

Supporting Information

Au nanoparticle synthesis

Au nanoparticles are synthesized by reacting 25 μmol of HAuCl_4 with 0.2 mmol of Sodium Citrate in 100ml of DI water¹. The colloidal solution is centrifuged at ~ 5000 rpm for 5 min to remove aggregates.

Linker molecule synthesis

The linker molecule is synthesized by reacting equal volume of 0.08M DMF (anhydrous 99.8%, Aldrich) solution of 1-pyrene butanoic acid, succinimidyl ester (Molecular Probes) and 0.1M DMF solution of Cysteamine (Biochemika $\geq 98.0\%$) in a microreactive vial for 2.5 hours in dark, ambient condition. The resulting solution is diluted 18 times with DMF for CNT device incubation (dark, ambient, 60 minutes).

Au nanoparticle attachment procedure

Carbon nanotubes are grown on 200nm SiO_2/Si n⁺⁺ wafers by chemical vapor deposition. CNT devices are pumped in a heated vacuum jar (55 C) for 5 hours and annealed in Argon atmosphere at 400 C for 20 minutes immediately before linker molecule solution incubation. After incubation, the chip is rinsed in DMF thoroughly and dried in a stream of dry N_2 . It is then dipped into Au colloidal solution for 60s, thoroughly rinsed in DI water and dried in dry N_2 . CNT devices show nearly identical V_g dependence in conductance measurement before and after Au nanoparticle attachment.

Parameters of the AFM cantilever

The AFM cantilever used in this experiment has a natural resonant frequency $\omega_0/2\pi = 61252$ Hz, a calibrated spring constant $k = 2.5$ N/m, and a high quality factor $Q = 39,500$.

FEMLAB simulations

We have used the following input parameters in the simulation (FEMLAB software): tip radius 30 nm, tip height above substrate 60 nm, CNT diameter 1.5 nm, Au nanoparticle diameter 10 nm, positioned 1 nm above the CNT. Results of the simulation: $C_g = 0.24$ aF, $C_{tip} = 0.11$ aF, $C_{cnt} = 0.53$ aF, $C_{Au} = 0.88$ aF.

Estimation of the tunneling current and power dissipation

The AFM tip perturbs the Au nanoparticle's electrochemical potential by $d\varepsilon = e \frac{dq_c}{dz} \frac{dz}{C_{Au}}$.

$d\varepsilon$ is estimated to be approximately 10 meV for particle No. 1 using the calibrated amplitude $dz = 2$ nm, $C_{Au} = 1.5$ aF and $dq_c/dz = 7.3 \times 10^{-12}$ C/m determined with the method described in Ref. 11. This ac perturbation induces tunneling charge $dq = ef'(0)d\varepsilon = 0.38$ e, which corresponds to a current $I = \omega dq = 23$ fA. The power dissipated in the tunneling process is given by $P_e = \frac{1}{2} \omega k dz_{\max}^2 \delta\left(\frac{1}{Q}\right)$. P_e ranges from 6-33 aW in Fig. 4(b). The accompanying

frequency shift signal varies between 0.1-0.44Hz, corresponding to a spring constant change of 0.8×10^{-5} - 3.6×10^{-5} N/m. These extraordinarily small values attest to the superb sensitivity

of our force-sensing techniques.

Estimation of the alkyl chain resistance

Per Ref. 13, we have used $\beta=1.1/\text{\AA}$, chain length $1.18 \text{ \AA}/\text{carbon}$ and a transmission rate 0.4 for Au-S bond to estimate the tunneling resistance of the alkyl chain.

Calibrating the Au nanoparticle nanoelectrometer

Potential change ΔV on both electrodes and hence the whole CNT shifts the electrochemical potential of the Au nanoparticle ε_{dc} by $\Delta V C_{cnt}/C_{Au}$ and causes the alignment between the CNT and the Au particle ($\Delta\varepsilon_{dc}$) to shift by $\Delta V (1-C_{cnt}/C_{Au})$. This shift is canceled by $\Delta V_g = -\Delta V (C_{Au}-C_{cnt})/C_g$ on the backgate. The calibration step determines the coefficient $-(C_{Au}-C_{cnt})/C_g$ for subsequent measurement of an arbitrary local potential change.

Perturbation on the CNT

Au nanoparticles linked to a CNT can potentially perturb the CNT by its static charges and single electron charging events. Below we provide additional data to demonstrate that this perturbation is weak in our setup.

