



Cite this: *Mater. Horiz.*, 2025, 12, 4016

Received 31st January 2025,  
Accepted 17th March 2025

DOI: 10.1039/d5mh00192g

rsc.li/materials-horizons

## Enlightening molecular logic: basics, tools and techniques for newcomers

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As silicon-based technologies approach their physical limits, the search for alternative computing paradigms becomes imperative. Molecular logic has emerged as a promising approach, particularly the systems based on trivalent lanthanide ions that exploit the unique photophysical properties of these ions to implement Boolean logic operations. This focus article provides a comprehensive introduction to the principles, methodologies, and recent advancements in luminescence-driven molecular computing. Designed for newcomers, it outlines the fundamental concepts, essential experimental techniques, and standardized protocols for characterizing luminescent molecular logic devices. The advantages of these devices, such as energy efficiency, multiplexing capabilities, and adaptability to complex environments, are also critically examined. Addressing some limitations of traditional electronics, molecular logic paves the way for innovative applications in diagnostics, sensing, and novel computational architectures, offering a transformative and sustainable pathway for next-generation information processing.

### Introduction

The rapid advancements in nanotechnology and molecular engineering have ushered in a new era of materials and devices operating at the molecular scale.<sup>1,2</sup> Currently, there is a critical need for novel computing approaches as silicon-based technologies near their physical and practical limits.<sup>3,4</sup> The exponentially increasing demand for higher computing power<sup>5</sup> is being driven by advancements in artificial intelligence,<sup>6</sup> big data,<sup>7</sup> and complex simulations.<sup>8</sup> As Moore's law, predicting the doubling of transistors on integrated circuits roughly every two years, reaches its limits,<sup>9,10</sup> new computational paradigms must be explored to bridge the widening gap between technological potential and existing capabilities.<sup>11</sup>

Fig. 1 provides an overview of historical and technological milestones that have shaped human progress and computing evolution. Intriguingly, the dates of key moments in human history, from the agricultural revolution to the sequencing of the human genome, marking turning points in our technological and scientific journey, follow closely an exponential law.<sup>12</sup> Similarly, the history of modern computers, starting with early mechanical devices demonstrates the same exponential growth in computational efficiency<sup>12</sup> (measured in computing cycles per unit of time and per thousands of dollars) from the early days of ENIAC to the current supercomputing architectures.

The trajectory in Fig. 1b underscores the limitations now faced by traditional silicon technologies, that are approaching their miniaturization limits due to scaling challenges at the nanoscale.<sup>13,14</sup>

Current fabrication technologies struggle to produce transistors smaller than 5 nm, prompting the semiconductor industry to explore alternative strategies.<sup>15,16</sup> The 5 nm technology node, representing the current smallest critical dimension of a transistor, poses significant challenges in terms of power consumption, switching speed, and transistor density.<sup>17</sup> Further reducing transistor dimensions leads to quantum tunnelling effects, leakage currents, and increased heat dissipation, hindering reliable performance and energy efficiency.<sup>18</sup> Fabrication processes for silicon devices, including photolithography and etching, become increasingly complex at the nanoscale, resulting in higher production costs and longer manufacturing times.<sup>19</sup> Additionally, defects in silicon wafers can adversely affect device performance and reliability,<sup>20</sup> complicating detection and correction efforts. Current manufacturing equipment is often ill-suited for sub-5 nm technologies, further limiting scalability.<sup>21</sup>

Notably, silicon fabrication is highly energy-intensive, requiring up to 100 kW h per wafer—equivalent to a household's daily energy consumption in the USA.<sup>22</sup> This elevated energy requirement not only escalates manufacturing expenses but also amplifies concerns regarding sustainability. In addition, constraints related to materials and integration difficulties with organic materials hinder the flexibility and adaptability of SiO<sub>2</sub>-based materials to fulfil the requirements of evolving applications. Silicon technology is inherently limited in handling complex multi-input operations and direct integration with biological and

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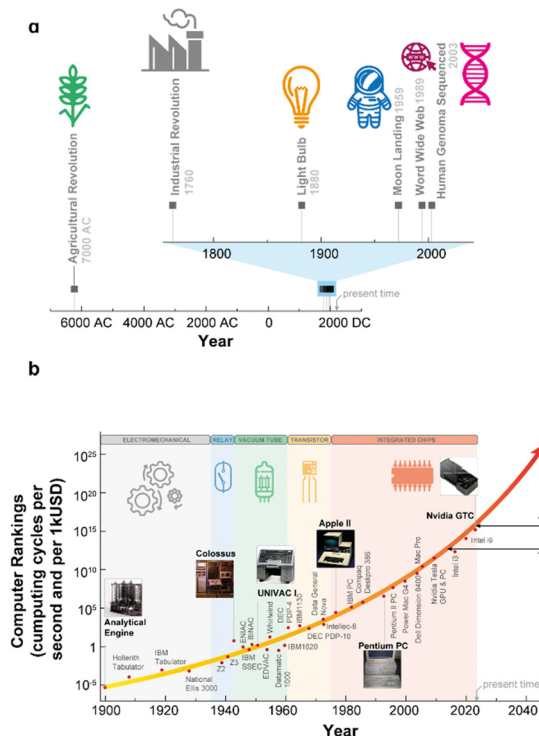


Fig. 1 (a) Timeline of key technological revolutions, from the agricultural revolution (~7000 AC) to modern human genome sequencing (2003), illustrating the exponential time scaling of the distinct technological breakthroughs. (b) Growth in computing power (cycles per second per 1000 USD) from early mechanical devices to modern integrated circuits, showing exponential gains.

chemical environments required for emerging applications.<sup>23</sup> The need for specialized instrumentation and exclusively electrical inputs hinders the deployment of silicon-based devices in dynamic, decentralized environments, particularly in sensing and diagnostic applications.

