Disentangling spin and valley dynamics in monolayer MoS_2 by non-equilibrium optical techniques

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The ability to control the valley degrees of freedom is the foundation of the emerging field of valleytronics [1]. Atomically thin Transition Metal Dichalcogenides (TMDs) are a promising platform for the implementation of new devices exploiting the valley degrees of freedom [2]. The lack of inversion symmetry, combined with a large spin-orbit interaction, leads to a conduction (valence) band with different spin-polarized minima (maxima) having equal energies [3]. These peculiar properties offer the possibility to develop a new valley-based electronics where information is carried not only by the spin, but also by the crystal momentum at multiple extreme points of the band structure. Any implementation of these concepts, however, needs to consider the robustness of valley and spin degrees of freedom, which are deeply intertwined. To this aim, here we measure separately the spin and valley relaxation dynamics of both electrons and holes in the prototypical TMD MoS_2. We disentangle the different processes by the combination of ultrafast optical spectroscopy techniques, i.e. Time Resolved Circular Dichroism (TRCD) and Time Resolved Faraday Rotation (TRFR). TRCD experiments are performed by exciting the sample with an ultrashort circularly polarized pulse, resonant with the optical gap (650 nm), and measuring the difference between the transient absorption response probed by co- and counter-circularly polarized pulses. The transient absorption is measured on a broad energy range including the A (λ = 650 nm) and B (λ = 605 nm) excitonic transitions at K and K'. These measurements reveal an extremely fast intravalley relaxation of the spin of the photoexcited electrons at the bottom of the conduction band. Furthermore, our data demonstrate that the two non-equivalent valleys K and K' are strongly coupled and the valley polarization is strongly quenched after few ps [4]. In TRFR experiments the pump pulse, in analogy with TRCD, creates a spin polarized population of electrons/holes in the conduction/valence band, while the rotation angle of the linearly polarized probe pulse is measured by a balanced photodiode bridge technique. We use a two-color TRFR configuration, in which the energy of the probe pulse is tuned well below the absorption gap. In these conditions, the TRFR signal is only sensitive to the helicity-dependent light scattering of the photoexcited electron and hole populations. Since the probe pulse couples with the carrier orbital degree of freedom, the Faraday rotation signal is related to an unbalanced distribution of the photoexcited carrier orbital degrees of freedom. The orbital momentum in MoS_2 single layer is locked with the valley index. Thus, the two color TRFR measurements probe exclusively the intervalley dynamics of electrons and holes. The combination of TRFR and TRCD allows us to disentangle intervalley and intravalley dynamics. Both TRCD and TRFR experiments are quantitatively explained by a set of rate equations which take into account intervalley and intravalley relaxation channels. These results are very useful for the engineering of spintronic and valleytronic devices based on single layer TMDs.

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