Supporting Information for

Thickness-Dependent Ultrafast Charge-Carrier Dynamics and Coherent Acoustic Phonon Oscillations in Mechanically Exfoliated $PdSe_2$

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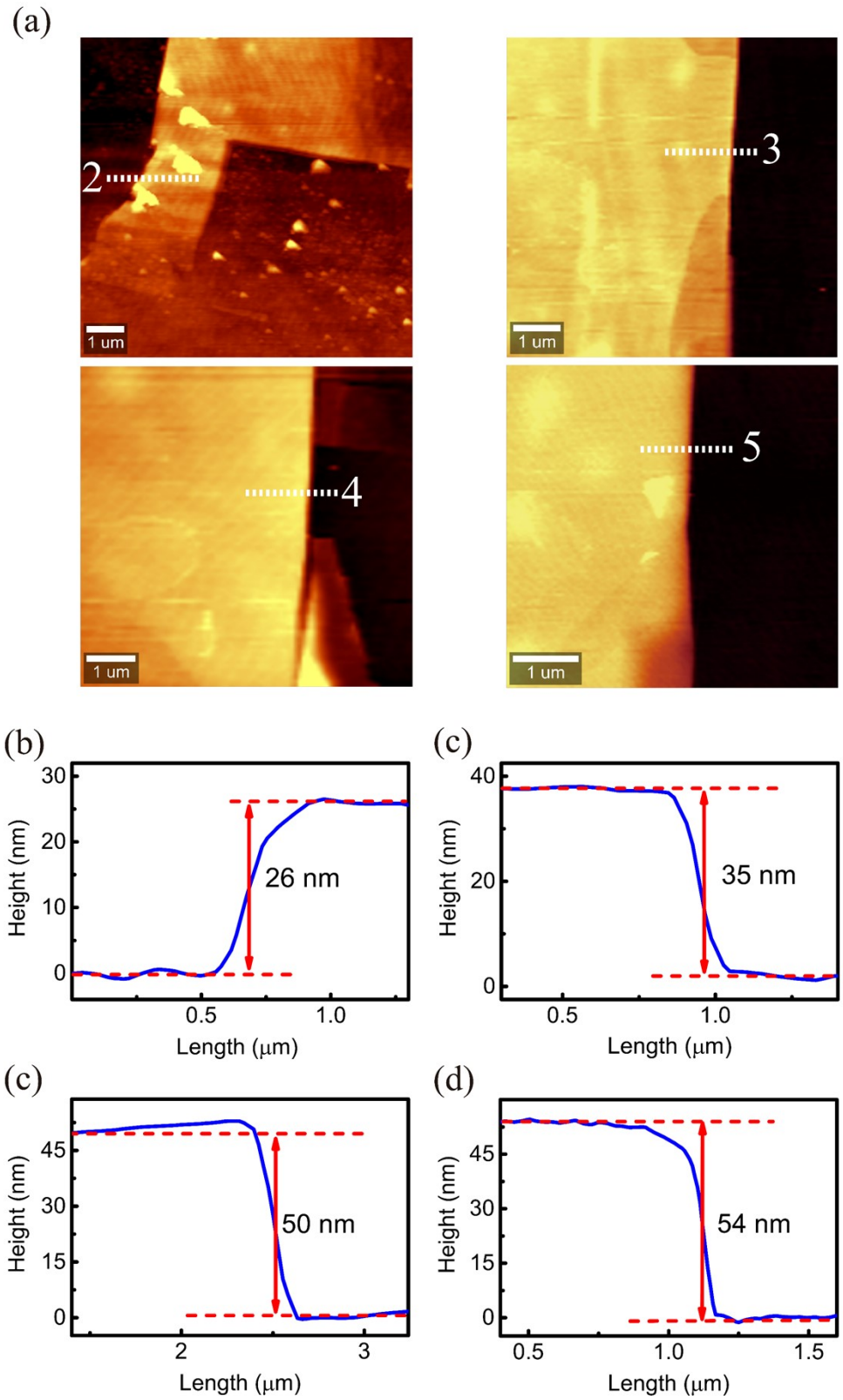

Figure S1. The figure sequence of the remaining four pictures shows the corresponding atomic force microscope (AFM) images to the red lines in image (b). The order of the labels is the same in image (b). The insets in (b)~(e) show the corresponding thickness of $PdSe_2$, respectively.