To address current and future challenges, the industry is transitioning beyond traditional CPU architectures, adopting new computational approaches like GPUs<sup>24</sup> and specialized hardware accelerators<sup>25</sup> to meet the growing demand for high-performance computing. GPUs leverage parallel processing architectures to perform multiple operations simultaneously,<sup>24</sup> approaching the prediction of Moore's law (Fig. 1b). Despite ambitious initiatives like the CHIPS for America in the USA<sup>26</sup> and CHIPS Act in the EU,<sup>27</sup> aimed at strengthening semiconductor production, they primarily focus on improving existing silicon-based technologies. Alternative computing approaches, such as quantum computing,<sup>28</sup> optical (or photonic) computing and molecular logic devices,<sup>29</sup> offer promising solutions that provide enhanced functionalities beyond the capabilities of traditional silicon systems (Fig. 2).

Molecular computing represents a crucial frontier for fostering further technological innovation,<sup>29–33</sup> exploiting changes in electron transfer properties, chemical reactivity, or optical features to process information.<sup>29,34</sup> The information processing in molecular logic is triggered by environmental stimuli, such as light, temperature, pH, or the presence of specific ions and molecules.<sup>31</sup>

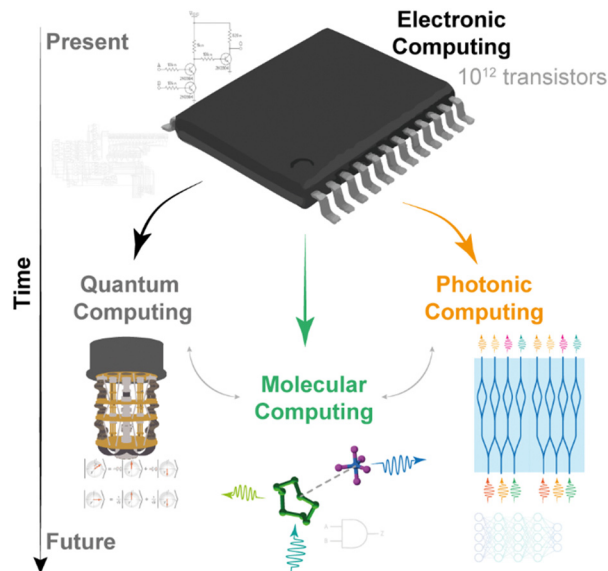
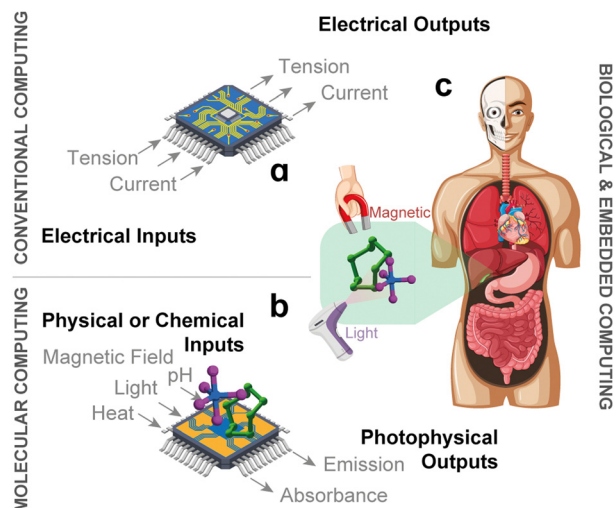


Fig. 2 Limitations of current silicon technology based on top-down lithography techniques are fostering the development of novel computing strategies such as quantum computing, photonic (or optical) computing and molecular computing.

The concept of molecular logic was first systematically introduced in the early 1990s by Prasanna de Silva and co-workers, who demonstrated that molecules could mimic Boolean logic operations.<sup>29,35,36</sup> His work established the foundation for molecular systems as computational units, demonstrating their capability to process information *via* chemical and physical interactions. Nowadays molecular logic devices can integrate sensing, diagnostics, and molecular-scale computation, effectively addressing the limitations of traditional silicon-based electronics.<sup>37,38</sup> Moreover, molecular logic devices can achieve ultra-sensitive detection at the molecular scale,<sup>39</sup> offering solutions that require less energy<sup>40</sup> and are responsive to a wider array of inputs.<sup>30,31,40</sup> Additionally, many molecular computing systems can integrate seamlessly into biological systems or function under challenging environmental conditions, which are critical in medical diagnostics, environmental monitoring, and smart materials.<sup>33,39</sup>

Mechanical computing is emerging as another alternative to computing, sharing a foundational principle with molecular logic: using system properties and interactions to process information.<sup>41</sup> Mechanical computing relies on nonlinearities and adaptable materials to perform logic operations, paralleling how molecular systems utilize physical or chemical stimuli to achieve logic functions. Both approaches emphasize distributed and environment-responsive computation, offering alternatives to traditional electronic systems (Fig. 3). Key differences lie in their operational scales and mechanisms: molecular computing operates at the nanoscale, leveraging quantum or molecular interactions, while mechanical systems function at macro or mesoscopic scales, emphasizing structural dynamics. Furthermore, molecular logic systems often achieve faster and more intricate processing *via* photonic or chemical inputs, whereas mechanical systems excel in robustness and integration with structural functionality.<sup>41</sup>





**Fig. 3** Schematic representation of different computing paradigms and their respective input–output relationships. (a) Conventional computing systems rely on electrical inputs and outputs, such as tension and current. (b) Molecular computing utilizes physical or chemical inputs (e.g., magnetic fields, light, pH, and heat) to generate photophysical outputs, such as emission and absorbance. (c) Biological and embedded computing extends these principles to unconventional environments, including the human body, where molecular logic operations can be actuated by physiological signals and external stimuli.

Although molecular logic devices hold significant promise, there remains a notable gap in standardized methodologies for characterizing luminescent molecules and nanoparticles for applications in information processing and storage. This work clarifies the core principles of luminescence-driven molecular logic, outlines essential experimental techniques, and provides guidance on interpreting luminescent signals as logic gate functions. By presenting standardized reporting protocols, we aim to make luminescence-based molecular logic more accessible, encouraging broader engagement and accelerating progress in this field.

