{\tau_3}) \frac{1}{1-\exp(-\frac{-t_{rep}}{\tau_3})} \tag{4}$$

We can solve for τ_3 (in the unit of nanosecond) from equation 3 and 4:

$$\tau_3 = \frac{11.96}{\ln(\frac{A(1)}{A(12.96)})} \qquad (5)$$

Here we present an example of extracting τ_3 at 5 K. Fig. S1(a)-(f) are six overlap Kerr rotation measurements. The τ_3 obtained using equation 5 are also shown in each figure. The mean value of τ_3 is 67.1 ns and the standard deviation is 21.0 ns, which are shown in Fig. 2(c) in the main text.

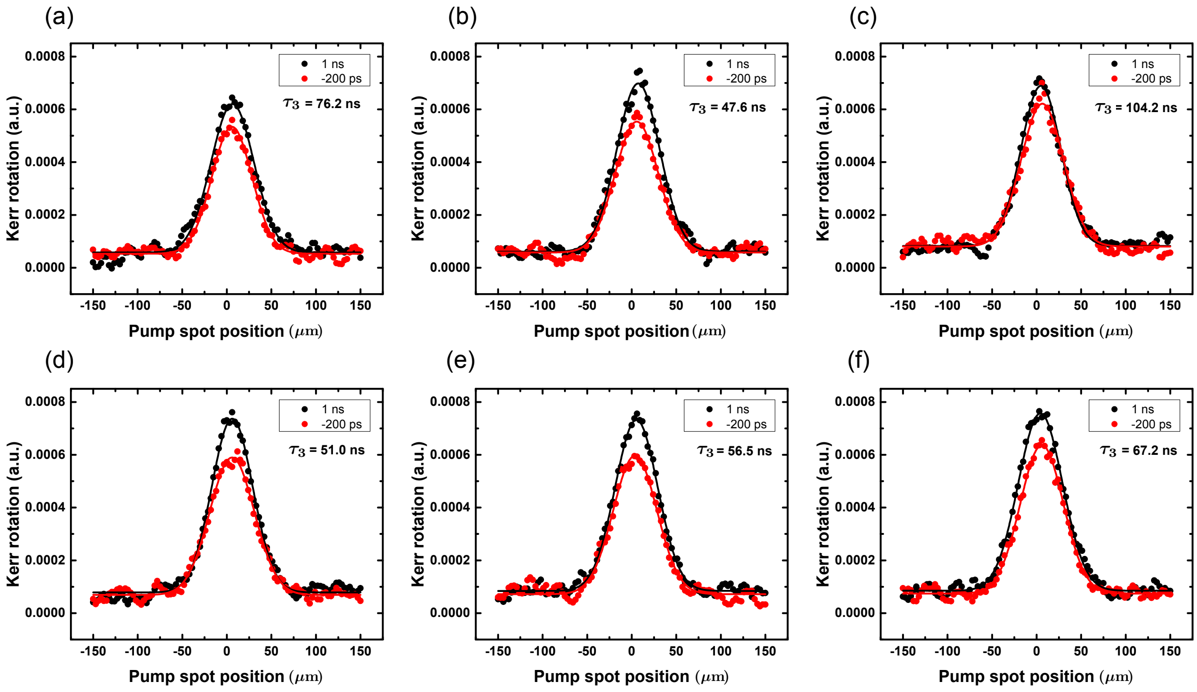

Figure S1: Six runs of overlap Kerr rotation measurement at 1 ns (black points) and -200 ps (red points) with corresponding Gaussian fits.

