Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Effect of Large Work Function Modulation of MoS_2 by Controllable Chlorine Doping Using a Remote Plasma

*Ki Hyun Kim,[a] Ki Seok Kim,[a e] You Jin Ji,[a] Inyong Moon,[b] Keun Heo,[c] Dong-Ho Kang,[c] Kyong Nam Kim,[d] Won Jong Yoo,[b] Jin-Hong Park,[c] and Geun Young Yeom[a,b,]**

[a]School of Advanced Materials Science and Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Republic of Korea.

[b]SKKU Advanced Institute of Nano Technology (SAINT), Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Republic of Korea.

[c]School of Electronic and Electrical Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Republic of Korea.

[d]School of Advanced Materials Science and Engineering, Daejeon University, Yongun-dong, Dong-gu, Daejeon 34520, Korea.

[e]Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, MA, USA.

*Corresponding Author : gyyeom@skku.edu

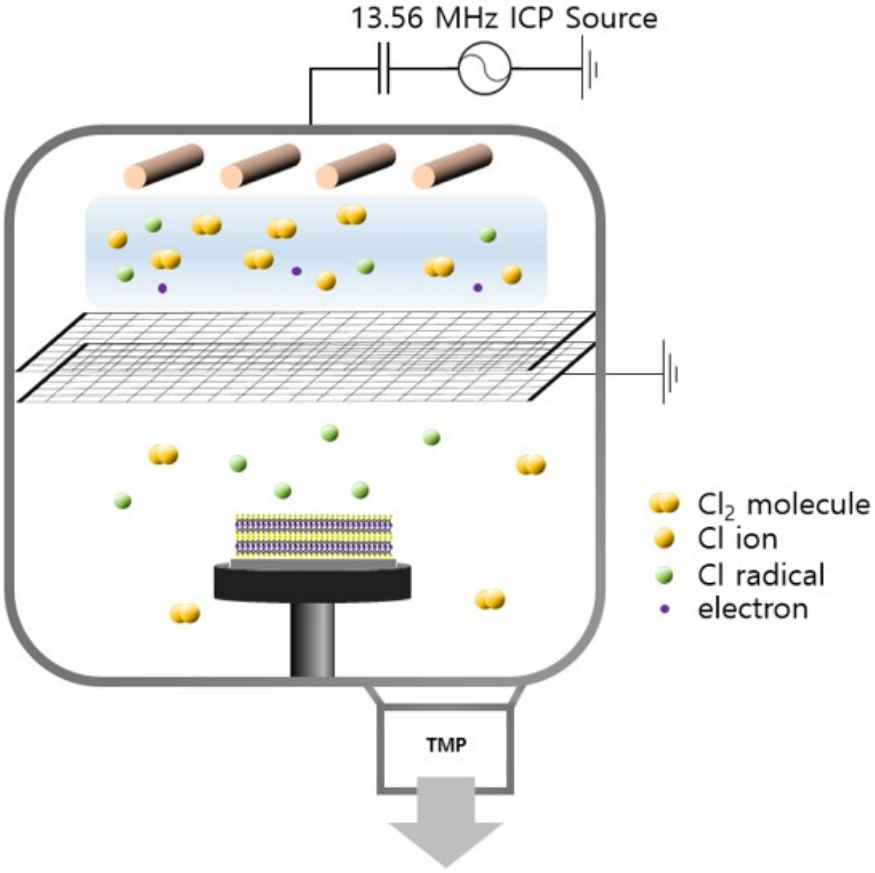

Figure S1. Schematic drawing of a 13.56 MHz remote ICP system. Double mesh-grid was installed at the center of the chamber to extract reactive radicals only. Almost all of ions were filtered at the grounded mesh grid.

