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## Beyond silicon: a roadmap for overcoming the five challenges (5Cs) in molecular computing

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Molecular electronics, which utilizes individual molecules as electronic components, presents a promising alternative to traditional transistors, especially as we approach the physical and economic limitations of silicon technology. This “bottom-up” approach offers several advantages over the conventional “top-down” methods employed in silicon-based transistors over the past seven decades. However, constructing a molecular computer from individual molecular components introduces unique challenges that must be addressed. This review highlights five critical challenges in molecular computing, collectively called the 5Cs: concatenation, connectivity, crosstalk, compatibility, and cost-effectiveness. For each challenge, we outline the current technological issues, discuss the latest advances, and explore future research directions. We conclude by proposing a potential roadmap for developing the world’s first molecular computer.

## Introduction

The latest developments in artificial intelligence (AI) and machine learning (ML) are fuelling increasing demands for computing power, pushing the limits of current silicon-based technology.<sup>1</sup> At the heart of every computer is the integrated circuit (IC), commonly known as a microchip, first developed by Kilby and Noyce in 1958.<sup>2</sup> An IC comprises a densely packed array of silicon semiconductor transistors performing complex calculations.<sup>3</sup> Gordon Moore stated in his 1st law that the number of transistors on an IC doubles every two years, this has been historically accurate (red line in Fig. 1A).<sup>4</sup> Miniaturization of silicon-based transistors has kept Moore’s 1st law alive despite significant technological obstacles.<sup>5</sup> Moore’s less well-known 2nd law (also known as Rock’s Law) states that the cost of IC fabrication doubles every four years<sup>6</sup> (blue line Fig. 1A), which has been observed over the last 50 years. The latest IC fabrication facilities range from \$10 billion to \$25 billion due to the complexity of generating sub-5 nm transistors.<sup>7</sup> Current fabrication involves a top-down approach (Fig. 1B), etching the silicon surface using extreme ultraviolet (EUV) lithography.<sup>8</sup>

A single EUV lithography machine is approximately \$400 million, and the global supply of these critical systems is exclusively controlled by a sole Dutch company ASML.<sup>9,10</sup> Furthermore, advanced IC manufacturing is highly localised with over 75% of the global supply of ICs produced in Asia (Fig. 2A and B).<sup>11</sup> The surging demand for artificial intelligence

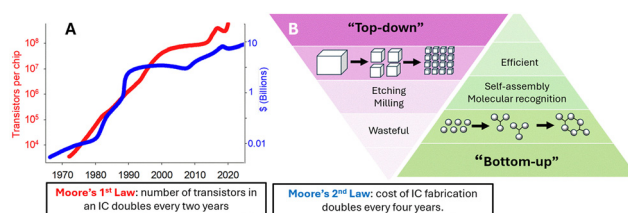


Fig. 1 Average number of transistors per IC (red line) and the estimated cost for IC fabrication facility (blue line) between 1970–2020 (panel A), “top-down” and “bottom-up” approaches to manufacturing (panel B).

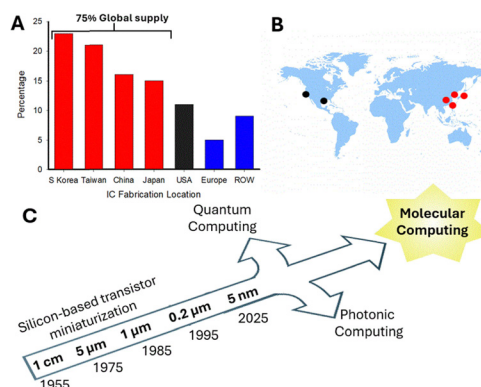


Fig. 2 75% of the global supply of advanced ICs are produced in Asia (panel A and B). The miniaturization of silicon transistors over the past 70 years and three possible successors (panel C).



critical semiconductor technologies.<sup>11,12</sup> This is exemplified by the recent CHIPS Act in the United States of America and the European Chips Act to secure supplies of these critical resources.<sup>12,13</sup> Alongside the financial implications of transistor miniaturization, the limits of silicon are rapidly approaching. As transistor size is reduced, reliability is impacted due to a variety of factors including quantum tunnelling,<sup>14</sup> leakage currents,<sup>15</sup> heat dissipation, and higher contact resistance.<sup>16</sup> A paradigm shift is inevitable, with quantum, photonic, and molecular computing emerging as potential successors to the silicon transistor (Fig. 2C).

This review will focus on the potential of single molecules to replace silicon-based transistors. This approach offers several advantages over existing technology however, not without unique obstacles of its own. We will introduce the early pioneers in this field, the first examples of molecular electronic components, and introduce the five key challenges facing molecular computing. Each challenge will be outlined with a brief description of the issue to be resolved, followed by the latest research in this area. Outstanding issues, current directions of travel, concluding with a possible roadmap for the world's first molecular computer, will be discussed.

## Molecular electronics

Richard Feynman proposed the concept of manipulating matter on the molecular scale to generate molecular devices in his 1959 lecture "There's plenty of room at the bottom" inspiring the field of nanotechnology.<sup>17</sup> One subset of nanotechnology is molecular electronics, which aims to replicate electronic components on the molecular scale.<sup>18</sup> This "bottom-up" approach offers several advantages over the traditional top-down approach to electronics. Aviram and Ratner proposed a single molecule, **1**, as an electronic rectifier, a device that converts alternating current into direct current in 1974.<sup>19</sup> Molecule **1** contained an electron-rich and an electron-poor region separated by a bridging sigma unit (Fig. 3A). Application of voltage results in electrons tunnelling across the molecule from electron-rich to electron-poor. Reed *et al.* demonstrated the first example of a single molecule conducting electrons while suspended between two gold electrodes discovering the first molecular wire **2** (Fig. 3A).<sup>20</sup> A single molecule transistor composed of a single C<sub>60</sub> between two gold electrodes, **3**, was reported in 2000.<sup>21</sup> Numerous other electronic devices have been replicated using single molecules over the past 50 years, validating the potential of molecular electronics (Fig. 3B).<sup>22</sup>

Silicon-based transistors serve two fundamental roles in integrated circuits: acting as switches that alternate between ON and OFF voltage states and functioning as amplifiers that increase signal strength. Applying a small voltage (1 V in Fig. 4A) to the base allows a higher voltage (5 V in Fig. 4A) to flow to the collector, switching a transistor to the ON state. The ON state carries a higher voltage, amplifying a smaller signal into a larger one. The OUTPUT of one transistor forms the INPUT for another, allowing multiple transistors to be combined to form complex circuits. In 1847, George Boole developed a system of

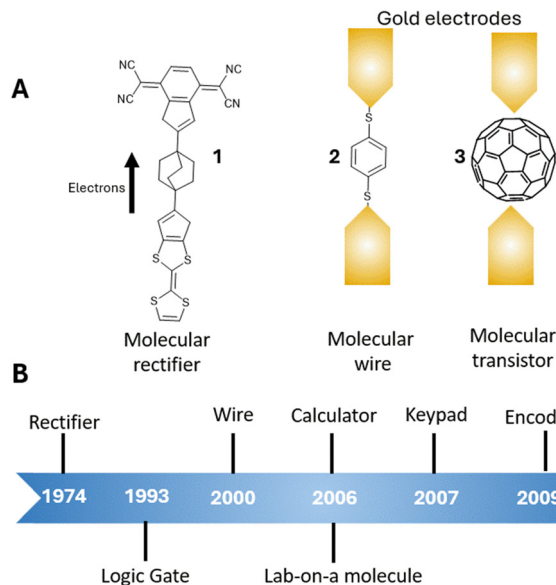


Fig. 3 Molecular electronic examples reported from 1974 to 2009.

