

Electronic Supplementary Information
for “Resistive switching in iron-oxide-filled carbon nanotubes”

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Current versus voltage for a regular not-filled MWCNT

We studied the electrical properties of a device based on a regular film of MWCNTs with a planar geometry (Fig. SI1). The device was assembled onto a well cleaned glass substrate, where a layer of titanium (10 nm thick) and gold (100 nm thick) was thermally evaporated in a low pressure chamber in order to produce the electric contacts. The channel was achieved from a carbon microfiber shadow mask. The obtained electrodes dimensions were: L=8 μ m; c=1 mm and h=0.5 μ m.

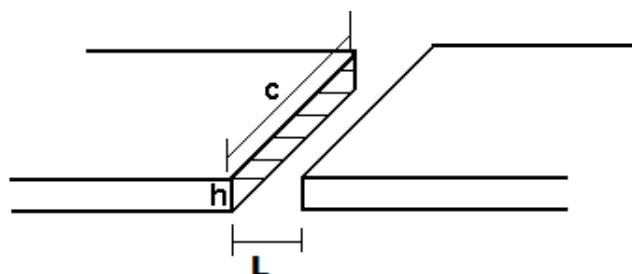


Fig. SI1 Planar device scheme.

The nanotubes dispersion was prepared from 0.3 mg of MWCNTs (commercialized by CNTCO Ltda.) dispersed at chloroform (20 mL) by an ultrasonic bath (90 min). After the dispersion, the MWCNTs were then deposited between the electrodes by casting, under a temperature of 70 °C. The electrical current versus

applied voltage (I-V) was obtained from an Agilent semiconductor parameter analyzer (4155C), with a limited compliance current of 100 μ A, in order to avoid any damage to the sample. The results showed at Fig. SI2 represent a characteristic I-V behavior from a film of MWCNTs.¹

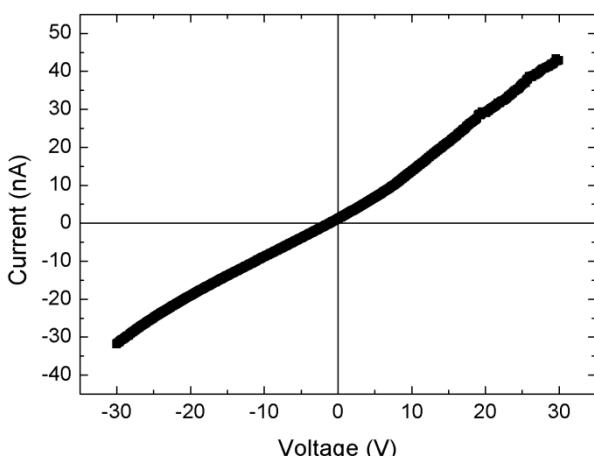


Fig. SI2 The characteristic current intensity versus voltage curve (I-V) for MWCNT planar device.

Lorentzian line shape analyses for non-filled MWCNTs

Using the device presented above, the same Raman analyses was performed for commercial non-filled MWCNTs using the same method as describe on the paper. The Lorentzian line shape analyses for this device are presented in Fig. SI3. The G center shift with a range applied from -40 to 40V is showed at Fig. SI3a. Due to the structural differences (e.g. diameter, length and chirality), a full comparison between non-filled MWCNTs and Fe-MWCNTs is not viable. However, it is possible to notice that the non-filled MWCNTs presented a similar behavior to those published by other groups (paper ref. 32, 33), where one MWCNT is tested in a tree-electrodes device. Different from Fe-MWCNTs, non-filled MWCNTs do not present a clear tendency which can be related with the absence of charge retention. The I_D/I_G analyses (Fig. SI3b) presented by the non-filled MWCNTs have a distribution of values around the value of 1.0. This behavior can also be attributed to the linear electric behavior presented by the non-filled MWCNT if compared to the Fe-MWCNTs.

