Wafer Scale Synthesis of Semiconducting SnO Monolayers from Interfacial Oxide Layers of Metallic Liquid Tin

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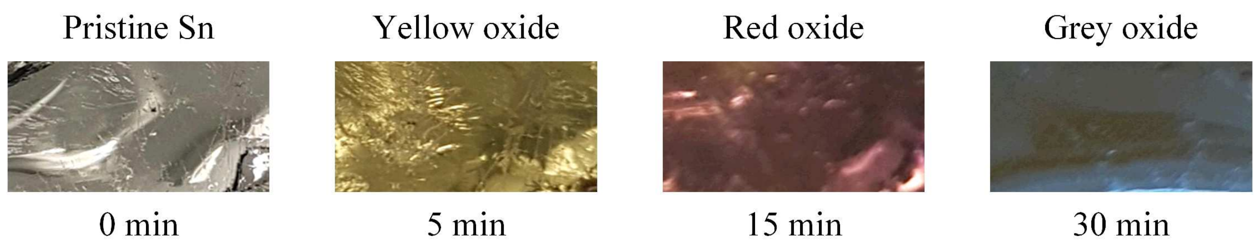

Figure SI-1: Photographs of liquid tin heated on top of a hotplate in an ambient atmosphere. The oxide layer was scraped off the liquid metal just prior to taking the first picture at 0 min. The photos were taken after the indicated time intervals.

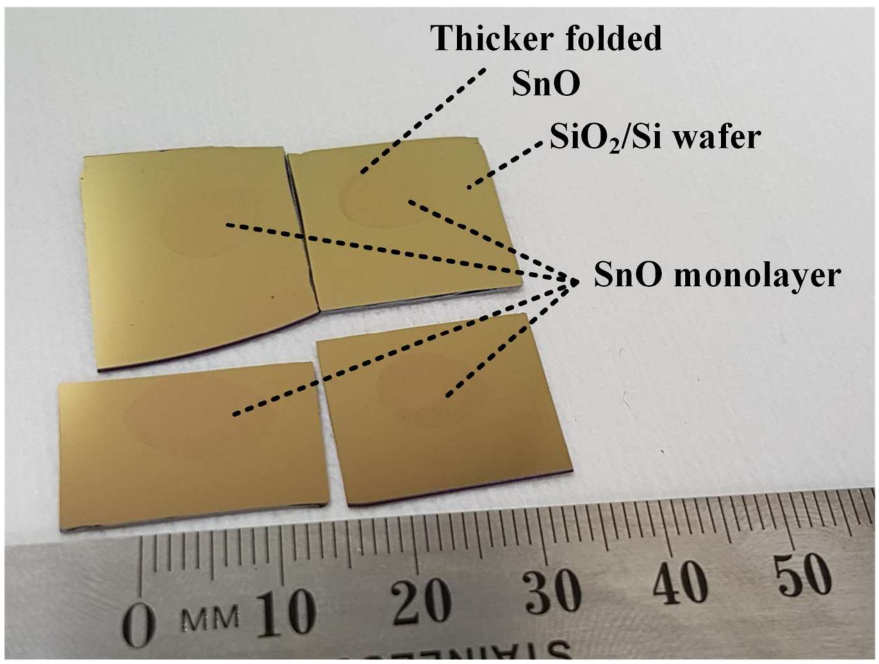

Figure SI-2: Centimeter sized SnO nanosheets deposited onto SiO_2/Si wafers. The color-contrast of the nanosheets against the SiO_2 coated wafer can be utilized as a proxy for the film thickness. This approach is commonly utilized for other 2D materials such as graphene and graphene oxide.[1] The results attest reproducibility between multiple successive depositions.

