Abstract for NANOMAN2024(500 words) An atomic-scale understanding of MoS_2 reactivity toward halogen atoms under ultrashort laser pulses: Implications for atomic layer etching

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Controlling the thickness of two-dimensional (2D) nanosheets, such as molybdenum disulfide (MoS_2), is critically important for the development of 2D-material-based devices. Halogenation-based atomic layer etching (ALE) presents a promising method for the layer-by-layer removal of both bulk materials and 2D nanosheets. In this study, density functional theory (DFT) and real-time time-dependent DFT calculations are employed to investigate the atomic-scale dynamics of MoS_2 reactivity towards halogen atoms under ultrashort laser pulses. The findings indicate that fluorine (F) atoms exhibit higher adsorption energy on MoS_2 compared to chlorine (Cl) atoms, resulting in greater electron transfer from the S-Mo bond to the halogen-S bond. The activation energy for F atom migration is higher than that for Cl atom migration, attributed to the stronger bonding energy between F and S atoms. Additionally, under ultrashort laser pulses, the adsorbed halogens can enhance the desorption of the adjacent S atom compared to the bare MoS_2 surface, with Cl demonstrating a more significant enhancing effect than F. During an oscillating laser cycle, the electrons in the halogen-MoS_2 system are excited and oscillate with the applied electric field. After a cycle, the electron density within the adsorbed S-Mo bond decreases, facilitating the desorption of the S atom. This study provides critical atomic-level insights into the reactivity of MoS_2 towards halogen atoms under laser pulses, thereby advancing the development of laser-based layer control techniques for 2D materials.