Ultrafast spin valley dynamics in single layer WS₂

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The single layer two dimensional transition metal dichalcogenides (TMDs), such as MoS₂ and WS₂, are widely considered as emerging valleytronic materials primarily due to the coupled spin-valley at K/K' points of the Brillouin zone [1]. Valley polarization carried by the circularly polarized light excited quasi-particles (excitons, trions or biexcitons) could be used as a discrete degree of freedom, therefore, valley depolarization dynamics of these photoexcitations in TMDs should be well understood.

Studies on electron spin/valley dynamics in single layer MoS₂ show a characteristic intervalley scattering time of ~200 fs for A exciton [2]. Different from MoS₂, single layer WS₂ has larger conduction band splitting with no band crossing [3], better spectrally resolved trion signature [4]. Such peculiar features have attracted intense studies, both experimentally and theoretically, on spin/valley relaxation mechanism in this material [5,6].

In this work, we study the spin/valley relaxation process in a mechanically exfoliated WS₂ sample at liquid nitrogen temperature using the Time Resolved Circular Dichroism (TRCD) and Time Resolved Faraday Rotation (TRFR) techniques with typical ~100 fs temporal resolution pump and probe laser pulses. By carefully tuning the laser photon energy resonant to WS₂ A, B exciton and trion peaks, we are able to access the various electron spin process in K and K' valley, specifically the inter- and intra-valley spin relaxation dynamics involving both neutral and charged excitons.

References

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Figure 1: (a) Time resolved circular dichroism measurements on single-layer WS₂. Both pump and probe are resonant to A exciton peak. Black and red curve: pump and probe have the same and opposite helicity, respectively. (b) Time resolved Faraday rotation of monolayer WS₂. Blue and cyan curve: pump with left and right circularly polarized light. Gray dashed curve: laser pulse shape.