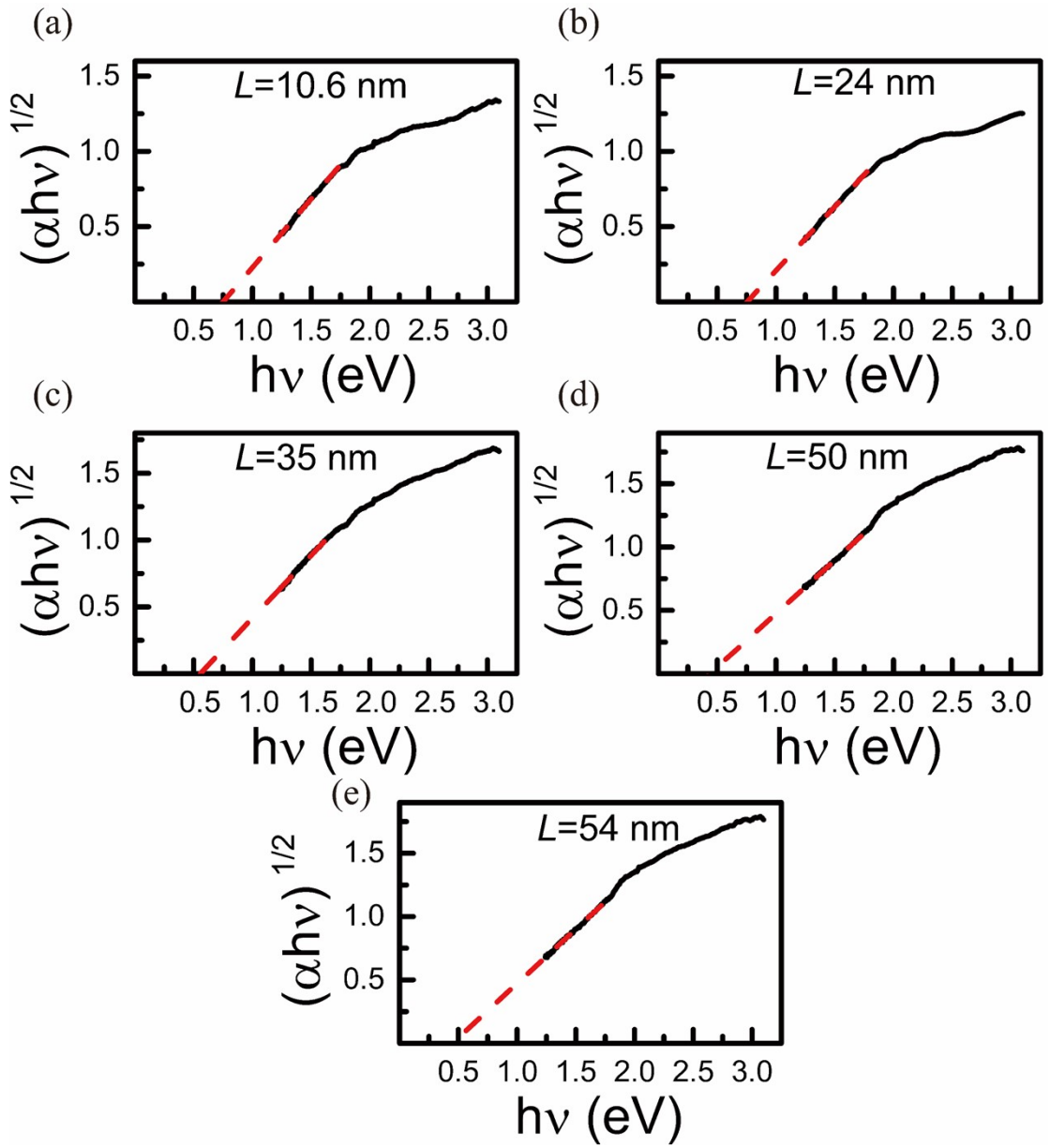

Figure S2. (a)-(e) Tauc plots of absorption spectra for $PdSe_2$ with thickness of 10.6 nm, 24 nm, 35 nm, 50 nm and 54 nm, respectively.