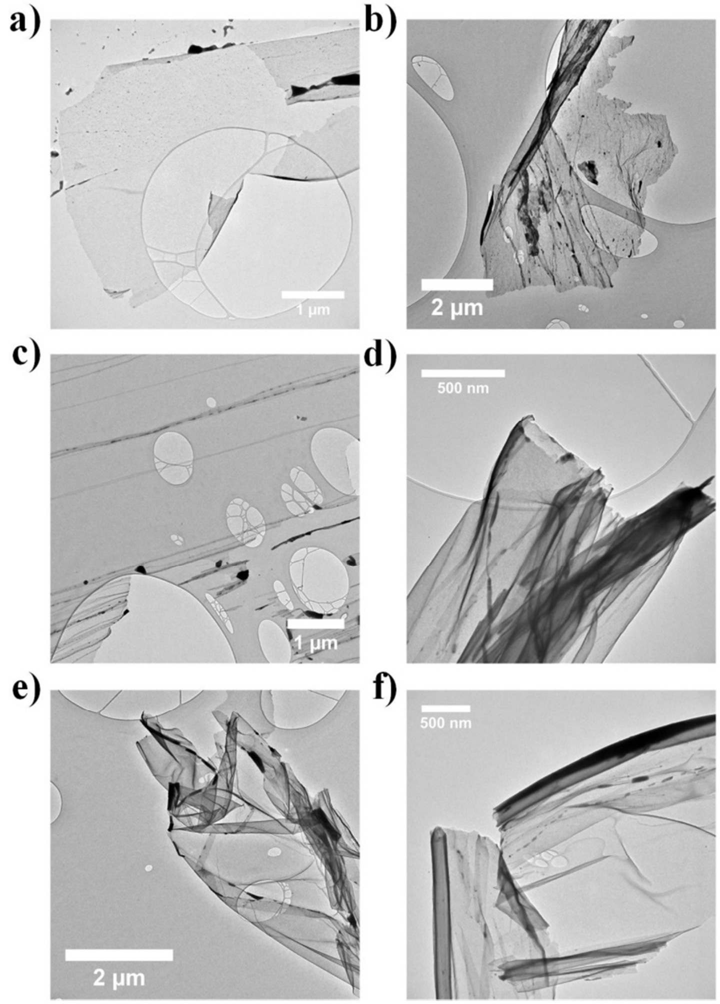

Figure SI-3: Additional TEM images.

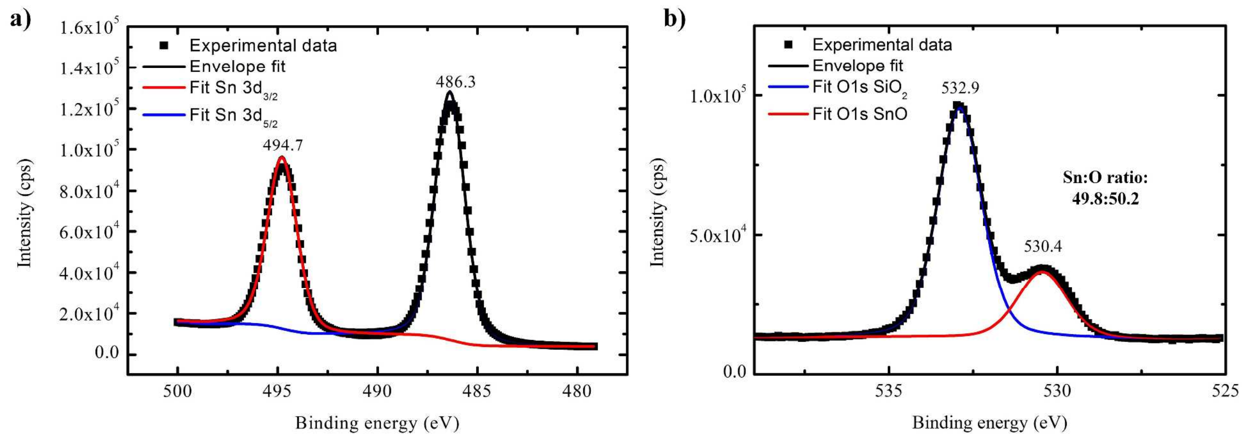

Figure SI-4: Representative XPS of the SnO nanosheets for the Sn 3d a) and O 1s b) regions. The oxygen peak at 532.9 eV can be attributed to the SiO_2 substrate, while the peak at 530.4 eV corresponds to SnO. Since the two observed oxygen peaks are well separated, the elemental ratio between Sn and O can be determined which is presented in b)

Table SI-1: Selected band transitions

	Indirect bandgap (eV)	Direct bandgap (eV)	Bandgap (Γ−Γ) (eV)
BULK	0.70 (Γ−M)	2.75 (M−M)	3.27
Monolayer	4.12(Γ−M)	4.21(Γ−Γ)	4.21
Bilayer	1.57(Γ−M)	3.09(M−M)	3.15

Brackets refer to regions in the first Brillouin zone where the top of the HOMO band and bottom LUMO band reside. For example (Γ−M) means the top of the HOMO band and bottom to the LUMO band are at the Gamma point (0,0,0) and M-point (1/2,1/2,0) respectively.

