Self-assembly of an Organic Radical Thin Film and its Memory Function Investigated Using a Liquid Metal Electrode

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ABSTRACT

In this work, the deposition of a persistent organic radical by thermal evaporation on gold, platinum and graphene is performed. The impact of the deposition parameters and the nature of the substrate on the molecular organization within the deposited film are investigated. The non-planarity of the molecule and the role of the molecule-molecule and molecule-substrate interactions are discussed. Ultraviolet photoelectron spectroscopy experiments demonstrate that the radical character, and hence its magnetic and redox properties, is preserved on the three surfaces. The optimized films are electrically characterized by top-contacting the film/substrate system using a liquid metal which permits to achieve a soft-contact avoiding damaging the layer. The hysteretic current versus voltage curves obtained from the electrical characterization points to the potential applicability of the studied system as an organic memory. Moreover, the demonstrated feasibility of using a liquid metal is an appealing approach towards the preparation of flexible devices.

1. INTRODUCTION

The use of organic compounds for the fabrication of electronic devices has emerged as a very appealing alternative to the traditional inorganic counterparts. Employing molecular systems for device fabrication have key advantages like the reduced cost, the synthetic versatility and the compatibility with a vast range of substrates such as transparent glass and flexible polymeric materials.¹ In this field, the design and the synthesis of the desired organic molecule is the first crucial step of the fabrication process. The intrinsic molecular properties (electronic and magnetic) can be tuned by adequate molecular engineering. When these molecules are deposited as thin film, the chemical structure will have a huge impact on the molecular orientation within the layer and with respect to the substrate which will highly influence the device performance.²⁻⁴ The orientation of the molecules will depend on the interplay between the molecule-substrate and the molecule-molecule interactions. If the molecule-substrate interaction is strong, the molecules will tend to organize in a face-on fashion, but if the molecule-molecule interaction is predominant, the edge-on organization will usually prevail.⁵ The face-on orientation is expected to be favorable for the out-of-plane charge transport whereas the edge-on orientation favors the in-plane charge transport. One or the other is preferred depending on the targeted device, for example, face-on is expected to be advantageous for solar cells or light emitting diodes, and edge-on for organic field-effect transistors.

Post deposition treatment, such as temperature and/or solvent vapor annealing,^{6,7} to induce molecular reorganization is a common methodology to improve the device performance. Furthermore, controlling certain experimental parameters during the deposition or using substrates of different nature can be a strategy to induce a certain preferential organization of the

molecules.^{8–10} For instance, for the films obtained by thermal evaporation, the temperature of the substrate during the deposition process is playing a very critical role. A distinct induced growth mode, 2D or layer-by-layer or 3D island growth, at different substrate temperatures was observed for different molecules such as copper phthalocyanine¹¹, tetracene and pentacene¹² and perylene derivatives.¹³ Furthermore, the intrinsic surface energy of the substrate can directly affect the structure of the film. It was shown previously that the dielectric surface-energy strongly influences the pentacene film morphology and its growth mode.^{14,15} Traditionally SiO₂ and metals (Cu, Au, Ag, etc.) have been used for these studies but, due to the demonstrated and predicted appealing applications of graphene, the assembly of functional small molecules on it is currently being intensively investigated.¹⁶

By virtue of their open-shell character, stable organic radicals possess attractive optical, electrochemical and magnetic properties which make them fascinating potential building blocks for molecular electronic applications and the interest on these systems is continuously increasing.¹⁷ They have been employed as light emitting diodes,¹⁸ biological imaging agents,¹⁹ catalysts,²⁰ electrochemical materials for batteries,²¹ magnetic materials,^{17,22} as well as in the field of molecular spintronics at a more fundamental stage.^{23,24} The grafting and assembly of several stable organic radicals on surfaces has been achieved following different methodologies.²⁵ Among other organic radicals, the tris(2,4,6-trichlorophenyl)methyl (TTM[•]) radical (Figure 1a)²⁶ has received special attention due to its remarkable stability which is due to the steric hindrance that confer the six chlorine atoms around the trivalent carbon. TTM[•] is very sensitive to electron-transfer reactions, i.e. it can be easily reduced to anion and oxidized to cation. Although the electronic, magnetic and electroactive characterization of TTM[•] based monolayers was previously performed,^{27,28} the exploitation of these properties on few

nanometers thick films has not been investigated. The redox character offers the intriguing possibility of fabricating organic memories in metal-insulator-metal diode-type structures.

