Spin Polarized Defect States Drive Valley Polarization Dynamics in V-Doped MoS_2

Nagendra S Kamath
Ashish Soni, Dipak Maity, T N Narayanan, Suman Kalyan Pal
Institute of Technology Mandi, Mandi, Himachal Pradesh 175005, India
Tata Institute of Fundamental Research Hyderabad, Hyderabad, Telangana 500046, India
nknagu24@gmail.com, suman@iitmandi.ac.in

Abstract Two-dimensional transition metal dichalcogenides (TMDs) exhibit valley-selective optical transitions which are promising for valleytronic applications [1,2]. Controlling and manipulating the valley degree of freedom in TMDs is essential for the development of valley-based electronics and optoelectronics [3]. Beyond the well-studied TMDs such as MoX_2 and WX_2 (where X = S, Se, Te), substitutional doping has gained significant interest due to the unique features that arise from structural modifications [4]. Here, we explore the valley physics in vanadium doped MoS_2 (V-MoS_2) using helicity-resolved transient absorption (HRTA) spectroscopy (Figure 1(a)). Our results reveal that defect states induced by vanadium show spin-polarized behaviour making this a three-valley like system (Figure 1(b)). Compared to pristine MoS_2, which exhibits a very fast valley depolarization time of ~0.5 ps, V-MoS_2 demonstrates an enhanced valley retention with an extended valley lifetime of ~12 ps. Additionally, intervalley scattering dynamics exhibit an energy transfer effect. When the B-exciton is pumped, excitons redistribute into the A-exciton valley while preserving spin polarization, yielding an enhanced degree of valley polarization of ~20% at room temperature. This behaviour emphasizes the role of vanadium doping in modifying intervalley coupling and exciton dynamics. The findings provide an effective strategy for governing valley lifetimes and intervalley dynamics, advancing spin-valleytronic applications.

References

[1] K. F. Mak, K. L. McGill, J. Park, and P. L. McEuen, Science, 6191 (2014)1489
[2] Nagendra S Kamath, Ashish Soni and Suman Kalyan Pal, J. Phys. Chem. Lett., 1 (2025) 25.
[3] John R. Schaibley, Hongyi Yu, Genevieve Clark, Pasqual Rivera, Jason S. Ross, Kyle L. Seyler, Wang Yao and Xiaodong Xu, Nat. Rev. Mater., 1 (2016) 16055
[4] Sreekant Anil, Dipak Maity, Snehith Adabala, Arpan De, Nagendra S Kamath, Raheel Hammad, Janmey Jay Panda, Ravi K Biroju, Viliam Vretenar, Rajalakshmi G, Suman Kalyan Pal, Soumya Ghosh and Tharangattu N Narayanan, NanoLett., 35 (2025) 13401.

Figures

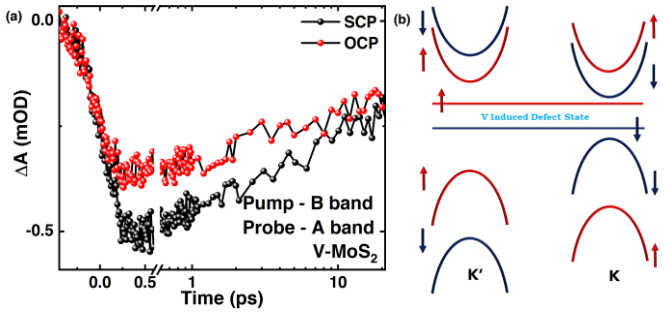

Figure 1: (a) HRTA kinetics of V-MoS2 with excitation wavelength corresponding to B band, probed at A band with same (SCP) and opposite circular polarizations of the pump and probe lights. (b) Schematic representation of band alignment in V-MoS_2.