Identifying and Probing Atomic Defects in 2D Semiconductors by Scanning Probe Microscopy

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Two-dimensional (2D) semiconductors provide an exciting platform to engineer atomic quantum systems in a robust, yet tunable solid-state system. In this talk, I will present our efforts to unravel the interesting physics behind single vacancies and dopant atoms in transition metal dichalcogenide (TMD) mono- and multilayers by means of high-resolution scanning probe microscopy [1-6].

Our recent studies on transition metal doped TMDs such as (n-type) Re-doped MoS_2 and (p-type) V-doped WSe_2 reveal the significance of the charge state in the spectroscopic signature of these defects [7,8]. By substrate chemical gating, we can stabilize three charge states of Re_{Mo} , where two of the charge states exhibit symmetry broken electronic orbitals and a distorted atomic configuration that we assign to a pseudo Jahn-Teller effect [7].

In the second part, I will share our recent advancements in probing ultrafast dynamics of single atomic defects in TMDs [9-10]. In monolayer WSe₂ (1ML), we observe rapid electron transfer from the lowest unoccupied orbital of the top Se vacancy to the graphene substrate within 1.2 ps. In multilayers, we find a sub-exponential increase of the charge lifetime of the top vacancy from 54 ps in 2ML, to 1.1 ns in 3ML, and 3 ns for 4 ML WSe₂. I will provide an outlook on our ongoing developments of ultrafast lightwave-driven scanning tunneling microscopy using single-cycle THz pulses to measure the sub-picosecond time dynamics at atomic spatial resolution [9].

References

- [1] B. Schuler et al., Phys. Rev. Lett. 123, 076801 (2019)
- [2] B. Schuler et al., ACS Nano 12, 10520 (2019)
- [3] S. Barja et al., Nat. Commun. 10, 3382 (2019)
- [4] B. Schuler et al., Sci. Adv. 6, eabb5988 (2020)
- [5] K. Cochrane et al., Nat. Commun. 12, 7287 (2021)
- [6] E. Mitterreiter et al., Nat. Commun. 12, 3822 (2021)
- [7] F. Xiang*, L. Huberich* et al., Nat. Commun. (2024)
- [8] S. Stolz et al. ACS Nano, 17, 23422 (2023)
- [9] J. Allerbeck et al. ACS Photonics, 10, 3888 (2023)
- [10] L. Bobzien et al. (under review)