Herein, the surface organization of the TTM* is investigated on gold, platinum and graphene. The targeted thin films are deposited by thermal evaporation under high vacuum conditions. Further, the post self-assembly of the active layers is studied with time and its evolution is correlated with molecule geometry, molecule-molecule and molecule-substrate interactions. Upon the film optimization and characterization, the most promising hybrid TTM*/substrate system, in terms of roughness, is electrically characterized by measuring the out-of-plane conductivity using a liquid metal as top electrode (Figure 1b). The gallium-indium eutectic alloy (^EGaIn) was chosen for this purpose because it permits to achieve a soft contact with the organic layer,²⁹ is a non-toxic liquid-metal and it is coated with a 0.7 nm thin layer of conductive GaO_x, mainly Ga₂O₃,²⁹ which makes possible to shape the liquid drop into a tip with a minimum radius of about 20 μm. In the present work, the possible memory function promoted by the redox process that the TTM* can undergo is investigated by measuring the charge transport across the ^EGaIn/Ga₂O₃/ TTM*/substrate sandwiched type structure. Previously, ^EGaIn or Hg tips were employed for contacting mostly self-assembled monolayers,³⁰ and scarcely employed for other functional molecular.^{29,31–33} Here we take advantage of this liquid metal not only to avoid damaging of the layer but also to avoid the penetration of the top electrode within the organic layer. This is a very critical aspect, since it has been shown that during the evaporation of some metals on organic layers to fabricate the top contact, metal atoms can penetrate the layer which could then act as charge carrier trapping centers or damage the layer.³⁴ Furthermore, it is worth taking into account that using liquid metals opens the possibility to fabricate flexible devices^{35,36}

and, hence,, the approach used in this work can be considered as a test bed methodology for evaluating the materials of interest.

Figure 1. a) Molecular structure of the TTM• and scheme of its thermal evaporation on Au, Pt, and graphene. b) Scheme of the “E-GaIn technique” employed for contacting the TTM• film.

2. EXPERIMENTAL METHODS

Synthesis of the TTM• derivative. The TTM• radical was synthesized as previously reported.²⁶

Sample preparation. Three different conductive substrates working as bottom electrode were used in this work: gold, platinum and graphene. The gold was template-stripped³⁷ to obtain a flat gold layer on glass, a platinum layer of 100 nm thick were deposited by thermally evaporation on SiO₂ surface, and finally the graphene (on quartz) was commercially purchased from Graphenea and used without further treatment. Monolayer graphene on Quartz (fully covered) was produced by CVD and transferred to a circular substrate of Quartz (500 μm thickness) by a wet transfer process. TTM• was deposited by thermal evaporation at rate of 0.04 $\text{\AA}/\text{s}$ and at pressure lower than 10^{-6} mbar on gold, platinum and graphene substrates (vacuum pressure decreases slightly $\sim 10\%$ during the deposition). The substrate temperature was set during evaporation at 25, 47 and

60 °C. Higher temperatures were not investigated to avoid material re-evaporation from the substrate.

As a general procedure for the three substrates, they were introduced in the evaporator chamber at least 15 hours previous to the deposition. TTM^{*} starts to evaporate at 80 °C and in order to keep controlled the evaporation, temperature was set above this temperature but in any case it did not overcome 105 °C.

The evaporation rate as measured by quartz crystal monitor was 0.04 Å/s. The substrates were maintained at room temperature (RT/25 °C), 47 °C and 60 °C during the deposition of TTM^{*}.

Surface morphology. The surface morphology of TTM^{*} films were investigated using scanning electron microscopy (SEM) with a Quanta FEI 200 FEG-ESEM and atomic force microscopy (AFM), with a Keysight 5100 working in tapping mode.