## Principles of luminescence-based molecular logic

Among the unique characteristics of molecular materials that can be employed for developing molecular logic devices, luminescence is particularly powerful (Fig. 3b).<sup>33</sup> Light-emitting molecules and nanomaterials emit photons in response to specific stimuli, making them highly suitable for molecular logic applications.<sup>42</sup> Variations in their emission properties—such as intensity, colour, or lifetime—can be directly linked to distinct logical operations, allowing luminescence to act as an effective readout for the output of a molecular computing device.<sup>43,44</sup> The luminescence-based approach offers advantages over electronic ones, namely (i) a non-invasive readout as the optical nature of luminescence allows for remote monitoring without physical contact or disruption of the molecular system, (ii) multiplexing capabilities, as designing molecules or nanoparticles with different emission properties enables multiple logical operations to be performed simultaneously within a single

system, and (iii) the environmental sensitivity of luminescent systems can be engineered to respond to subtle changes in their environment, making them incredibly useful for sensing applications.<sup>31,33,45</sup>

Molecular-scale computation represents an especially promising domain, offering the capability to perform computational tasks at scales that are several orders of magnitude smaller than those achievable with current silicon technologies. Unlike conventional systems (Fig. 3a), molecular logic devices harness the unique properties of molecules to operate efficiently in highly challenging environments. These include the intricate and dynamic milieu of living cells, where biochemical complexity, sensitivity, and adaptability are paramount, and extreme industrial conditions characterised by high temperatures, corrosive environments, or intense mechanical stress, where silicon-based systems often fail.

The ability of molecular logic systems to integrate seamlessly with biological and chemical environments is particularly significant for real-time diagnostics, targeted therapies, and biochemical sensing, as illustrated in Fig. 3c. In contrast to conventional computing (Fig. 3a), which relies on electrical inputs and outputs, and molecular computing (Fig. 3b), which responds to physical or chemical stimuli, biological and embedded computing (Fig. 3c) extends these principles to complex environments, including the human body.<sup>46</sup> Here, molecular logic operations can be actuated by physiological signals or external stimuli, enabling precision monitoring of disease biomarkers and intracellular processes. Beyond biomedical applications, their resilience and adaptability make them well-suited for remote sensing, environmental monitoring, and autonomous systems in extreme or inaccessible locations such as space applications or isolated off-grid regions of our planet, where conventional electronics may fail.

### Molecular responses to stimuli

A distinguishing feature of molecular computing devices in comparison with electronic counterparts is their ability to respond to a broad range of stimuli beyond electrical inputs, such as chemical species, mechanical, thermal, magnetic stimuli or light. This makes molecular logic devices ideal for integration into wearable or implantable diagnostic devices, offering real-time, remote monitoring of specific analytes (e.g., disease markers or physiological parameters in biomedical sciences). Moreover, a significant advantage in energy efficiency has already been reported for reversible molecular mechanical logic gates, thereby aligning with Landauer's principle and enabling thermodynamically reversible computations even in the presence of thermal noise.<sup>47</sup>

From the fundamental viewpoint, chemical, physical, and mechanical factors can all influence the emission of luminescent molecules (Fig. 3b). The resulting changes in luminescence can be interpreted as binary outputs. Common examples include changes in ion concentration (called chemical inputs), temperature, pressure or exposure to electromagnetic radiation (called physical inputs). The molecules respond by three primary photophysical features: emission intensity (referring to the number of emitted photons), emission energy, also known



as emission colour (consisting of the dominant colour of the emission spectrum), and the lifetime of the emitting state (telling on the dynamics of the physical mechanisms responsible for the photon emission). Understanding these photophysical features is key to designing effective molecular logic devices.<sup>31,33</sup>

### Designing logic gates

At the core of molecular computing lies the concept of logic operations, defining how molecular systems process information. By exploiting changes in electron transfer, optical signals, and chemical reactivity, molecules can execute Boolean logic functions as traditional silicon-based circuits do. This section explores the fundamental logic gates implemented in molecular systems, particularly those based on luminescence. The simplest approach to developing luminescence-based molecular logic gates involves modulating the emission properties of a material by a set of quantifiable external stimuli.

The logic gate defines a functional relationship between inputs and outputs, operating according to specific rules. The current computing systems rely on Boolean algebra, operating on binary values – 0 and 1. Binary logic gates establish the connections between these logical inputs and outputs, forming the foundation for computational processes.<sup>33</sup>

To elucidate the functionality of molecular logic gates, consider a simple AND gate. This gate outputs a logical '1' only when all inputs are simultaneously present. For instance, in a luminescent molecular system, two distinct chemical species or environmental stimuli can act as inputs, while light emission represents the output. By applying both inputs simultaneously, the system's emission exceeds a predefined threshold, resulting in a binary '1'. If one or both inputs are absent, the output remains below the threshold, corresponding to a binary '0'. In summary:

**AND gate.** The AND gate produces a logical output of 1 only when all required inputs (e.g., two or more) are simultaneously present. In luminescent molecular systems, light emission occurs exclusively under the simultaneous presence of specific inputs, such as a combination of ions or environmental conditions, ensuring the fulfillment of the AND operation.

This modular approach underpins the design of increasingly complex molecular logic circuits. Besides the AND logic gate, common gates include:

**NOT gate.** The NOT gate inverts the binary input; 0 becomes 1 and 1 becomes 0. In molecular systems, this is typically realized through quenching mechanisms. For example, a luminescent molecule emits light under default conditions but ceases to do so when a specific input, such as a quencher ion, is introduced. This suppression of emission represents the logical inversion.

**OR gate.** The OR gate produces a logic output of 1 if at least one of the multiple inputs is present. In luminescent systems, this gate is implemented by designing molecules or materials that emit light in response to any one of several stimuli, such as the presence of an ion or the application of light, enabling the detection of any positive input.

**XOR gate.** The XOR gate outputs a binary 1 when only one of the inputs is 1, but not both. In luminescent molecular systems, this behavior is achieved by engineering molecules that emit light

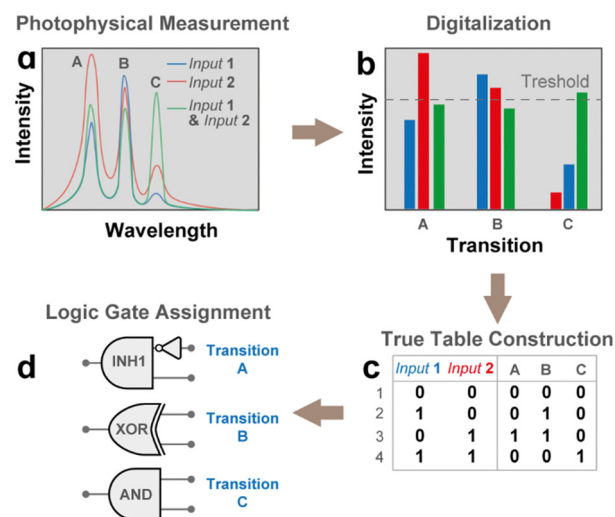
in response to a single input but suppress emission when both inputs are simultaneously applied. This precise modulation of emission ensures the exclusivity required for the XOR operation.