2. Determining the free exciton and localized exciton peak positions

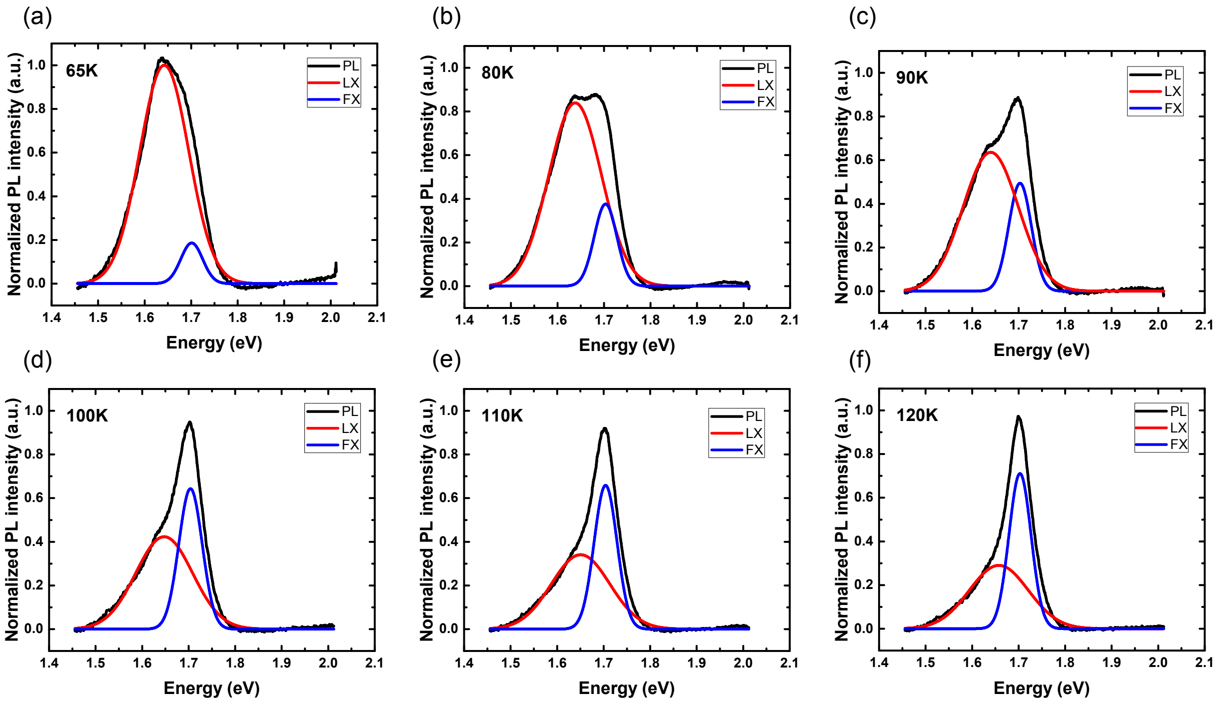

Figure S2: PL spectra at different temperatures. The localized exciton peak (red line) and free exciton peak (blue line) are obtained by fitting the PL spectra using the sum of two Gaussians. The primary PL emission peak changes from the localized exciton peak to the free exciton peak with the increase of temperature.

For temperatures above 120 K, the free exciton peak positions are obtained by fitting the PL spectrum using a single Gaussian. For temperatures between 65 K and 120 K, the free exciton peak positions and localized exciton positions are obtained by fitting the PL spectra with the sum of two Gaussians. For temperatures below 65 K, the localized exciton peak positions are obtained by a single Gaussian fit. These peak positions are label with diamonds in Fig. 3(a) in the main text.

3. Determining the Kerr rotation peak

An example is shown here to illustrate how to get the Kerr rotation peak positions (the green squares in Fig. 3(c) in the main text.) The overlap Kerr rotation measurements are conducted at 1 ns delay time with different pump photon energies (Fig. S3(a)). 1 ns is chosen so that only the polarization from the resident holes contributes to the Kerr signal. The peak height of each curve is extracted and normalized by the largest one (Fig. S3(b)). We define the upper and lower bound of the peak position as 90 % of the maximum height. In this example they are 1.717 eV and 1.712 eV, respectively. The peak position is defined as the average of the upper and lower bound (1.7145 eV) and the error bar is defined as the difference of the upper and lower bound (0.005 eV).

