

*Research Review*

# ELECTROCHEMICAL SYSTEMS AND MECHANICALLY INTERLOCKED MOLECULES. PART 2: NANO-TECHNOLOGY

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## ABSTRACT

Due to the difficulties encountered by moletronics in the realization of viable microcircuits based on microconductive bridges and nanoelectrodes, the current research aims to produce controlled deposited graphene microcircuits, in the form of lamellar multilayer, quantum coupled and functionalized with molecular machines, matrix configurations, which is an innovative approach compared to the present transversal heterojunction. From this assembled system, the resulted quantum clusters can stock the molecular energy using local effects, specific for spin quantum electrochemistry, and by photo-activation of integrated micro-circuit with logic gates in the form of deposited quantum graphene clusters, potential and quantum efficiency was investigated.

**Keywords:** spin quantum electrochemistry, graphene, molecular machines, graphenetrionics, logic gates

## 1. INTRODUCTION

Starting with the Nobel Prize for Graphene, latest studies regarding graphene materials are established on creative, innovative, disruptive dynamic technology [1]. These are directed on developing original producing methods [2-4], including nano-system topology, through the fundamental quantum mechanism of generating Stone-Wales defects on graphene [5], an essential effect both in the phenomena of magnetization to which graphene can participate, but also in its superconducting deca-nano-conductive functions [6].

According to the “Lemma of New Technology” stated by Herbert Kroemer, awarded in 2000 with the Nobel Prize in Physics for Pioneering Work in Information Technology and

Communication, it seems that “applications of any sufficiently new and innovative technology always have been – and will continue to be – applications created by that technology”. This postulate could be applied to the later discovery of graphene as a new state of matter (Nobel Prize in Physics 2010 awarded to Andre Geim and Konstantin Novoselov “for groundbreaking experiments regarding the two-dimensional material graphene”). From this point of view, graphene is still considered a rising star in the science of materials, with a strictly bi-dimensional nature (2D) and particularly efficient power and heat conduction ability. Consequently, graphene no longer requires further evidence of its importance in terms of physical fundamentals, because it “is probably the only system where ideas from quantum field theory can lead to patentable innovations“ – as Frank Wilczek said (awarded with Nobel Prize in Physics in 2004 for “the discovery of asymptotic freedom in the theory of the strong interaction”). From this point starts the possibility for the creation of smart material applications with nano-ecological energy functions in photovoltaic systems, with the aim to enhance the conversion of the sunlight energy into green electricity, based on its properties and a thin structure composed from a single layer of carbon atoms. Besides this remarkable ability, graphene structure could also be outstanding support for the concept of quantum-particle wave function/chemical bond field, bondon, in chemical bond field modeling. In its turn, is a boson, bondon can be associated in clusters, delocalized quantum dots, called bondots (bonding of quantum dots) – particles that describe the coherent links of quantum points occurring in condensed matter, including graphene. Since quantum dots (QD) have already proven their usefulness for solar cells through their sensitization potential, the present multi/matrices (bondotic) quantum dot – MQD approach, can generate a new production of amplification of light, this time at the photovoltaic level, promoting a new generation of solar cells of energy materials working on such a mechanism.

The Graphene study is very advanced in developed economies, based on a creative, innovative, disruptive dynamic technology. There is a comprehensive review in 2015 – with a projection for 2030 of graphene science and technology pioneers, including Nobel Prize winners for Graphene [1]. Also, there is the author’s series with the reputed Prof. Franco Cataldo, one of Italy’s few leading contractor manufacturers of graphene, developing an original method of producing fullerene base graphene [2-4]. Also, there is the author’s series with Dr. Ottorino Ori, a pioneer in the fullerene nano-systems topology, by the significant article on the quantum mechanism of generating Stone-Wales defects on graphene [5]. They are essential both in the magnetization phenomena to which graphene can participate, but also for its function conductive, superconducting [6], semiconductor, hole (h) or electron (e) deca-nano-systems, the base for the graphene quantum transistor.

Instead, molecular machines, rotaxanes, and catenanes have been designed and synthesized as dynamic systems stabilized by non-covalent bonds/interactions between components that contain complementary recognition centers [7-14]. In particular, rotaxanes are prototypes suitable to the construction of linear and rotational molecular machines, with the typical development of molecular shuttles allowing translational movements of rotating rings [15,16]. These types of molecular machines are dynamic, stable structures with functional integration capability in multicomponent systems [17,18], so also on/with graphene (even if this system is not yet studied). However, self-electrochemistry of molecular machines opens opportunities for what can be called (quantum) logic gates; thus, Credi et al. [19] proposed a structure of a pseudorotaxane formed in solution by the self-assembling of the 2,7-dibenzylidiazapyrene dication (as a wire type electron acceptor) and 2,3-dinaphtho-30-crown-10 (as an electron donor macrocycle). The compound may be unthreaded or rethreaded by chemical inputs, for example, the unthreaded process involves the addition of stoichiometric amounts of acids or amines, while the amine addition can achieve rethreaded, and vice versa. These threading/unthreading processes can be monitored taking into account

the essential changes in the fluorescence properties of the system. By two sets of different species with common and specific traits, two distinct and reversible unthreading/rethreading cycles can be achieved. The system can be considered as an XOR logic gate because of the system characteristics: input – chemical and output – fluorescence.

In Romania, studies about and with graphene, including its production in the laboratory, are becoming more common (including INCEMC-Timisoara) [20], but especially for its applications based on high electrical and thermal conductivity. In contrast, in terms of molecular machines, we have no knowledge of Romanian groups and researches – apart from those published by some of the authors of this paper – co-authors with Professor Margherita Venturi [21] (recognized co-worker of Nobel laureate for molecular machines, Sir J. Fraser Stoddart [22]) with the best intention to identify and collaborate with the active and potential national partners.