binary (0 or 1) algebra, forming the foundation of logic gates.<sup>23</sup> Logic gates trigger an OUTPUT only with specific INPUT signals. Logic gates serve a critical function in electronic devices, enabling complex calculations to be performed. Simple molecules replicate this function, for example, De Silva *et al.* reported the first molecular AND gate **4**, which provides an OUTPUT of 1 if both INPUT 1 (H<sup>+</sup>) and 2 (Na<sup>+</sup>) are present (Fig. 4B).<sup>24</sup>

Pyrazoline OR gate **5** will only provide an output if INPUT 1 (Ca<sup>2+</sup>), INPUT 2 (Mg<sup>2+</sup>) or both are present (Fig. 4B).<sup>25</sup> The first inhibit gate **6** gives an OUTPUT with INPUT 2 (O<sub>2</sub>) only; if INPUT 1 (H<sup>+</sup>) is also present, then INPUT 1 inhibits the gate.<sup>26</sup> The opposite of an AND gate is a NAND gate, for example, **7**, in which an OUTPUT of 1 is always present until both INPUT 1 (H<sup>+</sup>) and 2 (O<sub>2</sub>) are 1 (Fig. 4B).<sup>27</sup> Molecule **8** requires three specific INPUT signals in a predefined sequence to trigger an OUTPUT (Fig. 5).<sup>28</sup> Molecule **8** is an example of a molecular-scale keypad lock<sup>29</sup> demonstrating that molecular logic gates can perform complex functions. This is further confirmed for molecule **9**, which detects multiple analytes concurrently and was the first example of a "lab-on-a-molecule" (Fig. 5).<sup>30</sup> In summary an AND gate will only trigger an OUTPUT when all INPUTS are present, an OR gate will activate with one or more INPUTS and an INH gate will trigger in the presence of the required INPUT, but only in the absence of any inhibitory INPUT. These highly versatile building blocks are used for more complex molecular device construction. For example, molecules that perform simple arithmetic, referred to as "molecular calculators",<sup>31</sup> encoders, decoders, multiplexers, and encryption devices have all been mimicked using single molecules. These early pioneers have gifted the world with a molecular toolkit of electronic components for even more complex device construction. The ultimate goal of molecular electronics is the development of the world's first fully functional molecular computer ushering in a new paradigm in computing. However, five major



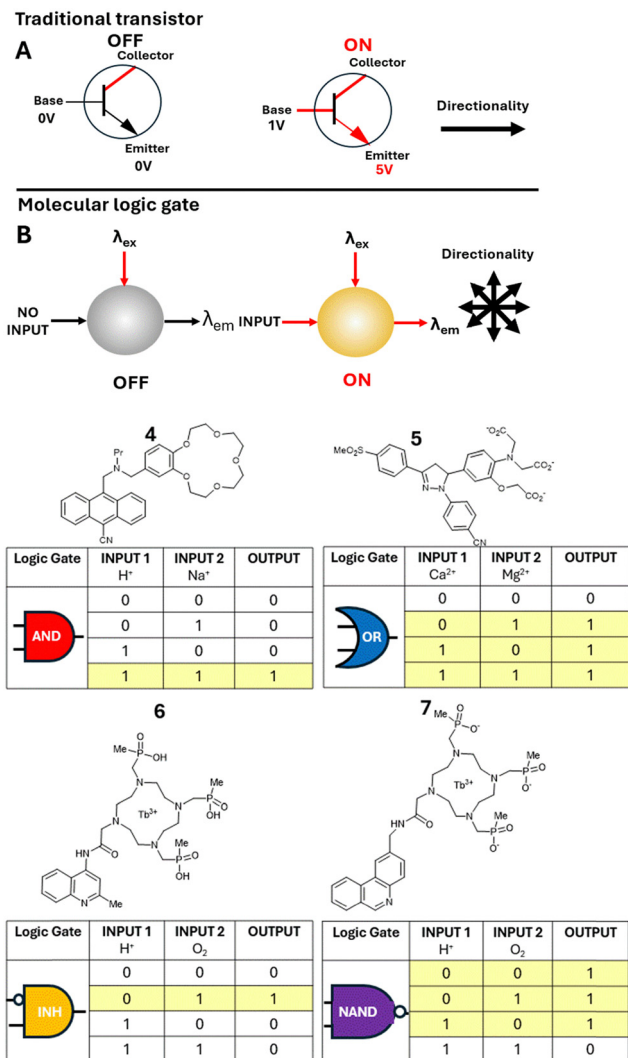


Fig. 4 The first reported molecular AND gate **4** in 1993, OR gate **5** in 1994, INHIBIT gate **6** and NAND gate **7**.

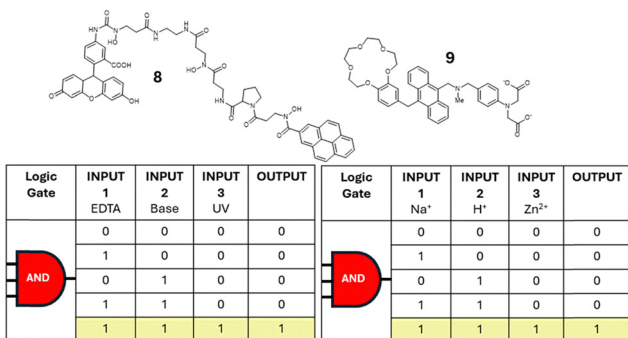


Fig. 5 The first reported molecular keypad lock **8** and "lab-on-a-molecule" **9**.

challenges must be overcome, we will outline each with a brief description of the problem, followed by the latest research in this area. Outstanding research gaps, current directions of travel, and a potential roadmap will be outlined.

### Challenge one: concatenation

Electrons are both INPUT and OUTPUT for silicon-based transistors, enabling the OUTPUT of one transistor to act as INPUT for another. Electrons have the advantage of moving in a single direction, allowing multiple transistors to be connected in series, allowing complex circuit construction. For molecular logic gates to perform a similar function, we require multiple molecular logic gates to be connected, a process known as concatenation.<sup>32</sup> This presents the first challenge: how to link different molecular logic gates together so the OUTPUT from one is the INPUT for the next, and so on. The INPUT (typically metal ions) differs from the OUTPUT (often fluorescence emission) in molecular logic gates. It's also essential that signal intensity is maintained during concatenation and multiple gates function in unity under a range of environmental conditions (Fig. 6). Undaunted by these challenges, researchers have investigated multiple strategies to overcome these obstacles. One promising approach by Akkaya *et al.* involved using click chemistry to connect two AND gates into a single molecule **10** (Fig. 7).<sup>33</sup>

An OUTPUT at  $\lambda_{em}$  660 nm is only triggered when all three INPUT signals ( $\lambda_{ex}$  560 nm, Zn<sup>2+</sup>, and Hg<sup>2+</sup>) are present. Of note is that this molecule exhibits high selectivity for the target INPUT and no crosstalk. Molecule **10** is an excellent example of

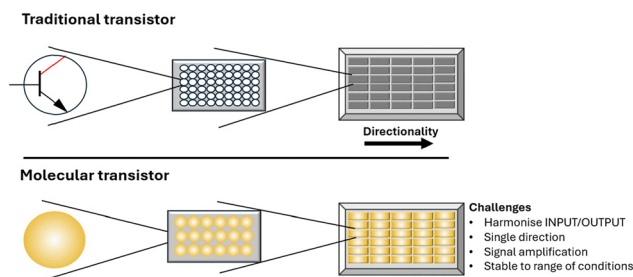


Fig. 6 The first challenge: connecting logic gates together.

