Supporting Information

From N-type Doping to Phase Transition in Large-Area MoS_2 via Controlled Sulfur Vacancy Formation

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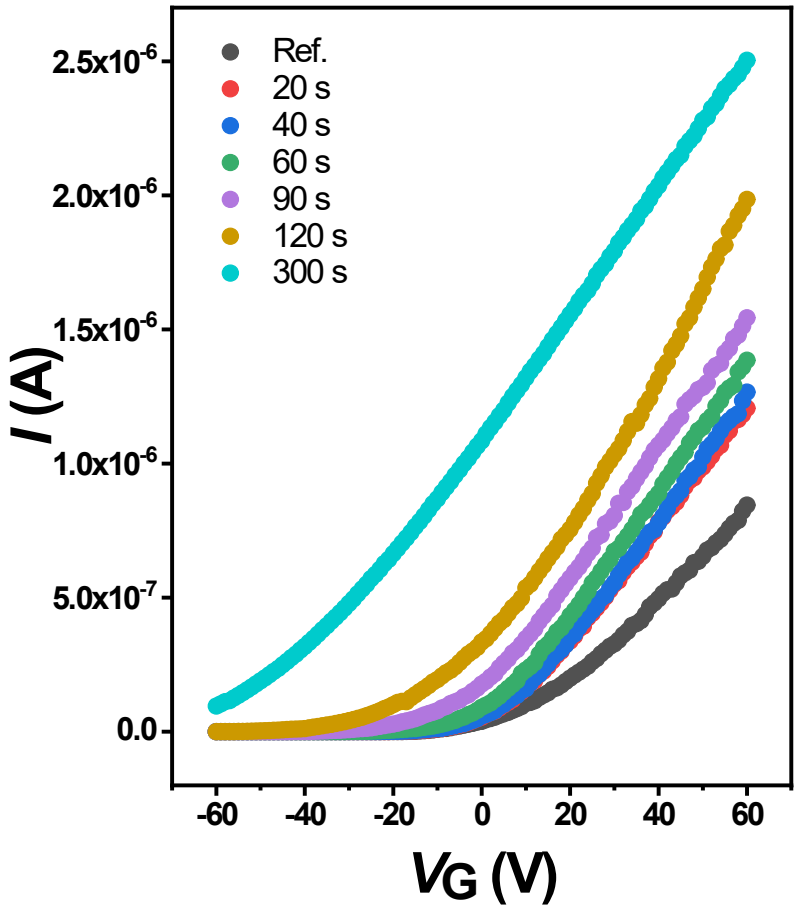

Figure S1. Transfer characteristics (I–V curves) of MoS_2 FETs under varying Ar^+ ion treatment duration plotted on a linear scale.

The drain current (I_D) increases progressively with longer Ar^+ ion exposure times, indicating enhanced channel conductivity due to increased electron concentration. This trend supports the conclusion that Ar^+ ion treatment effectively introduces n-type doping in monolayer MoS_2, resulting in a clear enhancement of on-current across the tested durations (20 s to 300 s).