Structural and Electronic characterization of the film. X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) measurements were performed with a Phoibos 150 analyzer (SPECS GmbH, Germany) instrument with mono- chromatic AlK α X-ray (1486.74 eV) and HeI UV (21.2 eV) sources under ultra-high vacuum conditions (base pressure 3×10^{-10} mbar). The X-ray radiation is at an angle of 55° to the sample with a spot size of 3.5 x 0.5 mm in ellipsoidal arrangement. A pass energy of 50 eV, a dwell time of 500 ms, and an energy step size of 1 eV were employed for the general scan. For specific element scanning, pass energy of 20 eV and step size of 0.05 eV were used. The binding energies were calibrated with respect to Au 4f_{7/2} at 84.0 eV for TTM^{*}/Au, Pt 4f_{7/2} at 71.0eV for TTM^{*}/Pt and C1s at 284.8 eV for TTM^{*}/Gr-q (CVD graphene on quartz). The sample for UPS was held at an angle of 50° with respect to the UV radiation and the spot size was 0.5 mm diameter. The overall resolution for XPS, as

measured by the full width at half-maximum intensity (FWHM) of the Ag 3d_{5/2} peak for a sputtered silver foil, is 0.6 eV and for UPS as measured by FWHM of Fermi level is 0.14 eV.

Electrical characterization of the TTM[•] thin films. A ^EGaIn drop hanging from a Hamilton syringe is molded in the form of cone with a tip of approximately 20 μm of diameter. The ^EGaIn cone, working as top-electrode, is biased while the bottom electrode is grounded. The junctions are formed when the ^EGaIn cone contact with the sample surface. An electrometer Keithley 2604B was used to perform the electrical measurements. For each measured junction, 12 traces were acquired in the low voltage regime (<|±1.5 V|) and 6 traces when the voltage range was higher (>|±1.5 V|). In each trace the voltage is swept from 0 V to the maximum voltage, then towards the minimum voltage applied and finally returns to 0 V. The integration time of the measured current was 250 ms and the waiting time between two consecutive points (voltage steps) was 100 ms.

3. RESULTS AND DISCUSSION

3.1 Surface growth mode and self-assembly on gold, platinum and graphene. TTM[•] was thermally evaporated under high vacuum conditions. First, the substrate temperature induced growth mode on gold was investigated, and secondly, keeping the temperature as a constant parameter the influence of the substrate nature was explored. The deposition and organization of the TTM[•] molecules are studied on gold, platinum and graphene substrates. In this study, the non-planarity of the TTM[•] molecules is expected to have an important impact on the obtained structures.

Different deposition conditions were tested to prepare TTM[•] films with smooth morphology and low roughness, which is aimed for practical applications. First, the deposition was performed

keeping the substrate at different temperatures, 25 °C (RT), 47 °C and 60 °C, during the evaporation. The optical images of the TTM[•] on Au are depicted in Figure 2a-c and clearly show that the film morphology is greatly affected by the substrate temperature. The films obtained keeping the substrate temperature at 25 °C and the ones obtained at 47 °C show some aggregates with a dendritic structure which are rarely present when the substrate temperature is 60 °C. For more clarity see Figure S1a-b in the Supporting Information (SI). The roughness differences of the non-dendritic regions are much evident in the AFM images (Figure 2d-f). The roughness of the film is here determined by the root mean square value (R_{ms}/nm) (extracted from an area of $5 \times 5 \mu\text{m}^2$) which decreases from 36 nm to 1.6 nm and finally to 0.6 nm at 25, 47 and 60 °C, respectively (Figure S2). The thickness of the film prepared at 60 °C was measured to be 22 nm (Figures S3). Several attempts to obtain thinner films were performed unsuccessfully (see Figure S4 in the SI). The AFM images, shown in Figure 2d-f, correspond to the areas far from the aggregates observed in Figure 2a-b. Distinctly, the images illustrate that at RT the TTM[•] has a lower interaction with the Au substrate and that the intermolecular interactions between TTM[•] dominate the observed structures. At higher substrate temperatures the molecule-molecule interactions become weaker and the formation of a smoother film is promoted. These results are in agreement with a previous work where a polychlorinated triphenylmethyl (PTM) radical derivative was deposited on Au (111) and Ag (111) and characterized by several spectroscopic techniques.³⁸ The optimized configuration by Density Functional Theory calculations corroborated that the radical molecules were at certain tilting angle with respect to the gold surface.

Figure 2. Microscope optical images of TTM* films thermally evaporated on Au^{TS} substrates kept at a) 25 °C b) 47 °C and c) 60 °C. The scale bar is 100 μm. The corresponding AFM topographical images of the samples d) 25 °C b) 47 °C and f) 60 °C. The AFM images were acquired on the film areas without the presence of the aggregates observed in the corresponding microscope optimal image (a, b and c).