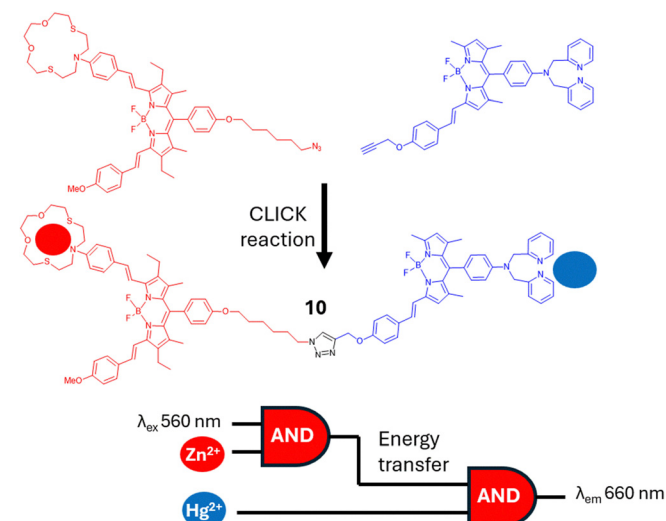


Fig. 7 Click chemistry to combine two AND gates in molecule **10**.



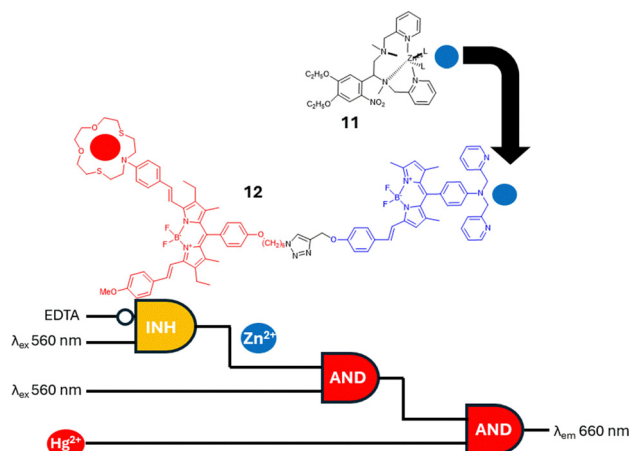


Fig. 8 Molecular logic gate cascades using molecules **11** and **12**.

coupling two well-established logic gates and using the latest click reactions to meet the concatenation challenge. Cascading molecular logic gates are an alternative strategy of cooperation between logic gates. In a cascade system, the presence of specific INPUT signals triggers an OUTPUT which in turn acts as an INPUT for another logic gate. This approach is demonstrated with molecule **11** which acts as an INHIBIT logic gate.<sup>34</sup> In the presence of INPUT 1 ( $\lambda_{em}$  560 nm) an OUTPUT ( $Zn^{2+}$ ) is released (blue dot in Fig. 8) which acts as an INPUT for an AND gate. If INPUT 2 (EDTA) is present it chelates  $Zn^{2+}$  and inhibits the gate. If INPUT 1 ( $Zn^{2+}$ ) is present alongside INPUT 2 ( $Hg^{2+}$ ) (red dot in Fig. 9) then the second AND gate is triggered, and OUTPUT ( $\lambda_{em}$  660 nm) is observed. Dey *et al.* reported a simple, yet highly tuneable molecule **13** composed of two rhodamine B molecules connected by a phenanthroline bridge (Fig. 9).<sup>35</sup> On excitation of the phenanthroline at  $\lambda_{ex}$  350 nm, energy transfer to the rhodamine B resulted in  $\lambda_{em}$  at 585 nm under certain conditions. A range of logic gates were devised using INPUT such as  $Cu^{2+}$ ,  $Hg^{2+}$  and  $I^-$  and a variety of OUTPUT signals. A particular advantage with molecule **13** is the use of well-studied and wide availability of the rhodamine B fluorophore. The utilization of inexpensive and widely available molecules is a key feature in challenge five, the cost-effective production of molecular computing.

One proposed solution to overcome the concatenation challenge is to move to all photonic molecular logic gates in which the INPUT and OUTPUT are exclusively light. This approach

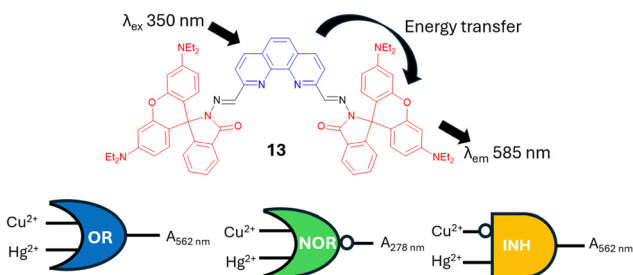


Fig. 9 Molecular cascade logic gates.

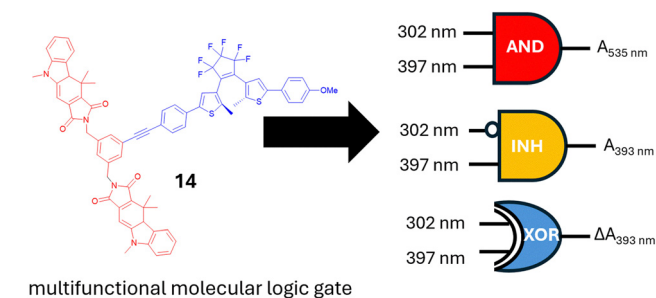


Fig. 10 Fully photonic molecular logic gate **14**.

offers several advantages, including lightspeed ON/OFF switching and bypassing the delivery of metal ions, binding and unbinding events and their subsequent removal. A pioneering example of a fully light-driven (photonic) logic gate is molecule **14** reported by Andréasson *et al.* (Fig. 10).<sup>36</sup> This molecule is constructed from two well-studied chromophores, dithienylethene, and fulgimide and displays a multitude of different logic gate configurations. The major advantage of **14** is the homogeneity of the INPUT and OUTPUT signals ensuring the logic gates can be concatenated. Majumdar *et al.* recently reported an elegant method of concatenating three well-established fluorophores to produce a fully photonic molecular logic gate platform.<sup>37</sup> Excitation of coumarin (Cou1) at  $\lambda_{ex}$  360 nm resulted in  $\lambda_{em}$  415 nm, which in turn activates curcumin (Cur), resulting in  $\lambda_{em}$  480 nm exciting rhodamine 123 (Rh123) (Fig. 11). This system can be configured to display a range of logic gate functions, including AND, OR, NAND, INH, and XOR. This is a further utilisation of well-established inexpensive and widely available fluorophores to perform sophisticated functions. Such systems provide proof-of-concept for challenge one and challenge five. The concatenation challenge can be overcome through a variety of different strategies (Fig. 12), such as chemical concatenation (Fig. 8). This approach may be suitable for connecting a small number of molecular logic gates in series however its large-scale application may be more challenging. An alternative is cascade concatenation in which the OUTPUT from one molecular logic gate forms the INPUT for another, for example **11** and **12** (Fig. 8).

Molecule **13** composed of the well-established fluorophore rhodamine B provide proof-of-concept that extensively studied molecules can be repurposed for use in molecular logic gates.

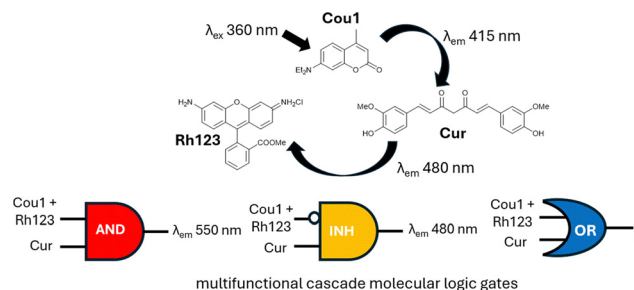


Fig. 11 Fully photonic, three component logic gate platform.