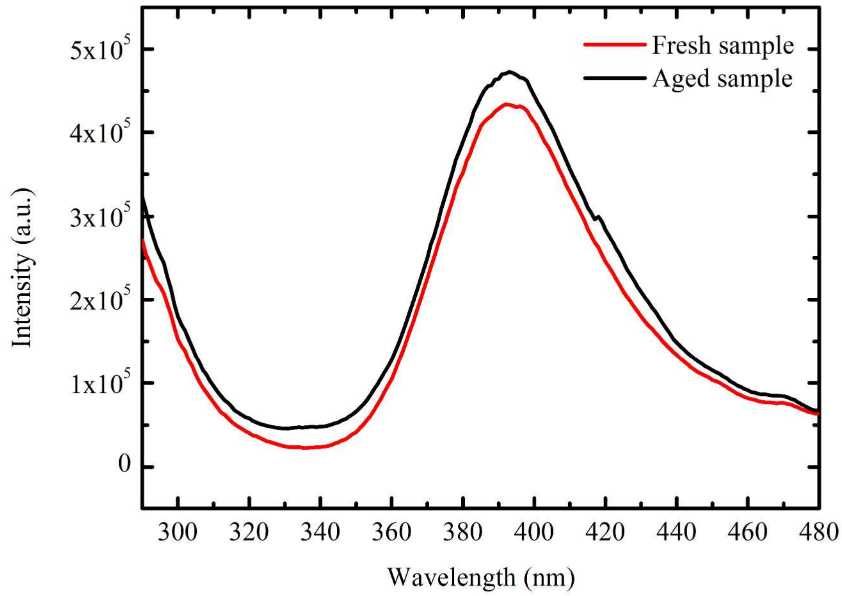

Figure SI-5: Photoluminescence spectra of monolayer SnO samples on quartz excited at 250 nm. The aged sample was measured after storing in ambient atmosphere for approximately 100 hours. The fresh sample was measured quickly after synthesis. All samples were prepared inside the glove box.

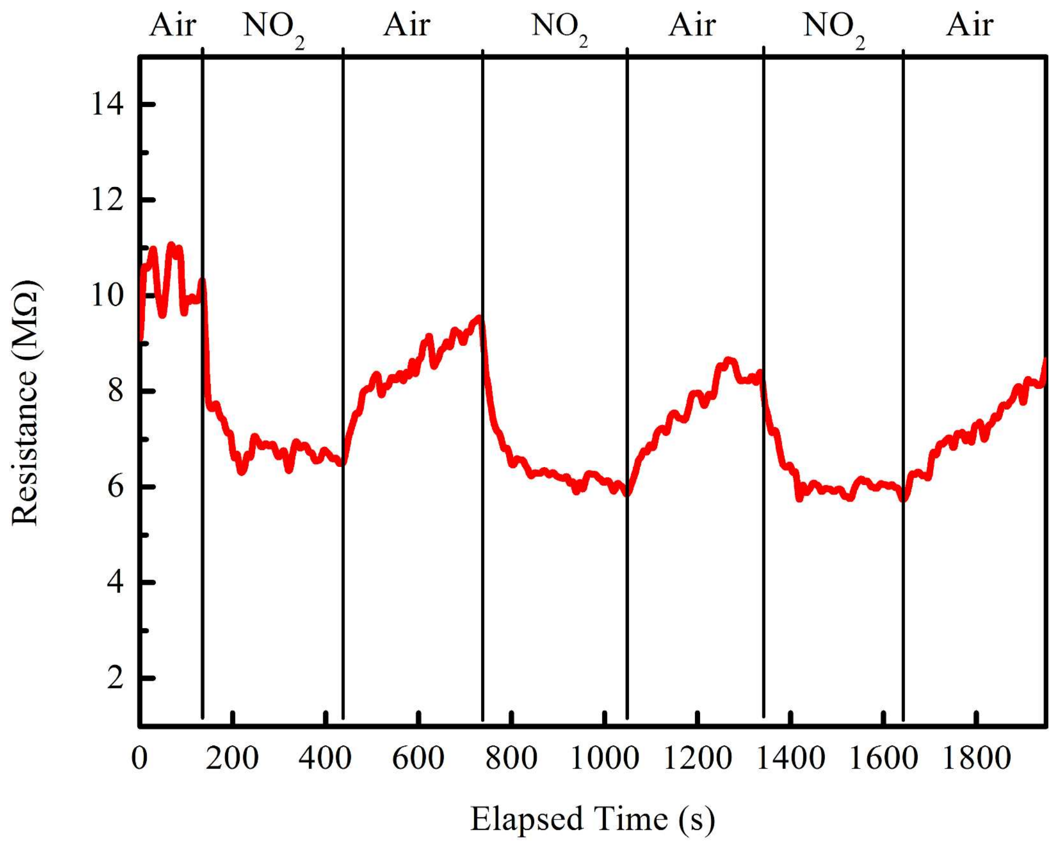

Figure SI-6: Resistance response of the SnO nanosheet exposed to alternating gas flows of dry air and 11.1 ppm NO_2 in dry air. The gas sensing device was prepared using SnO deposited in the glove box. Two contacts were prepared on the nanosheet using silver paste with an approximate distance of 1 mm. The flow rate was 200 sccm and the measurement was conducted at room temperature. The drop in resistance of the semiconductor when exposed to NO_2 indicates p-type conductivity.