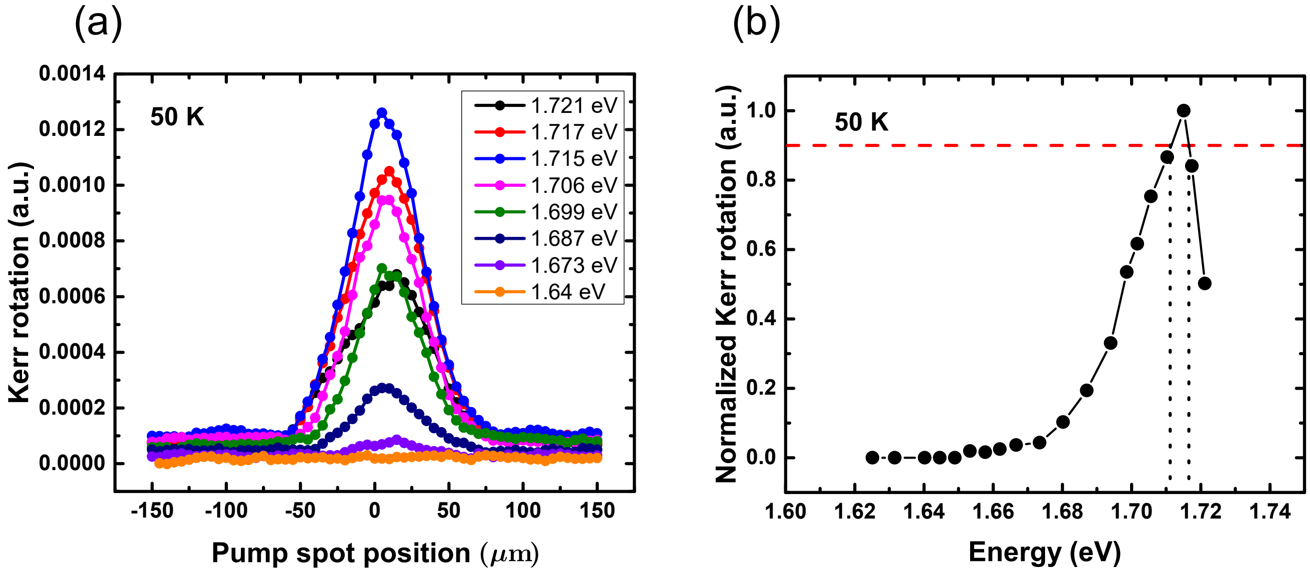

Figure S3: Overlap Kerr rotation measurements (1 ns delay time) at different pump energies at 50K. The pump induced Kerr signal decreases when the pump energy is tuned away from the free exciton emission energy (1.703 eV at 50 K). No Kerr rotation signal is observed when the pump energy is tuned close to the localized exciton emission energy (1.654 eV at 50 K). (b) Normalized peak heights of each curve from (a) versus pump photon energy. The horizontal red dashed line shows 90 % of the maximum Kerr rotation and the black dashed lines indicate the upper (1.717 eV) and lower bound (1.712 eV) of the peak position.

4. TRKR scans around zero delay time

By doing pump energy dependent overlap Kerr rotation measurements at 1 ns delay time, we show that the resident hole polarization cannot be generated with pump energy close to the localized exciton emission energy (see Section 3). However, this does not rule out the possibilities of generating short-lived exciton or trion polarizations, since they die out in approximately 200 ps (see the main text). In order to confirm this, we perform TRKR around zero delay time with the pump energy close to the localized exciton emission energy. As shown in Fig. S4, there is no Kerr signal around zero delay time when pumping at 1.632 eV. This verifies that no spin/valley polarization can be generated with excitation close to the localized exciton emission energy. TRKR with 1.706 eV pump energy is also shown as a comparison.