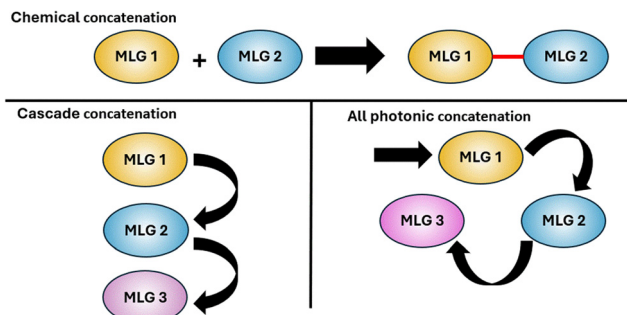


Fig. 12 Concatenation challenge summary.

Furthermore, by careful design, these systems can display multifunctional behaviour and display a range of different logic gate configurations. Concatenated molecular logic gates **11–13** utilise metal ions as INPUT signals, which result in additional obstacles involving metal ion transport and removal. There is a significant shift towards homogenous systems in which both INPUT and OUTPUT are the same. Light is emerging as the most promising option, with **14** a leading example (Fig. 10). The ability of a single molecular structure, or a small library of compounds to function as multiple logic gate configurations is encouraging novel approaches to this challenge. The molecular logic gate platform from Majumdar *et al.* demonstrates that even well-established fluorophores can be concatenated to perform advanced logic gate functions. The use of inexpensive and widely available reagents will be a key component in overcoming challenge five, cost-effective manufacturing. The platform reported by Majumdar *et al.* offers the potential to meet both challenge one and five simultaneously. It is highly likely future work will continue with fully photonic systems with the aim of developing more complex logical functions while minimizing structural complexity (Table 1). One key feature to integrate into future research would be to transition away from solution-based systems to substrate-bound applications. This will be essential if these systems are to find real-world applications and introduces challenge two, connectivity to solid supports.

### Challenge two: connectivity

The second significant challenge involves transitioning from the solution to the solid phase to enable the organised arrangement of multiple logic gates in series (Fig. 13). This is essential if complex architectures of molecular logic gates are to be assembled into compact areas to replicate the function of silicon-based transistors. This requires attachment of molecular logic gates to solid substrates with complementary functional groups without loss of photophysical properties.

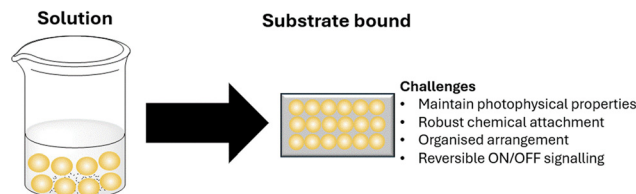


Fig. 13 The second challenge: attachment to substrates.

Previous work on single molecules held between two electrodes proves proof-of-concept of this approach, for example, the thiol group is a well-established anchor for gold electrodes, as observed in molecular wire **2** (Fig. 3A). Other reported anchors include carboxylic acid, amine, cyan, and  $\text{NO}_2$  groups<sup>38</sup> however for practical industrial applications we must use larger more cost-effective substrates. Van der Boom *et al.* bound molecular logic gate **15** (metal was either Os or Ru) onto glass substrates and demonstrated a range of logic gate operations including OR and INHIBIT operations (Fig. 14B).<sup>39</sup> This study demonstrated gate to gate communication with the concatenation of an OR and INHIBIT logic gates. These systems were stable for up to 200 °C in air for 48 h confirming robust connection to the glass substrate. A recent study by Brites *et al.* demonstrated doping the lanthanides  $\text{Eu}^{3+}$  and  $\text{Dy}^{3+}$  directly onto a glass substrate<sup>40</sup> enables simple logic gates such as AND and OR to complex devices such as molecular calculators to be constructed.

Araki *et al.* demonstrated molecule **16** (Fig. 15) attached to  $\text{TiO}_2$  via an ester linkage which retained XOR and INHIBIT logic gate functions.<sup>41</sup> Redmond *et al.* reported a similar configuration of a transition metal logic gate **17** bound to  $\text{TiO}_2$  (Fig. 15) which displayed NOR logic gate functionality.<sup>42</sup> Gunnlaugsson *et al.* developed molecular logic gate **18** bound to 5 nm gold nanoparticles via a thiol linker (Fig. 16).<sup>43</sup> Two different systems, one with an Eu and a second with Tb were devised and these were confirmed to display a multitude of logic gate properties in aqueous solution under a range of pH conditions. Furthermore, lanthanide based molecular gates are emerging as promising candidates for logic gates due to a range of properties, for example well-defined  $\lambda_{\text{em}}$  due to f-f electronic transitions, particularly centred on  $\text{Eu}^{3+}$  and  $\text{Tb}^{3+}$ .<sup>44</sup> Magri *et al.* recently reported the attachment of two different logic gates to different substrates, pyrazoline **19** to TentaGel<sup>®</sup> polymer beads<sup>45</sup> and Cinchona alkaloid **20** to an acrylamide polymer (Fig. 17).<sup>46</sup> Gunnlaugsson *et al.* reported the incorporation of logic gates into polymers,<sup>47</sup> these important studies demonstrate the feasibility of connecting logic gates to solid supports, vital for widespread industrial applications. Molecular logic gates are well-suited to biological applications particularly

Table 1 Concatenation solution summary

Challenge	Solution	Advantages	Disadvantages	Ref.
1 Concatenation	Click chemistry Cascading molecular logic gates Fully photonic molecular logic gates	Efficient synthesis Repurposing of established fluorophores Lightspeed ON/OFF signalling	Limited large-scale use Signal degradation between gates Require multiple $\lambda_{\text{ex}}$ concurrently	33 and 60 35 36 and 37



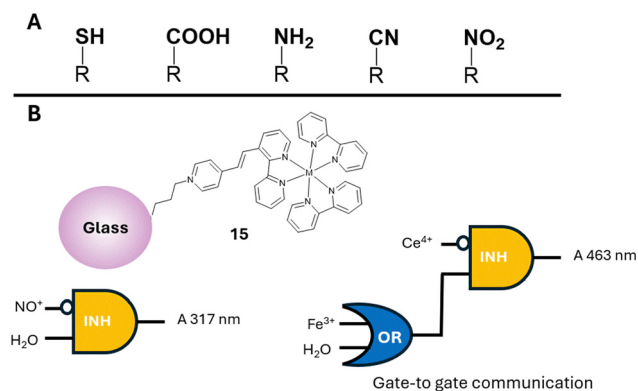


Fig. 14 Logic gate **15** attached to a glass substrate.

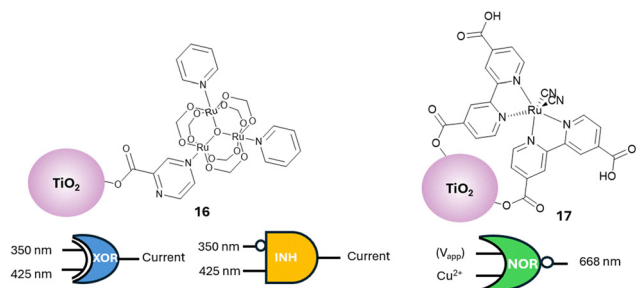


Fig. 15 Molecule **16** and **17** attached to  $\text{TiO}_2$ .

