Anisotropic Ultrafast Carrier Dynamics Observed in Bulk ReS_2 Using Femtosecond Time- and Energy-Resolved Photoemission Electron Microscopy

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Transition metal dichalcogenides (TMDs) are promising candidates for optoelectronic applications, attributed to their properties such as high in-plane carrier mobility. The well-studied group 6 TMDs such as MoS_2, $MoSe_2$, WS_2 and WSe_2 exhibits isotropic in-plane optical response due their symmetric crystal lattice. In contrast, group 7 TMDs, such as ReS_2, demonstrate anisotropic in-plane optical behavior with respect to the b-axis because of their distorted crystal lattice. This optical polarization anisotropy could serve as an additional tuning parameter in device applications using TMDs.

In this study, we investigate the polarization-anisotropic ultrafast carrier dynamics of AA-stacked bulk ReS_2 with time- and energy-resolved photoemission electron microscopy (TR-PEEM) with sub-50-fs pump and probe pulses, and sub-200-meV energy resolution. In the experiments, 2.41-eV pump pulses first initiate above-gap excitation. The electron population in the conduction band is subsequently probed by 4.81-eV probe pulses at various delay times. Dynamics that are dependent on both pump- and probe-polarization are observed. When keeping the probe polarized parallel to the b-axis, at a photoexcitation carrier density of $\sim 3.3 \times 10^{12}$ cm^{-2}, for example, excitation perpendicular to the b-axis results in a fast decay of $\tau_{fast} = 0.49 \pm 0.05$ ps, whereas excitation parallel to the b-axis yields a slower $\tau_{fast} = 1.3 \pm 0.3$ ps (Fig. 1a), with both cases exhibiting a shared slow decay time constant. Increasing the carrier excitation densities results in a decreasing τ_{fast}, suggesting that the fast decay arises from carrier-carrier scattering. When the pump polarization is fixed parallel to the b-axis, an additional rise component of $\tau_{rise} = 0.10 \pm 0.03$ ps is observed only when probing perpendicular to the b-axis (Fig. 1b).

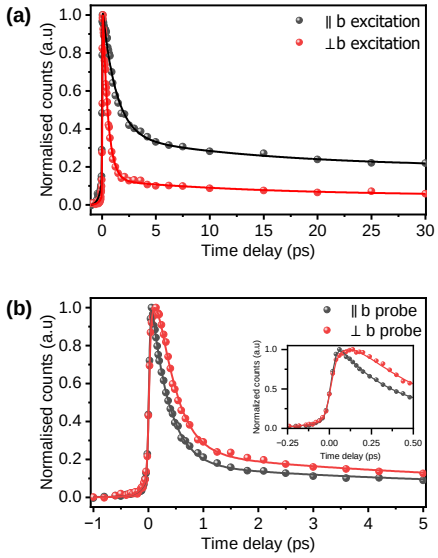

Figure 1. (a) Time trace with probe polarization fixed parallel to the b-axis, varying pump polarization. (b) Time trace for pump polarization fixed parallel to the b-axis, varying probe polarization, with inset showing the early time delays.

These preliminary results demonstrate that bulk ReS_2 possesses anisotropic ultrafast carrier dynamics and could serve as a promising platform for optoelectronic device applications requiring polarization selectivity.