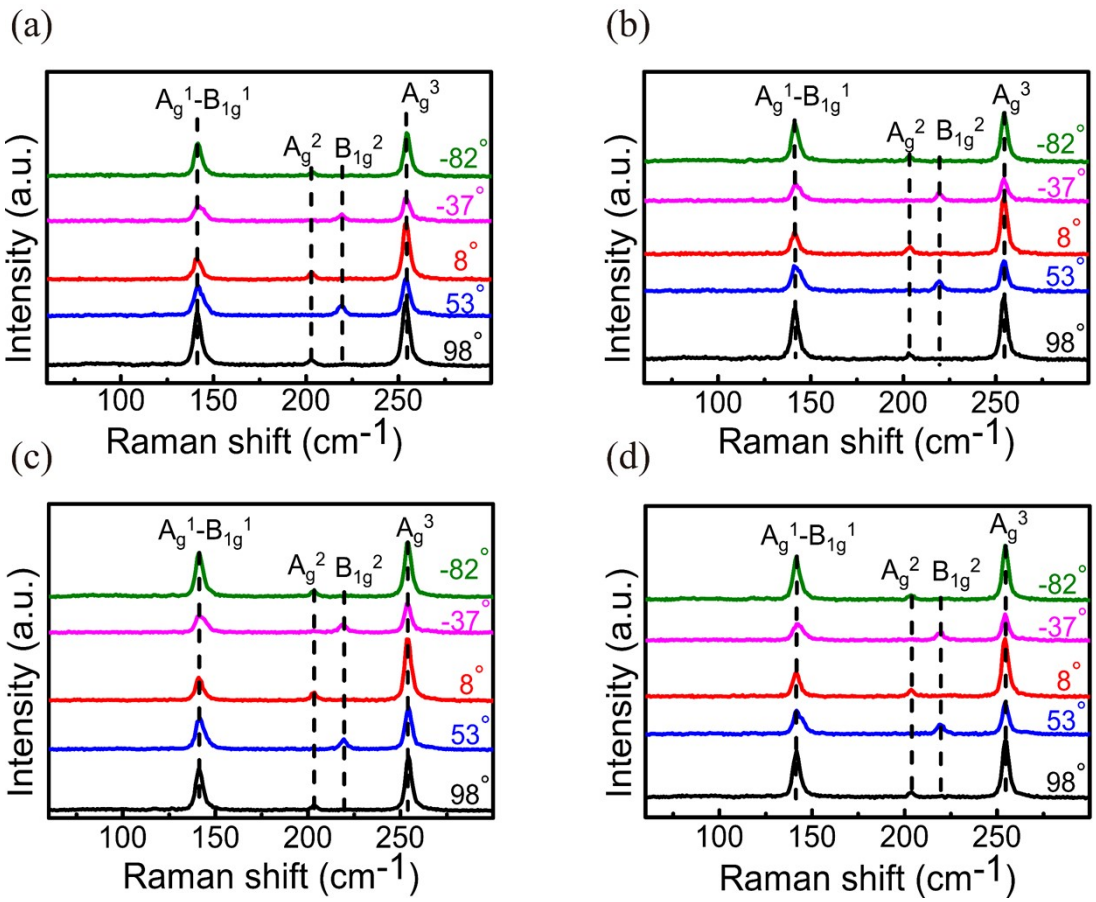

Figure S3. (a)-(d) The polarization dependent Raman spectra of $PdSe_2$ with thickness of (a) 24 nm, (b) 35 nm, (c) 50 nm and (d) 54 nm, respectively.