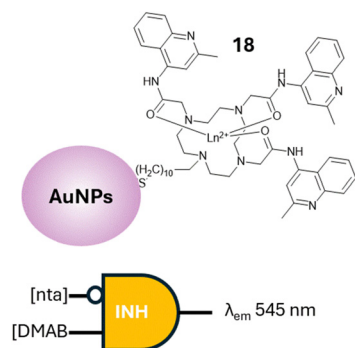


Fig. 16 Logic gate **18** attached to a gold nanoparticles.

when attachment to hydrogels and polymers.<sup>48</sup> Self-assembly monolayers (SAMs) offer a further approach to attach molecular logic gates to substrates.<sup>49</sup> The use of SAMs in the semiconductor industry is well-established<sup>50</sup> with the ability to attach molecules in organized patterns onto a surface in an automatic manner. There are three components to a SAM, the head group, a hydrophobic chain and a terminal group (Fig. 18A). Spiropyran **23** was attached to a gold electrode *via* SAM is an excellent example of this approach (Fig. 18B).<sup>51</sup>

On addition of UV light **23** converts to **23A** which can chelate  $\text{Zn}^{2+}$  ions and generate a voltage. In this situation **23** is functioning as a substrate bound AND gate. Of note is the strong signal transduction observed in this system, vital for providing a clear ON/OFF signals.

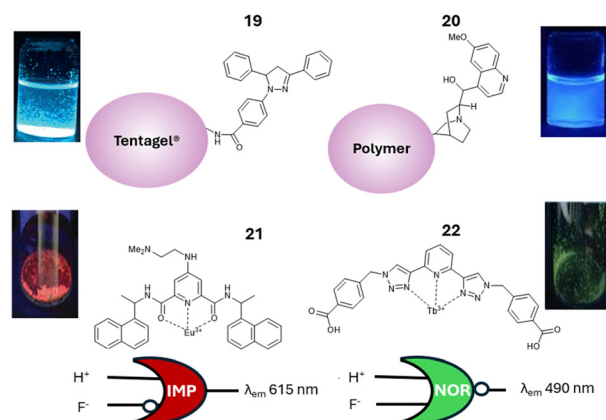


Fig. 17 Attachment of molecules **19–22** to various substrates, image reproduced from ref. 45 and 47 with permission from RSC, copyright 2021 and 2025.

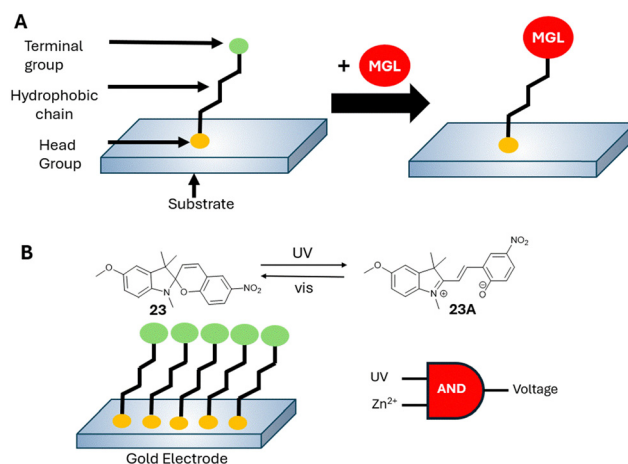


Fig. 18 Self-assembly monolayers to attach molecular logic gates (panel A), for example spirogyran **23** (panel B).

An alternative method of connecting molecular logic gates to substrates are nanowires. The “bottom-up” synthesis of nanowires is performed by nanocluster-catalysed vapour liquid solid (VLS) consisting of three stages.<sup>52</sup> A catalytic nanocluster directs the formation of a nanowire in a single direction with gold commonly employed as a catalyst (Fig. 19A). Silicon nanowires are widely used with diameters of approx. 20 nm and lengths in the micron range. The diameter can be controlled by the use of different metals and the nanowires themselves doped to adjust for specific electrical and optical properties. The attachment of molecules to the surface is widely reported, for example molecular logic gate **24** on a silicon nanowire displays INHIBIT and NOR properties (Fig. 19B).<sup>53</sup> In summary, many researchers have answered the call of challenge two, a variety of molecular logic gates have been connected to numerous substrates without loss of photophysical function (Fig. 20). The careful selection and incorporation of a chemical handle combined with a complementary functional group on the substrate is vital to this process. Fabrication



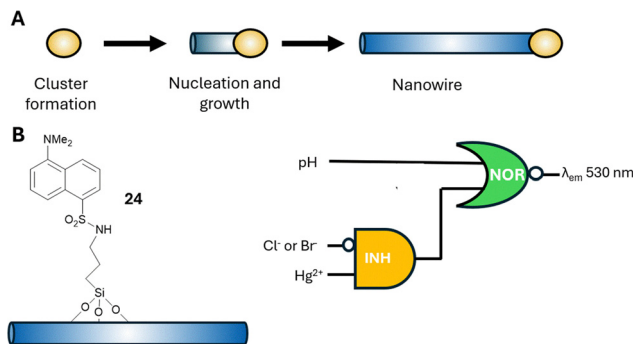


Fig. 19 Nanowire formation (panel A) and molecule **24** attached to nanowires (panel B).

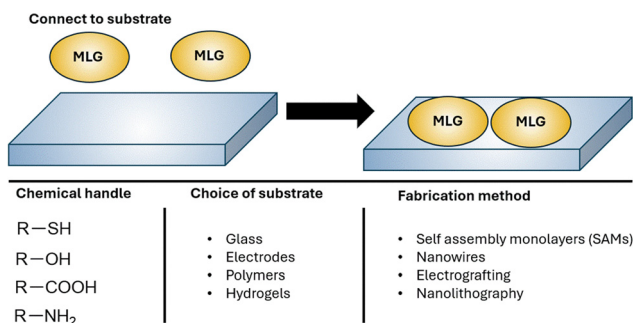


Fig. 20 Connectivity challenge summary.

methods from the semiconductor industry such as SAMs, nanowires, and electrografting<sup>54</sup> are promising tools to connect logic gate to substrate using autonomous self-assembly (Table 2). Future work will involve building in complex architectures and hierarchies with the aim of developing more sophisticated devices. However, as more logic gates are concentrated into smaller areas a third challenge arises, the problem of increased interference and crosstalk.

### Challenge three: crosstalk reduction

The first two challenges involved connecting (concatenation) several molecular logic gates together and transitioning from solution phase to substrate-bound systems. This is critical if molecular logic gates are to function as a single unit. The OUTPUT from molecular logic gates is typically fluorescence emission ( $\lambda_{em}$ ), and this is multidirectional. This presents the third challenge: how to ensure the OUTPUT from one molecular logic gate does not interfere unintentionally with its surroundings? (multiple red and blue arrows in Fig. 21). This is often

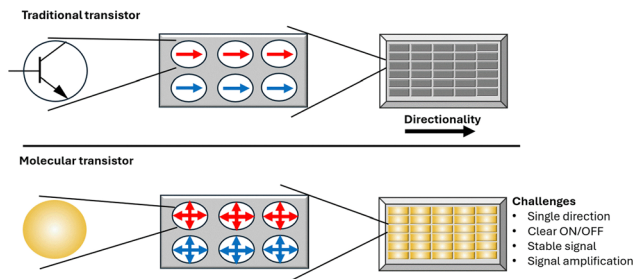


Fig. 21 The third challenge: reducing crosstalk.

described as crosstalk and is becoming a significant problem for 5 nm silicon-based transistors due to quantum tunnelling.<sup>55</sup> Furthermore, we require a clear ON/OFF signal to activate each logic gate predictably and reliably. Activation must be reversible and switched ON/OFF rapidly without signal interference or deterioration. Light is a leading candidate with all photonic molecular logic gates a clear contender. Crosstalk is a significant challenge in photonics<sup>56</sup> and there is much we can learn from this field to advance molecular logic gate development. We will examine these strategies and how to implement them in molecular electronics. Waveguides are commonly employed to direct the flow of light in specific orientation.<sup>57</sup> An elegant study by Zhang *et al.* demonstrated the attachment of spiropyran **25** to a polydiacetylene microtube (Fig. 22).<sup>58</sup> Upon excitation at  $\lambda_{ex}$  532 nm at 90° through the microtube,  $\lambda_{em}$  is observed at the tip with minimum emission from the body of the microtube. Zhang *et al.* confirmed the additional functionalisation with spiropyran **25** did not affect the waveguiding properties of the microtube. Furthermore, this system displayed both OR and INHIBIT logic gate properties.

