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Periodic current oscillations observed in organic MIM junctions

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Abstract We reported here the fabrication of an all-organic transistor built with metal–insulator–metal (MIM) junctions and its preliminary results. These planar samples consist basically of three interconnected tunnel junctions each one formed by an insulating film of poly(methyl methacrylate) (PMMA) sandwiched between two electrodes of polyethylene dioxythiophene/polystyrene sulfonate (PEDOT/PSS) assembled on a polyester flexible substrate. Electrical measurements were taken at room temperature, with a Keithley 2400 programmable semiconductor measuring system. The junctions presented individual capacitances of 1.40 pF. We observed an oscillatory variation of the drain current with the gate voltage. All associated capacitances were calculated for characterizing the transistor and the bottom electrode material.

Introduction

Recently a great attention has been given to organic materials for applications in electronic, microelectronic, and photonic devices [1–3]. Also, single electron transistors (SETs), based on tunnel junctions—which are metal–insulator–metal structures—and operating on the principle of Coulomb blockade (CB) in nanostructures are promising candidates for future ultralow power and high-density integrated devices [4–11]. Many features of the SET are discussed in many papers [12–15]. However, the research of SET able to operate at room temperature is not yet concluded, because observing tunneling phenomena of one single electron at room temperature is difficult. Although the word "SET" is put side by side with the word "room temperature", what the authors observed was a tunneling current formed by more than one electron [12–15]. Most of these papers are about SETs based on inorganic materials.

Typically single-electron tunneling devices operate in the full Coulomb blockade regime, where thermal energy can be neglected on electrical transport characteristics; at low-bias voltages current is blocked due to the charging energy of single electron. Otherwise, the weak Coulomb blockade operates in a different regime, where single-electron effects are still observed but temperature can influence the electrical transport characteristics [16]. Here, we present an approach to fabricate an organic three-terminal device—that we called organic transistor—based on MIM junctions that exhibit room temperature operation. The planar samples consist basically of three interconnected tunnel junctions each one formed by an insulating film of PMMA sandwiched between two electrodes of PEDOT/PSS assembled on polyester flexible substrate. The *I*–*V* characteristics of the fabricated device show an oscillatory variation of the drain current with the gate voltage at room temperature.

Experimental

The main feature of working with polymers is the fact that it does not require special conditions for preparation. The

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This manuscript is an expansion of the results presented in the work "Periodic current oscillations observed in organic MIM junctions" at ICAM 2009 in the Symposium Q "Materials and Process for Large-Area Electronics".

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PMMA is low molecular weight, $\rho = 1.188$ g/mL, acquired from Bayer Co. and was dissolved at room temperature in analytical reagent grade chloroform. The PEDOT/PSS is commercialized with the name Baytron® P obtained from Bayer (Germany). The planar samples consist basically of three interconnected MIM junctions each one formed by an insulating film of PMMA sandwiched between two electrodes of PEDOT/PSS. The polyester flexible substrate was used to assemble the sample and did not receive any special preparation. The bottom electrode and the insulating film were deposited by spin coating (6000 rpm) and the top electrode of the solution PEDOT/PSS was painted on the PMMA films as showed in Fig. 1. The thickness of the insulating layer of PMMA is 70 nm and this measure was taken by Dektak 150. The junction suitable for this work is large in area, typically 200 μm^2. The three junctions are identical and can be named drain, source, and gate as show in Fig. 2. Each junction is represented by its equivalent capacitance and the common terminal is the bottom layer of PEDOT/PSS.

Electrical measurements were taken at room temperature with a Keithley 2400 programmable semiconductor system with stainless needle. No variations were noticed due to the effect of light, and measurements were done in a lit room. The $I \times V$ curves presented here were obtained with 500-Hz ramps. Results have shown stability, having been repeated in measurements taken days apart with no special storing precautions. The capacitance per junction was measured directly on the sample with an Agilent 4284A precision LCR meter. The junctions presented individual capacitances of approximately 1.40 pF.

