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Unfolding potential and challenges in molecular field-coupled nanocomputing

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Abstract

Molecular Field-Coupled Nanocomputing (MolFCN) represents a revolutionary approach to computational technology, exploiting single molecules for encoding and processing logical information. MolFCN permits zero-current logical operations to achieve ultra-low power and hyper-miniaturized computing units. This perspective article explores the current state and future potential of MolFCN, highlighting recent technological advancements, potential applications, and the significant challenges that lie ahead. Despite the challenges, the pathway to practical implementation holds significant promise, with obstacles such as scalability, stability, integration, and practical considerations offering opportunities for innovation and advancement. MolFCN can shape the future of nanocomputing and contribute to current major challenges in nanoelectronics by opening key research directions.

1. Molecular electronics and molecular field-coupled paradigm for future nanocomputing

Molecular electronics was born in 1974 with the theoretical work of Aviram and Ratner, who proposed that a donor-bridge-acceptor molecule would behave like a rectifier [1]. This initial pioneering work sparked a long history of scientific interest, and several important milestones have been reached. Among them, the first experimental evidence of an effective current modulation by an external gate voltage [2] opened the way to molecular transistors and to the enticing opportunity of replacing silicon transistors with molecular ones [3–5]. Nowadays, experimental techniques are mature enough to reliably and reproducibly study charge transport through single-molecule devices. The main challenge remains to contact a single molecule successfully with a satisfactory yield. Several proposed techniques span from crack-defined and mechanically controllable break junctions, electromigrated gold junctions and electro-burned graphene junctions, rolled-up soft contact, STM-based break junctions, and nanolithography-defined junctions [6]. The STM-break junction technique has become the most established tool to investigate the electronic properties of single molecules thanks to accessible equipment and experimental setup, ease of automation, and favoured repeatability. Looking towards the future of integrated molecular electronics, fabrication challenges in nanopatterning and molecular deposition are, however, still the main limiting factor. Nevertheless, in 2022, the fabrication of the first molecular integrated circuit through nanolithographic processes marked a significant milestone, paving the way for the hybrid integration of CMOS-molecular systems [7]. In addition to current-driven molecular components, MolFCN technology emerges as a promising alternative, offering new opportunities for developing functional molecular electronics devices. In this perspective review paper, we present an overview of the MolFCN state-of-the-art. We critically highlight the challenges and the open gaps in the fields of design and simulation, as well as in technology and characterization. MolFCN