It is well-known that the surface energy (γ_s) can have an impact on the growth mode of the film.^{14,15} Taking this into account, and to go further on the TTM* surface organization investigation, besides gold, two additional substrates were chosen, platinum (Pt) and Chemically Vapor Deposited (CVD) graphene transferred on quartz (Gr-q). The reported γ_s values are 2.48 and 0.115^(ref.39) J/m² respectively. This value is reported for CVD grown-graphene on copper. The γ_s of gold is 1.50 J/m² ^(ref40) which lies between the values reported for Pt and CVD-graphene. For these experiments the temperature of the substrate was kept at 60 °C. Graphene has been chosen for its low γ_s as well as for its relevance in the development of graphene-based

organic devices.¹⁶ Also, it is known and expected that the interaction of organic molecules between metals and carbon based substrates is drastically different.

As observed in the tapping mode AFM topographic images and in the SEM images shown in Figure 3, larger aggregates are formed when the TTM• is evaporated on Gr-q at 60 °C. That is, taking into account the non-planarity of the molecule and the lower surface energy a more 3D-growth mode is produced. These aggregates have a clear tendency to form fibrillar like structures with a height of 300-600 nm and a length of around 2-5 μm . The AFM image depicted in Figure 3a corresponds to some isolated fibers at the edge of the film (see SI, Figure S5 for more AFM images of TTM• film on graphene and height profile). The film morphology grown on Pt is similar to the one on gold (see SI, Figure S6 for optical microscope images and AFM images of the TTM• film on Pt).

Figure 3. a) Topographic AFM images and b) SEM images of the TTM• aggregates forming the evaporated film on CVD graphene/quartz.

From these experiments it seems that the γ_s difference between Au and Pt does not have a huge impact on the morphology of the resulting film and its metallic nature is dominating the film growth, but when this value is largely decreased by employing graphene as substrate, the

influence is definitely observed leading to the formation of TTM[•] radical ordered structures due to a stronger molecule-molecule interaction versus a lower molecule-substrate interaction. These results are in agreement with previous studies, where it was observed that the perchlorinated trityl radicals formed well-ordered 3D structures on graphite sustained by intermolecular interactions such as Cl...Cl and Cl...Ph.⁴¹

3.2 Defects as seeds for molecular self-reorganization of the TTM[•] film. After 24 hours at room temperature, the film morphology has completely changed compared to the initial one, from a smooth amorphous and homogenous film to a film composed by crystalline fibers. It has been observed that, when some defects are present in the film after the evaporation, they clearly serve as nucleation points to promote the growth of the TTM[•] fibers (see SI, S7 and S8), which can be accelerated by annealing the sample at relatively low temperatures (70 °C for 30 minutes) (Figure S8). The same phenomenon is observed for the films grown on Pt (See SI, Figure S9). In addition to the strong intermolecular interactions, the non-planar geometry of the TTM[•] can certainly favor a higher mobility of the deposited molecules on the surfaces due to the weak molecule-substrate interaction. Based on this observation and, taking into account the precedents on the self-assembly of these radicals, we have intentionally introduced defects in films obtained at 60 °C as nucleation points in order to promote the self-reorganization and crystallization of the amorphous film to lead to the most stable TTM[•] self-assembly. As shown in Figure 4, after scratching the film, this serves as a nucleation seed for the crystallization of the film. The topographical seeding of crystallization has been reported as a methodology for the coherent and directional crystallization of surrounding amorphous material.⁴² As expected, there are no observable modifications with time for the films prepared on graphene (See SI, Figure S10).

Figure 4. a) TTM[•] film on Au after 50 minutes of its evaporation at 60 °C. b) After 6 hours of introducing the defect in the film and c) after 24 hours.

3.3 X-ray and Ultraviolet photoelectron spectroscopy (XPS and UPS) characterization.

TTM[•]/Au, TTM[•]/Pt and TTM[•]/Gr-q were characterized by XPS and UPS. XPS survey spectra were collected for the three hybrid systems (Figure S11). As a most representative element for the TTM[•] films, Figure 5a shows the high resolution XPS spectra for Cl2p for the three modified surfaces. The doublet expected for the Cl2p_{3/2} and Cl2p_{1/2} (at lower and higher binding energy (B.E.), respectively) which is characteristic of the deposited polychlorinated radical is observed for the three samples. A shift of the Cl 2p_{3/2} peak towards lower B.E. from graphene and Au to Pt (200.5, 200.5 and 200.2 eV, respectively) is observed. Although being small (0.3 eV) the shift direction is in accordance with the increasing surface energy.