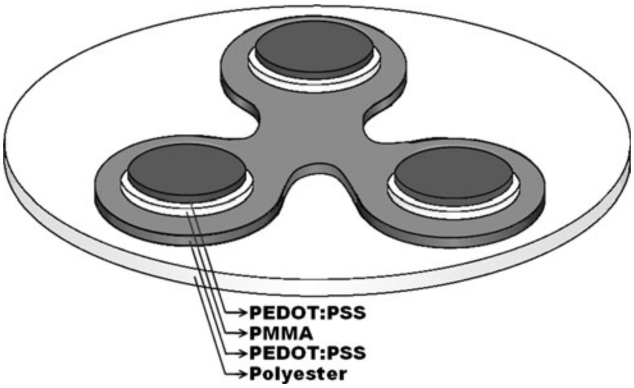

Fig. 1 Structure of the three planar junctions

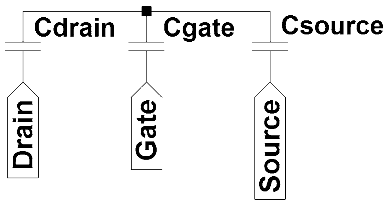

Fig. 2 Organic transistor equivalent circuit schematic

Results and discussion

The room temperature current–voltage (*I*–*V*) characteristic of the device with the source–drain voltage (V_{SD}) swept from −10.0 to +10.0 V is shown in Fig. 3. It can be seen from the non-linear I_D–V_{SD} characteristics of the device in Fig. 3 that the current remains constant for several voltage ranges. The drain current of the device is on the order of nanoamperes.

We can obtain from Fig. 3 the capacitance values for the three external junctions. In this device, we have $ne/2C_j = V$, where n is the number of electrons and C_j is the capacitance of the individual external junctions. The value of n is obtained by $n = \Delta q/e$, where $\Delta q = I\,\Delta t$. The current I is obtained from Fig. 3 and Δt is related to the frequency of the measurements, in this work 500 Hz. The capacitance individual junction thus obtained was $C_j = 1.25$ pF and it is in agreement with the directly measured values of 1.40 pF.

Figure 4 shows the room temperature current–voltage (*I*–*V*) characteristics of the device with V_G swept from −3.0 to 0 V for a fixed $V_{DS} = 50$ mV. Current oscillations with gate voltage V_G are visible for the applied source–drain voltage; the drain current, which is in nanoampere range, oscillates periodically with the increases of V_G.

Following the idea proposed by Nakazato et al. [16] for individual atomic crystal lattice, the charging energy will finally correspond to the Hubbard model [17]. These considerations immediately link electronic device miniaturization and single-electron effects to molecular electronics,

