

Supporting information:

Highly efficient and stable MoS₂ FETs with reversible *n*-doping using a dehydrated poly(vinyl-alcohol) coating

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Electrical characterization of PVA

To ensure that the PVA coating makes no substantial contribution to the source drain current of the PVA-doped MoS₂ FET, we measured the current characteristics of a PVA film in a similar transistor configuration. The transfer characteristics of this PVA device are shown in Figure S1. The total current, I_D , which is composed of the current flowing through PVA and the back gate leakage current, was obtained from a device with L_{CH} 2.5 μm and W_{CH} of 28.1 μm with 47 nm of PVA bridging the source and drain contacts. The current was orders of magnitude smaller than the corresponding value of the PVA-doped MoS₂ FET ($|I_D/W| < 500$ fA), as expected for an insulating material.^{S1}

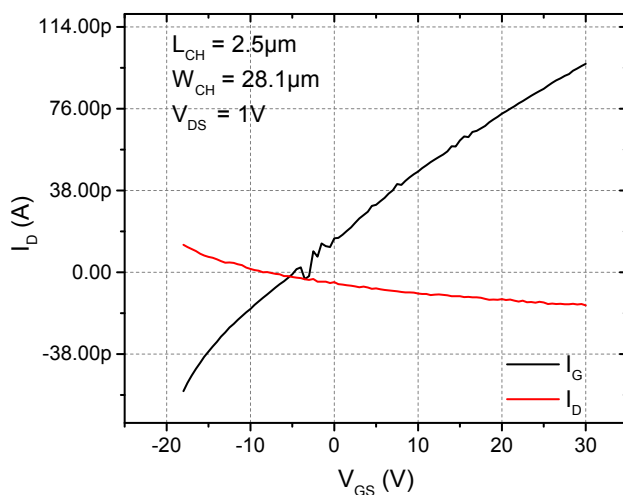


Figure S1: Plot of I_D against V_{GS} for a FET containing 1% PVA as a channel.

FTIR spectroscopic and XRD characterization of PVA films

Fourier transform infrared (FTIR) spectroscopy was used to extract information about the composition of the deposited PVA films. First a thick film ($\sim 100 \mu\text{m}$) deposited by drop casting on a 55-nm-thick SiO₂ film on top of a silicon wafer was assessed for reference. The FTIR spectrum of this PVA film (Figure S4) clearly contained absorption peaks from O-H, C-H and C=O chemical groups.^{S2}

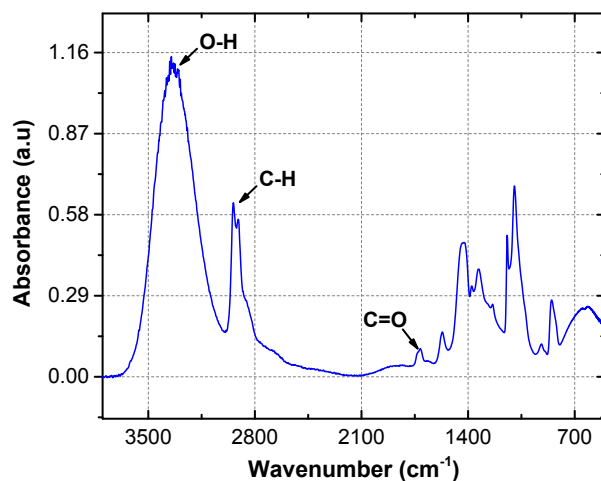


Figure S2: FTIR spectrum of a thick ($\sim 100 \mu\text{m}$) PVA film.

Next, attenuated total reflectance FTIR (ATR-FTIR) spectroscopy was used to evaluate a 20-nm-thick PVA film spin coated on a 55-nm-thick SiO_2/Si substrate under the same conditions that were used in the experiments in the main paper. This ATR-FTIR spectrum is presented in Figure S5.

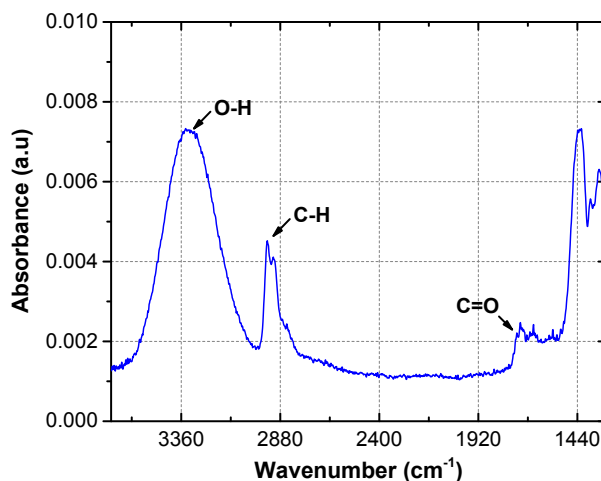


Figure S3: ATR-FTIR spectrum of a 50-nm-thick PVA film.

The same chemical groups can be identified in the ATR-FTIR spectrum of the 20-nm-thick film as in the FTIR spectrum of the thicker film. This is further proof of the formation of a thin PVA film by the described process. However, the peaks at lower wavenumbers were poorly resolved because of the small thickness of the material being sampled.

Next, grazing incidence X-ray diffraction (GI-XRD) patterns of thick PVA films ($\sim 100 \mu\text{m}$) were collected to obtain information about the crystallinity of the material. Three different PVA films were assessed: A) as-deposited, B) as-deposited + annealed for 20 min at 90°C in N_2 atmosphere, and C) as-deposited + annealed for 20 min at 90°C (N_2) + aged for 1 day at room temperature (Figure S4).