Importantly, the presence of the peak corresponding to the highest singly occupied molecular orbital (SOMO) observed in the UPS spectra of the three systems demonstrates that the radical character of the TTM[•] derivative is preserved after the evaporation. It can be noticed that SOMO level is placed around ≈ 1.9 eV below the Fermi energy in the case of Au and Gr-q while in the case of Pt, the SOMO orbital lies ≈ 1.6 eV below the Fermi energy (Figure 5b). These values are in agreement with previous results.^{27,28,43} Furthermore, the resulting work function (WF) of the

TTM[•]/substrate system was determined to be 4.4 eV for Au and Gr-q and 4.7 eV for Pt as substrate.

Figure 5. a) High resolution Cl₂p XPS spectra for the three hybrid systems (doublet corresponding to Cl₂p_{3/2} and Cl₂p_{1/2} at higher and lower binding energy, respectively). b) UPS spectra of the three samples.

3.4 Electrical characterization. As mentioned above, the ^EGaIn was employed as top electrode to evaluate the electrical properties of the vertically structured ^EGaIn/Ga₂O₃//TTM[•]/substrate device. Taking into consideration the observed self-reorganization of the TTM[•] films with time explained above, the density current *versus* the applied potential (*J-V*) curves were always acquired immediately after the evaporation of the TTM[•] samples. Smooth homogenous morphology, uniform coverage over a relatively large surface area and a few nanometers thick film are desired to fabricate the device. As seen above, this is the case for the film prepared on Au and Pt. For TTM[•]/Gr-q, the tendency of the TTM[•] molecules to form fibers led to the formation of an incomplete layer on the substrate. Due to the minimum size of the ^EGaIn electrode (around 20 μm), in most of the cases it was difficult to understand the role of the organic layer in the device performance in front of the direct contact ^EGaIn/Ga₂O₃/Gr-q. For this

we could not perform a representative statistical analysis of the electrical measurements and, hence, they were not conclusive and are not discussed here.

Figure 6a shows the mean values of the J in the voltage range of ± 1.5 V for the ${}^E\text{GaIn}/\text{Ga}_2\text{O}_3//\text{TTM}^*/\text{Au}$ and Pt. In these experiments the bottom electrode is grounded. The error bar corresponds to the standard deviation of the distribution. The statistical analysis was performed on 19 robust junctions in the case of Au and 21 in the case of Pt. The yield of successful junctions formed was 90 % for both substrates. In this operation voltage we did not observe any significant hysteretic effect upon sweeping the bias. But, it is remarkable that a significant current rectification is observed. At negative voltages the J values for both systems are very similar but at positive voltages the J values for the TTM^*/Au are higher and, this difference is enlarged when increasing the applied bias. This difference is clearly depicted in the box plot shown in Figure 6b. Figure 6c shows the histograms of the rectification ratio values (R , $R=J(+1.5\text{V})/J(-1.5\text{V})$) obtained for each J - V curve. The rectification mean value for the TTM^* on Pt is 28 ($10^{1.45}$) and in the case of the TTM^* on Au is more than six times higher, i.e. 177 ($10^{2.25}$). Such a statistical difference in J at positive voltages between the two different systems (${}^E\text{GaIn}/\text{Ga}_2\text{O}_3//\text{TTM}^*/\text{Au}$ and ${}^E\text{GaIn}/\text{Ga}_2\text{O}_3//\text{TTM}^*/\text{Pt}$) suggests that the transport is mainly governed by an injection-limited conduction mechanism, although a certain contribution of Poole-Frenkel type transport mechanism, i.e. bulk controlled, cannot be completely discarded for a 20 nm thick film.⁴⁴ When a positive voltage is applied, electrons are injected from the metal substrate (Au or Pt) into the TTM^* . In accordance, in the case of Au the charge injection barrier must be lower than in the case of Pt. Here a deeper work function implies a higher electron injection barrier to the lowest singly unoccupied molecular orbital (SUMO) which is translated in a lower measured J . The SOMO-SUMO gap in these type of radicals has been measured and

theoretically calculated to be around 2.0 eV.⁴⁵ On the other hand, when a negative voltage is applied there is no statistical difference in J because electrons are injected from the ^EGaIn electrode (WF = 4.2 eV)³⁰ into the TTM*. This is further supported by the experiments performed changing the polarity of the electrodes, since then the current rectification is inverted, i.e., a higher J is measured at negative bias (see Figure S12). Supplementary detailed study of the charge transport mechanism such as thickness dependent and/or temperature dependent measurements were not considered to be plausible here, because the film may suffer a morphology modification in the elapse time of the data acquisition.