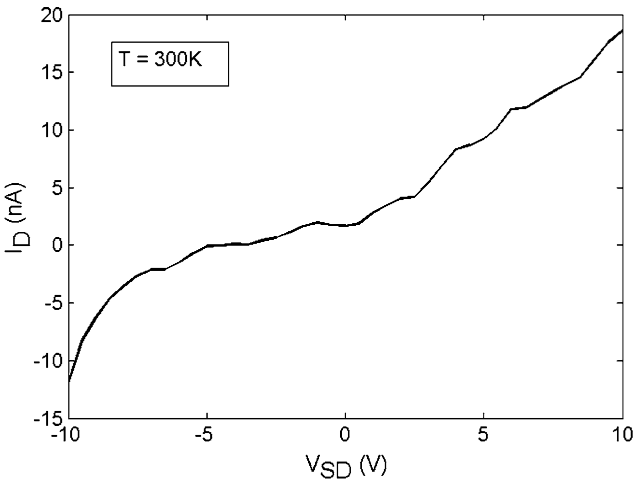

Fig. 3 Source–drain characteristic of the device at room temperature

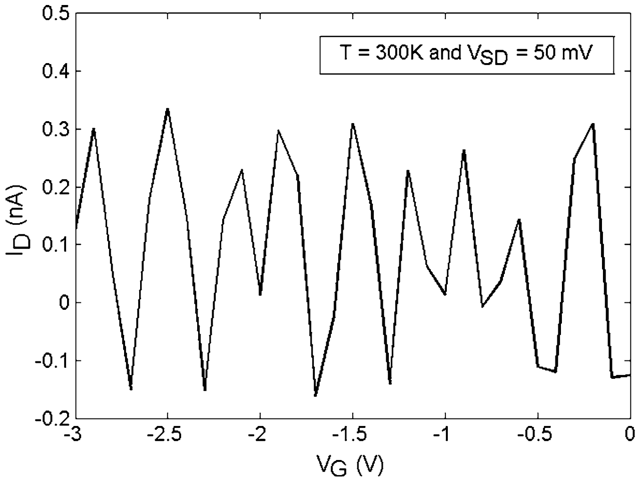

Fig. 4 Periodic current oscillations measured at room temperature for a source–drain voltage of 50 mV

and indeed, articles on "multiple tunnel junctions in GaAs" do not fail to point out that their ideas are also applicable to polymers [18, 19]. Taking that into account the observed periodic current oscillations can be compared to Coulomb oscillations in nanoscale devices. Specifically, the obtained results can be related to the weak Coulomb blockade which is noted at room temperature [19] as can be seen in Fig. 3. For a weak Coulomb blockade regime, K_BT is greater than E_C, where E_C is the charging energy of the system. The capacitance is proportional to the cross section of a particle and, in polymers several nanometers in diameter, the charging energy is comparable to the thermal energy at room temperature.

In a weak Coulomb blockade regime it is convenient to measure not the *I*–*V* directly, but the differential conductance, i.e., the slope of the *I*–*V* curve $G = \mathrm{d}I/\mathrm{d}V$ versus *V*. The result is nearly dip in conductance like shown in Fig. 5. In order to obtain the PEDOT/PSS capacitance we must determinate the number of internal junctions. In our case, considering a polymer structure we can possibly admit that we have a lot of internal junctions where the conduction mechanism is hopping (tunneling assisted by phonon). Here, we supposed that the PEDOT/PSS film consists of a N-junction symmetric linear array of series capacitance C as shown in Fig. 6. So, the full width at half minimum of the conductance dip has the value $V_{1/2} = 5.439\ NK_BT/ne$ [19]. G_V is the conductance at half minimum $V_{1/2}$ and G_T is the maximum conductance value. The depth of the dip $\Delta G_V/G_T$, where which, in the lowest order in E_C/K_BT is proportional to the inverse temperature $\Delta G_V/G_T = (E_C/3K_BT)[N - 1/N]$, where $N = 9 \times 10^7$ junctions. In all case we used $\Delta G_V/G_T = ½$, so the E_C value could be obtained. In implemented organic transistor $E_C = 78$ MeV.