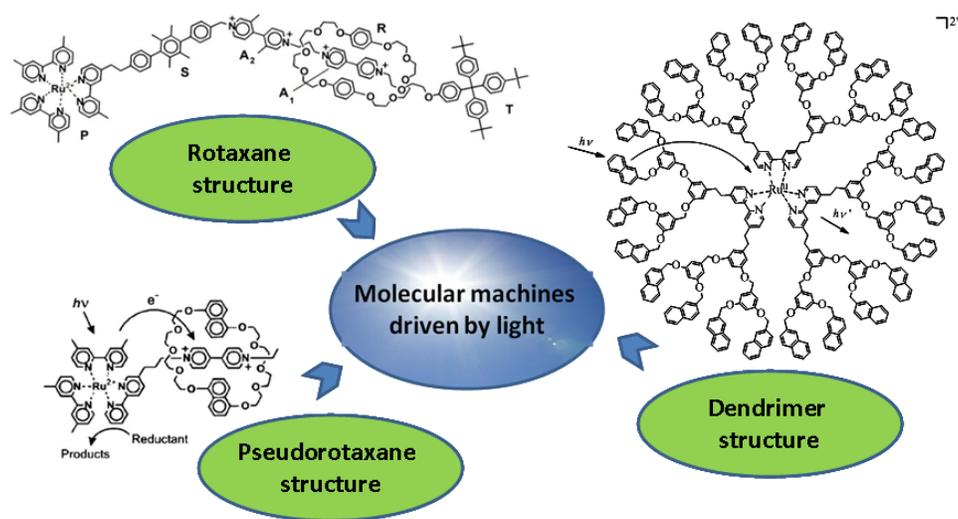
The present paper, advances the combined graphene + molecular machine system with the applicative research of photo-energy generated and stored by the resulting deca-nano-system, by electro-quantum spin activation, as follows.

## 2. METHODS AND MODELS

From this point, the present paper proposes the systematic study of graphenetrionics through innovative, theoretical and experimental methods, with a high potential to create a new solar cell prototype (based on the quantum deca-nano-transistor/semiconductor). Also the paper advance the graphene-integrated circuits, functionalized with photoactivated, spin-coupled molecular machines, arranged as a matrix in a controlled manner, which facilitates redox phenomena by quantum tunneling (spin quantum electrochemistry).

The proposed research is divided into the following stages regarding the design, testing and calibration of nano-systems with graphene with photoelectric energy conversion function and their storage in molecular machines/motors:

1. **Graphenetrionic Engineering** (concerning *Electronic algebra on Graphene*, and *Graphenic Matrix Heterojunctions*) aimed to guide the modeling and experimental observation of the “green” over-current (on graphene support) with the help of graphenetrionics, through innovative matrix heterojunction configurations.
2. **Molecular Machines with Photoactivated Graphene** (concerning *The second kind Storage of Energy for Molecular Machines with Graphene*, and *Electrochemical Reactivity with Spin Molecular Machines*) regarding the storage of the molecular current (over-micro-current) with molecular machines (Figure 1) functionalized on graphene support.
3. **Graphene Quantum Electrochemistry** (concerning *Nano-Imaging Control of Quantum Tunneling on Multilayer Graphene*, and *Electrochemical Quantum Metrology*) controlling the electrochemical flow with defective graphene in quantum logic gates configurations.
4. **Spin Density Photoactivation on Semiconducting Graphene** (concerning *Activation of Quantum Logic Gates on Defective Graphene*, and *Superconductivity of Magnetized Deca-Nano Graphene Semiconductor*) controlling the electrochemical photo-current with the aid of defective and spin coupled (magnetized) graphene in matrix configurations such as quantum logic gates.



**Figure 1.** Fundamental types of molecular machines, self-electrochemical structures under the action of light [23]

Regarding all the previously described statements, this research also contributes significantly to the present and future sustainability of Econo-ecological nano-energy efficiency as follows:

- ✓ Scientifically – by advancing a mechanism (graphene + molecular machines + bondonic and bondotic photoactivation) of energy generation and storage at the nanoscopic level (smart bottom-up approach);
- ✓ Technologically – by advancing a prototype of photovoltaic energy storage on graphene functionalized with molecular machines, respectively replacing silicon-based integrated circuits with other technologies based on graphene functionalized with molecular motor capacitors, that exhibit magnetic response, with a spin coupling on matrix depositions (the effective top-down approach);
- ✓ Socio-economic – by creating and storing green, non-toxic, carbon-based (graphene) energy with electrochemical control and autonomy (viz. molecular machines), with the perspective of technological transfer and leap from the scientific-technological disruption to innovation of devices and accessories (including household, but also medical, educational, info-communication, encryption and cyber-security) based on solar cells and photoactivated integrated circuits (sustainable multi-stakeholder approach).

## 2.1. Conceptual Approach

Graphene has already entered in the leading scientific flow as a support for both fundamental research (band structure, heterojunctions) and applicative research (photovoltaic, electrochemical effects, graphene environment both as an electrode and sensitization medium, replacing, at least from a non-toxic perspective, the generation of solar cells based on organic sensitizers and dyes). On the other hand, molecular machines, although very recently recognized as producing the smallest autonomous systems at the molecular, nanoscopic level, are barely at the stage of synthesis and observation; however, they can control the auto-oxidation-reduction mechanism under the influence of light. Including both, graphene and molecular

machines, the present research highlights two significant debuts. First of all is the functionalization of molecular machines supported on graphene, as a premise of a combined deca-nano-system that can generate (by graphene) + storage (by molecular machines) photo-energy produced by light activation or spin coupling (magnetization), or by both conjugated mechanisms (simultaneous or stepwise). The other is the phenomenological combination in various systems acting as quantum logic gates (on/off by electronic tunneling at mono-atom or mono-ion level charge carrier from the system). Graphene functionalized with Molecular Machines Integrated Circuits (acting as molecular capacitors, based on self-electrochemical processes) in matrix-designed configurations will substitute the Silicon Integrated Circuit Technology. Also, they will replace the bi- and multi-graphenic cross heterojunctions (staking type), attributed to the van der Waals interaction and hence Coulomb blockade, acting as electronic transfer speed limiters.