Zou *et al.* reported a configurable amine functionalized polydiacetylene microtube which underwent thiol-ene click reaction to produce microtube bound **26** (Fig. 23).<sup>59</sup> Like molecule **10** (Fig. 7), this is a further example of a click reaction to develop novel fluorescence devices.<sup>60</sup> Microtube bound **26** displayed a range of logic gate operations including AND and INHIBIT using heat and voltage as INPUT and fluorescence emission at 650 nm as the OUTPUT. Polydiacetylene microtubes have been reported to display polarisation and this was also observed in **26**. Further studies revealed that by altering the polarisation state of the excitation beam, the emission at the tip of the microtube could be easily controlled and modulated. This is an excellent additional function for optical computing applications ensuring crosstalk between parallel systems is minimised. Xia *et al.* reported spiropyran doped

Table 2 Connectivity challenge solution summary

Challenge	Substrate	Advantages	Disadvantages	Ref.
2 Connectivity	Glass	Low-cost	Brittle	39 and 40
	TiO <sub>2</sub>	Industrial feedstock	Restricted UV range	41 and 42
	AuNPs	Well-defined geometries	Expensive	43
	Polymers	Biological applications	Temperature limited	45 and 46
	SAMs	Well-established process	Poor stability	51
	Nanowires	Surface functionalisation	Limited scale up	53 and 58–63



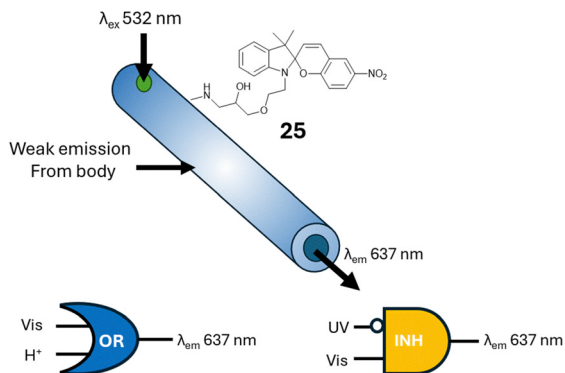


Fig. 22 Spiropyran-functionalised polydiacetylene **25**.

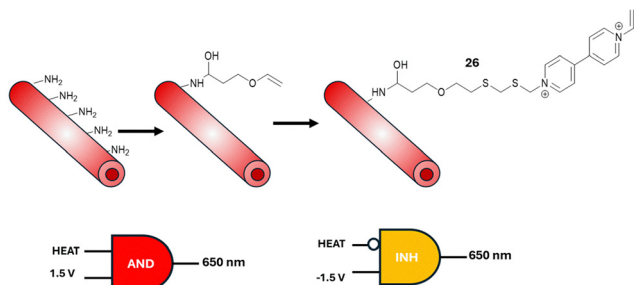


Fig. 23 NH<sub>2</sub> functionalised microtube **26**.

microwire system **27** and **28** (Fig. 24).<sup>61</sup> This study used polystyrene microwires with spiropyran **27** as a donor unit which could undergo FRET to excite a second type of microwire doped with fluorescein **28** (Fig. 24). These microwires were then constructed into a range of configurations to demonstrate the transfer of light between complex microwire shapes.

Takazawa *et al.* reported thiacyanine sub-100 nm organic nanofiber **29** which displayed optical waveguide properties

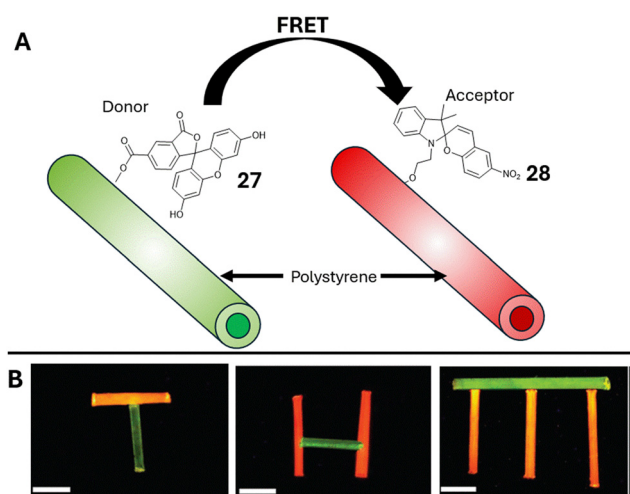


Fig. 24 Fluorescein and spiropyran-functionalised polydiacetylene microwires **27** and **28** (panel A) and assembly into different configurations (panel B), image reproduced from ref. 61 with permission from RSC, copyright 2021.

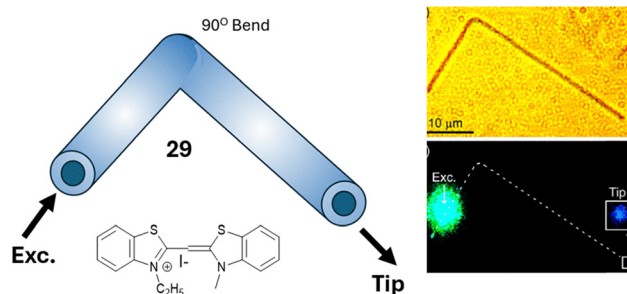


Fig. 25 Nanofiber **29**, image reproduced from ref. 62 with permission from ACS, copyright 2016.

(Fig. 25).<sup>62</sup> This study demonstrated that **29** successfully transmitted light through a nanometre fibre containing 90° bends opening the possibility of complex shape waveguides in the future. While this application was limited to liquid nitrogen temperatures, it does confirm that nanoscale optical waveguides can be a useful method of transmitting lights while minimising crosstalk. In 2018, Sun *et al.* reported a fully optical nanowire network composed of indium phosphide (InP) and aluminium gallium arsenide (AlGaAs) which could perform logic gate functions.<sup>63</sup> A grid of intersecting InP and AlGaAs nanowires was easily constructed using mechanical combing resulting in crossbars where the two meet (black boxes in Fig. 26). Each nanowire emitted fluorescence emission at different wavelengths depending on the polarization of the excitation wavelength. This enabled AND, OR, NAND, and NOR logic gate construction. Furthermore, this system operated at room temperature. This proof of concept clearly demonstrates nanowires have a valuable role to play in photonics and also molecular logic gates.

In summary, microtube and nanowires are proving to be pivotal in advancing nanophotonic applications, offering practical solutions to challenges two and three in the development of molecular logic gates (Fig. 27 and Table 3). The concatenation of distinct logic gates has been effectively demonstrated using microwire-bound systems (compounds **27** and **28**, Fig. 24). Moreover, crosstalk—a significant obstacle to signal fidelity—can be substantially mitigated by encapsulating fluorescence within nanotube structures, thereby minimizing unwanted emissions from the tube walls (compound **25**,

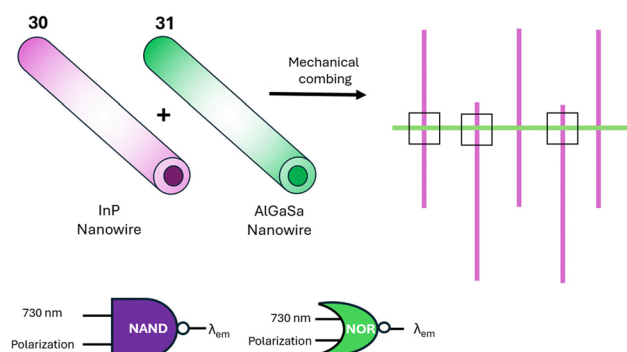


Fig. 26 Nanowire networks displaying logic gate properties.