Figure 6. a) Mean values of the density current as a function of applied voltage. b) Box plot comparing the distribution of J at -1.5 and +1.5 V. c) Histogram of the rectification ratio obtained for all the J - V acquired.

To further explore the TTM[•] functionality in this type of junction, electrical measurements at higher applied voltages were performed with the TTM[•]/Au system (Figure 7). Interestingly, when the applied potential window is extended to ± 2.5 V hysteretic J - V curves were obtained. The graph shown in Figure 7 corresponds to a single junction that is representative of the film behavior. In each junction, 6 J - V curves were acquired. According to the definition of ON as the more conductive state and OFF the lower conducting state, the ON/OFF ratio reach values of hundreds at -1.2 V as can be observed in the inset of Figure 7. Although better performances have been found for other systems such as the nitronyl-nitroxide radicals⁴⁶ radical-polymer based,⁴⁷ and organic polyradicals,⁴⁸ the ratio measured here for the TTM[•]/Au system is large enough to be exploited to read the state of the memory. It is striking that the measured ON/OFF ratio is one order of magnitude higher at negative voltage than at positive voltages and that the rectification observed in Figure 6a vanishes when the devices are swept to -2.5 V. The redox process, i.e. the charging-discharging process, that the TTM[•] radical can undergo,⁴⁹⁻⁵¹ in addition to some charge trapping phenomena at the interfaces or at the grain boundaries of the film are attributed here to be the responsible for the observed hysteresis. It is important to highlight that thanks to the soft contact offered by the liquid metal with the film, the observed hysteretic behavior is ascribed to the intrinsic properties of the organic film and not to other contributions such as those coming from the penetration of metallic atoms or filaments into the film.^{50,51}

Figure 7. Representative plot for the J - V curves recorded for the junction GaIn/Ga₂O₃//TTM[•]/Au. Inset: Plot of the ON/OFF ratio extracted from the hysteretic curves in the applied voltage range of ± 2 V. In this plot the ON state is the retrace curve (from + 2.5 V to - 2.5 V) and the OFF state is the trace curve (from - 2.5 V to + 2.5 V).

4. CONCLUSIONS

In summary, in this work we report the preparation of a thin film based on a non-planar persistent organic radical by thermal evaporation. In the case of gold and platinum, by increasing the substrate temperature during the evaporation, molecule-substrate interactions dominate and lead to the formation of low rough films over a large area with a thickness of ~ 20 nm. In the case of employing CVD graphene on quartz, molecule-substrate interactions become weaker and the TTM[•] radical molecules tend to be in a standing-up orientation forming fibrillar structures. It has been observed that defects in the smooth films on gold and platinum serve as seeds for the

crystallization of the film. The liquid metal gallium-indium eutectic alloy (^EGaIn) electrode is used to softly contact the layer and to electrically characterize the film. This helps to avoid the penetration of the metallic top electrode in case of being evaporated. The electrode is used in the shape of a cone with a minimum contact area of 300 μm^2 . Interestingly, it is observed that in the case of the TTM[•] film prepared on gold and platinum, in the applied voltage range of ± 1.5 V, a current rectification is observed at positive voltages. Furthermore, it is striking that when the voltage is swept between ± 2.5 V hysteretic curves are measured with maximum ON/OFF current ratio over 100. This phenomenon is attributed to the charging/discharging of the TTM[•] and/or to the charge trapping at the metal/TTM[•] interface. We believe that these findings contribute to reinforce and to highlight the potential of using persistent organic radicals as electrical active material in thin films in vertical structured molecular electronic devices. Based on these results, molecular systems containing polychlorinated triphenylmethyl radicals that could permit to obtain non-crystalline films will be investigated in the future. Furthermore, the use of liquid metals is a very attractive alternative for fabricating flexible memory devices with organic materials.

ASSOCIATED CONTENT

Supporting Information. Additional optical microscope and atomic force microscopy images and further electronic and electrical characterization of the films are included in the Supporting Information.

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