MOCVD of WS_2 monolayers on conductive 4H-SiC for transfer-free vertical device fabrication

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Abstract

Two-dimensional (2D) transition metal dichalcogenides (TMDCs), such as WS_2, with a direct bandgap near 2 eV for monolayers (MLs), are promising for next-generation optoelectronics. Uniform TMDC films can be reproducibly synthesized on wafer-scale by metal-organic chemical vapor deposition (MOCVD). This process, typically at high temperatures, is usually performed on sapphire or SiO_2/Si. However, direct TMDC growth on conductive substrates for transfer-free vertical device fabrication remains a challenge. SiC, as a wide-bandgap semiconductor with optical transparency and high conductivity when doped, is ideal for e.g. light-emitting devices. Its crystalline structure and thermal robustness render it a suitable MOCVD substrate, enabling the integration with 2D WS_2. In this work, using a commercial AIXTRON multi-wafer MOCVD reactor, we establish a direct-growth method for WS_2 MLs on conductive 4H-SiC to demonstrate the transfer-free fabrication of vertical hybrid inorganic-organic p-n diodes. Tungsten hexacarbonyl ($W(CO)_6$) and di-tert-butyl sulfide (DTBS) serve as precursors with N_2 as carrier gas. 2" double-side-polished n-type 4H-SiC wafers (resistivity≤0.05 Ω·cm) are used as conductive substrates. The Si-terminated front side is CMP (chemical mechanical polishing)-finished, while the C-terminated backside is only mechanically polished. Preliminary attempts revealed that the direct WS_2 nucleation on the Si-terminated face was unsuccessful, likely due to its high surface energy and poor adatom adhesion. In contrast, WS_2 nucleation was possible on the C-terminated face, attributed to its lower surface energy and enhanced oxidation kinetics for improved sticking. However, WS_2 film quality was limited by the inferior backside preparation.
To achieve growth on the high-quality front side, the Si-terminated face was modified via in-situ carbonization (1050 °C, 30 min) using CH_4 (5%)/Ar. Compared to pristine SiC, the carbonized surface exhibits additional carbon-related Raman modes and prominent C-C bonds in X-ray photoelectron spectroscopy, demonstrating a successful carbonization. On such surface, WS_2 nucleation (nuclei size up to 130 nm) and ML coalescence were successful. The coalesced ML features a strong excitonic photoluminescence at 1.98 eV.
For vertical diode fabrication, the carbonized SiC and as-grown WS_2 serve as the n-type side. Poly[N,N'-bis(4-butylphenyl)-N,N'-bis(phenyl)-benzidine (poly-TPD) is spin-coated onto WS_2 modified by hexamethyldisilazane (HMDS), followed by electron beam evaporation of Ni under an oxygen atmosphere at room temperature to fabricate NiOx, serving as a hybrid inorganic-organic hole-transport and electron-blocking stack. 20 nm Ni and 50 nm Au are evaporated through a 250 µm-diameter shadow mask to form the top electrode. This vertical structure displays diode-like current-voltage characteristics, with a turn-on voltage of around 2.5 V. The highest forward current density is around 27.1 A/cm^2 at 5 V, while the reverse current remains as low as 0.04 A/cm^2 at −5 V, corresponding to a rectification ratio up to 678 at ±5 V. However, the electrical characteristics are not uniform across the entire wafer, primarily due to insufficient adhesion of poly-TPD on HMDS-treated WS_2. Light emission could not be detected, most likely attributed to an electron-dominated diode current. Further research will be performed to optimize polymer adhesion and to tune the electron-hole balance in favor of hole injection to demonstrate light emission of WS_2 and/or poly-TPD.