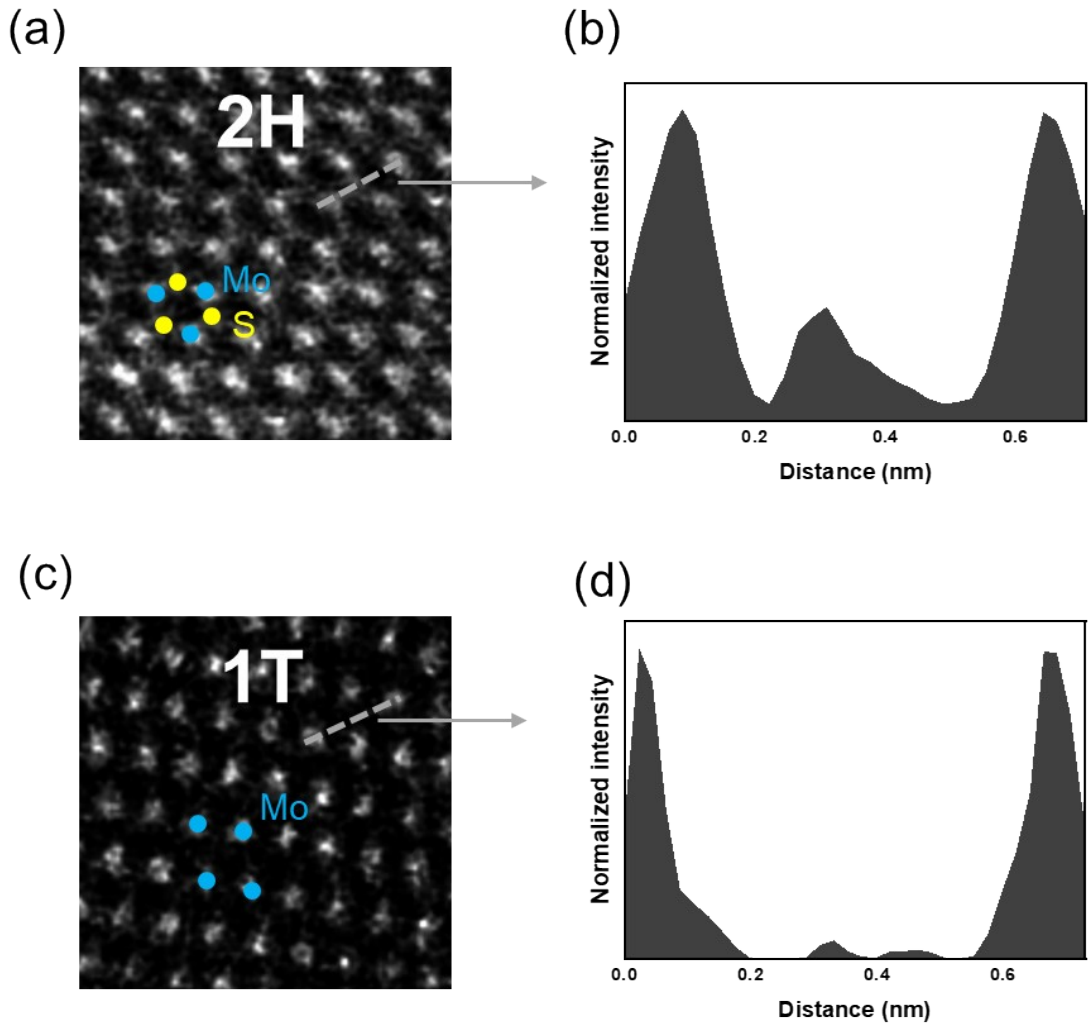

Figure S2. High-resolution STEM images and corresponding intensity profiles of monolayer MoS_2 in 2H and 1T phases. (a) Atomic-resolution HAADF-STEM image of monolayer MoS_2 in the 2H phase, showing a well-ordered hexagonal lattice with Mo (blue) and S (yellow) atoms arranged in a trigonal prismatic coordination. (b) Normalized intensity profile extracted along the atomic column indicated in (a), clearly resolving Mo and S atoms as distinct peaks with alternating contrast. (c) HAADF-STEM image of Ar^+ ion treated MoS_2, showing a distorted lattice consistent with a structural transition to the 1T phase, where Mo atoms are expected to adopt octahedral coordination. (d) Corresponding intensity profile taken from the region indicated in (c), exhibiting reduced contrast and broadened features, indicative of atomic rearrangement and possible sulfur vacancy formation.

To investigate the atomic-scale structural transition of MoS_2 induced by Ar^+ ion treatment, high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) was employed. Figure S2a shows the pristine monolayer MoS_2 in the semiconducting 2H phase, with a well-ordered hexagonal lattice composed of Mo atoms in trigonal prismatic coordination with surrounding S atoms. The normalized intensity profile in Figure S2b clearly distinguishes the alternating contrast between heavier Mo and lighter S atoms. After Ar^+ ion exposure, a structural transformation is observed, as shown in Figure S2c. The lattice becomes distorted, and the original symmetry is altered, consistent with a phase transition to the metallic 1T phase, in which Mo atoms are octahedrally coordinated. The corresponding intensity profile (Figure S2d) exhibits decreased peak contrast and broadened features, suggesting atomic rearrangement and the formation of sulfur vacancies.[1] These results support the conclusion that ion-induced lattice distortion is a key mechanism driving the 2H-to-1T phase transition in a monolayer MoS_2.