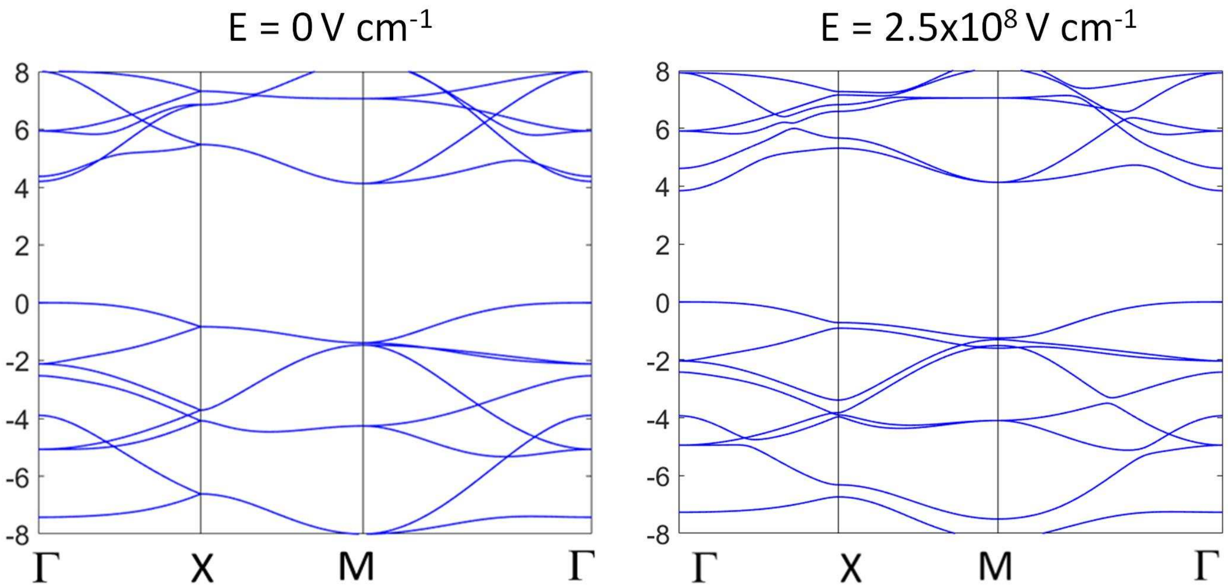

Figure SI-7: Calculated band structures of monolayer SnO under applied electric field in comparison to no electric field.

Table SI-2: Selected band transitions of monolayer SnO under different electric field strength.

	Indirect band-gap (eV)	Direct band-gap (eV)
E = 0 V cm^{-1}	4.127	4.200 (Γ–Γ)
E = 2.5×10^{8} V cm^{-1}	Not dominant	3.844(Γ-Γ)

Table SI-3: BULK Structure: SnO: Tetragonal, Space Group 129, (P 4/nmmS)

	a	c/a
This work	3.79	1.19
Experiment[2]	3.796(6)	1.27

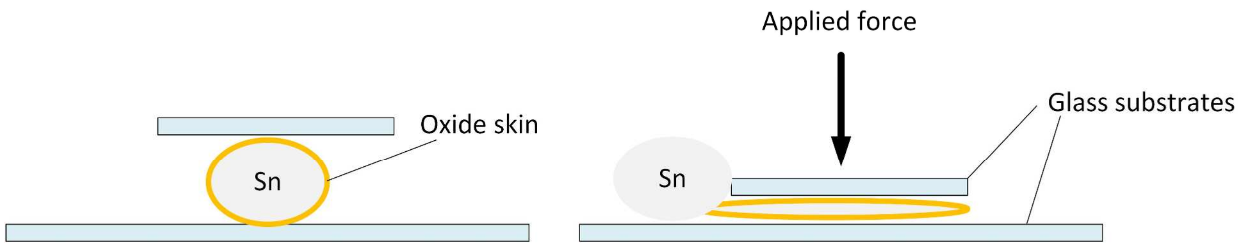

Figure SI-8: Schematic diagram of the preconditioning process used inside the glovebox. The elemental tin was melted on a hot plate while being placed on top of a microscopy glass slide. A second smaller glass slide was placed on top of the molten metal. The second glass slide was preheated to avoid freezing the tin on contact. Force was then applied to the top glass slide, squeezing the liquid metal. The liquid metal was found to penetrate through its oxide layer, leaving the oxide as residue between the two glass slides. A shiny metal droplet was obtained which could be used for the synthesis of 2D SnO nanosheets.

References

1. Inhwa, J.; Jong-Soo, R.; Jong Yeog, S.; Rodney, S. R.; Kyong-Yop, R. Colors of Graphene and Graphene-Oxide Multilayers on Various Substrates. *Nanotechnology* 2012, 23, 025708.

2. Moore, W. J.; Pauling, L. The Crystal Structures of the Tetragonal Monoxides of Lead, Tin, Palladium, and Platinum. *J. Am. Chem. Soc.* 1941, 63, 1392-1394.