$I-V_g$ measurements of the NT junction used in our manuscript before and after the attachment of Au nanoparticles are shown in Fig. S1. This plot shows that charges present on the attached Au nanoparticles do not introduce significant scattering to the NT. Similar behavior is seen in all 4 NTs to which we attached Au nanoparticles. In addition, we have also performed extensive scanned gate measurements (e.g., Fig. S2, also see ref. 12 for details of the technique) to monitor the conductance of the NT simultaneously while measuring the single electron charging events of the Au nanoparticles. We did not see any conductance change of the NT associated with these events. These experiments also ruled out any possible formation of quantum dots on the NT (see ref. 13) due to the electrostatic influence of the charges present on the Au particles.

A recent study on charging Au nanoparticles deposited on NTs showed conductance jumps and slow time dynamics in the NT, likely due to the combination of a low conductance of the NT (10^{-8} - 10^{-6} S) and the very high resistance of the junction ($10^{19} \Omega$)².

References

1. Enustun, B. V.; Turkevich, J. *Journal of the American Chemical Society* **1963**, 85, (21), 3317-3328.
2. Gruneis, A.; Esplandiu, M. J.; Garcia-Sanchez, D.; Bachtold, A. *Nano Letters* **2007**, 7, (12), 3766-3769.

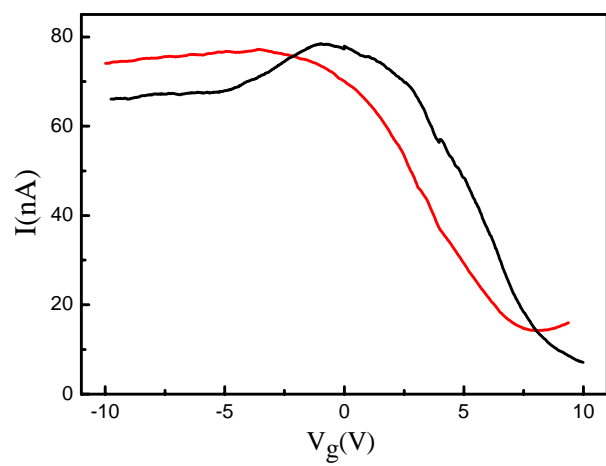


Fig. S1

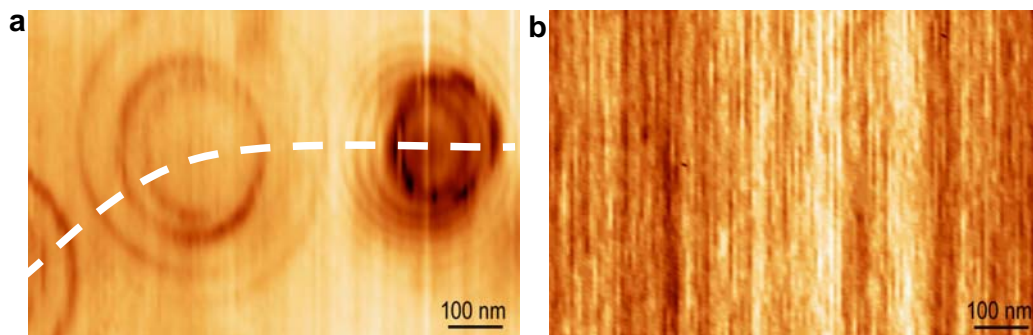


Fig. S2

Supporting figure captions:

Fig. S1: The current-backgate voltage (I - V_g) dependence of the NT junction before (red trace, room temperature, ambient condition) and after (black trace, 77K, vacuum) the attachment of Au nanoparticles. The slight difference between the two traces is commonly seen in CNTs and is most likely due to the rearrangement of charged impurities in the substrate during the cooldown process. In the measurement setup, $V_{sd}=10\text{mV}$ and a $10\text{k}\Omega$ resistor is in series with the NT junction. The high conductance level corresponds to a resistance of $\sim 115\text{k}\Omega$.

Fig. S2: (a) Coulomb oscillations of the Au nanoparticles as described in Fig. 2. Overlaid white dashed line indicates the location of the NT. (b) Simultaneous measurement of the current through the NT demonstrating the lack of conductance change associated with the charging events of the Au nanoparticles and the negligible gating effect of the AFM tip. RMS roughness in (b) corresponds to 0.05nA . Imaging parameters: $V_{tip} = 2.5\text{ V}$, $V_g = -2\text{ V}$, tip height $z = 60\text{ nm}$, $V_{sd}=10\text{mV}$.