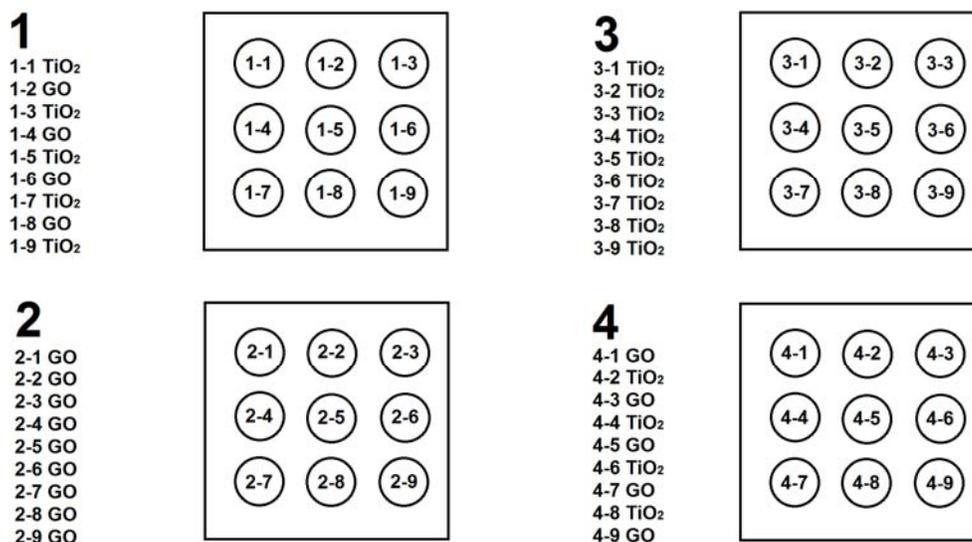
As a result, it can define (and possibly impose, through this research), the new electronic approach (including integrated circuits) with graphene belongs to GRAPHENETRONICS, and by functionalization with molecular machines and matrix combinations with them, having an electronic control mean – SPIN QUANTUM ELECTROCHEMISTRY.

## **2.2. Experimental Idea of Graphenetrionic Engineering. *Electronic Algebra on Graphene***

The deposition of the selected materials (such as graphene oxide systems GO and TiO<sub>2</sub>) was carried out by drop casting (Figure 2) as follows:

- Several dripping methods were tested to obtain the smallest deposition diameter (using, for example, needle, pen, and micropipette). All preliminary tests were applied on the glass, and only the most regular shaped depositions were optimized.
- The second step consisted in depositing GO and TiO<sub>2</sub> on ITO as presented in the Figure 2.
- The diameter and distances between the droplets were determined using the microscope (Olympus BX51 halogen lighting system – 100W, focusing capability of 0.1 mm = focus sensitivity with Nikon LU Plan Fluor 10x/0.30 A, OFN25 WD 17.5 lens).
- Next step, alternative graphene/oxide configurations will be realized, aiming to create a matrix of periodical deposits, bi- or multi-component, to replace cross heterojunctions. These will be further studied, concerning quantum contacts, short circuit, open circuit, and photo-activation, with selection and comparison of response signals.

The unique/distinct responses in the electro-/photo-current recorded on the matrix plates will be associated with nano-energetic conversion matrices, precursors of the graphene integrated circuits.



**Figure 2.** Matrix positions of GO / TiO<sub>2</sub> depositions on ITO glass

### 3. FIRST RESULTS AND DISCUSSIONS

Determination of coefficients by measurement, comparison, interpolation, metrological calibration (with Hall, Josephson, and tunneling quantum effects) combines the algorithm for each matrix multi-graphene deposition and provides the basis for the projection and realization of the quantum transistor with photo-activated graphene.

#### 3.1. Graphenetronic Engineering. *Graphene Matrix Heterojunctions*

Transforming the response of electro-voltaic matrices for the graphene/oxide deposits from the previous phase into the quantum matrices associated with the applied potential  $V(x, y, z)$ , through the densities of generated states  $\rho_n(x, y, z)$ , to a given (or varied) thermal energy according to Boltzmann's relationship  $\beta = 1/Tk_B$ , with the observation of the current density provided by the Eq.(1):

$$j_n[V(x, y, z)] = (\eta\beta V(x, y, z))^{-1} \iiint \rho_n(x, y, z) V(x, y, z) dx dy dz \quad (1)$$

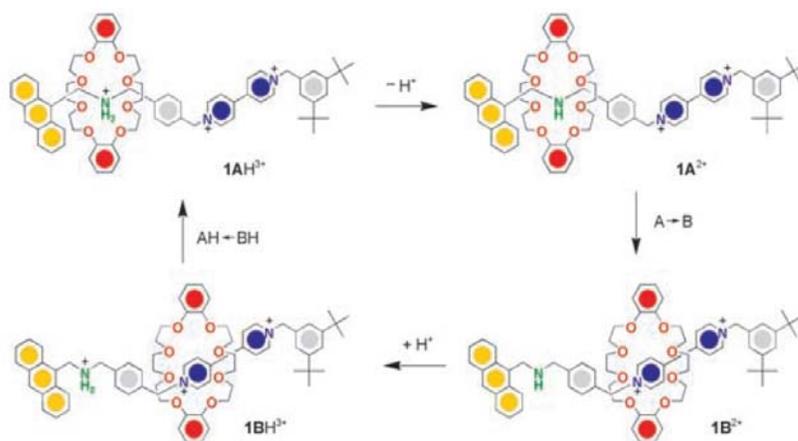
The quantum supercurrent identified corresponds to the critical temperature for which the electronic fermion spin (fermion electron)  $\rho_{Fermi-Dirac}(x, y, z)$  passes into the bosonic states of the bondons  $\rho_{Bosonic-Bondon}(x, y, z)$ , with the identification of the respective boson accumulation by clusters identified in Atomic Force Microscopy (Figure 3) for different graphene structures (and origins).