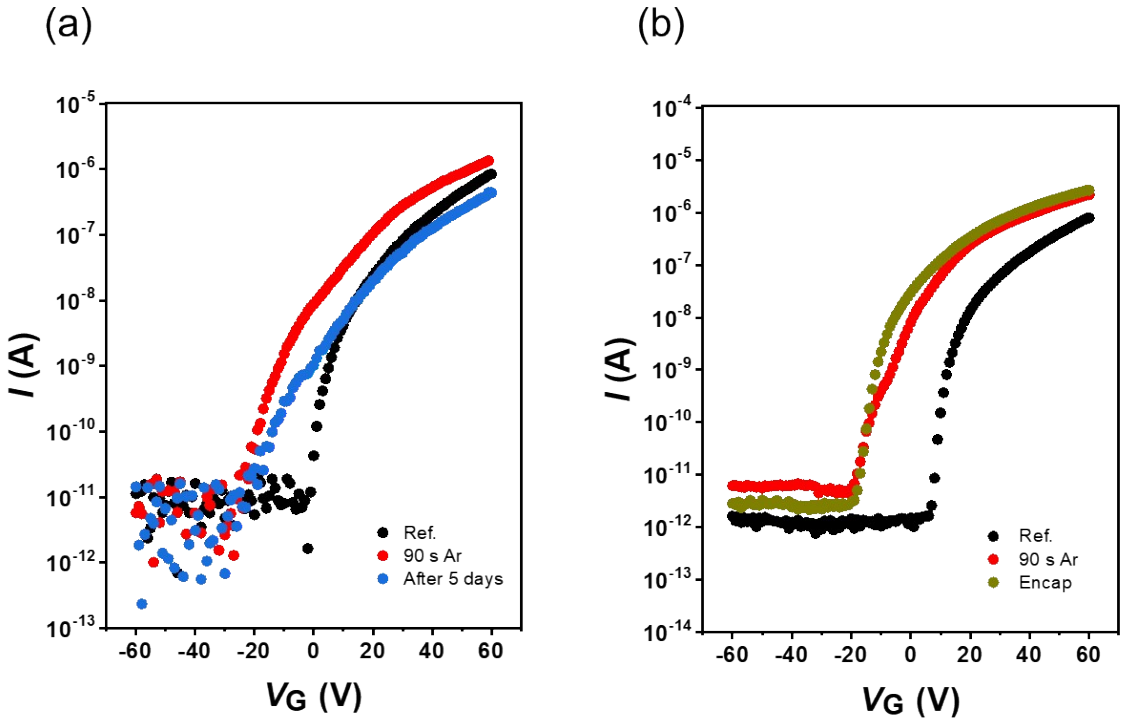

Figure S3. Effects of ambient air exposure on Ar^+ ion treated MoS_2 FETs and the role of Al_2O_3 encapsulation in suppressing degradation. (a) Transfer characteristics of devices measured immediately after Ar^+ ion treatment and after 5 days of ambient exposure, showing clear degradation including reduced on-current and threshold voltage shift. (b) Transfer characteristics of devices encapsulated with ALD-grown Al_2O_3, exhibiting stable performance even after 5 days of ambient exposure, confirming the protective effect of the encapsulation layer.

To evaluate the degradation behavior of MoS_2 FETs after Ar^+ ion treatment, we compared transfer characteristics measured immediately after the ion exposure and after 5 days of ambient air exposure. As shown in Figure S3a, unencapsulated devices exhibited significant degradation, including a noticeable decrease in on-current and a positive shift in threshold voltage. These changes are attributed to oxidation and moisture-induced degradation on the exposed MoS_2 surface. In contrast, devices encapsulated with ALD-grown Al_2O_3 retained stable transfer behavior under the same ambient conditions, as shown in Figure S3b. The

negligible variation in electrical performance confirms that the Al_2O_3 encapsulation layer effectively protects the MoS_2 channel from environmental degradation, thereby preserving the doping state and ensuring long-term device stability after the doping.

REFERENCES

1 S. S. Chou, N. Sai, P. Lu, E. N. Coker, S. Liu, K. Artyushkova, T. S. Luk, B. Kaehr and C. J. Brinker, *Nat. Commun.*, 2015, **6**, 8311.