

Supplementary material: Ultrafast valley-selective coherent optical manipulation with excitons in WSe₂ and MoS₂ monolayers

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SUPPLEMENTARY NOTES

SUPPLEMENTARY NOTE 1: Experimental setup

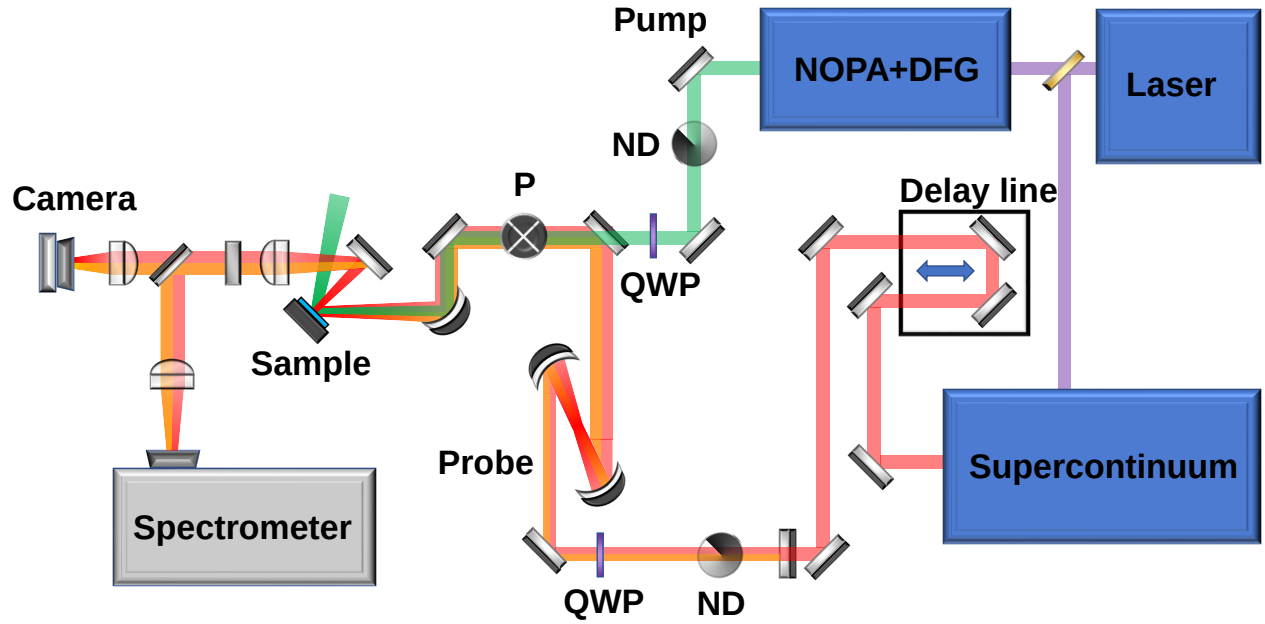
The layout of the experimental setup is shown in Supplementary Fig. 1. The delay between the pump and probe beams is controlled by an optical delay line. The polarizations of both the pump and the probe pulses are controlled using broadband quarter wave plates. While for the probe we find that the quality of circular polarization in the sample plane is preserved after the focusing by the parabolic mirror, the polarization was strongly elliptical for the mid-infrared pump pulse. For this reason we added a periscope with two silver mirrors, at which the beams are twice reflected with the incidence plane in the vertical direction. During these two reflections, the phase shift between the s- and p-polarization components acquired at the last two mirrors is compensated and we reach circular polarization of the infrared pump beam.