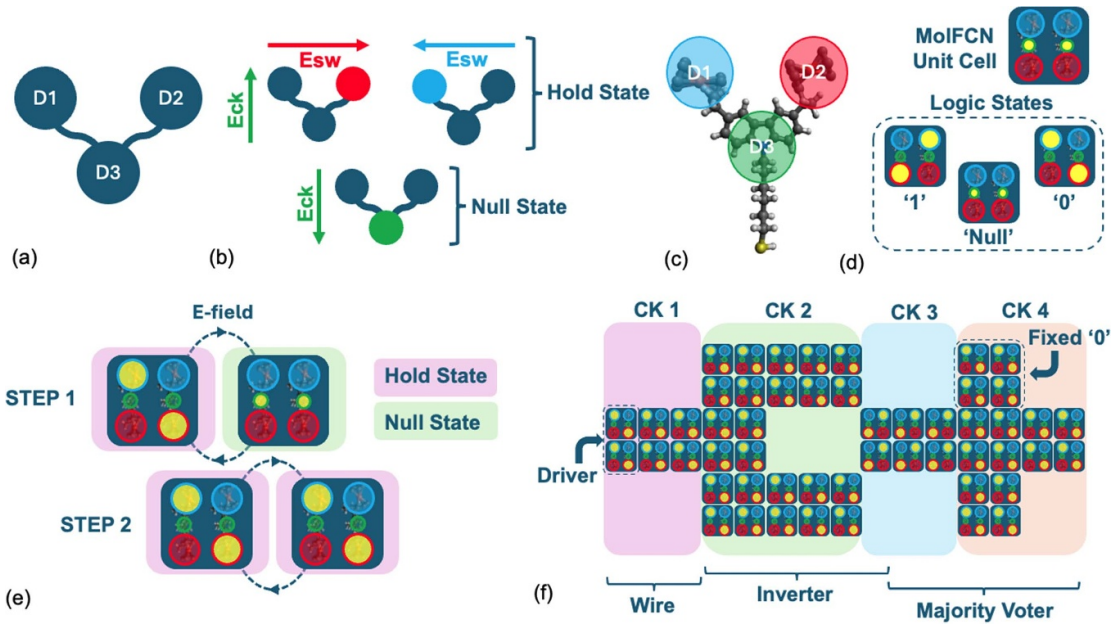

Figure 1. (a) Sketch of a three-dot Y-shaped candidate molecule for MolFCN applications; (b) schematic representation of the possible configurations that can be forced with the external electrical control; (c) an example of a Y-shaped candidate molecule: the bisferrocene, and its aggregation regions; (d) the MolFCN unit cell and its logic encoding capability; (e) an example of the coupling and propagation mechanism between two adjacent cells under the clocking control mechanism; (f) an illustrative example of a MolFCN circuit containing the state-of-the-art basic blocks: the wire, the inverter, and the majority voter. The circuit is divided into four clock regions, enabling robust and pipelined information propagation and computation.

implements the quantum-dot cellular automata (QCA) paradigm at the molecular scale. Specifically, it encodes logic information in the aggregation of charge in specific regions of ad hoc molecules [8]. Figure 1(a) shows the schematic of a molecule suitable for MolFCN applications. It contains three charge aggregation regions, each known as Dot (D). As depicted in figure 1(b), charge localization within a molecule is controlled by two guiding fields: the switching field (E_{sw}), applied between D1 and D2, and an orthogonal clock field (E_{ck}), applied between the upper D1 and D2, and D3. When the charge is localized in the upper region, the molecule is in the so-called *Hold State*, allowing the charge to move between D1 and D2 in response to E_{sw}. Conversely, when the charge is localized in D3, the molecule is in the so-called *Null State*, which inhibits charge movement between D1 and D2. The Y-shaped molecular structure is particularly convenient in MolFCN applications because of the possibility of two-directional charge movement control and integration of a clocking mechanism [9, 10]. The chemical structure can also be optimized to ensure isolation of the aggregation centers from the substrate, for instance, by adapting the design principles used to prevent quenching of photoexcited states [11]. So far, various molecular candidates have been identified [12–14]. Amongst those, bis-ferrocene derivatives, depicted in figure 1(c), have shown promising results, from synthesis and theoretical characterization to successful adsorption on a gold substrate [15, 16]. The design of digital circuits based on MolFCN starts with the so-called MolFCN unit cell, formed by electrostatically coupling two molecules. When in the *Hold State*, the molecular charge can arrange into two energetic minima configurations, associated to the logic values '0' and '1'. When the molecules are forced in the *Null State*, the information is canceled, as depicted in figure 1(d). Arranging cells appropriately permits the creation of circuits that enable information propagation and digital computation. Cells are grouped into clock regions to ensure stable and robust information propagation, where consecutive cells are guided to the same induced state [17–19]. The mechanism of enabling or disabling information propagation within these regions is called *clocking*. When the molecules are in the *Hold State*, information can propagate through electrostatic interactions, allowing each cell to reach its energetic minimum [20]. Conversely, when in the Null State, propagation is blocked, as shown in figure 1(e). Appropriately arranging these clocked cells permits information propagation and digital computation by associating different sets of consecutive cells with either the *Null State* or *Hold State*.

Figure 1(f) shows an illustrative example of a MolFCN circuit presenting the essential elements of MolFCN circuits to show the state-of-the-art of available MolFCN basic cells. The circuit is composed of a wire, an inverter, and a majority voter. Information propagates from the driver to the consecutive circuit cells through the sequential activation of clock regions, which guarantees robust and pipelined transport. Information propagates from the driver to the consecutive circuit cells through the sequential activation of

clock regions, which guarantees robust and pipelined transport [21]. Most importantly, MolFCN offers theoretical advantages over other technologies, such as low-power consumption, high operating frequencies, and functionality at room temperature [22–25].