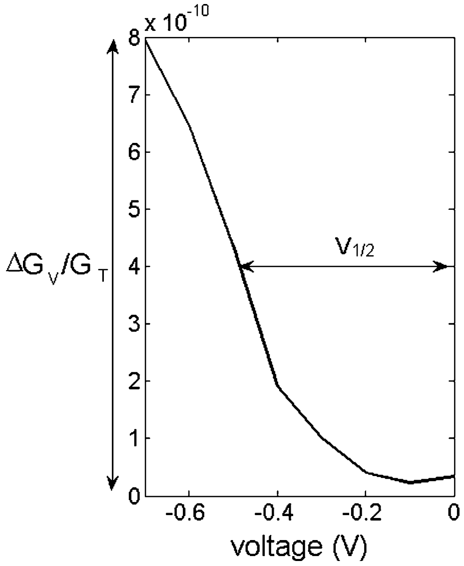

Fig. 5 The differential conductance as a function of bias voltage *V*

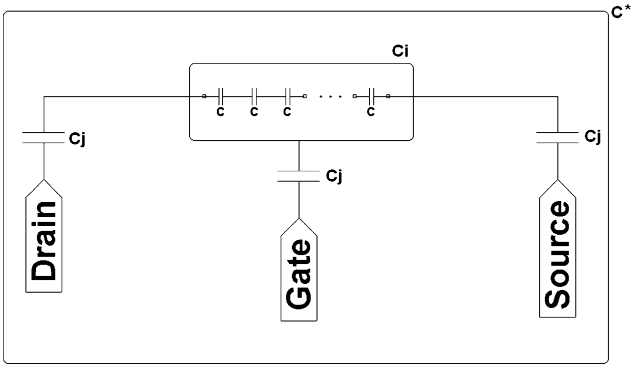

Fig. 6 The equivalent transistor circuit model

We could also determine the C^* equivalent capacitance $E_C = (ne)^2/2C^*$ where $C^* = 1/(1/C_j + 1/C_j + 1/C_i)$ and C_i is the PEDOT/PSS equivalent capacitance. With this value of C_i we can determinate the individual capacitance of the several junctions in the PEDOT/PSS film: $C_{ii} = 2C_i(N - 1)/N = 124$ pF.

Conclusions

An organic transistor organic using MIM junctions is investigated by *I*–*V* characteristics. The drain current oscillates periodically with the gate bias swept from −3.0 to 0 V. The conductance obtained is a function of the gate voltage. The device parameters were calculated by approximation from the measured data using "orthodox theory". The estimated E_C is higher than the room temperature thermal energy, resulting in the observation of periodic current oscillations at room temperature when compared to Coulomb oscillations. Based on these results we can see that many electrons are possibly tunneling through the PEDOT/PSS and the device consist of a multiple-tunnel junction structure.

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References

1. Durães JA, Araujo Sales MJ, Souto RF, Romariz A, da Costa JC, Ceschin AM, Moreira SGC (2006) Appl Phys Lett 89:133502
2. Bandeira MC, Guimarães JG, Durães JA, Silva LM, Ceschin AM, Sales MJ, da Costa JC (2007) ECS Trans 9(1):363
3. Fan X-L, Wang C, Yang D-L, Wan L-J, Bai C (2002) Chem Phys Lett 361:465
4. Majima Y, Azuma Y, Nagano K (2005) Appl Phys Lett 87: 163110
5. Armour AD, Blencowe MP, Zhang Y (2004) Phys Rev B 69:125313
6. Lahaye MD, Buu O, Camarota B, Shwab KC (2004) Science 304:74
7. Scheible DV, Blick RH (2004) Appl Phys Lett 84:4632
8. Nakajima A, Futatsugi T, Kosemura K, Fukano T, Yokoyama N (1997) Appl Phys Lett 71:353
9. Sakamoto T, Kawaura H, Baba T (1998) Appl Phys Lett 72:795
10. Augke R, Eberhardt W, Single C, Prins FE, Wharam DA, Kern DP (2000) Appl Phys Lett 76:2065
11. Tilke A, Blick RH, Lorenz H (2001) J Appl Phys 90:942
12. Takahashi Y, Ono Y, Fujiwara A, Inokawa H (2003) Phys E 19:95
13. So H-M, Kim J, Yun WS, Park JW, Kim J-J, Won D-J, Kang Y, Lee C (2003) Phys E 18:243
14. Azuma Y, Hatanaka T, Kanehara M, Teranishi T, Chorley S, Prance J, Smith CG, Majima Y (2007) Appl Phys Lett 91:053120
15. Karre PSK, Bergstrom PL, Mallick G, Karna SP (2007) J Appl Phys 102:024316
16. Nakazato K, Blaikie RJ, Leaver RJA, Ahmed H (1993) Adv Mater 5:668
17. Ding GH, Ye F, Xu BW (2003) Commun Theor Phys 39:105
18. Roth S, Carrol D (2004) One-dimensional metals, 2nd edn. Wiley-VCH, Weinheim
19. Pekola JP, Hirvi KP, Kauppinen JP, Paulanen MA (1994) Phys Rev Lett 73:2903