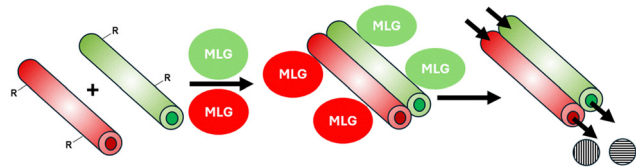


Fig. 27 Crosstalk challenge summary.

Fig. 22). Further reduction of crosstalk can be achieved by leveraging nanowire polarization; arranging nanowires orthogonally (at  $90^\circ$  angles) serves as an effective geometric strategy to minimize signal interference. Beyond these optical benefits, nanowires serve as highly versatile substrates for molecular logic systems. Their surfaces can accommodate a wide array of functional group attachments, effectively addressing challenge one—connectivity to substrates. Furthermore, nanowires themselves can exhibit logic gate behaviour, enabling synergistic enhancement of photophysical properties such as signal amplification and directional propagation. Incorporating the latest advancements in nanowire research into molecular logic architectures paves the way for constructing increasingly complex devices, marking a critical step toward realizing a functional molecular computer. This progression naturally leads to the next major challenge: integrating molecular logic devices with conventional electronic components.

#### Challenge four: compatibility with electronics

The fourth challenge facing molecular computing involving integration into existing electronic systems (Fig. 28). Current silicon-based computing is predominantly electron-based, with silicon transistors at its core.

As we advance toward next-generation computing paradigms, the develop of hybrid technologies enabling emerging systems to coexist with established silicon-based architectures is required. The photonics industry is a market leader in this field and offers valuable examples for molecular based systems. Hybrid integration of light-based technology with conventional silicon electronics has been achieved with commercial systems providing valuable improvements to data centres and accelerating AI development by harnessing the potential of light for computation.<sup>64,65</sup> This demonstrates integration is possible and is paving the way for the next generation of computing. These advancements provide valuable insights that can be adapted to address the challenge of developing molecular logic gates compatible with current electronic systems. A variety of different photonic integrated circuits (PICs) are available however on silicon photonics is the current leader in commercial systems.<sup>66</sup> On silicon PICs devices are manufactured using

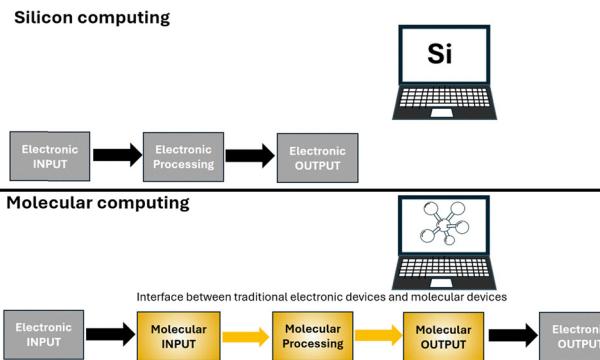


Fig. 28 The fourth challenge: compatibility with existing electronics.

existing fabrication technology for silicon-based transistors preventing the requirement for new facilities and technology. This is a significant advantage enabling side-by-side construction with existing silicon-based transistors. Furthermore, the integration is more easily achieved as they share similar substrates. The first generation of PICs involved pluggable optics which were positioned off the central IC (Fig. 29).<sup>66</sup> This was then replaced with on-board optics, further miniaturization through generations III and IV to the latest generation V technology with 3D optics and integrated lasers (Fig. 29). The integration of photonic and electronic components onto a single chip is now possible. The progress of on silicon PICs provides an excellent roadmap for the construction of molecular logic gate integrated circuits (MLGICs). There is much we can learn from the four major challenges in silicon PICs (design, integration, reliability, and fibre attachment, Fig. 29B) which are likely to be experienced in MLGICs development. It is highly likely the first MGLICs will follow a similar approach to the 1st generation PICs in which logic gates functional are performed off the electronic circuit. A pluggable optical receiver is used to interface between the two components, and this leads to the first obstacle, how to transform the output from a MLG into a suitable signal for a receiver.

The latest research in nanoscale lasers and detectors provides a valuable resource in this regard. Yang *et al.* reported the first example of a nanowire laser in 2001 based on a zinc oxide (ZnO) nanowire with emission at 385 nm.<sup>67</sup> Numerous applications have been reported since including sensing, biological probes and nanoengineering taking advantage of the coherent light generated from nanolasers.<sup>68</sup> A recent example reported by Kim *et al.* involved micro-transfer printing of a silicon based nanolaser which was directly mounted on silicon waveguides.<sup>69</sup> This nanolaser operated at room temperature and could be used at the excitation source for molecular logic gates. A further

Table 3 Crosstalk solution summary

Challenge	Substrate	Advantages	Disadvantages	Ref.
3 Crosstalk	Polydiacetylene	Surface functionalisation	Limited scale-up	58 and 59
	Thiacyanine	$90^\circ$ bends possible	Low temperature only	62
	Polystyrene	Complex shapes	Limited scale-up	61
	InP + AlGaAs	Logic gate properties	Limited scale-up	63



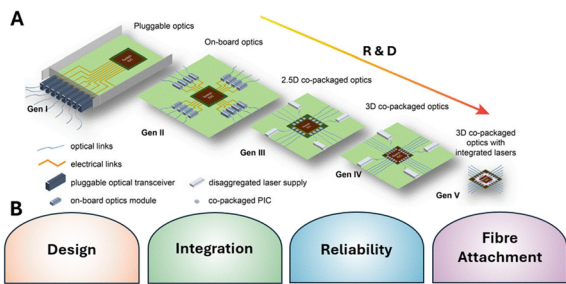


Fig. 29 Miniaturization of photonic integrated circuits (PICs) (panel A) and the four challenges experienced (panel B), image reproduced from ref. 66 with permission from AIP publishing, copyright 2021.

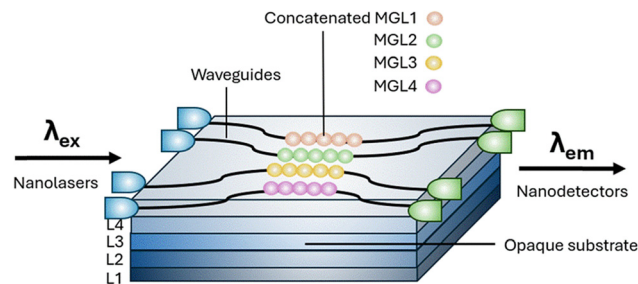


Fig. 32 Compatibility with electronics summary, a possible design for a molecular logic gate integrated circuit.

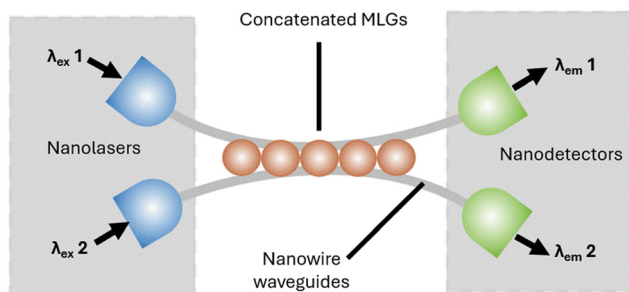


Fig. 30 A possible design for concatenated molecular logic gates within a nanowire with multiple  $\lambda_{ex}$  and  $\lambda_{em}$  connections enabling lightspeed ON/OFF signalling at multiple wavelengths.

example reported Kim *et al.* also demonstrated the mounting of a nanolaser onto a silicon wavelength and upon activation, 88% of the laser emission was transferred to the waveguide.<sup>70</sup> Osada *et al.* reported integrating two quantum dot based nanolasers directly onto a silicon PIC.<sup>71</sup> This enabled the use of multiple excitation wavelengths and the ability to multiplex on a single chip. A possible design for molecular logic gates is shown in Fig. 30. Numerous other nanolaser<sup>72</sup> and nanodetectors<sup>77</sup> have been reported (Fig. 31) allowing a nanoscale toolbox of components to be developed.<sup>72</sup> Such a supply of various devices will be vital to meet the needs of challenge four. In summary, the fourth challenge is possibly the most difficult yet, involving a

complex interplay between nanodevices (nanolasers and nanodetectors) and nanoscale manufacturing. As these two research fields develop, it is likely reports of early prototype molecular logic gate integrated circuits (MLGICs) within the next 5–10 years emerge.