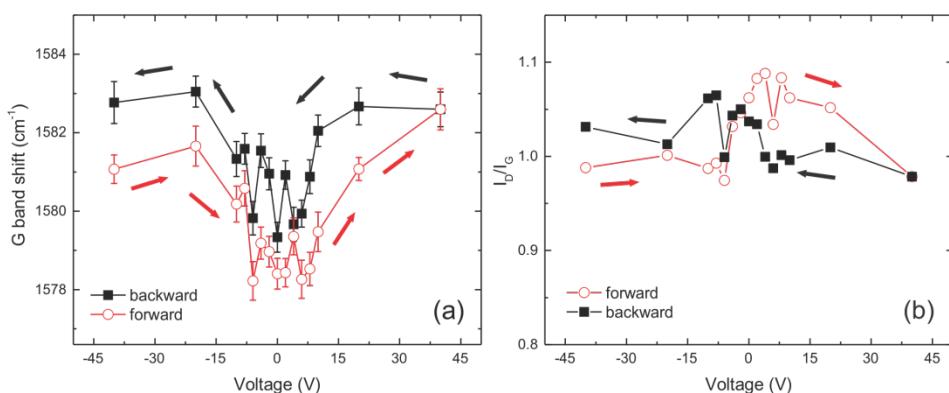


Fig. SI3 Raman mode energies as a function of the applied voltage in both directions for non-filled MWCNTs. The solid square represents -40 to 40 V (backward), and the empty circle represents 40 to -40 V (forward). (a) G band applied voltage dependence and (b) I_D/I_G relative intensity applied voltage dependence.

Polarization test of Fe-MWCNTs

In order to confirm the time dependence of the electrical behavior of the Fe-MWCNTs, the polarization versus voltage (P-V) measurements were carried out on Fe-CNTs samples. The polarization test consists in a Sawyer-Tower circuit using a capacitor ($0.1 \mu\text{F}$) as a reference to achieve the charge on the ferroelectric device at room temperature. In this test we assemble a device with a higher amount of Fe-MWCNTs in order to achieve a lower resistivity. The polarization tests showed at Fig. SI4 demonstrate the effect of the electric applied field and the frequency. The figure clearly demonstrates the hysteresis loop behavior characteristic of a polarization effect induced by an electric field applied, without the characteristic saturation of a ferroelectric.

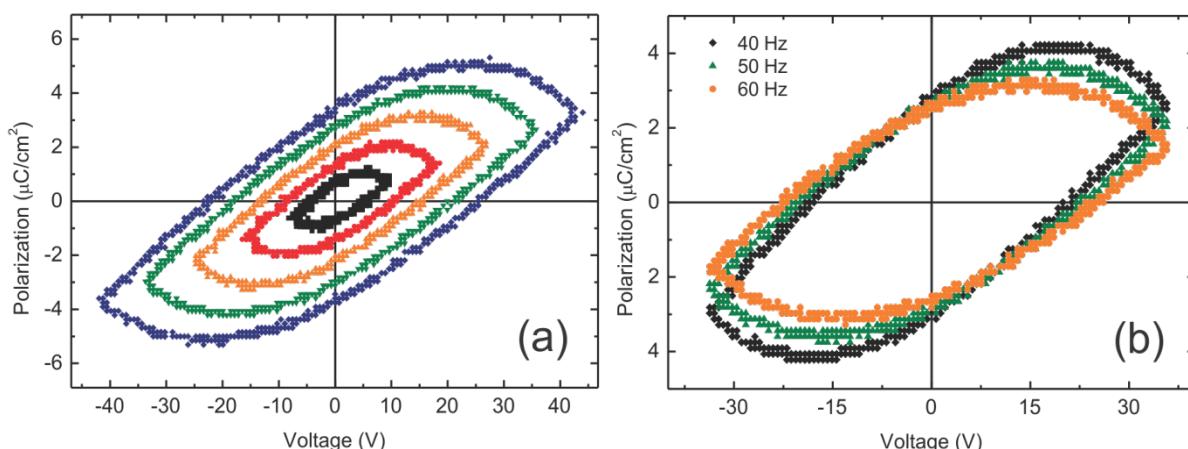


Fig. SI4 (a) Polarization versus electric applied field: (a) voltage dependence; (b) frequency dependence.

For the Fe-MWCNT device it is possible to note the polarization external voltage dependence with a fixed frequency applied of 40 Hz at room temperature. From the Fig. SI3a, it can be seen that there is no saturation with these values of voltage applied. The Fe-MWCNT time dependence is demonstrated at Fig. SI3b. The frequency applied was increased in small steps and the device presented a small reduction of the remnant polarization from $2.8 \mu\text{C}/\text{cm}^2$ at 40 Hz to $2.5 \mu\text{C}/\text{cm}^2$ at 60 Hz. These results are characteristic of devices with a nonlinear electric behavior. This is not a usual ferroelectric behavior, but indicates a complex process of leakage current.²

References

1. G. F. Close and H. S. P. Wong, *Nanotechnology, IEEE Transactions on*, 2008, **7**, 596-600.
2. M. Dawber, K. M. Rabe and J. F. Scott, *Reviews of Modern Physics*, 2005, **77**, 1083-1130.