**Figure 3.** Phase transition on combined fullerene graphene systems through the AFM technique. (a) Left - Graphene oxide dissolved in water – “Graphenea” in an isopropyl alcohol solution. (b) Center - Graphene oxide dissolved in water – “Graphenea” in ethylene glycol solution. (c) Right - Graphene oxide dissolved in water – “Graphenea” in water

### 3.2. Molecular Machines with Photoactivated Graphene. *The second kind Storage of Energy for Molecular Machines with Graphene*

It is considered as a working system a rotaxane-based molecular machine, in fact [2]rotaxane  $1H^{3+}$  containing a  $\pi$  electron donor macrocycle, dibenzo[24]crown-8 ether (DB24C8), along with a “dumbbell” component consisting of a secondary ammonium ( $-NH_2^+$ ) and a 4,4'-bipyridine unit ( $bpy^{2+}$ ); the stopping system consists of an anthracene unit on one side and the 3,5-di-tert-butylphenyl group on the other side. The system is stable to charge transfer (CT) due to the weak interaction of the electron-p donor macrocycle with the  $bpy^{2+}$  p-electron acceptor unit, compared to the interaction of the hydrogen bond [ $N^+-H \cdots O$ ] between the DB24C8 macrocycle and the ammonium center, when the macrocycle surrounds the center (Figure 4).



**Figure 4.** Schematic representation of a molecular shuttle process of a rotor-crown ether by deprotonation and reprotonation of the ammonium center [13]

However, if a base such as tributyl amine is added, the ammonium center is converted to an amine function with a  $1A^{2+}$  transition state, and after the macrocycle dislocation in the  $bpy^{2+}$  unit is switched to the stable  $1B^{2+}$  state. The process is reversible if an acid, such as trifluoroacetic acid, is added when the system returns to its original state by the  $1BH^{3+}$

transition state. If the rotaxane is deprotonated, for example by uni-electronically reducing of the bipyridine unit, the charge transfer channel is destroyed, and the macrocycle ring can be detached from the  $\text{bpy}^{2+}$  station. This step consists of calibrating a self-electrochemical process with molecular machines, testing and measuring of the thermodynamic functions (free enthalpy, formation enthalpy, and Gibbs energy) for the case where the process is carried out on graphene at locations selected from the matrix deposits in the earlier stages of the research.

### 3.3. Molecular Machines with Photoactivated Graphene. *Electrochemical Reactivity with Spin Molecular Machines*

The spin chemical reactivity is modeled in the auto-electrochemical system from the previous stage for estimating the storage energy (at each such internal electrochemical cycle) following the energies released by electronic destabilization and restoration, in an isolated state (Figure 5) – respectively on graphene support.

As an acid-base process, the “Hard and Soft Acids and Bases” (HSAB) principle in the Parr-Pearson variant is applied, whereby the energy transferred/stored in an acid-base transformation cycle, the induced electrochemical phenomenon is written in an additive way (see quantum superposition principle):

$$\Delta E = \Delta\Omega_A + \Delta\Omega_B \quad (2)$$

but with mixed contributions (see quantum interference) of the type:

$$\Delta\Omega_A = -\frac{\eta_A}{4(\eta_A + \eta_B)^2} (\Delta\chi)^2 \quad (3)$$

$$\Delta\Omega_B = -\frac{\eta_B}{4(\eta_A + \eta_B)^2} (\Delta\chi)^2 \quad (4)$$

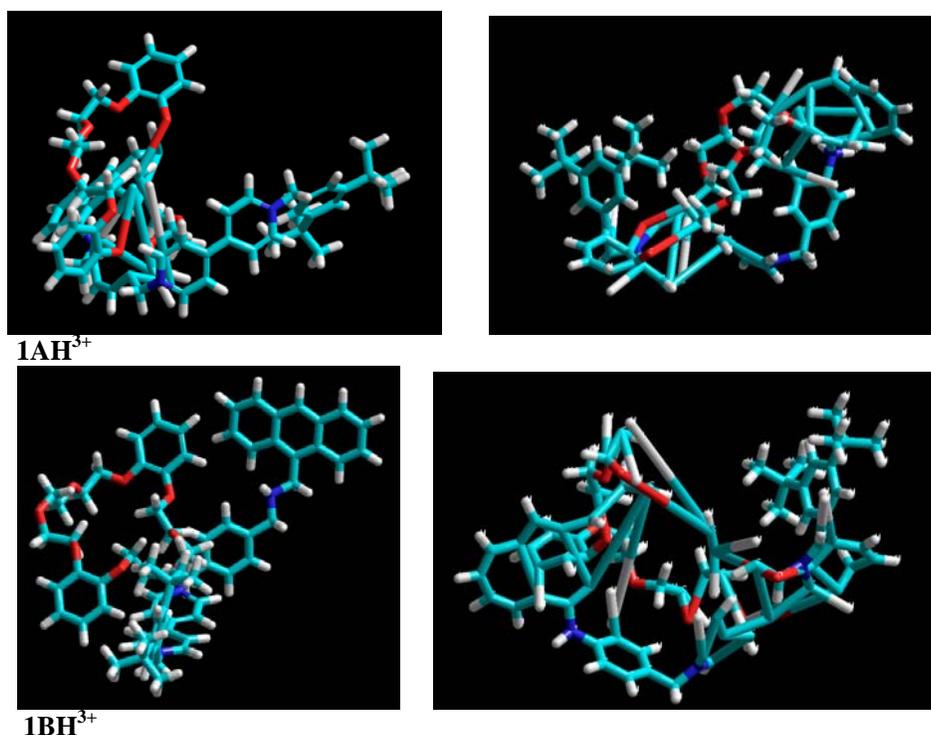
with the determinant as the difference in electronegativity of the reactants:

$$\Delta\chi = \chi_A - \chi_B \quad (5)$$

The optimal energy condition (storage) implies, for example, the minimization of the contribution  $\Delta\Omega_A$  concerning the chemical strength  $\eta_A$ , while the electronegativity variation  $\Delta\chi$  and the chemical strength of the counterpart  $\eta_B$  are kept constant (stabilized).

It should be noted that the HSAB principle implies the optimal transfer (excess energy storage, possibly via functionalization with the graphene support) until the HOMO-LUMO differences for the electrochemically reactive species are equilibrated  $\eta_A = \eta_B$ , respectively coordinate weak acids with weak bases and strong acids with strong bases.

The computational modeling in HyperChem [24], as in Figure 5, allows the obtaining of a complete energy panel (HOMO and LUMO spectral calculation) to optimize inter-band energy couplings to the cycle functionalization of the molecular machines with the graphene structure matrix deposited as above.



**Figure 5.** HyperChem representations [24] of molecular shuttle structures and internal electrochemistry from Figure 4

### 3. 4 Graphene Quantum Electrochemistry. *Nanoscience Control of Quantum Tunneling in Multi-Graphene Systems*

In the multigraphene context, as the result of electronic coupling, bondonic forms occur in three structural forms. First of all is the chemical-graph  $|0\rangle$  corresponding to the isolated state, then the state of a grid/node (in Figure 2) as vacation type or defective hole  $|h\rangle$ , respectively the pre-bosonic topological/electronic defective state  $|e\rangle$ , corresponding to the “quantum sea” which incorporates the space of the topological isomers in the multigraphenic matrix node. Given that these states can simultaneously exist in a multigraphene matrix arrangement of the specific type shown in Figure 2, the interaction between them defines the quantum electrochemistry or the tunneling spin that can occur between them, namely:

The transition/tunneling  $|0\rangle \leftrightarrow |h\rangle$  is associated with the so-called “nuclear mass defect”, and corresponds to the quantum-electrochemical potential given by the law of variation:

$$\Delta\Pi_B = \alpha_1 \exp(-\beta_1 \Delta H) \quad (6)$$

with the activation parameters and the reaction rate limiters to be determined for each kind of matrix adjacent beside the bondon formation enthalpy;

Similarly, the transition  $|0\rangle \leftrightarrow |e\rangle$  correlates the quantum-electrochemical rate and potential with Gibbs bonding energy  $\Delta G$  in this adjacency:

$$\Delta\Pi_\chi = \alpha_2 \exp(-\beta_2 \Delta G) \quad (7)$$

and corresponds to the specific activation (selection) of an isomeric topology from the set of topo-electron graphene isomers in the system;

Finally, the quantum tunneling of a defective hole – topological defect  $|h\rangle \leftrightarrow |e\rangle$  corresponds to a “cellular” transition with the creation/annihilation topological potential:

$$\Delta\Pi_{EC50} = \alpha_3 \exp(-\beta_3\Delta A) \quad (8)$$

associated with the rate of creation/boson annihilation  $\Delta A$  (in bondon, or inter-bosonic in bondots).

### 3.5 Graphene Quantum Electrochemistry. *Quantum-Electrochemical Metrology*

The previous phase is completed with experimental studies vs. the conceptual-computational estimates of the quantum tunneling current (in the quantum deca-nano-transistor  $|0\rangle \times |e\rangle \times |h\rangle$  obtained by matrix multigraphene configurations), with the estimated intensity:

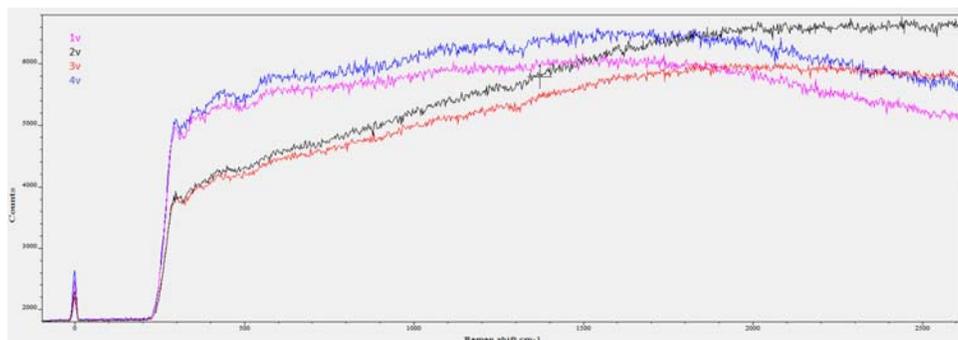
$$i(\varepsilon) = -\frac{2e}{\eta} \int_{\varepsilon}^{\infty} f(E_D) dE_D \quad (9)$$

for the clustering of quantum dots (bondons) in bondots ( $\mathcal{D}$ ).

Here occurs the “comb” function of the Dirac multi-function specific for the bondots:

$$f(E_D) \equiv \sum_{q=-\infty}^{q=+\infty} \delta(q - E_D) \quad (10)$$

with the multi-energy tunneling effect of the Coulomb blockade specific to isolated (fullerene) electrons, respectively observable by the Raman specific displacements for different assemblies of the graphene oxide fullerene (Figure 6).