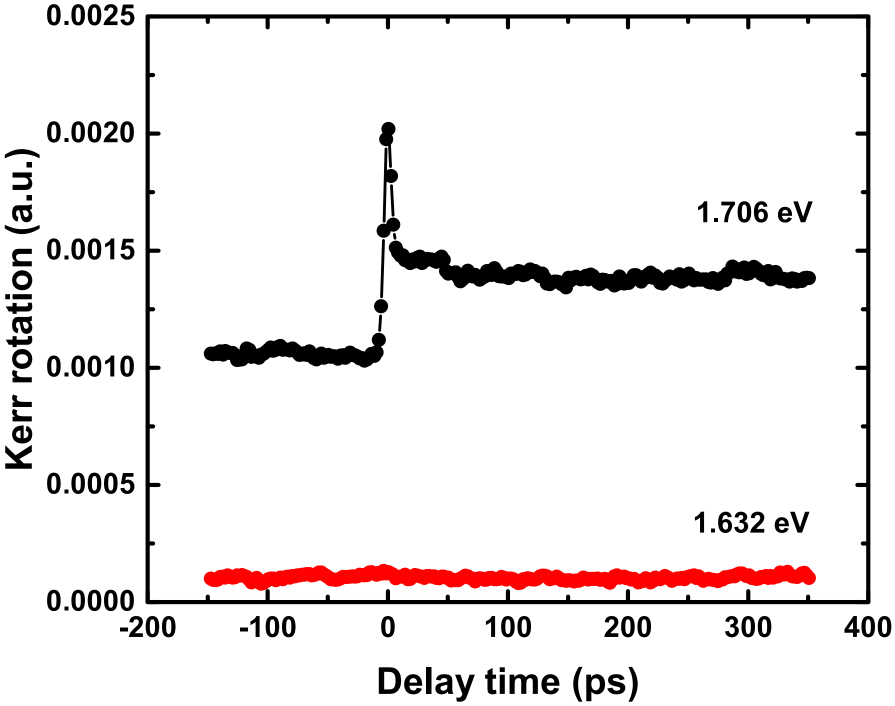

Figure S4: TRKR scans around zero delay time with different pump energies (1.706 eV and 1.632 eV) at 10 K (curves are not shifted).

5. Pump power dependence

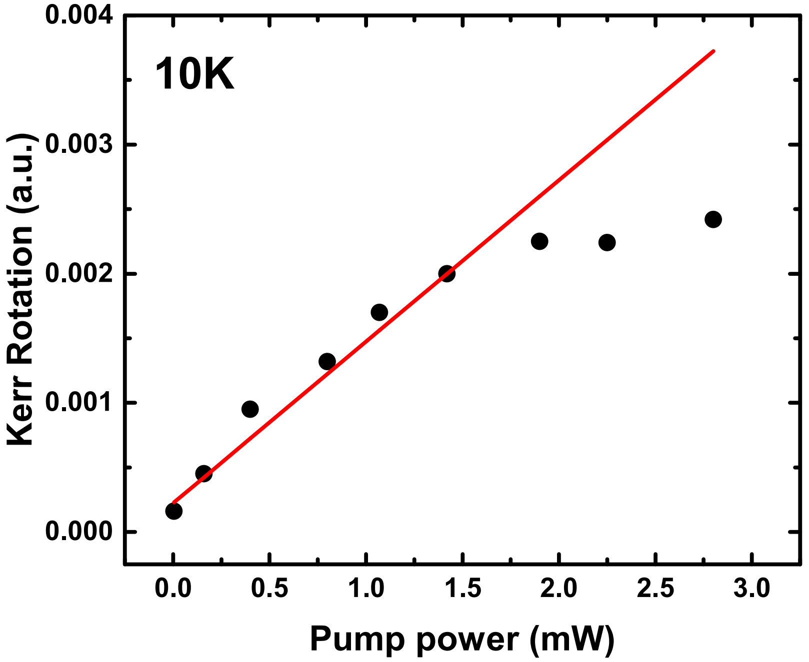

Figure S5: Kerr rotation amplitude (obtained from the overlap Kerr rotation measurements at 1 ns delay time) versus pump pulse power at 10 K. The pump and probe photon energy is 1.708 eV and the probe pulse power is fixed at 0.37 mW. The Kerr rotation amplitude has a linear dependence with pump power when the pump power is below 1.5 mW. The red line is a linear fit of the data points below 1.5 mW. The pump pulse power is ∼ 1 mW and the probe pulse power is ∼ 0.4 mW for all the measurements in this paper.