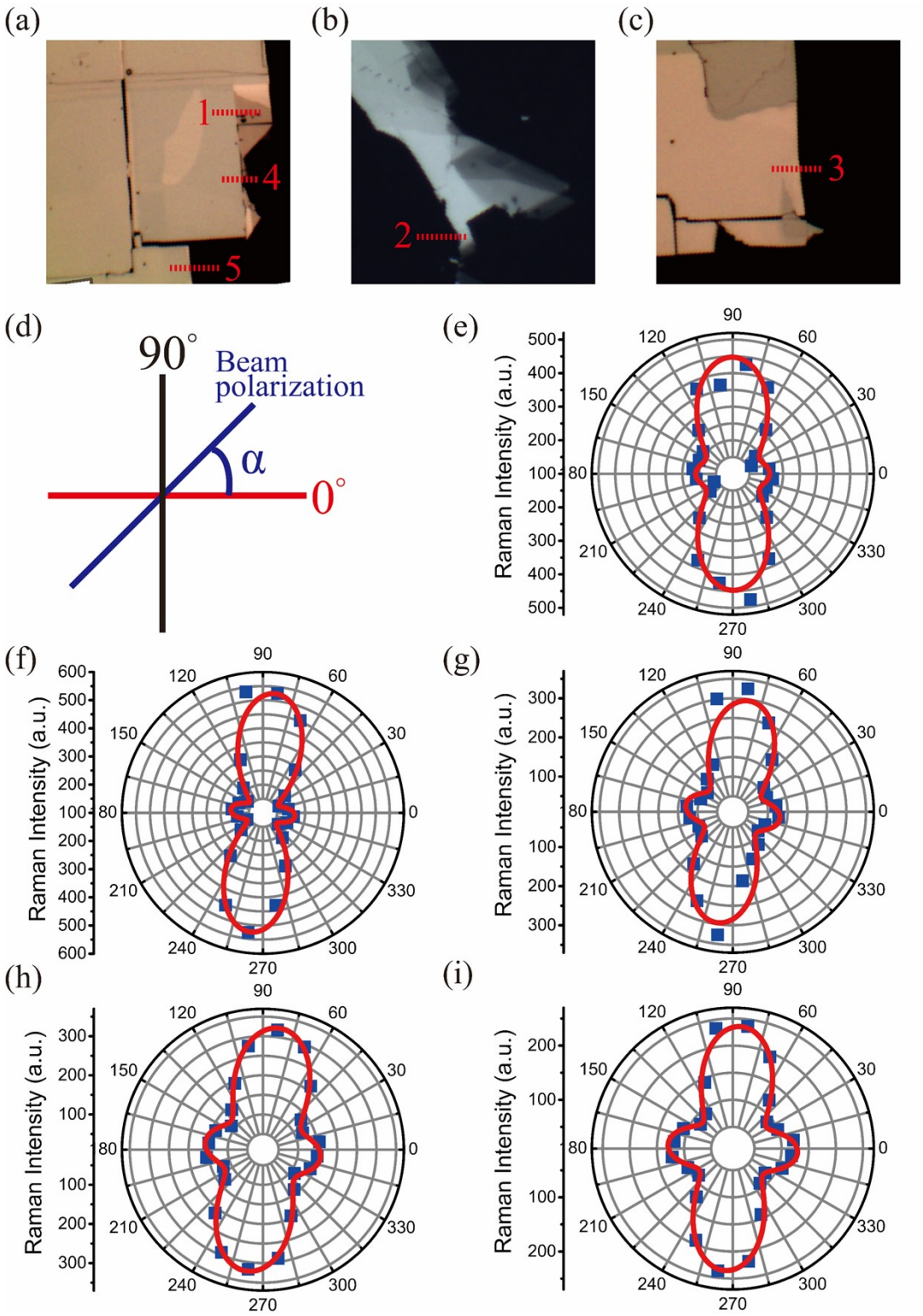

Figure S4. Anisotropy in the Raman intensity of $PdSe_2$ samples. (a)-(c) optical microscopic images of $PdSe_2$ on the fused silica. All scale bars are 10 μm. (d) Red line and black line indicate the direction of 0° and 90° in (a), (b) and (c), respectively. Polarization diagrams of the Raman intensities of A_g^1-B_{1g}^1 (~143 cm^{-1}) mode was extracted through the fitting of the Raman spectra of each polarization angle under the parallel configurations for thickness of 10.6 nm (e), 24 nm (f), 35 nm (g), 50 nm (h)

and 54 nm (i), respectively. The blue markers represent the experimental data. The red curves indicate the fitting results according to the Raman tensors [1].