**INH gate.** The INH gate, or inhibitory gate, outputs a binary 1 only when a primary input is present, and a secondary inhibitory input is absent. In luminescent systems, this can be realized through a molecule that emits light when exposed to a specific stimulus (primary input) but has its emission suppressed in the presence of an inhibitory factor (secondary input), such as a quencher ion. This behavior makes the INH gate critical for selective and conditional logical operations.

Some molecular systems avoid the need to perform singular logic gate operations by directly executing complex computational tasks. These systems integrate multiple inputs and process them simultaneously, enabling the performance of intricate operations such as arithmetic functions, signal multiplexing, or combinatorial logic without the intermediate steps of singular gate functionalities. This capability stems from the inherent multifunctionality of molecular architectures, where changes in their emission properties can reflect the combined influence of various stimuli, effectively encoding higher-order logical operations within a single system.

## Reporting luminescent molecular logic devices

A fundamental challenge in molecular logic is converting raw photophysical data into unambiguous binary outputs that align with standard Boolean operations. The methodology, illustrated in Fig. 4, provides a systematic workflow of four steps



**Fig. 4** Schematic representation of molecular logic implementation using photophysical measurements. (a) The luminescence spectra are measured under distinct environmental conditions, corresponding to different inputs. (b) It is digitized based on an intensity threshold, allowing the assignment of logic gates to specific transitions. (c) The resulting truth table correlates photonic inputs with Boolean logic operations, demonstrating the feasibility of molecular logic processing. (d) The logic gate assignment follows the conventional logic circuitry, allowing a direct comparison with conventional electronic gates.



for designing, characterizing, and validating luminescent molecular logic systems.

### 1. Photophysical measurement

The process begins with the photophysical measurement of the molecular system under various controlled input conditions such as changes in temperature, ion concentration, pH, or the application of multiple excitation wavelengths (Fig. 4a). Emission spectra are recorded using common spectroscopic techniques, which yield intensity-*versus*-wavelength data for each combination of inputs. In these measurements, distinct emission peaks (denoted as transitions A, B, and C in Fig. 4a) correspond to different electronic transitions, components of the same transition or emission originating in distinct luminescent centres within. The instrument calibration and baseline corrections are key to ensure that the recorded signals accurately reflect the response of the luminescent systems.

### 2. Digitalization of inputs and outputs

Following the collection of emission spectra, the continuous intensity values are transformed into discrete binary outputs through digitalization. This critical step involves establishing threshold values derived from calibration experiments and statistical noise analysis (Fig. 4b). In our approach, the threshold for each photophysical parameter is defined as at least two times the uncertainty in the determination of that parameter. Emission intensities that exceed the determined thresholds are assigned a logical '1', while those falling below are designated as '0'. It is important to note that if the measured intensity falls within the uncertainty range, the logic output of the system is ambiguous and thus is considered undefined. The method described here minimizes the impact of experimental noise and variability, ensuring that analogue signals are reliably converted into a digital format compatible with Boolean logic.

### 3. Truth table construction

Once the binary outputs are determined through digitalization, the next step is to organize these results into a comprehensive truth table. In this table, every possible combination of input conditions is listed in a systematic manner alongside the corresponding binary outputs for each luminescent transition. (Fig. 4c). Each row in the truth table represents a unique set of experimental conditions (*e.g.*, the presence or absence of specific stimuli), and documents the resulting output for transitions A, B, and C. This detailed mapping provides a clear, structured overview of the logic response of the material. Repeated experiments under identical conditions should yield the same row entries, confirming the reproducibility of the response of the molecular system. Moreover, any deviations in the expected output can be readily identified and further investigated, allowing for refinement of the threshold settings or other experimental parameters.

### 4. Logic gate assignment

In the final step, the binary output patterns observed in the truth table are matched with canonical Boolean logic gates

(Fig. 4d) such as AND, OR, NOT, XOR, and INH. For instance, if the luminescence at a given transition occurs only when all inputs are present, the system is assigned an AND gate functionality; if luminescence is observed when at least one input is present, the behaviour is analogous to an OR gate. It is not uncommon for different transitions within the same system to correspond to distinct logic gates, thereby allowing for parallel information processing. Validation is achieved by performing repeated measurements under slightly varied conditions, which confirms the reproducibility and robustness of the assigned logic functions (Fig. 4d).

### Stability of logic operations

Understanding the relationship between thermal fluctuations and the stability of molecular logic operations is crucial for the practical deployment of luminescent logic devices. To comprehensively characterize this relationship, additional analyses such as activation energy calculations and temperature sensitivity studies should be carried out. Activation energies can be calculated using Arrhenius-type analyses, where the emission intensity (or the rate constants) associated with the luminescent transitions are measured across a range of temperatures. By plotting the logarithm of the observed intensities (or rates) against the inverse of temperature (so-called Arrhenius plots), the energy barriers for the transitions governing the logic switching processes can be accurately determined by its slope. Higher activation energies indicate increased robustness of the molecular logic against thermal variations, providing a useful metric to evaluate the suitability of specific materials for real-world applications.

Moreover, systematic temperature sensitivity studies are essential to validate the stability of logic gate functionality. Such studies involve measuring luminescence intensities at multiple temperatures over the desired operational range. These measurements enable the assessment of whether the binary output signals ('0' or '1') remain stable or become ambiguous as temperature fluctuates. Thresholds should be carefully defined, typically at least twice the experimental uncertainty, to reliably discriminate between logical states. If luminescent responses fall within the defined uncertainty range, the logical output should be considered ambiguous and not clearly defined, highlighting conditions under which the system may fail to reliably perform logic operations.