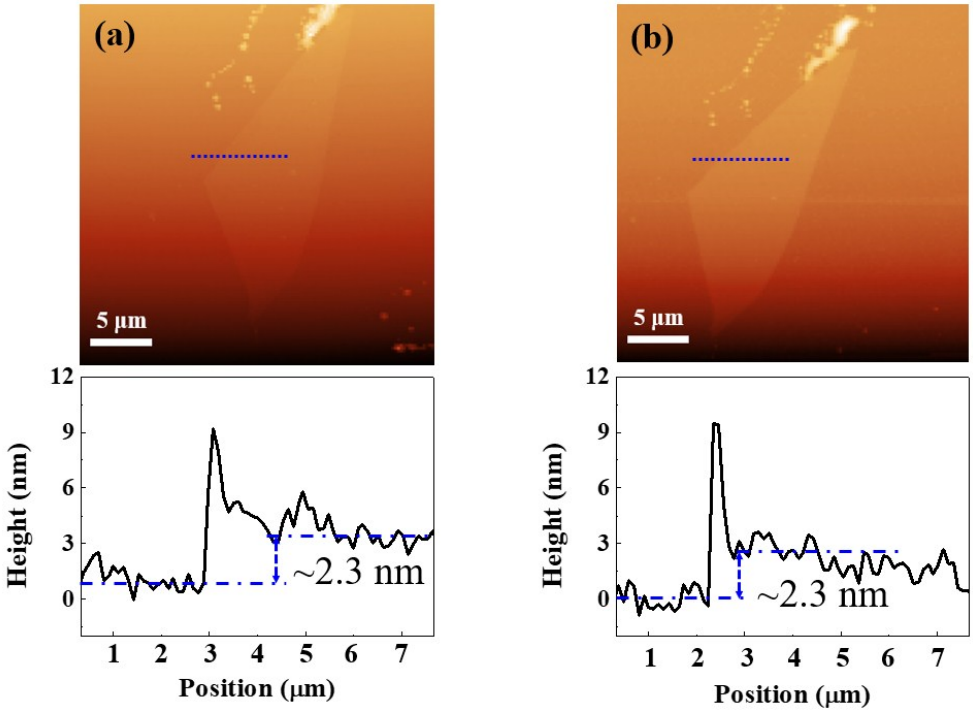

Figure S2. AFM profiles of (a) before and (b) after chlorine doping for 60 s. The thickness of ~ 2.3 nm is well accord with Raman spectroscopic data in Figure 2a. Any thickness change of MoS_2 flake was not observed after the chlorine radical treatment.

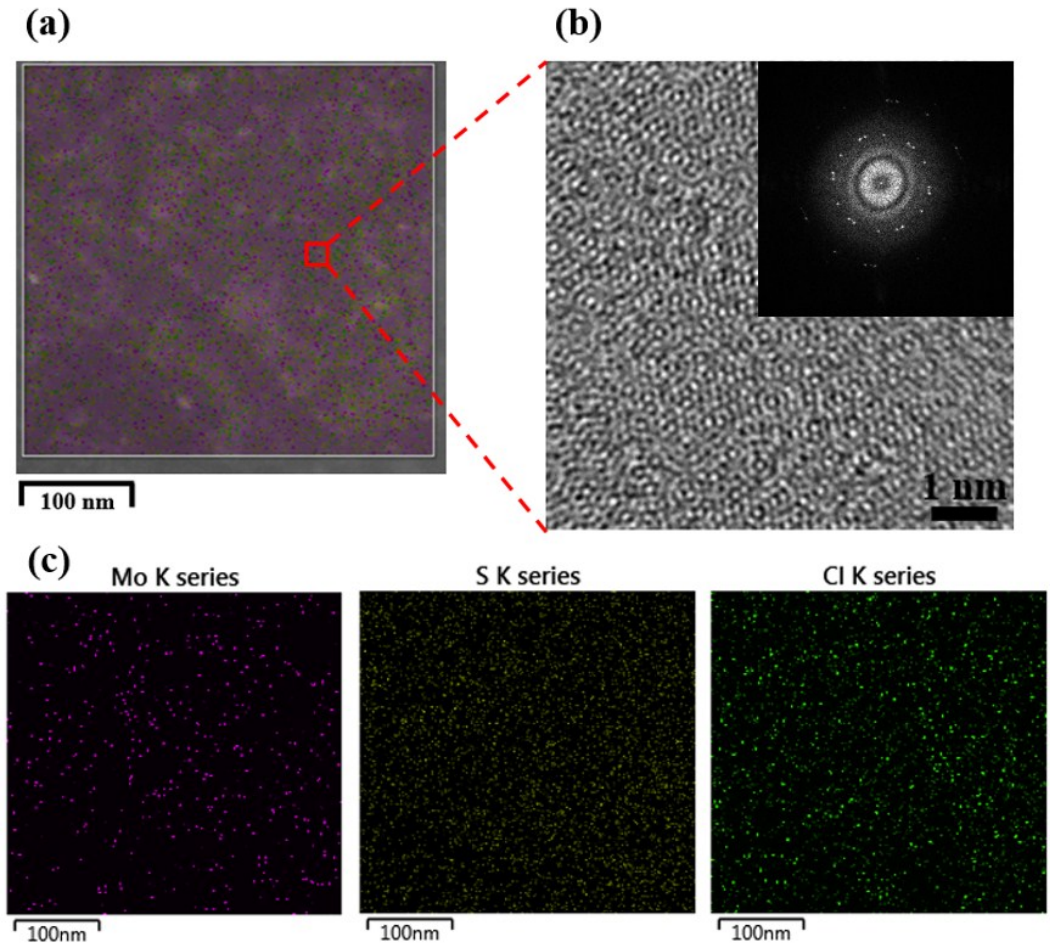

Figure S3. (a) EDS mapping image of MoS_2 in TEM after the chlorine radical doping for 30s. (b) High resolution TEM image of MoS_2. The inset shows a fast Fourier transform (FFT) image as a moiré pattern that appears for the crystalline multilayer-MoS_2[1]. (c) The mapping images of each element (Mo, S, and Cl) was illustrated separately. Surface distribution of Cl atoms on MoS_2 after the treatment was observed using an energy dispersive spectroscopy (EDS). As shown in (a), signals of Mo, S and Cl were observed on a surface of MoS_2 after treatment for 30 s. We observed no significant disorder of MoS_2 lattice after the chlorine radical doping, which was confirmed through high resolution TEM image in (b). Elemental images of Mo, S and Cl in (a) were separately illustrated in (c).