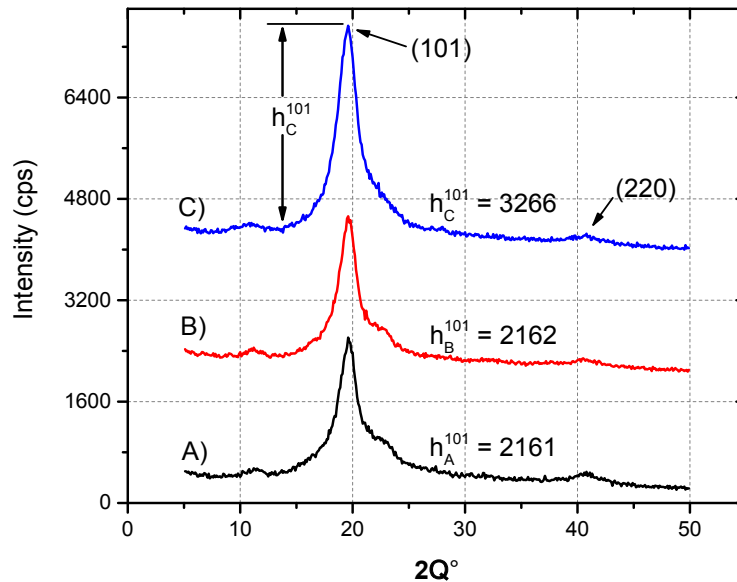


Figure S4: GI-XRD patterns of a thick ($\sim 100 \mu\text{m}$) PVA film under different post-treatment conditions. The figures are shifted for better visualization.

The peak at around $2\theta = 20^\circ$ corresponds to the 101 plane of semi-crystalline PVA.⁵³ Thus, the height of this peak can be used as an indication of the degree of crystallization of the PVA film.⁵⁴ The height of this peak did not change drastically when the sample was annealed at 90°C for 20 min. However, when the annealed sample was aged for 1 day, the relative height of the peak increased almost 51%, indicating that the crystallization of the PVA film increased. This observation supports the argument made in the main text that the device performance is enhanced by the crystallization of PVA.

Influence of surface passivation on doping of MoS_2

It has been shown that surface passivation of thin-film MoS_2 can partially alter its electrical performance⁵⁵. Thus, to ensure that the charge modulation observed in our PVA-treated MoS_2 FETs is mainly originated from doping effect rather than simply from surface passivation, two tests were performed:

First, as described in the methodology section, all the devices are measured in N_2 atmosphere. This minimizes the time that the device is exposed to air. Thus, quenching of the intrinsic n-doping of the MoS_2 film is not likely. Moreover, to demonstrate that the ambient air does not affect the device substantially, it was measured in vacuum condition (10^{-7} mbar) after degassing for 2 days. As shown in figure S5-a, the threshold voltage (V_{TH}) shows a negligible variation when switching from N_2 to vacuum condition, while after coating with PVA and measuring the device in N_2 we observe a large negative shift in V_{TH} , which indicates n-doping.

Next, to ensure that the doping is not originating from a passivation effect of PVA but rather from its doping effect, we applied poly-trifluoroethylene (TrFE), a highly non-reactive thermoplastic fluoropolymer, on our MoS_2 devices and compare them with PVA doped ones. As shown in figure S5-b TrFE passivation has minimum impact on the device V_{TH} (and thus its doping level) while the PVA doped device shows a strong V_{TH} shift. We believe these results rule out substantial contribution arising from surface passivation.

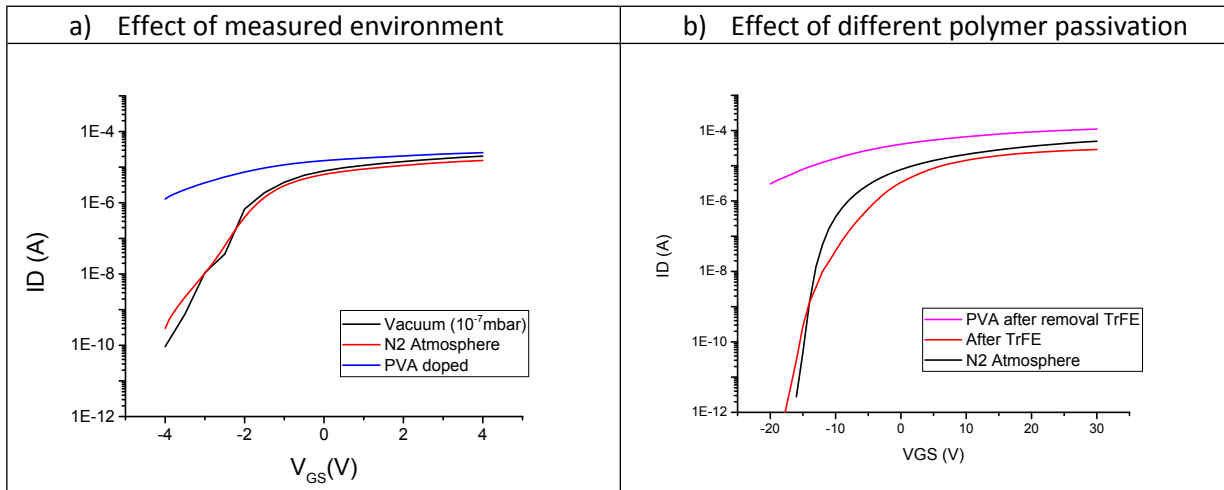


Figure S5: I_d - V_g of a MoS₂ FET measured in N₂ and vacuum conditions before and after PVA doping. (b) PVA vs TrFE coated MoS₂ FET

References:

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