The spectrum of the probe beam reflected from the sample, which is illuminated by the pump beam, is collected and detected by a spectrometer with cryo-cooled CCD camera (Andor iDus 420). Alternatively, the probe light serves as illumination source for imaging of the sample in a microscope setup with standard CCD camera. To enhance the signal/noise ratio in the experiments we split the probe supercontinuum pulse before the sample to two replicas using a neutral density filter. The second replica is sent to the parabolic focus-

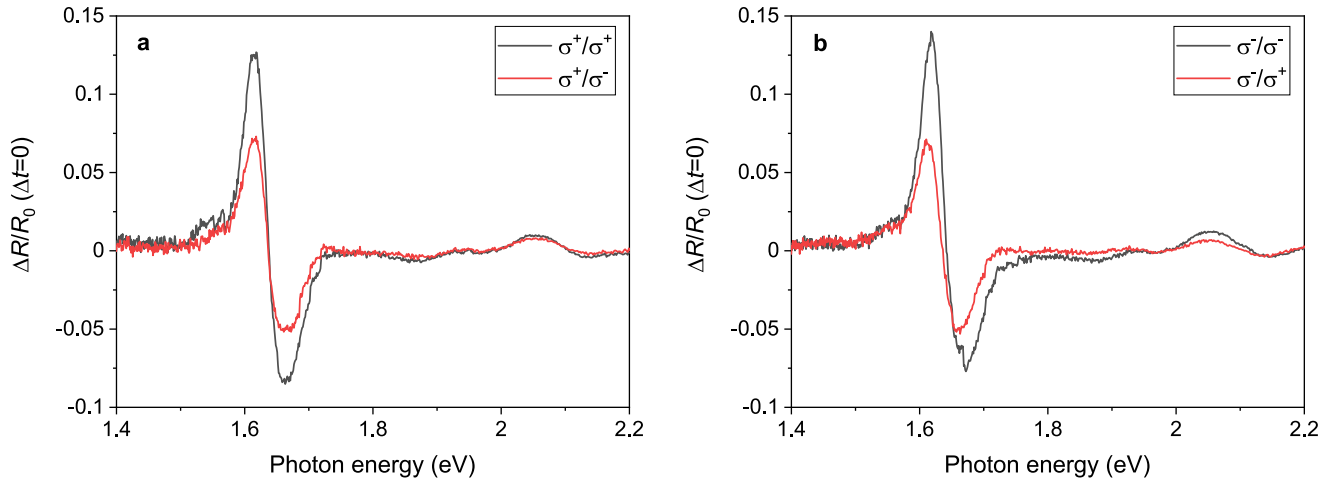
ing mirror under a small angle leading to its focusing away from the region illuminated by the pump beam. The CCD camera in the spectrograph is used in the multitrack mode, which allows to detect both spectra of the probe in the same time. By dividing the spectrum of the probe transmitted through the illuminated region by the reference spectrum we suppress the noise due to the supercontinuum fluctuations.

SUPPLEMENTARY NOTE 2: Verification of signal symmetry for different combinations of circular polarizations

In Fig. 2 we show the transient reflectivity spectra of the WSe₂ monolayer in zero time delay between the pump and probe pulses measured with different combinations of circular polarizations of both beams. By this measurement we exclude any experimental artifacts to play role in the observed valley-selective signals. This could potentially come from a small displacements of one of the beams when the half wave plate generating the circular polarization is rotated by $\pi/2$, which could influence the spatial overlap or the position on the sample. However, because the signals for co-rotating (black curves in Supplementary Fig. 2) and counter-rotating (red curves in Supplementary Fig. 2) polarizations are virtually the same for both handednesses of the circular polarization of the pump, such experimental artifacts are excluded.



Supplementary Figure 1. Layout of the experimental setup used for pump probe experiments. P-periscope, QWP-quarter wave plate, ND-neutral density filter, NOPA+DFG-noncolinear optical parametric amplifier with subsequent difference frequency generation.



Supplementary Figure 2. Measured transient reflectivity spectrum of WSe₂ monolayer with different combinations of circular polarizations of the pump and probe pulses. Black curves show measurements with co-rotating polarizations (σ^+/σ^+ and σ^-/σ^-) and red curves show counter-rotating polarizations (σ^+/σ^- and σ^-/σ^+).