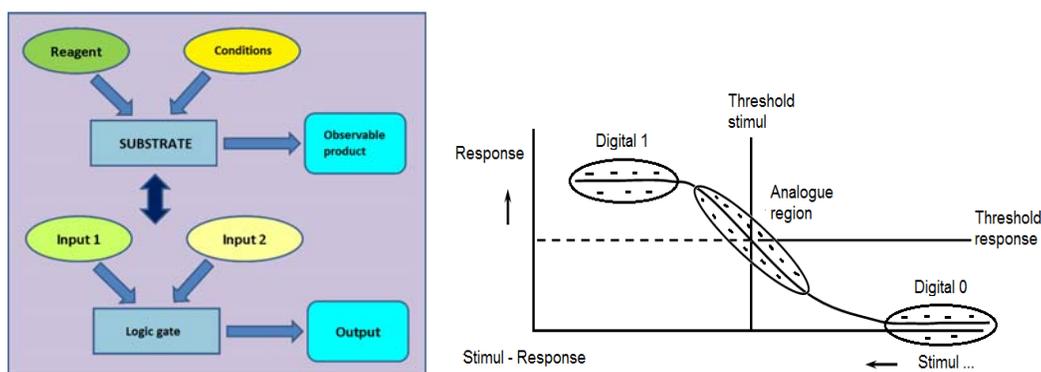


**Figure 6.** The Raman spectroscopy of quantum currents for bondots (bondon aggregates) formed in fullerene graphenes (nominated as 1v, 2v, 3v, and 4v)

### 3.6 Photo-Activation of Spin Density on Graphene Semiconductor. *Activation of Quantum Logic Gates on Defective Graphene*

The deca-nano graphene transistor from the previous stage can be in the last phase “enriched” with quantum logic-gate properties by functionalization (on a graphene substrate) with molecular machines (motors) (Figure 7-left) in the 2nd stage. Furthermore, the acid-base

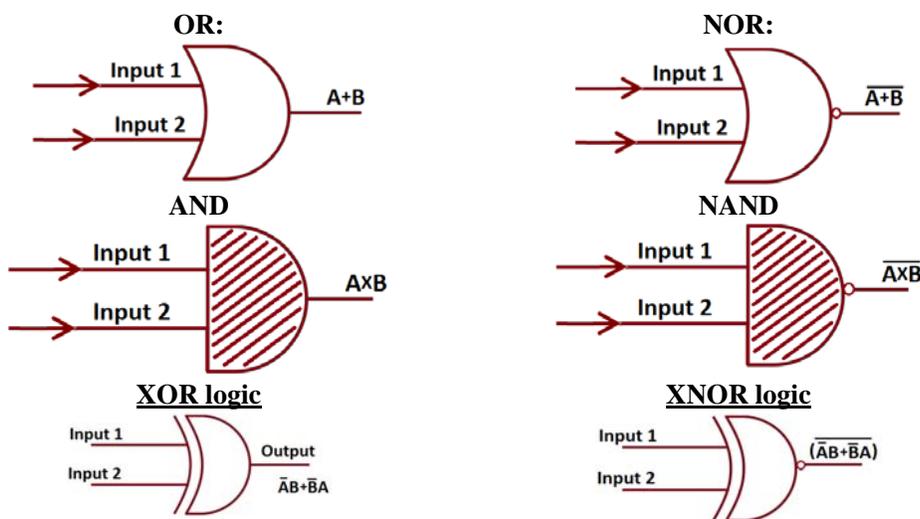
(deprotonation, reprotonation) of the molecular motor can be associated with distinct signal inputs in the conjugate system (graphene + molecular machines) with the specific signal (isolated or by photoactivation or spin coupling respectively in the quantum graphene transistor) of the analog type (direct current) or digital (“comb” current signal), see the last phase bondotic current, Figure 6, and Figure 7-right.