2. Advances in MolFCN modelling and simulation

Accurate modeling from the molecular to the circuit levels is crucial for advancing MolFCN technology. Traditional modeling based on *ab initio* simulations, i.e. on Quantum Chemistry, offers high accuracy but with inflated computational costs, especially for circuits with numerous molecules. Lu and Lent introduced the Two-State approximation, useful to approximate the electronic behavior of bistable molecules such as mixed-valence compounds [26]. Recent works also pointed out the possibility of creating MolFCN computation with monostable molecules [19, 20, 27]. We introduced the MOlecular Simulator QCA TOrino (MoSQuiTO) as an efficient methodology to speed up the MolFCN circuit simulation without compromising on the reliability of results [16, 28, 29]. The MoSQuiTO framework—figure 2(a)—provides a structured methodology for modeling MolFCN, encompassing three key stages: (1) *ab initio* geometry optimization and analysis of the candidate molecule under several electrostatic conditions. By applying external electric fields, denoted as E_{sw} and E_{ck}, the resulting charge distributions within the molecular dots are evaluated; (2) definition of the figures of merit characterizing the electrostatic properties of the molecule of interest. V_{in}-to-Aggregated Charge Transcharacteristic (VACT) function is central in this step. The VACT links the input voltage (V_{in}) applied between D1 and D2 to the AC within the molecule, evaluated as the sum of atomic charges in the molecule dots. The AC is determined at a constant electric field (E_{ck}), providing a comprehensive interpretation of how the molecule's charge state responds to various V_{in} and E_{ck}. This step effectively translates the molecular-level electrostatic behavior into a usable form for subsequent analysis; (3) simulation of circuits using the Self-Consistent ElectRostatic Potential Algorithm (SCERPA) [30, 31]. SCERPA is designed to iteratively resolve the electrostatic interactions between molecules in the circuit by calculating the V_{in} for each molecule and then evaluating the charge distribution through the VACT. Specifically, SCERPA relies on the self-consistent solution of the following equation:

$$V^s_{\mathrm{in},i} = V^s_{D,i} + \sum_{j \in \mathrm{IR}} V^s_{j,i}\left(V^s_{\mathrm{in},j}, E^s_{\mathrm{ck},j}\right) \tag{1}$$

where $V^s_{\mathrm{in},i}$ is the input voltage of the molecule i generated by the $E_{\mathrm{sw}_{i-1}}$, s indicates the current clock step, $V^s_{D,i}$ is the driver input voltage effect on each molecule i, and $V^s_{j,i}$ represents the cumulative effect of the molecules j, within a specific distance from i, on the considered one i. This iterative process continues through each propagation step, allowing for a detailed assessment of information propagation within the MolFCN circuit.

With the three steps, MoSQuiTO provides a comprehensive and computationally efficient approach to model the electrostatic behavior and information propagation capabilities of MolFCN, facilitating the design and analysis of advanced molecular devices [32, 33]. Using the SCERPA algorithm, MoSQuiTO can simulate complex MolFCN circuits containing hundreds of molecules within short time frames while ensuring a DFT-level molecule modeling detailed *ab initio* characterizations [18]. This capability is crucial for practical applications where understanding the behavior of large-scale molecular networks is essential. The constraints provided by SCERPA empower designers with crucial information for layout optimization and understanding the characteristics of individual molecules [10, 18].

Figure 2(b) shows the results of the SCERPA simulation for the MolFCN circuit composed of bis-ferrocene, previously discussed in figure 1. In particular, it depicts the clock signal timing adopted in the simulation and illustrates the main steps of propagation, where the logical value '0' is transmitted from driver D to the output. As observed, the circuit operates correctly; the logical value '1' is first carried along the input wire, inverted by the inverter, and processed by the output majority voter. In this case, the majority voter has two fixed inputs, one at '1' and the other at '0'. Since the logical value derived from the inverter is '0', the final output of the circuit remains '0'. Various insights can be pulled from such a SCERPA simulation. Notably, during the majority voting operation, there is diagonal coupling between the input branches of the majority voter, leading to a loss of information in the branch carrying the logical value '1', which is less prevalent in this case. Additionally, border effects can be observed in the last cell of the output circuit, resulting from the reduced influence of molecules on the molecules forming the circuit's output. Such considerations, including the need for multi-line circuits for inversion operations, are crucial for the proper design of MolFCN circuits, as they directly stem from the electrostatic nature of the involved molecules [21, 33].