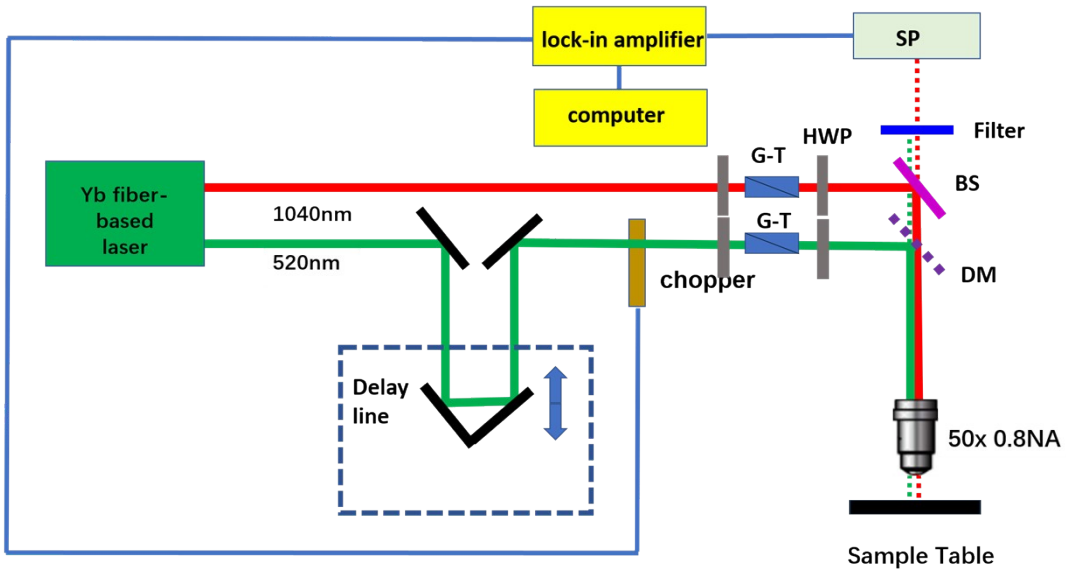

Figure S5. Schematic diagram for the home-built time-resolved transient reflection measurement system. BS, beam-splitter; HWP, half-wave plate; DM, dichroic mirror; SP, Silicon photodiode detector; G-T, Glan-Taylor prism.

