Supporting Information

Microfluidic Approach for the Detection of Uric Acid through the Electrical Measurement of Atomically Thin MoS₂ Field-Effect Transistor

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1. Device fabrication process

The substrate (285 nm SiO2/p++Si) was first cleaned with acetone before being subjected to a 5-minute ultrasonic treatment. After that, it was cleaned with isopropanol solvent and then put through another five minutes of ultrasonication in water. Afterward, it was dried with an N₂ gun and kept on the hotplate at 110 ° C for 2 minutes. Ultraviolet (UV)-O₃ treatment was applied for 30 minutes to remove the remaining organic impurities. Next, the bulk MoS₂ flake was separated layer by layer by the scotch-tape method, followed by pasting onto the cleaned Si/SiO₂ substrate. This technique is widely described as the mechanically exfoliated method. After that, resist coating was performed by methyl methacrylate (MMA) and poly (methyl methacrylate) (PMMA A2) onto the substrates, followed by sophisticated electron beam lithography to form the metallic source (S) and drain (D) contacts by operating ELS-F125 ultra high precision E-beam lithography system. Finally, 10 nm Nickel (Ni) and then 1500 nm gold (Au) were deposited by an automatic E-gun evaporator followed by lift-off using N-methyl-2-pyrrolidone (NMP), acetone, and IPA solvents. The process is depicted in Fig. S1.



Fig. S1 Schematic illustration of the MoS₂-FET fabrication process.

2. Contrast method for the flake thickness measurement

It is possible to estimate the thickness of the MoS₂ layer by measuring the contrast of the reflected light from the SiO₂ surface with and without the presence of the MoS₂ flake.¹ The contrast is defined with the following equation: $C(\lambda) = \frac{R_0(\lambda) - R(\lambda)}{R_0(\lambda)}$

, where $R_0(\lambda)$ and $R(\lambda)$ are the intensities of the reflected light without and with MoS₂ for the wavelength of λ , respectively. It was suggested that the use of λ in the red region is more efficient than using the blue and green lights.¹ For the device used in this report, the $C(\lambda)$ of the red light was found to be 0.5, which indicates the number of layers of the flake to be four from the table calibrated previously.¹

3. Raman Spectroscopy Analysis

Raman spectra were obtained from a 3D Laser Raman Micro spectroscopy analysis system (Nanofinder 30) with $100 \times$ and $20 \times$ object lens. Excitation laser, grating selection, and wavenumber resolution were 532 nm, 1800 G mm⁻¹, and 0.5 cm⁻¹ to 20 cm⁻¹, respectively. The power and the laser exposure time were 5 mW and 5 s, respectively.

The Raman spectroscopy data is used to verify the doping effect physically. Fig. S2 Shows the Raman spectroscopic data of a few layered pristine MoS₂ before (black line) and after (red line) adsorption of uric acid on its surface. The number of layers of 2D chalcogenides can be estimated from the Raman frequency of in-plane vibration mode E_{2g}^1 and the out-plane vibration mode A_{1g} which is represented by Δ .² The Raman shift (Δ) of pristine MoS₂ was found to be $[A_{1g} - E_{2g}^1] = [403 \text{ cm}^{-1} - 384.5 \text{ cm}^{-1}] = 18.5 \text{ cm}^{-1}$, which reflects that our Raman analyzed MoS₂ was mono layered materials as shown by other reports.³ It is considered that the A_{1g} peak has a higher capacity for softening and is more sensitive to the deposited foreign molecules onto the MoS₂ substrate than the E_{2g}^1 peak due to the stronger electron-phonon coupling ^{4,5}. In our uric acid case, it is seen from the Fig. S2, that A_{1g} peak has shifted little lower wavenumber direction after depositing 1 μ M uric acid molecular solution onto the MoS₂ substrate surface, while the E_{2g}^1 peak is unchanged even after charge transfer happens from the molecule to the MoS₂.



Fig. S2 Raman spectrum of bare MoS₂ surface and after functionalization of uric acid.

4. Time of flight secondary ion mass spectroscopy analysis

Time of flight secondary ionization mass spectroscopy (SIMS) data was taken to validate the possibility of uric acid adsorption onto the bare MoS_2 surface. Fig. S3 (a) and (b), obtained using TOF-SIMS, show the presence of Mo^+ and $C_5H_5N_4O_3^+$ ion on the bare and adsorbed surface of MoS_2 , respectively. The color scale represents the relative ion counts, and brighter colors indicate higher secondary ion concentrations. Fig. S3 (c) and (d) are typically positive and negative TOF-SIMS spectrums where it is clear the presence of uric acid molecule both as a $[M+H]^+$ and $[M-H]^+$ peak at ~ 169.04 m/z and 167.02 m/z, respectively. The bright part is not homogeneous throughout the measured place may be due to the different-sized droplet formation on the MoS_2 during the drop cast time in the drop-casting process.



Fig. S3 (a) TOF-SIMS elemental mapping of Mo $^+$. (b) TOF-SIMS elemental mapping of $C_5H_5N_4O_3^+$. (c) - (d) TOF-SIMS spectrum of uric acid.

References

1 Y. Y. Wang, R. X. Gao, Z. H. Ni, H. He, S. P. Guo, H. P. Yang, C. X. Cong and T. Yu, *Nanotechnology*, DOI:10.1088/0957-4484/23/49/495713.

2 A. S. Pawbake, M. S. Pawar, S. R. Jadkar and D. J. Late, *Nanoscale*, , DOI:10.1039/c5nr07401k.

3 C. Lee, H. Yan, L. E. Brus, T. F. Heinz, J. Hone and S. Ryu, ACS Nano, , DOI:10.1021/nn1003937.

4 B. Chakraborty, A. Bera, D. V. S. Muthu, S. Bhowmick, U. V. Waghmare and a. K. Sood, *Phys. Rev. B*.

5 G. Kukucska and J. Koltai, *Phys. Status Solidi Basic Res.*, , DOI:10.1002/pssb.201700184.