Currently, the SCERPA simulation and the characterization of MoSQuiTo provide a clear understanding of the digital design of MolFCN circuits. Looking ahead, the MolFCN simulation setup will evolve to incorporate dynamic aspects and frequency studies, unlocking new insights. Initial work on MolFCN's

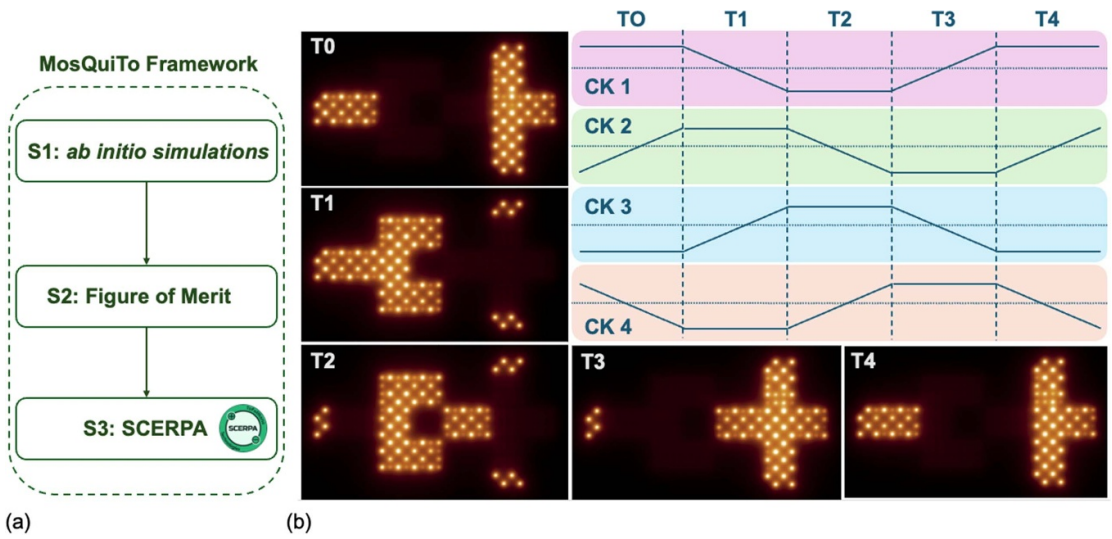

Figure 2. (a) The three steps of the MosQuiTo Framework: first, *ab initio* simulations for geometry optimization and electrostatic characterization of the candidate molecules; second, from the electrostatic characterization, are extracted figures of merit to describe the molecule as an electronic device; third, the FOM is used to simulate MolFCN circuits. (b) The SCERPA simulation results for the circuit presented in figure 1(f). Each clock zone is controlled by a specific signal, enabling/disabling the propagation of the information encoded within the region cells. At each time step (Tx), the activated regions encode the information and prepare it for propagation into the subsequent clock region.

dynamic analysis has shown that a single molecule can achieve maximum switching frequencies exceeding hundreds of GHz [34]. Future studies should investigate more in details the dynamic aspects, and the effects of temperature on information encoding and processing, with the perspective to furtherly optimize circuit designs for an improved and more comprehensive understanding of performance and functionality.

Furthermore, QCA optimization methodologies can be exploited to enhance the MolFCN circuit design even more. The already developed QCA methods can enhance design by addressing placing, routing, post-layout optimization, and synchronization challenges. For example, placing and routing techniques minimize delays and improve spatial efficiency [35], while post-layout refinements mitigate cross-coupling effects [36]. Parameter optimization, demonstrated in one-bit full adders via QCA Designer, underscores the potential for robust molecular circuit designs [37–39]. Incorporating these strategies strengthens the feasibility of MolFCN systems and enriches the design framework.

The future direction from the simulative standpoint is to have a comprehensive toolchain composed of well-engineered, technology-aware, and inter-compatible tools, allowing many degrees of freedom, from the choice of the molecules to the choice of the substrates, that incorporate simulations from the single molecule DFT to the basic unit cell simulations, and to the design of an entire complex circuit based on molecules. The perspective is of a robust and comprehensive toolchain that enables the possibility of engineering MolFCN circuit design and verification from single molecule physics to high-level circuit design.