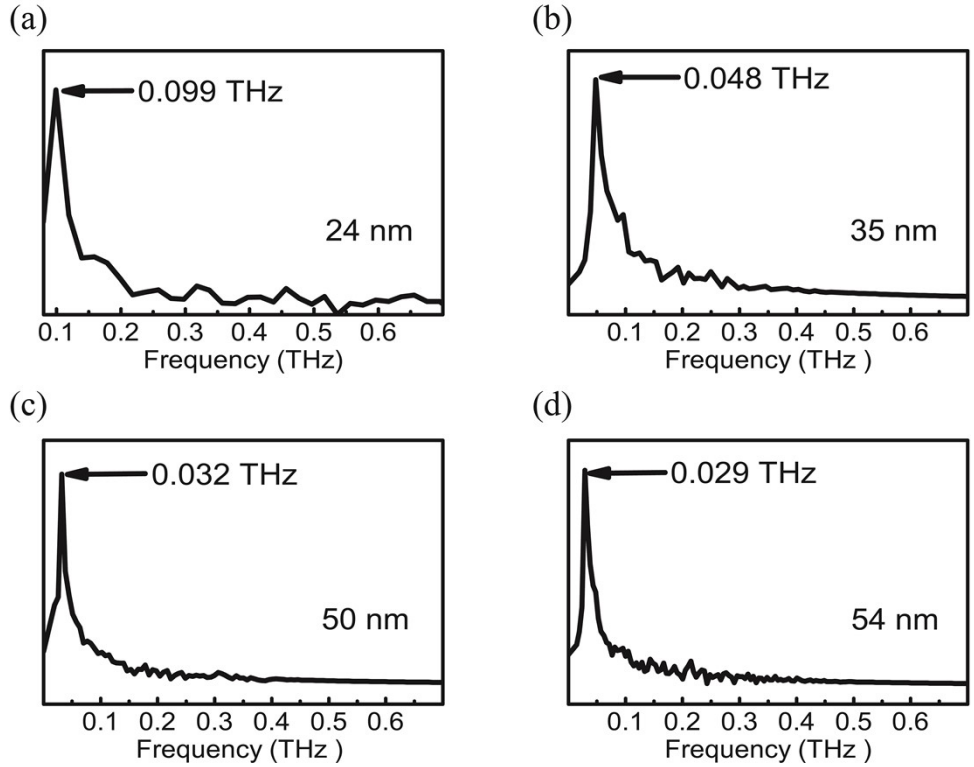

Figure S6. (a)-(d) The fast Fourier transform (FFT) of the oscillations from the TR traces of $PdSe_2$ regions with thickness of 24 nm, 35 nm, 50 nm and 54 nm, respectively.

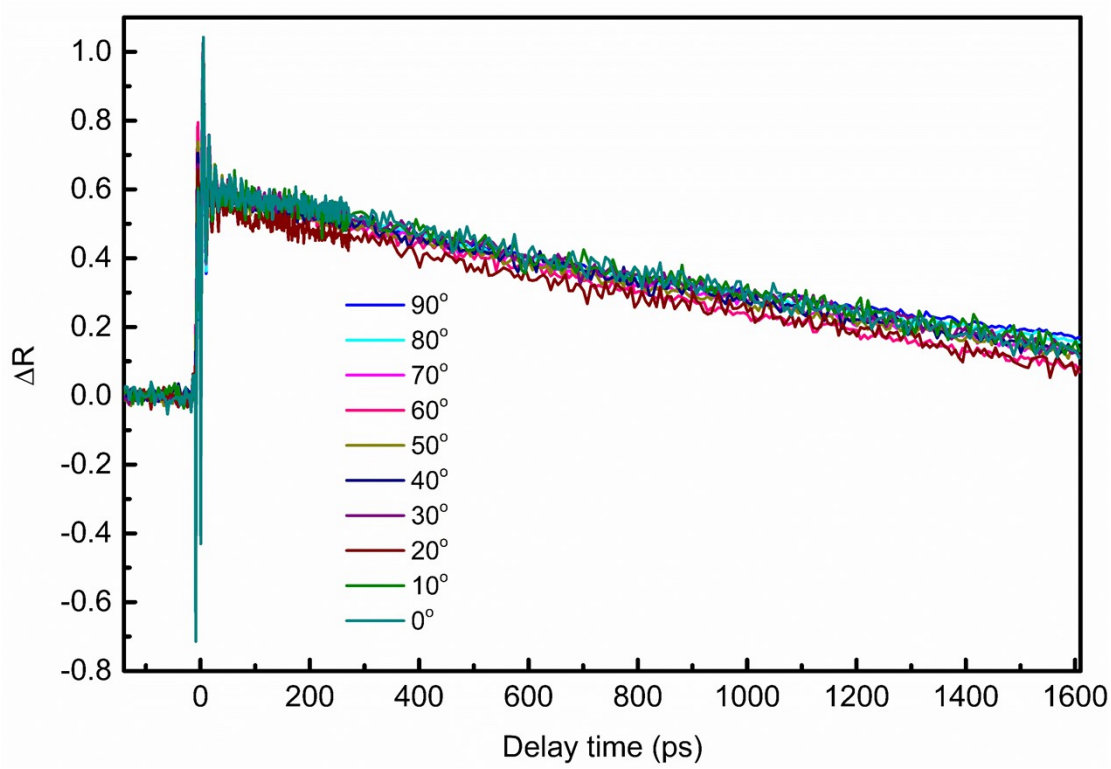

Figure S7. Polarization dependence of normalized time-resolved ΔR curves.

References

1. J. Yu, X. Kuang, Y. Gao, Y. Wang, K. Chen, Z. Ding, J. Liu, C. Cong, J. He and Z. Liu, *Nano Letters*, 2020, **20**, 1172-1182.