## Luminescent molecular logic devices

With established protocols for reporting luminescent molecular logic, researchers have developed various innovative applications. Recent studies have demonstrated the versatility of molecular logic in fields ranging from biosensing to information encryption. The following section highlights key advancements in the field, illustrating how different materials and molecular architectures contribute to functional logic devices.

The study of molecular logic gates has traditionally centered on synthetic molecules, but fluorescent natural products offer

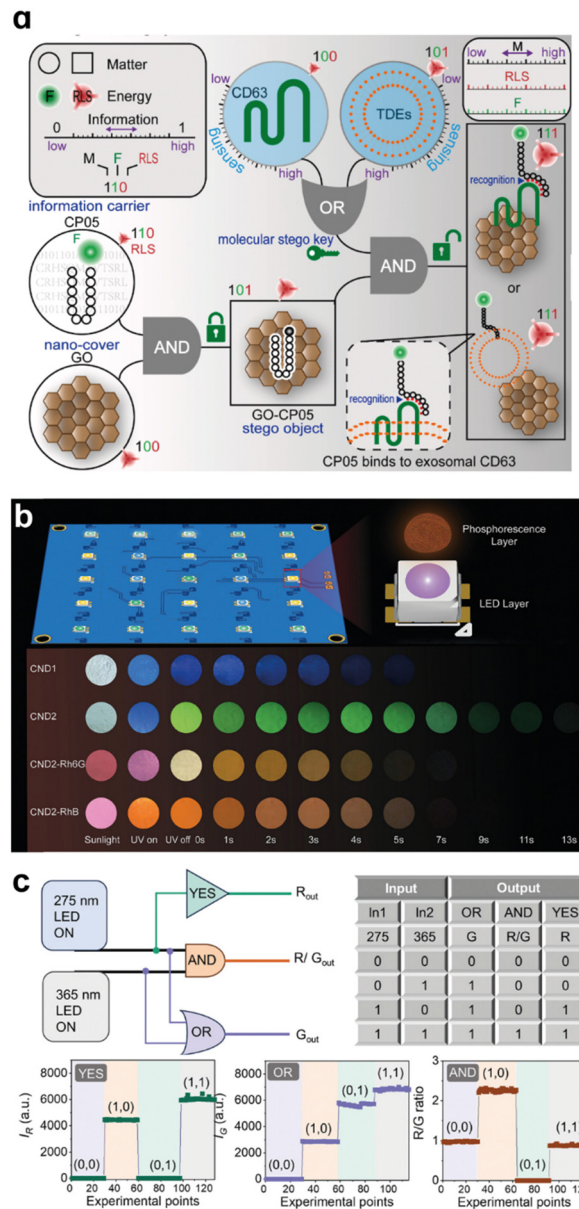


an untapped reservoir for these applications. Agius & Magri showcase that many naturally occurring fluorophores, such as quinine and calcimycin, exhibited logic behaviours like INHIBIT and NOR gates before the formal definition of these concepts.<sup>48</sup> Leveraging natural products can save synthetic effort and inspire new strategies for molecular logic gate design. This includes directly identifying intrinsic molecular architectures, modifying fluorescent cores through semi-synthetic approaches, and synthesizing gates from natural product components. Exploring this avenue can uncover overlooked fluorescent switches and logic functions, bridging natural and synthetic systems for molecular computing.<sup>48</sup>

In a significant advancement in the integration of natural and synthetic systems, Lue *et al.*<sup>49</sup> developed an innovative peptide-graphene logic sensing platform for the dual-channel detection of tumour-derived exosomes and their marker CD63 (Fig. 5a). This work exploits the sequence-dependent properties of peptides alongside the exceptional electrical conductivity and computational capacity of graphene oxide. By combining these elements, the authors engineered a hybrid system capable of executing multiple logic functions—ranging from simple gates such as YES, AND, OR, and INHIBIT to more complex circuits that operate in parallel or batch modes. Moreover, the platform extends its utility beyond sensing applications by demonstrating its efficacy in molecular cryptography and steganography, where the peptide-graphene interactions are harnessed for secure encoding and information hiding. This approach not only represents a breakthrough in disease diagnostics but also lays the groundwork for integrated systems in intelligent sensing, biomedicine, and advanced information technology by leveraging the inherent versatility of peptides and the powerful processing capabilities of graphene-based materials.

Parallely, Zhang *et al.*<sup>50</sup> reported the development of photochromic diarylethene molecules that can reversibly switch between two isomeric states under UV and visible light exposure, thereby enabling optical data encoding (Fig. 5b). Their study highlights the precise control of molecular switching when these diarylethene molecules are immobilized on polymer beads, a configuration that permits reversible writing, reading, and erasing of information at the microscale. Through selective addressing of individual beads and optimization of the illumination parameters, the authors achieved high contrast and enhanced photostability—critical factors for reliable photonic logic operations. This strategy opens up new avenues for the design of advanced photonic logic circuits that utilize light for information processing, offering a scalable and energy-efficient approach to molecular computing. Together, these studies not only illustrate the diverse methodologies employed in the field of molecular logic but also underscore the potential for integrating different molecular architectures to achieve sophisticated and multifunctional logic systems.

Exploiting inorganic materials Wang *et al.* reported significant advancements in the development of dual-emission zone semiconducting materials.<sup>51</sup> The single crystal  $[\text{NH}_3(\text{CH}_2)_2\text{NH}_3]_2\text{ZnCl}_6$  exhibiting multiple emissions under different UV excitation wavelengths (275 nm and 365 nm), generate unique emission profiles in the visible spectral range attributed to different luminescence



**Fig. 5** (a) Schematic illustration of peptide-graphene logic sensing system for dual-mode detection of CD63 and TDEs, molecular logic, and information protection. CP05 as a recognition probe and information carrier, is combined with GO as a nano-quencher and nano-cover to form a nanocomplex (stego object). When CD63 or TDEs (molecular stego key) is added, the fluorescence and RLS responses can be used for quantitative detection of CD63 or TDEs, molecular logic and information coding, encryption and hiding. Reproduced with permission from ref. 49 Copyright 2024 Elsevier. (b) Schematic illustration of the programmable information encryption circuit system with an enlarged view. (b) Images of four CND powders under 365 nm UV lamp on and off with time. Reproduced with permission from ref. 50 Copyright 2024 Wiley. (c) Design principle of all-optical logic gate operations based on multicolour emission performance of  $[\text{NH}_3(\text{CH}_2)_2\text{NH}_3]_2\text{ZnCl}_6$ . Schematic of the logic AND, OR, YES gates with all-optical inputs and outputs, together with the corresponding truth tables. The bottom row shows the emission intensity for four input states of the logic YES, OR and AND logic gates. Reproduced with permission from ref. 51 Copyright 2024 Wiley.