3. Technology: promising achievements and future challenges

Despite the promising theoretical results, a prototype for MolFCN has yet to be demonstrated. Specifically, there are four critical steps required for the realization of such a device: (i) the synthesis and (ii) deposition of the candidate molecule, (iii) the fabrication of the host device containing the molecular circuit, (iv) and the development of both the structure for generating the clocking mechanism and a readout system for hybrid MolFCN-CMOS interfacing [29].

Candidate MolFCN molecules must satisfy specific technological, functional, and environmental requirements criteria to ensure the technology feasibility and sustainability. From a technological perspective, the molecule must have a stable anchoring point, allowing for robust and precise adsorption on the target substrate. The molecule geometry also plays a significant role in determining its suitability. As an example, Y-shaped structures are promising for stably localizing charge, allowing for both encoding and clocking [13–16]. Nevertheless, they could be more susceptible to unwanted rotations. Functionally, the molecule's electrostatic properties should enable precise control of its internal charge distribution through external electric fields. Key characteristics such as high polarizability, a low native dipole moment, and chemical stability under ambient conditions are essential to facilitate the intramolecular charge controllability and ensure efficient, responsive behavior within molecular devices while maintaining

electronic properties and structural integrity during circuit operation [19, 29]. Furthermore, considering the current trends and attentions in sustainabile and green electronics, the environmental impact is becoming a critical aspect in defining new molecules for MolFCN. It is primarily determined by two key factors: the molecule synthesis process and how the molecule affects the environment in the afterlife. The search for the optimum molecules meeting the characteristics mentioned above has to derive from synergetic efforts between engineers, chemists, and physicists working on the future of this technology.

Concerning prototype fabrication, MolFCN devices should be built on atomically flat and uniform substrates to guarantee correct intermolecular interaction [32, 40, 41]. Another critical aspect of MolFCN fabrication is the pattering of the network providing the clock, i.e. the clocking structure [40]. At present, the patterning could be achieved through nanoscale lithographic techniques like focused ion beam , electron beam lithography , nano-imprinting, secondary sputtering lithography, and SPM-based lithography [42–44]. Then, the molecule adsorption can be obtained through the exploitation of self-assembled monolayer (SAM), uniform parallel unimolecular templating deposition, or through SPM-based single-molecule manipulation [45–48].

Interfacing MolFCN with CMOS requires systems that directly convert the logical information encoded by charge position into an electrical signal, such as voltage or current. Currently, promising results have been achieved using single-electron transistors (SETs); however, these devices face constraints related to operating temperature [49]. Therefore, research must focus on developing new types of charge-position sensors capable of operating at room temperature and scalable at the single-molecule level. As a potential solution, we introduced the bend-boosted MolFCN paradigm in [50]. Our simulation demonstrates that using the position of molecules to encode logical information enables an efficient readout mechanism via molecular junctions. This approach can be seamlessly integrated with charge-position-based MolFCN.

The three main challenges outlined in this section represent the key points to address a functional MolFCN prototype. Given the highly specific and complex fabrication processes involved, a synergistic approach combining simulation and experimentation is essential to address each challenging aspect effectively.

4. A workflow toward prototyping: modelling connects with experiments

In the path towards prototyping MolFCN systems, an efficient approach may be to exploit a virtuous loop between DFT-based modeling and experimental characterization, schematically proposed in figure 3 [29]. Indeed, such an approach has been demonstrated successful to achieve the first carbon nanotube-based microprocessor [51]. The MoSQuiTo framework can be integrated with fabrication simulations and technological feedback for comprehensive MolFCN device design. Fabrication simulations may include molecular deposition results, molecule-substrate interface considerations, process simulations for hosting devices fabrication, and electric field generation modeled via finite-element modeling (FEM). For the latter, in [18, 40], we proposed and integrated in SCERPA the FEM-evaluated clock and switching field generated by a target structure hosting the molecular circuit. Specifically, we studied a dielectric nano-trench with metallic electrodes on top and in the middle. The analysis highlighted the necessary characteristics for clock electrode generation, multi-line systems, and input mechanisms, emphasizing the importance of incorporating technological aspects into simulations [52–54]. Regarding molecular deposition, SCERPA can account for deposition substrate defects [32]. Thus, deposition results of candidate molecules, obtained through molecular dynamics methods or SAM characterization, can be integrated into the SCERPA simulation framework. Furthermore, molecule-substrate interface analysis could be incorporated into the characterization of the molecule itself to account for charge exchanges, potentially impacting the molecule VACT [55–58]. Moreover, results from advanced nanoscale process simulations, such as atomic layer deposition (ALD) and area-selective ALD, can be integrated into SCERPA to provide precise inputs to consider process variations possibly occurring in the fabrication of either the hosting device and molecular patterns [59, 60]. Finally, fully understanding the technological and fabrication aspects of the materials involved in MolFCN device construction should enable the study of accurate and comprehensive power consumption models. These models will consider, in particular, the need to generate clock electric fields and input driving, allowing for a direct comparison of MolFCN technology with current CMOS technologies, as preliminarily done in [22].

