Supporting Information for "Electrical and Optical Polarization Responses of Composite Films Based on Aligned Carbon Nanotubes"

Firstly, we prepared vanadium alkoxide via an organic sol-gel method,[S1] in which a mixture of V_2O_5 powder, benzyl alcohol and isopropyl alcohol with a molar ratio of 1:4:40 reflux reacted at 110 ^oC for 4 hours to generate vanadium alkoxide. In this work, silicon and glass substrates were used as the substrates for the preparation of composite films. Before film preparation, the substrates were pre-treated by the following process: 1) Ultrasonic baths in acetone and methanol for 20 minutes, respectively; 2) Ultrasonic bath for 15 minutes in a mixture of concentrated H_2SO_4 and hydrogen peroxide solution with a ratio of 3:1; 3) Chemical clean at 70 ^oC for 1 hour in a mixture of deionized water, ammonia, and hydrogen peroxide solution with a ratio of 5:1:1; 4) Finally, the substrates were rinsed with deionized water, and then they were dried by N_2 gas. After treatment, VO_x sol was spin coated onto these cleaned substrates. The spin coating conditions for preparing VO_x sol include: first low-speed spin coating at 1000 r/min for 5 seconds, and then high-speed spin coating at 2500 r/min for 30 seconds, repeating for four times. Secondly, in order to yield solvable and dispersed SWCNTs, SWCNTs with carboxyl groups (1–2 nm diameter, 1–5 μm length, Xianfeng Nano Inc., China) were functionalized by ODA for 7 days.[S2,S3] Thirdly, 0.2 wt % functional single-walled carbon nanotubes (F–SWCNTs) and resin epoxy (Sigma-Aldrich, Z105937) were resolved in Tetrahydrofuran to obtain a SWCNT solution, after then hardener was added and stirred until the viscosity was increased to a range of 15–25 Pa·s that is suitable for blowing bubbles.[S4] Fourthly, 0.5–1.0 g SWCNTs were transferred to the polished the top surface of the die, and then high-purity N_2 gas (pressure of 150 kPa) was introduced to initiate bubble expansion. Meanwhile, a ring adhered on the middle top of the bubble was vertically driven by hand to keep the grow direction continuously moving

upward with a certain speed until getting a 25−30 cm diameter bubble. Fifthly, the BBF were transferred to the silicon or glass substrates that have been pre-treated and pre-coated with VO_x sol by spin coating, as described above. Finally, after BBF transferring, the films were annealed in furnace at 400 ^oC for 1 hour. Thus, the VO_x–aligned CNT composite films were yielded.

The as-prepared composite films were characterized by scanning electron microscopy (SEM, JEOL JEM-6610LV), dark-field optical microscope (OM, OLYMPUS BX51), high-resolution transmission electron microscopy (HRTEM, JEM-2100F), high resistance meter (KEITHLEY 6517A), and UV-Vis spectroscopy (SHIMADZU UV-1700), respectively. Particularly, a polarizer was employed to generate polarized light for UV-Vis measurements.

We further employed Raman spectroscopy to investigate the chemical structures of the SWCNTs used in this work. The following Figure SI-1 shows the Raman spectrum of the SWCNTs deposited on a Si substrate. Two peaks appearing at ~302 cm^{-1} and ~939 cm^{-1} are ascribed to the Raman signals from the Si substrate. The G^+–peak at ~1585 cm^{-1} and D–peak at ~1310 cm^{-1} are assigned to the in-plane vibrations along the tube axis and disorder induced phonon vibrational mode of the hexagonal Brillouin zone in the graphitic structure, respectively. The peaks at 265–156 cm^{-1} in Figure SI-1 are the radial breathing modes (RBM) of SWCNTs, from which the diameters of SWCNTs were estimated to be 0.9–1.6 nm. Notably, these data agree well with those (1–2 nm) provided by the suppliers as well as those roughly estimated from the SEM image (Fig. 2 in the manuscript). Accordingly, we believe that the diameters of the SWCNTs used in this work are about 1–2 nm.

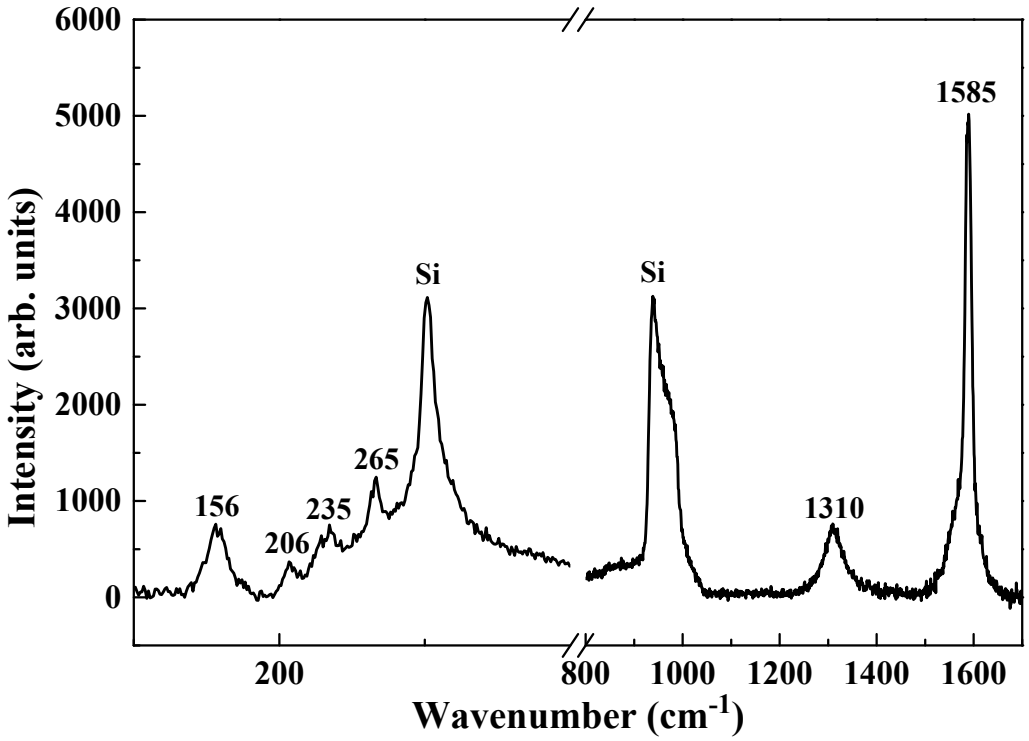

Figure SI-1. Raman spectrum of the SWCNTs used in this work on Si substrate.

S1 X. Xu, S. Yang, Y. Jiang, D. Zhou, W. Dan, Vanadium oxide film applied in infrared detector and method for manufacturing the same. *Chinese Patent*, CN 101900607 B, 2012.

S2 G. Yu, A. Cao, C. M. Lieber, Large-area blown bubble films of aligned nanowires and carbon nanotubes. *Nat. Nanotechnol.*, 2007, 2(6), 372-377.

S3 G. Yu, X. Li, C. M. Lieber, A. Cao, Nanomaterial-incorporated blown bubble films for large-area, aligned nanostructures. *J. Mater. Chem.*, 2008, 18(7), 728−734.

S4 A. Javey, S. Nam, R. S. Friedman, H. Yan, C. M. Lieber, Layer-by-layer assembly of nanowires for three-dimensional, multifunctional electronics. *Nano. Lett.*, 2007, 7(3), 773-777.