centres, including charge transfer states and  $\text{Mn}^{2+}$  ion transitions. This unique emission profile was explored to develop logic gates performing fundamental logical operations such as “YES,” “AND,” and “OR” (Fig. 5c).<sup>51</sup> The logic gates are driven by the intensity of red and green emissions or their intensity ratio, transforming optical signals into binary logic outputs. By dynamically controlling input signals with UV light and tracking the interaction of light with objects, such as the shape and transit time of a toy car, the system was able to process information in real time. These findings demonstrate the potential of these materials for advanced image processing applications.<sup>51</sup>

Trivalent lanthanide ions ( $\text{Ln}^{3+}$ ) have emerged as highly effective active centres in luminescent logic devices due to their unique optical properties. These ions exhibit sharp, well-defined emission bands, typically in the visible and near-infrared regions, owing to their characteristic f-f electronic transitions.

Additionally, they possess long-lived excited states, often with lifetimes exceeding 1 ms, which enable clear, distinguishable signals for logic operations. The ability of  $\text{Ln}^{3+}$  ions to emit in ultraviolet, visible and near-infrared spectral ranges with narrow emission bands (<10 nm) further enhances their suitability for applications in molecular logic, where precise control over optical signals is required. Moreover, when combined with ligands the  $\text{Ln}^{3+}$  ions present high emission quantum yields, that can exceed 0.8. Their versatility in response to various stimuli, such as light, ions, or chemical changes, makes them ideal candidates for implementing logic gates in molecular systems.

Different molecular logic devices can be found in the literature, exploiting both chemical and physical stimuli, that have been recently reviewed.<sup>33</sup> As the field is under constant development, new examples reporting the development of luminescent logic gates and molecular equivalents to electronic devices such as filters and transistors were published and will be revised next. Practical implementations of luminescent logic gates have demonstrated the versatility of these systems. For instance, luminescent materials doped with trivalent lanthanide ions have been used to create temperature-dependent reconfigurable gates. Under controlled excitation conditions, these systems transition between AND and XOR operations based on thermal stimuli. Such adaptability highlights the potential for molecular logic in dynamic and multi-functional computing environments.<sup>30</sup>

Our group reported a reprogrammable molecular logic system based on a  $\text{Ln}^{3+}$ -doped organic-inorganic di-ureasil hybrid material ( $\text{dU6EuTb}$ ).<sup>52</sup> Unlike previous works of the same group requiring two fixed temperatures (14 and 298 K), this approach utilizes the temperature-dependent transfer function ( $H$ ) relative to the detection frequency ( $f$ ), offering greater flexibility in defining logic operations (Fig. 6a). In this system, the output changes in response to temperature, with  $H$  increasing when the temperature is above the characteristic temperature ( $T_c$ ) and decreasing below it. This behaviour enables the realization of different logic gates: in the regime where  $T < T_c$ , the system operates as a NOT gate, while in the  $T > T_c$  regime, it mimics a PASS 1 gate (Fig. 6b). The key breakthrough lies in the ability to modulate the functionality of the logic gates through temperature control,

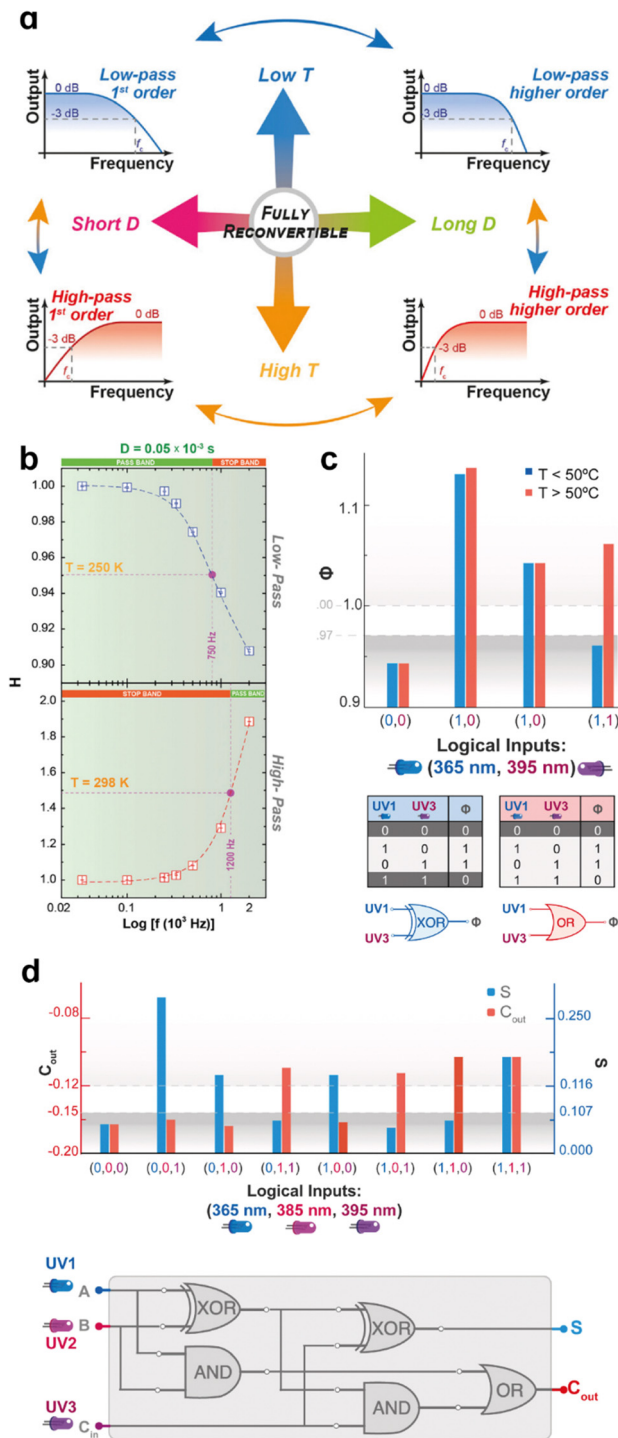


Fig. 6 (a) Flowchart scheme for designing molecular filters. (b) Dependence of  $H$  with the frequency in semi-logarithm scale for at 250 and 298 K. Reproduced with permission from ref. 52 Copyright 2023 Wiley. (c) Bar plot of the logic output for the several excitation wavelengths. The horizontal lines define the threshold values. Truth table and corresponding logic gates are shown at the bottom. (d) Bar plot of the logic outputs  $S$  and  $C_{out}$  for the several inputs. The lines mark the threshold values. The diagram representation of the Full-Adder is presented at the bottom. Reproduced with permission from ref. 53. Copyright 2025 Elsevier.



