## **Opto-Valleytronic Spin Injection in Monolayer MoS<sub>2</sub>/Few-Layer Graphene Hybrid Spin Valves**

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Two dimensional (2D) materials provide a unique platform for spintronics and valleytronics due to the ability to combine vastly different functionalities into one vertically-stacked heterostructure, where the strengths of each of the constituent materials can compensate for the weaknesses of the others. Graphene has been demonstrated to be an exceptional material for spin transport at room temperature, however it lacks a coupling of the spin and optical degrees of freedom [1]. In contrast, spin/valley polarization can be efficiently generated in monolayer transition metal dichalcogenides (TMD) such as MoS<sub>2</sub> via absorption of circularly-polarized photons, but lateral spin or valley transport has not been realized at room temperature [2]. In this talk, we fabricate monolayer MoS<sub>2</sub>/multilayer graphene hybrid spin valves and demonstrate, for the first time, the opto-valleytronic spin injection across TMD/graphene interface [3]. We observe that the magnitude and direction of spin polarization is controlled by both helicity and photon energy. In addition, Hanle spin precession measurements confirm optical spin injection, spin transport, and electrical detection up to room temperature. Finally, analysis by a one-dimensional drift-diffusion model quantifies the optically injected spin current and the spin transport parameters. Our results demonstrate a 2D spintronic/valleytronic system that achieves optical spin injection and lateral spin transport at room temperature in a single device, which paves the way for multifunctional 2D spintronic devices for memory and logic applications.



**Figure 1. (a)** Illustration of optical spin injection, lateral spin transport, and electrical spin detection in a monolayer  $MoS_2/few$ -layer graphene hybrid spin valve structure. (b) Electrical spin signal  $V_{NL}$  as a function of  $B_y$  exhibits clear antisymmetric Hanle spin precession signals which flip polarity with the Co magnetization direction (M+ vs M–).

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## **Supplementary Information:**

**Figure 2.** Photon energy dependence of opto-valleytronic spin injection. (a) Schematic band structure of monolayer  $MoS_2$  at the K and K' valleys. The valence band of monolayer  $MoS_2$  has a large spin splitting with opposite spin orientation for the A and B optical excitations within the same valley. (b) Representative antisymmetric Hanle curves at four photon energies (1.87, 1.93, 1.96, and 2.06 eV). (c) Spin signal  $\Delta V_{NL}$  as a function of photon energy. Starting from low photon energy,  $\Delta V_{NL}$  reaches a maximum positive signal near the A resonance (1.90–1.95 eV) and then decreases with increasing photon energy until  $\Delta V_{NL}$  flips sign around 2 eV.  $\Delta V_{NL}$  reaches a minimum near the B resonance at ~2.06 eV. This photon energy dependence clearly reflects the nondegenerate spin-split structure of the valence band, which results from strong spin–orbit coupling and the broken inversion symmetry of the monolayer  $MoS_2$  lattice. Inset shows zoom-in detailed features around the A exciton resonance, consistent with the double peak structure of the A<sup>-</sup> trion and A exciton.



**Figure 3.** Room-temperature characteristics of opto-valleytronic spin injection. (a) Photon energy dependence of the spin signal  $\Delta V_{NL}$ .  $\Delta V_{NL}$  at room temperature exhibits a similar dependence on photon energy as at low temperature, with the positive peak at the A resonance red-shifted to around 1.86 eV, and the negative peak at the B resonance red-shifted to around 1.99 eV. (b) (c) Two Hanle scans with photon energies near the A and B peaks are measured to confirm room temperature spin orientation switching from the A resonance to the B resonance. The room temperature signal is about 5 times smaller than at 10 K.