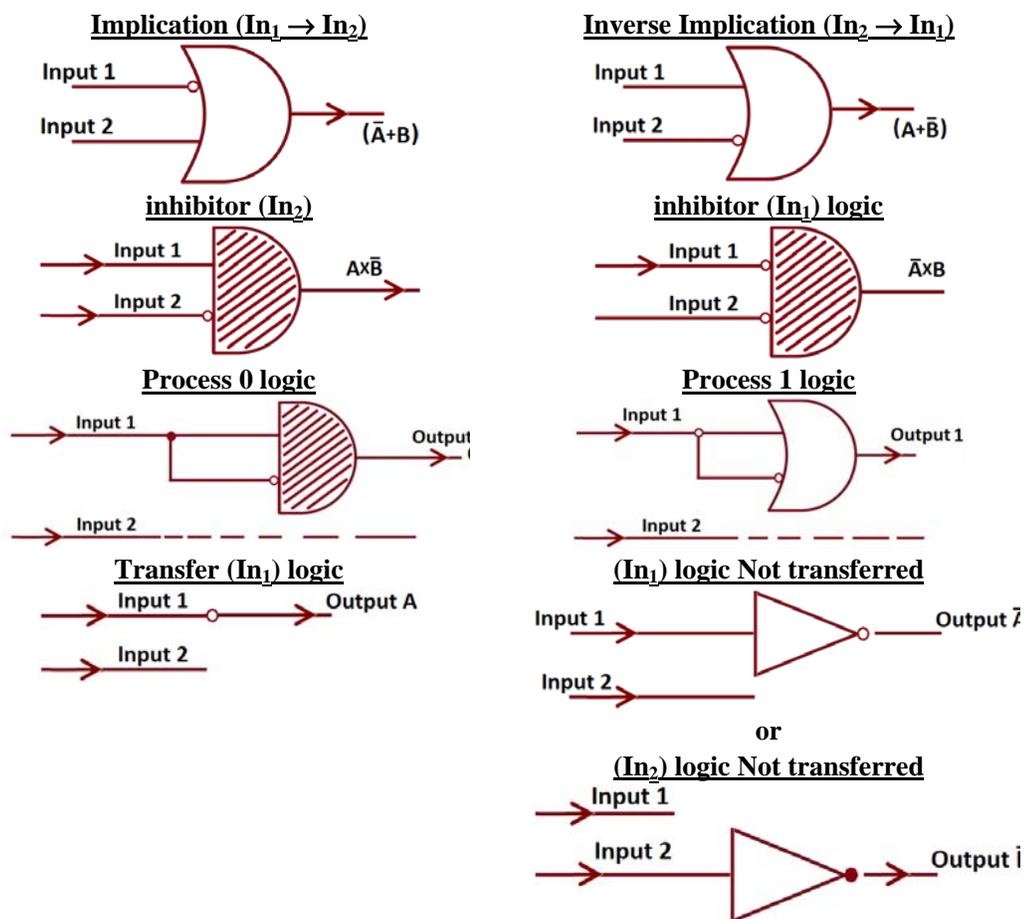


**Figure 7.** (a) Left - The conceptual design of a logic gate functionalization [24] (for example, molecular machines, protonation input processes, deprotonation, reprotonation, etc.) on a graphene substrate with an obvious response. (b) Right – conceptual design of the analog type (non-stationary, continuous), or of the digital type (stationary, quantum, Dirac “comb”)

These unique characteristics of the complex graphene system + the molecular machines make the bondots ideal candidates for further development for what is called molecular quantum algebra, in this case with graphene layers, in junctions and logical combinations that allow the obtaining of filtered, intelligent, controlled signals, and finally integrable into quantum moletronics, semiconductor, diode, quantum transistor and their applications.

It should be noted that logic gates are switches that have an output (0 or 1) that depends on the input (0 or 1). The list of logic gates, respectively digital switches for the molecular machine systems to be implemented and tested in this project is shown below in Figure 8.





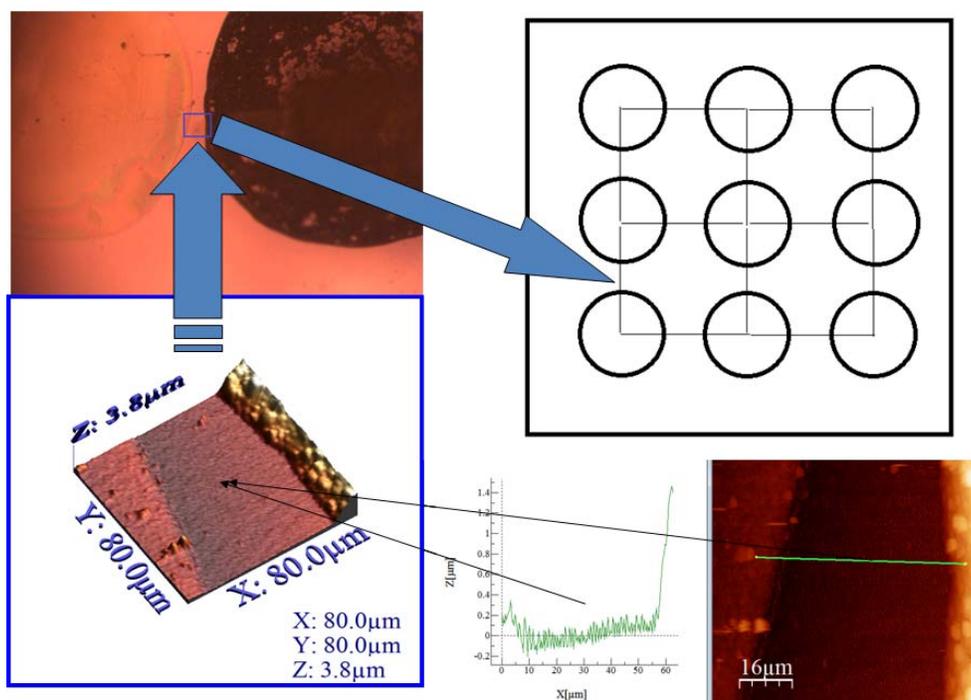
**Figure 8.** The list of logic gates, respectively digital switches for the molecular machine systems to be implemented and tested

The current estimations, based on the first atomic microscopy tests on graphene, non-functionalized with molecular machines, suggests the formation of stable over-micro-currents (by about 15%), thus analogous, as a result of increasing defects from the first photoactivation cycle, based on the collective aggregation of bondots, bondons, quantum couplings of electrons in the graphene conduction band. Thus, a digitalization effect of these analog signals (Figure 7-right) is expected in the case of the functionalization of these multi-graphene systems, matrix-deposited, with molecular motor machine-logic gates.

This way, Bose-Einstein's collective condensation can be controlled on the functionalized graphene with molecular machines, with the formation of super-micro-currents, integrable in moletronic circuits, and molecular algebraic calculus, with the establishment of quantum logic gates, conversion, but also intelligent storage and filtering, possibly activated by multi-graphene solar cells.

### 3.7. Photo-Activation of Spin Density on Graphene Semiconductor. *Supraconductivity of Deca-Nano-Magnetized Graphene Semiconductor*

The last phase, integrating the present research, extends the activation mode of the graphene system + molecular machines through the quantum spin coupling, along with the effect of magnetization, possibly induced by the specific form of adhesion in matrix multigraphene depositions (Figure 2). The specific quantum spin effect (magnetization) is shown in Figure 9 (combined detail of Figures 2 and 3, first phases of research) and the data of the material analysis in Table 1.