6. TRKR measurements of Sample 2

The TRKR measurement results of Sample 2 are shown here. A long-lived Kerr rotation signal is also observed and no obvious magnetic field dependence is observed.

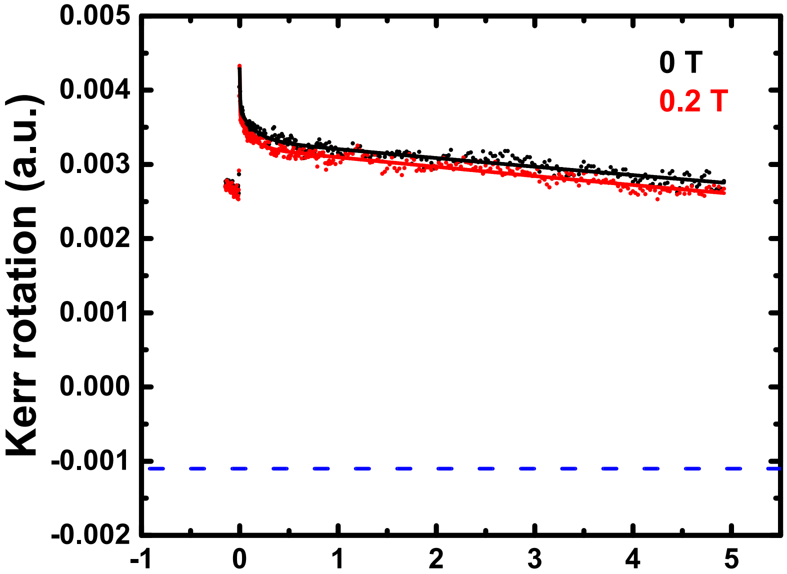

Figure S6: TRKR measurements of Sample 2 with 0 and 0.2 T transverse magnetic field at 10 K. The blue dashed line indicates the background level (when the pump and probe spot are totally separated). A large portion of pump-induced spin/valley polarization persists beyond 5 ns. The lifetimes extracted from the fittings are similar for 0 T and 0.2 T (approximately 35 ns).

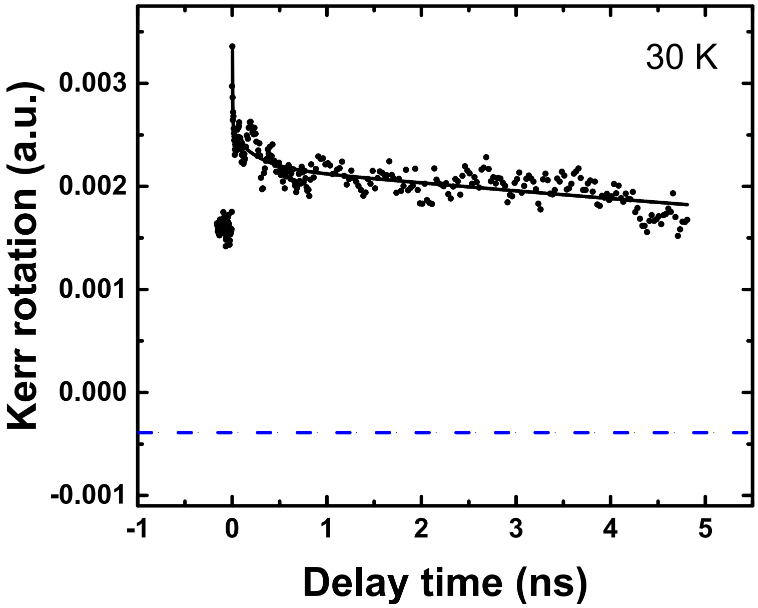

Figure S7: TRKR scan of Sample 2 at 30 K with zero magnetic field. The blue dashed line indicates the background level. The lifetime extracted from the fitting is approximately 30 ns.