allowing for two distinct operational modes with a single material. This innovation significantly enhances the practical applicability of molecular devices in real-world scenarios, as it moves beyond temperature-specific solutions to a more versatile, reprogrammable platform. The work demonstrates how molecular materials, particularly those doped with lanthanide ions, can be engineered to perform complex computational tasks, pointing toward future developments in molecular photonics and molecular computing systems.<sup>52</sup>

In the present year, Salgueiro *et al.* reported  $\text{Eu}^{3+}$  and  $\text{Dy}^{3+}$ -doped glass for developing molecular logic systems actuated by physical inputs.<sup>53</sup> Exploiting the luminescence properties of these lanthanide ions under various excitation wavelengths and temperatures, the authors constructed a diverse array of logic – gates. Specifically, they implemented basic gates like AND and OR (Fig. 6c), as well as more complex systems such as full adders (Fig. 6d) and full subtractors. This work represents the first instance of using  $\text{Eu}^{3+}$  and  $\text{Dy}^{3+}$  ions exclusively for molecular logic *via* physical stimuli, showcasing their robust and stable optical properties. Distinct excitation pathways were studied for each ion, allowing for selective excitation. Recording the steady-state emission spectra of the  $\text{Dy}^{3+}/\text{Eu}^{3+}$  co-doped oxide glass between 30 °C and 60 °C the emission intensities quench. Ten photonic outputs, defined on the base of diverse combinations of the integrated areas were applied to define various logic gates using the photoluminescence properties of  $\text{Eu}^{3+}$  and  $\text{Dy}^{3+}$ -doped glasses.

The logic gates were defined by converting continuous photophysical outputs into binary logic values. Basic logic gates such as AND, OR, XOR, and INH were constructed using different combinations of excitation wavelengths and temperature as inputs (Fig. 6d). Using LED excitation with peak emissions at 365 nm and 395 nm as inputs, the system behaved as an XOR gate below 55 °C and as an OR gate above 55 °C, yielding temperature reconfigurable logic gates. Beyond basic logic gates, full adders and full subtractors were also reported (Fig. 6d). The operational temperature range for these circuits was between 40 °C and 55 °C, highlighting the potential of  $\text{Eu}^{3+}$  and  $\text{Dy}^{3+}$ -doped glasses for constructing stable and versatile molecular logic systems.<sup>53</sup>

### Comparative analysis of luminescent molecular logic materials

Several series of luminescent molecular logic materials have been explored, each offering unique advantages and presenting distinct challenges.

Pure organic materials are derived from well-characterized organic fluorophores, offering significant synthetic versatility and tunable optical properties at relatively low cost.<sup>29,40</sup> Their chemical structure can be modified to optimize luminescence and logic functionality. However, these materials can be prone to photobleaching and may exhibit limited stability under harsh conditions, which can restrict their long-term performance in practical applications.

Inorganic–organic hybrids combine the merits of both organic and inorganic components. The integration of inorganic elements often enhances chemical and thermal stability and can improve optical performance through synergistic

interactions.<sup>30</sup> Yet, the synthesis of these hybrid materials is generally more complex, and issues such as phase separation or interfacial incompatibility between the organic and inorganic parts may arise, potentially affecting device consistency.

Nanomaterials, including semiconductor quantum dots and carbon nanodots, offer high brightness, broad absorption spectra, and narrow, well-defined emission profiles.<sup>45</sup> Their nanoscale dimensions facilitate integration into miniaturized devices and enable multiplexing capabilities for complex logic operations. Despite these benefits, concerns regarding toxicity, limited biocompatibility, and challenges in controlling surface chemistry can hinder their widespread adoption, especially in biological applications.<sup>46</sup>

Lanthanide ion-based systems rely on the unique photophysical properties of lanthanides, such as sharp emission bands, long excited state lifetimes, and high luminescence efficiency when appropriately coordinated.<sup>33</sup> These attributes make them particularly attractive for applications that require precise spectral resolution and energy transfer mechanisms. However, the practical implementation of lanthanide-based logic devices often demands sophisticated ligand design and synthetic procedures to overcome issues like inherently low absorption cross-sections and potential quenching effects.<sup>45</sup>

Each class of materials thus presents a trade-off between factors such as synthetic complexity, stability, optical performance, and biocompatibility. By understanding these differences, researchers can tailor the choice of luminescent molecular logic material to the specific requirements of their intended application, whether it be in biosensing, data encryption, or the development of advanced computing systems.

One should note that luminescent molecular computing devices represent one of several emerging paradigms in the broader landscape of molecular computing, competing notably with DNA computing and semiconductor quantum-dot-based logic systems. Each of these paradigms offers distinctive advantages and faces specific challenges, making their comparison particularly relevant for guiding future research and technological development.