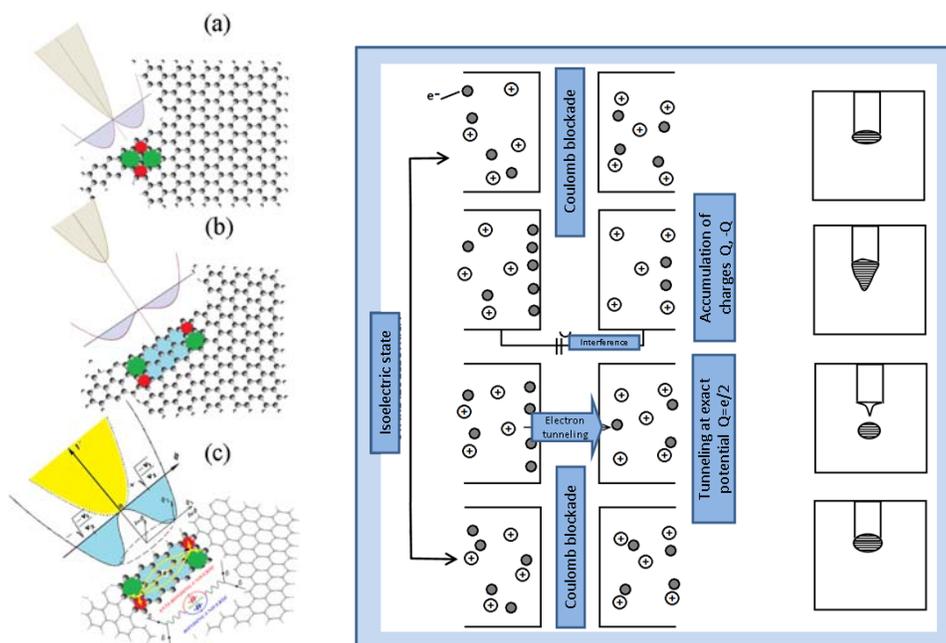


**Figure 9.** (a) Top-left - The microscope image at the closest distance between GO (left) and  $\text{TiO}_2$  (right) as a detail of the matrix configuration in Figure 2. (b) Bottom-right - AFM and 3D analysis. (c) Bottom-middle - Material roughness on selected area (with global data in Table 1)

**Table 1:** The data obtained from the AFM image for the GO- $\text{TiO}_2$  adjacent configuration in Figure 2 and details from Figure 9

Sample name	Area [ironed surface] ( $\mu\text{m}^2$ )	Sa [roughness average] (nm)	Sq [RMS roughness] ( $\mu\text{m}$ )	Sp [maximum value] ( $\mu\text{m}$ )	Sv [minimum value] ( $\mu\text{m}$ )	Sy [peak to peak] ( $\mu\text{m}$ )	Sku [surface kurtosis]	Ssk [surface skewness]
1	6875.48	0.39	0.63	3.59	-0.23	3.82	9.49	2.69

The AFM technique observation was performed on the smallest distance between the GO and  $\text{TiO}_2$  deposition to obtain a complete AFM image including both deposited and non-deposited area (ITO).



**Figure 10. (a)** Left - Tripartite vibrational instability on Graphene Network with Stone-Wales topological rotation vs. the breakage of symmetry in the spin coupling and the bosonization of the fermions, with the appearance of specific bondot: 1) the Jahn-Teller effect (through the canonical potential intersection), 2) the effect of the pseudo Jahn-Teller effect, 3) the Renner-Teller effect (see also the text for details).

**(b)** Right - schematic representation of the Coulomb blockade [24], in which the change in the electrostatic potential expresses the charge tunneling/redistribution. The Coulomb blockade allowed precise (metrological) control of the number of electrons and pairs of bondot and formed bondot trains (as a digital signal) in logic switches/gates (such as graphene complex system + molecular machines), allowing design advanced graphene integrated circuits (see graphenetrionic engineering)

The phenomenological basis of this type of internal activation (in addition to photo-activation) lies in the tunneling capacity and the type of adhesion in the multi-graphene matrix system (Figure 2). Thus, a gradual change of spin - electron spin coupling occurs in the bondonic boson, with the activation of magnetism (spin alignment or spin anti-alignment) on controlled areas of the multi-graphene matrix, depending on the type of potential coupling (double fermion vs. parabolic bosonic unit) described in Figure 10-left. Even more interesting, a correlation can be established between the three types of magnetic activation, the Jahn-Teller effect (symmetry breaking effect in the bosonization of the fermions, the first coupling of the quantum degenerated states), the pseudo-Jahn-Teller effect (the second order coupling of non-degenerate quantum states), and the Renner-Teller effect (the second order coupling of degenerate quantum states) with the three types of graphene states specifically to the quantum deca-nano-transistor,  $|0\rangle \times |e\rangle \times |h\rangle$ , studied in the 3rd stage. Furthermore, these three phases (quantum phase transitions, concerning the production of digital signals) correspond to the three phases of the Coulomb blockade (Figure 10-right), respectively surpassed, induced by magnetic activation and thus finally controlled.

## 4. CONCLUSIONS

The present research devoted to Graphenetrionics with Quantum Spin Electrochemistry develops disruptive applicative algorithms in photo-electro-chemistry with multiple possibilities for the development of efficient nano-energy solutions. Regarding the contribution of the recent studies in the field, the proposed research is provided with an innovative character, which is even more pronounced by the proposed magnetically activation (via the electrochemistry of the coupling by spinning) following an original multi-graphene matrix type deposition. Thereby, the matrix logic gates at the graphene neighborhood level areas supported by quantum induction (quantum electrochemistry) the activation of molecular machines as local quantum logic gates in the multi-graphene matrix nodes, thus achieving gate-in-gate systems with multi-potential electronic control, transfer in electronic pairs (bondons, bondots), digital signal communication and deca-control of nano-energy efficiency.

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