Investigation of MoS_2 Exciton Dynamics Using State-Selective Multidimensional Spectroscopy

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The A and B excitonic states of MoS_2 thin films were investigated using state-selective multidimensional spectroscopy. Interband electron transfer and intraband carrier relaxation is observed on a sub-70 fs timescale, while A and B excitons are observed to decay on a ~680 fs timescale.

Transition metal dichalcogenides (TMDs) have gained considerable attention in recent years as materials with promising solar energy and spintronics/valleytronics applications [1,2]. The electronic structure of TMDs are dominated by strongly absorbing excitonic resonances associated with spin-orbit split valence and conduction band states at the K and K' points in the Brillouin Zone [3].

Here we use state-selective multidimensional spectroscopy to investigate A and B exciton dynamics in MoS_2. Our findings show that excitation of the A exciton induces a B exciton bleach, suggesting ultrafast A→B interband electron transfer. Sub-70 fs formation of vertical features centered near the $\hbar\omega_1$=E_A(E_B) states, arise due to ultrafast intraband relaxation of hot-A(B) excitons. The prominent A and B excitonic features decay along a ~680 fs timescale suggesting an ultrafast trapping mechanism associated with grain-boundary defects [4]. Expansion of this technique into fully coherent pulse orderings will allow us to investigate the coherent coupling of quantum states in MoS_2 and other TMDs, providing key insights into the potential of these materials in solar energy and spintronics/valleytronics applications.

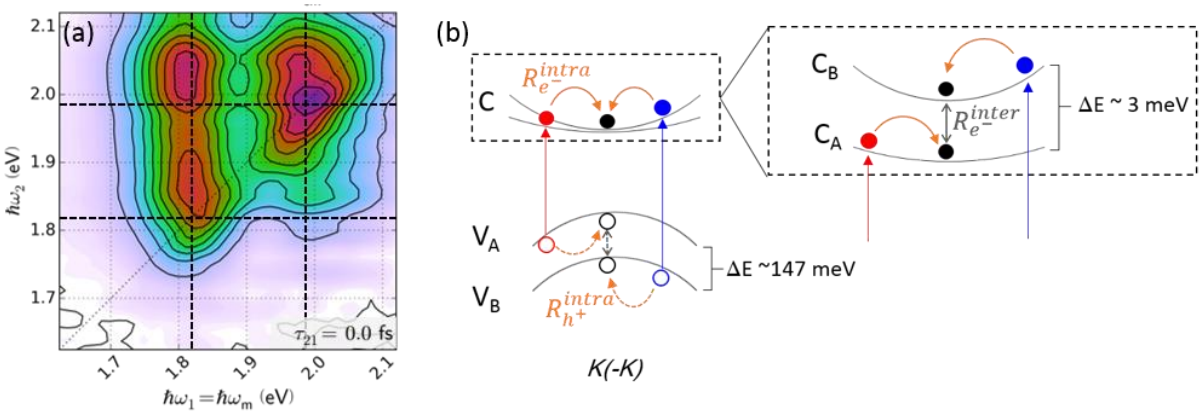

Fig. 1 (a) 2D-frequency spectrum of MoS_2 thin film at τ_{21} = 0 fs, where $\hbar\omega_2$ and $\hbar\omega_1$ correspond to the excitation and probe energies, respectively. A and B exciton features appear as on-diagonal and off-diagonal peaks and are indicated by intersecting dashed lines. (b) Expected relaxation pathways including intraband electron and hole relaxation, R^{intra} (left), as well as interband electron relaxation, R^{inter} (right).

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[3] D. Xiao *et al.* Phys. Rev. Lett. **108**, 196802 (2012).
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