DNA-based logic systems capitalize on the intrinsic programmability, biocompatibility, and parallel processing capabilities inherent to nucleic acids. They offer unmatched molecular recognition specificity and have demonstrated sophisticated logical and computational functions. However, DNA computing typically requires carefully controlled aqueous conditions and complex enzyme-assisted processes for logic operations, which may limit their broader applicability in harsh or dynamic environments. Furthermore, readout methods often involve fluorescence labeling or electrophoresis, presenting challenges in miniaturization and integration with existing electronic systems.<sup>54–56</sup>

Quantum-dot-based logic devices benefit from their exceptional optical properties, including tunable emission spectra, high brightness, and excellent photostability. Their optical signals provide robust multiplexing potential for complex logic operations, which is advantageous in multiplexed sensing and high-density data storage. However, quantum dots often pose concerns regarding cytotoxicity, stability, and reproducibility of



synthesis, alongside difficulties in consistent surface functionalization, raising questions about their biocompatibility and suitability for biological or environmental applications.<sup>57,58</sup>

In contrast, luminescent molecular logic devices—particularly those based on lanthanide ions and hybrid organic–inorganic materials—combine several strengths of these competing systems while addressing some of their critical limitations. They offer stable, sharp emission bands, long excited-state lifetimes, high luminescence quantum yields, and versatility in responding to diverse physical and chemical stimuli.<sup>33</sup> Importantly, these systems typically exhibit improved stability and reproducibility under variable environmental conditions. Their optical readout is non-invasive, does not require direct electrical connections, and allows easy integration into existing optical detection platforms. Additionally, their inherent multiplexing capability through emission wavelength or lifetime modulation enables parallel processing of multiple signals simultaneously, advantageous for complex, real-time sensing applications.<sup>45</sup>

## Conclusions

The advancements in luminescent molecular logic underscore its potential as a viable alternative to traditional silicon-based computation. By leveraging light-driven processes and molecular-scale interactions, these systems pave the way for energy-efficient, adaptive, and multifunctional computing paradigms. As research progresses, standardization, scalability, and integration with existing technologies will be crucial in translating these molecular devices into real-world applications.

Molecular logic represents a groundbreaking approach to designing computational systems at the molecular scale. By responding to diverse stimuli—including chemical, thermal, magnetic, and optical inputs—these systems offer a level of flexibility and energy efficiency that surpasses traditional silicon-based electronics. Their ability to process multiple signals simultaneously and execute logic operations in a thermodynamically efficient manner positions them as key enablers of next-generation computing technologies.

A particularly promising class of molecular logic devices is based on luminescent materials, where lanthanide ions serve as active centres, providing highly reliable and well-defined optical signals for logic operations. By modulating emission properties such as intensity, energy, and lifetime, these systems can replicate classical Boolean logic gates (AND, OR, NOT, XOR, *etc.*), making them suitable for a wide range of applications, from portable diagnostics to advanced information processing. Furthermore, integrating molecular design with natural fluorophores, inorganic semiconductors, and hybrid materials opens new avenues for sustainable, cost-effective, and multifunctional logic architectures. Notably, some molecular systems can perform computational tasks directly, eliminating the need for intermediary logic gates and simplifying circuit design.

The future of luminescent molecular logic devices is marked by exciting opportunities for integration, miniaturization, and the development of intelligent biosensing platforms. Recent

advances are beginning to bridge the gap between proof-of-concept studies and practical applications in fields such as diagnostics, environmental monitoring, and security.<sup>54,55</sup> A recent contribution exemplifies this trend by demonstrating a fully integrated platform that couples advanced nanomaterials with molecular logic operations, achieving real-time processing of multiple biochemical signals with high sensitivity and specificity.<sup>59</sup>

Building on such breakthroughs, future research will increasingly focus on systems that harness multi-input molecular logic cascades—such as AND–OR configurations—for enhanced biosensing and environmental monitoring. Recent literature examples illustrate that multi-input logic systems can process complex signal patterns and simultaneously analyse several biomarkers or analytes. This capability not only improves sensitivity and selectivity but also offers significant advantages over conventional single-signal systems by mitigating background noise and enabling robust error correction. The superior performance and flexibility of these cascades will be pivotal in developing advanced diagnostic platforms and environmental sensors capable of real-time, high-fidelity signal processing.<sup>59</sup>

Moreover, further miniaturization is expected to yield portable and implantable devices, critical for point-of-care diagnostics and personalized medicine. Key challenges include optimizing signal-to-noise ratios through improved threshold definitions and noise reduction techniques, enhancing surface immobilization methods to preserve the activity and stability of the molecular components, and developing robust error-correction strategies for multi-signal networks. Additionally, integrating luminescent molecular logic devices with conventional electronic transducers could give rise to hybrid systems that combine the specificity of biochemical interactions with the rapid processing capabilities of digital electronics, thereby creating platforms that interface seamlessly with existing technologies while maintaining low-power, high-fidelity performance. Furthermore, the development of smart, adaptive systems that incorporate stimuli-responsive actuators offers a pathway toward fully integrated sensor-actuator networks. Such systems could, for example, trigger therapeutic responses or environmental remediation actions upon detecting specific biochemical signals, ultimately leading to next-generation “sense/act/treat” devices.<sup>59</sup>

Despite recent advances, molecular logic remains an emerging field, with current research largely focused on proof-of-concept devices rather than fully integrated molecular processors. However, these challenges also present opportunities for innovation, particularly in bridging the gap between laboratory-scale demonstrations and real-world applications.

As the field evolves, molecular logic devices are poised to drive a paradigm shift in computing technologies. The integration of photonic inputs offers exciting prospects for novel applications in computing, energy, health, and data storage. Recent progress in energy transfer mechanisms, computational modelling, and molecular gate design underscores the potential of these systems to address key limitations of conventional electronics.

Equally important is the role of new researchers in advancing the field. The fresh perspectives and innovative methodologies



brought by early-career scientists, including PhD students and postdoctoral fellows, are essential for driving breakthroughs and shaping the future of molecular computing. Moving forward, the successful translation of molecular logic devices into practical, scalable, and sustainable technologies will require strong interdisciplinary collaboration between experienced researchers and newcomers alike.

## Author contributions

C. D. S. Brites: conceptualization, data curation, funding acquisition, project administration, visualization, and writing – review & editing.

## Data availability

No new data were created or analysed during this study. Data sharing is not applicable to this article.

## Conflicts of interest

There are no conflicts to declare.

## Acknowledgements

This work was developed within the scope of the project CICECO-Aveiro Institute of Materials, UIDB/50011/2020, UIDP/50011/2020, and LA/P/0006/2020, and LogicALL (PTDC/CTM-CTM/0340/2021) financed by Portuguese funds through the FCT/MECI (PIDDAC).

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