Supporting Information

Large-scale Assembly of "Type-switchable" Field Effect Transistors based on Carbon Nanotubes and Nanoparticles

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1. Experimental Procedure

Surface molecular pattering: 3-Aminopropyltriethoxysilane (APTES), octadecyltrichloro-silane (OTS) SAM molecules and solvents were purchased from Sigma-Aldrich. For the patterning of OTS SAM on SiO_2 or Al_2O_3, photoresist (AZ5214) was first patterned on the substrates via photolithography using a short baking time (<10 min in 95^{o}C). The patterned substrate was placed in the OTS solution (1:500 (v/v) in anhydrous hexane) for 100 sec. Then, the photoresist was removed with acetone. For the patterning of OTS and APTES, the substrate with OST SAM patterns was placed in APTES solution (1:500 (v/v) in ethanol) for 7 min to backfill remained oxide surface. Finally, the substrate was thoroughly rinsed with ethanol.

Assembly of swCNTs and Au NPs: For the preparation of single walled carbon nanotubes (swCNTs) suspensions, purified swCNT purchased form Carbon Nanotechnologies Inc. were dispersed in 1,2-dichlorobenzene via ultrasonic vibration for 20 min. The typical concentration for swCNT suspension was 0.05 mg/ml. For the swCNT assembly, the patterned surface was placed in the suspension usually for 10 sec and rinsed thoroughly with 1,2-dichlorobenzene. Au NP solution was purchased from British Biocell International. For NP assembly, the surface patterned comprised of OTS and APTES SAM was placed in the NP solution for 20 min and rinsed thoroughly with de-ionized water.

Thin film deposition and electrode fabrication: Al_2O_3 layers were deposited at 150°C via ALD process (Quros Company, plus-150) using trimethylaluminum and water. For the electrode fabrication, the photoresist was first patterned on the substrate. And then

Au (30nm) on Ti (10nm) was deposited on it and the photoresist was removed with acetone for the lift-off process.

2. Theoretical Simulation

The model system is composed of an Au NP (30 nm in diameter), two swCNTs (1 nm in diameter), a top and a bottom gates (located at z = 180 nm and 0 nm, respectively) within a unit cell of 100 nm × 100 nm × 180 nm depicted in **Figure 4** in the manuscript. Periodic boundary conditions were used along the x- and y-axes, while fixed boundary condition was used along the z-axis.

Figure 4a and **4b** show color-coded electric potentials with equipotential contour lines for V_T = 10 V and when the top gate voltage is turned off (V_T = 0 V), respectively, corresponding to **Figure 3b**. Two different configurations were considered in each case. At V_T = 10 V, the potentials of Au NPs and the swCNTs are saturated to $V_{Au} = V_c + V_{CNT}$ and $V_{CNT} = V_g/2$, respectively, where V_g is the potential corresponding to the semiconducting swCNT band gap and chosen to be 1 V. V_c is the critical voltage for tunneling between Au NP and swCNTs and is chosen to be 1.5 V, based on the breakdown field (0.3 ~ 0.5 V/nm) of the Al_2O_3 layer. The average spacing between Au NPs and swCNTs estimated to be 3~5 nm in our modeling and experimental configurations. When saturated, the potential difference between the Au NP and the swCNT is held to be V_c. If the top gate voltage is turned off from V_T = 10 V, the potentials of the Au NP and the swCNTs drop to $V_{Au} \approx$ -1.86 V and V_{CNT} = -0.5 V, respectively, but their potential difference is smaller than V_c, thus keeping the excess charges on the Au NP (**Figure S2** in Supporting Information).

The voltage saturation can be explained as following. Since the numbers of electronic states of the Au NP and the swCNT near the Fermi level are much larger than their respective excess charges calculated with the given geometry, the excess charge barely changes their Fermi levels. Therefore if the potential difference between the NP and swCNT reaches the critical value $\pm V_c$, then it is held at that value even at larger gate voltage than $\pm V_c$. Especially, for the semiconducting swCNTs, the Fermi level can change, but only within the electronic band gap; once the Fermi level meets the lowest energy of the conduction band or the highest energy of the valence band, electronic self energy will dominate the total energy preventing further change in the Fermi level within the band structure. Therefore, a nanotube network can be treated as a nearly-conductor aside from the voltage shift due to their effective semiconducting gap chosen to be 1 eV in our calculations.

3. Supplementary Figures

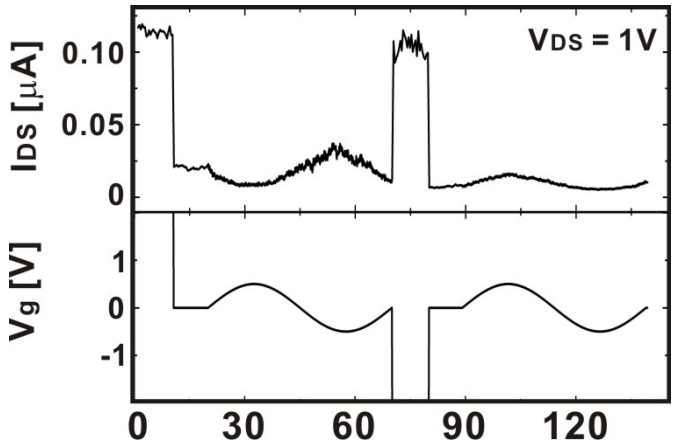

Figure S1. Real-time switching of device types using a single gate. After applyi ng a large *positive* (or *negative*) top-gate voltage (+*10*V (or -*10*V)), we obtained *p*-type (or *n*-type) behavior of the type-switchable FET.

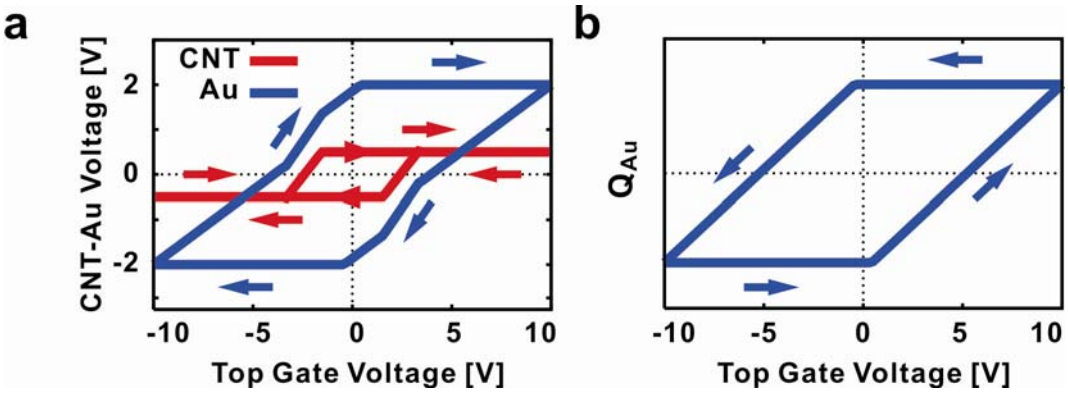

Figure S2. a, Hysteresis behaviors in potentials of the Au NP and the semicond ucting swCNT as a function of the top gate voltage. **b**, Excess charges on the A u NP as a function of the top gate voltage. This plot also shows the hysteresis behavior. The arrows indicate the hysteresis direction along the gate voltage swee ping.