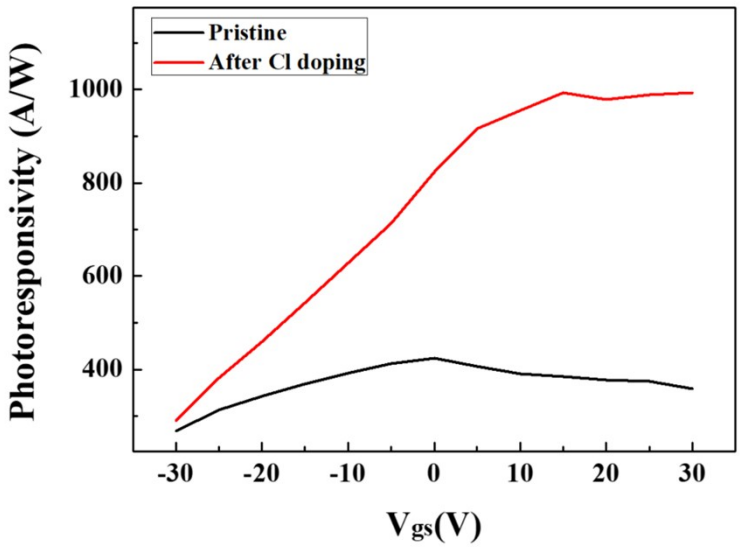

Figure S4. Photoresponsivity of pristine and chlorine doped MoS_2 FETs measured at λ = 520 nm while varying gate voltage from -30 to 30 V. The photoresponsivity of MoS_2 FETs was measured with gate bias (from V_{gs} = -30 to +30 V) at λ = 520 nm. The photoresponsivity of pristine MoS_2 FET showed the saturation point at V_{gs} = 0 V. For the chlorine doped MoS_2 FET, however, gradual increase of photoresponsivity was observed until V_{gs} = 30 V.

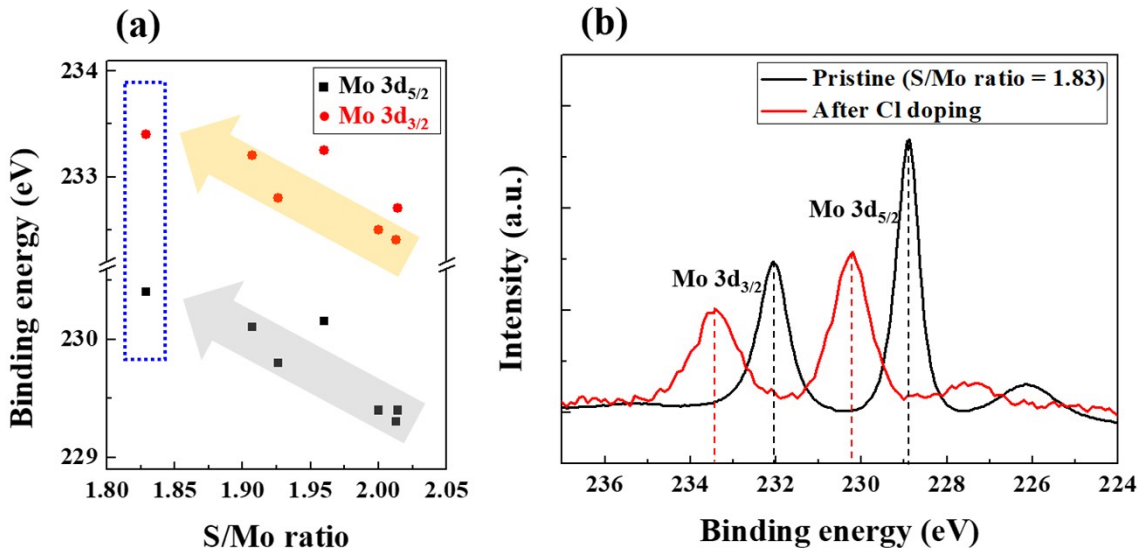

Figure S5. XPS analysis of trilayer MoS_2 grown by Chemical Vapor Deposition (CVD) process with various S/Mo ratio from 2.02 to 1.83. (a) The binding energy of two Mo 3d peaks for chlorine doped CVD MoS_2 having S/Mo ratio from 2.02 to 1.83. The MoS_2 having S/Mo ratio close to stoichiometric number of 2 showed relatively small blue-shift of Mo peaks because little substitutional doping of Cl atoms was occurred. However, those having lower S/Mo ratio under 2 have more probability for Cl atoms to bind on S vacancy sites resulting in more noticeable blue-shift of Mo peaks. (b) Narrow scan XPS data of Mo 3d measured on CVD MoS_2 having S/Mo ratio of 1.83 [the blue box in Figure (a)] before and after chlorine radical treatment for 30 s.

Reference

1. J. H. Jeon, S. K. Jang, S. M. Jeon, G. W. Yoo, Y. H. Jang, J. H. Park, S. J. Lee, *Nanoscale*, 2015, **7**, 1688-1695