Overall, a fabrication-enhanced MoSQuiTo framework will enable the evaluation of increasingly specific fabrication requirements. The integration of the framework with nanoscale manufacturing will drive progress towards the prototyping stage.

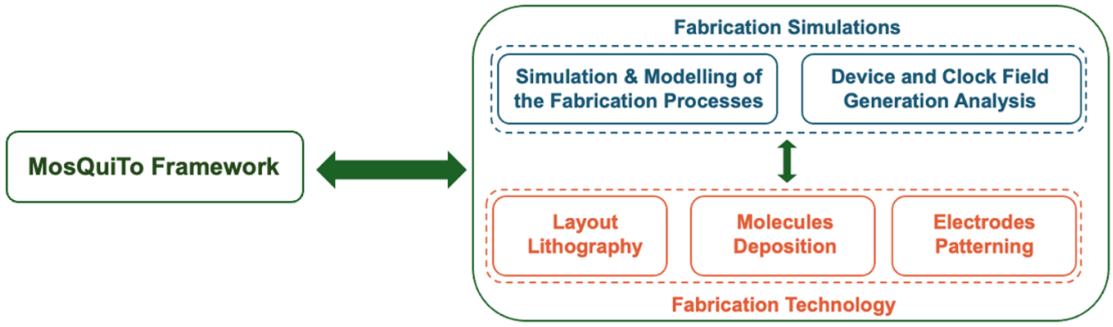

Figure 3. Schematic of the possible integration of DFT-based modeling and experimental feedback in the MoSQuiTo framework for MolFCN prototyping advancement. Fabrication simulations, clock field generation, and process variations must be incorporated. This comprehensive approach would accelerate MolFCN development and comparisons with CMOS-based technologies through exhaust device modeling.

5. Conclusions: overcoming challenges to implementation

MolFCN represents a transformative step in nanoelectronics, offering potential for ultra-dense, high-speed, and power-efficient hardware across applications such as logic gates and neural networks [18, 33, 61, 62]. However, its development faces significant hurdles in nanofabrication, characterization, and CMOS integration. Achieving molecular precision requires scalable and reproducible techniques. scanning tunneling microscopy (STM)-based lithography, extreme ultraviolet lithography, and dip-pen nanolithography show promise but face scalability issues. Hybrid approaches combining lithography with self-assembly may provide a viable path forward. Research should prioritize controlled prototypes to evaluate these methods [63–65]. Non-destructive characterization tools are essential for studying molecular circuits. STM and synchrotron-based techniques offer insights into molecular interactions but lack resolution and adaptability. Coupling these with complementary tools like Near-field scanning optical microscopy could enhance characterization while preserving molecular circuit integrity during prototyping [63, 64, 66]. CMOS integration poses challenges in interfacing molecular components with electronic architectures. SET, proven effective in sensing applications, could bridge the molecular-CMOS scale. Demonstrating their viability in hybrid configurations is crucial for MolFCN implementation [67, 68]. Advanced simulation tools are needed to integrate molecular-scale phenomena with fabrication constraints. These tools must optimize molecular stability, material compatibility, and interfacing, expediting the transition from theory to practical devices [69, 70]. Overcoming these challenges requires multidisciplinary collaboration among chemists, physicists, and engineers. By focusing on scalable prototypes, refined interfaces, and innovative methodologies, MolFCN can progress from concept to a revolutionary computing technology.

Data availability statement

No new data were created or analysed in this study.

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