Such MLGICs require a convergence of multiple technologies to solve a single problem (Table 4). A potential design for the first MLGIC incorporating the latest designs from challenges one to four is displayed in Fig. 32. The photonic industry is an exemplar of this approach, through several rounds of iteration, commercially available systems have been developed. As technology advances and the uptake increases, this will drive down the manufacturing costs and help solve the fifth challenge: cost-effective manufacturing.

#### Challenge 5: cost-effective manufacturing

To advance molecular logic-based devices from laboratory to commercially viable technologies, it's essential to develop cost-effective and scalable manufacturing processes. Repurposing well-established fluorophores, such as molecules 13 and 14 (Fig. 9 and 10 respectively) are leading examples. The use of modular molecular scaffolds<sup>82</sup> allows for fine-tuning of molecular logic gates through minor adjustments using a pool of common reagents. This introduces design flexibility and facilitates the customization of logic gates for specific applications. The use of inexpensive, readily available reagents is vital for reducing production costs. Establishing a flexible and resilient supply chain ensures a steady supply of these reagents, minimizing the risk of bottlenecks and enabling consistent manufacturing at an industrial scale. Molecular logic devices offer the potential to decentralize the IC fabrication industry, which is currently heavily reliant on facilities based in Asia. By diversifying the materials and methods used in logic device fabrication, it's possible to reduce the political and economic uncertainties associated with traditional semiconductor manufacturing. Its highly likely the cost of early MLG devices will be



Material	$\lambda_{em}$ nm	Diameter nm	Ref.	Material	$\lambda$ Range nm	Ref.
MALH	540-555	238	73	NiO/ZnO	365-750	78
CLB	533	120	74	ZnO/CuSCN	355-750	79
ZnO	372-386	30	75	ZnO/ZnTe	325-800	80
MALH	768-796	200	76	PtSe2/GaAs	365-800	81


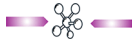


Fig. 31 Recently reported nanolasers and nanodetectors.

Table 4 Compatibility with electronics solution summary

Challenge	Solution	Advantages	Disadvantages	Ref.
4 Compatibility with electronics	Nanolasers Nanodetectors Waveguides	Low power usage Established use in photonics Established use in photonics	Difficult scale-up Difficult scale-up Difficult scale-up	67–76 7–81 57 and 58



Table 5 Current methods of molecular electronic device production

Method	Process	Molecular control	Precision	Scalability	Ref.
Mechanical break junction		High	High	Low	86
Nanogap electrodes		Medium	High	Medium	87
SAMs		Low	High	High	88
Dielectrophoresis		Medium	Medium	Medium	89

high due to the novelty of the technology and limited production volumes. One further area for cost-effective manufacturing involves the methodologies of fabrication itself. Current “bottom-up” approaches offer several advantages over the traditional “top-down” methods.<sup>83</sup> For example “bottom-up” methods are efficient with minimal wastage<sup>84</sup> and allow precise control and positioning of molecular components.<sup>85</sup> However these methods are often bespoke and limited to small scale production of proof-of-concept devices (Table 5). For example, the most employed method is a mechanical break junction in which a molecule is held between two electrodes.<sup>86</sup> While providing a high level of precision and molecular control, this method involves producing molecular devices one at a time and cannot be scaled easily to mass production. A recent develop in nanogap electrodes demonstrated some improvement however this is far from industrial applications.<sup>87</sup> Surface assembly monolayers (SAMs) offer scalability, however with a cost to molecular control.<sup>88</sup> Dielectrophoresis is a recent development which may hold promise for the future due to its ability to generate single molecules and scalability.<sup>89</sup> As manufacturing processes improve and market adoption increases, economies of scale are expected to drive down costs, making these devices

more accessible fuelling further adoption. This has been proven with the silicon-based IC and recently in the photonic industry. By focusing on these strategies (Fig. 33 and Table 6), the development of cost-effective production can be realised. The 5Cs are integral to this process ensuring continual advancement in molecular electronics and bringing a step closer towards the world's first molecular computer.

## Conclusion

Silicon-based integrated circuits (ICs) have been the foundation of technological advancement for the past seven decades, underpinning the information age and shaping our modern world. As we approach the limits of silicon-based transistor miniaturization, a paradigm shift in computing is enviable. The concept of molecular electronics introduced in 1974 by Aviram and Ratner is now well-established with many research groups contributing to an ever-expanding toolbox of molecular electronic components (Fig. 3). The next chapter involves integrating these individual components into more complex devices to construct the world's first molecular computer. We have highlighted five main challenges to achieving this goal and discussed the ongoing research to address each challenge (Fig. 34). Future molecular logic gates should be designed specifically to be 5C compliant with each challenge carefully considered as part of the design process. There are clear trends toward fully photonic systems which display multifunctional logic properties. The repurposing of well-established fluorophores and using widely available reagents is also emerging. This will ensure cost-effective manufacturing on an industrial scale is possible, vital if molecular electronics are to become commercially available. The use of nanowires both as substrate and to overcome crosstalk, combined with incorporating within existing electronics is an emerging field of research. Advancements in nanotechnology and photonics are providing valuable case studies to assist the development of more sophisticated molecular devices. As we go through this technological transformation, we have a valuable

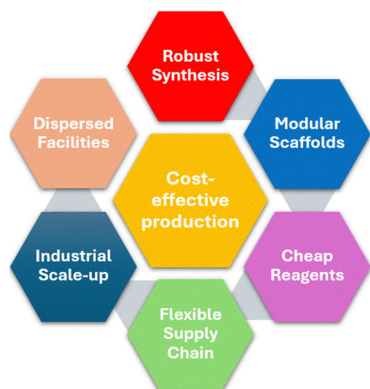


Fig. 33 Six components to cost-effective production.

Table 6 Cost-effective manufacturing solution summary

Challenge	Approach	Solution	Advantages	Ref.
5 Cost-effective manufacturing	Modular design	Heterocycles	Fine-tune properties	82
	Repurpose fluorophores	Well-defined properties	Wide availability	35–37
	Short robust synthesis	Use established chemistry	Efficient scale-up	82
	Low-cost reagents	Industrial feedstocks	Reduce bottlenecks	82



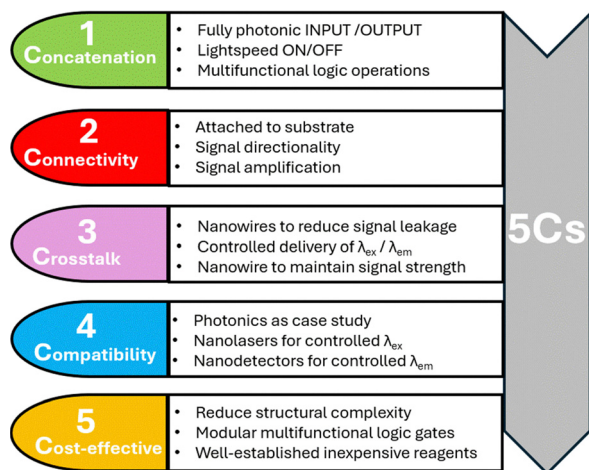


Fig. 34 Summary of the 5Cs and current directions of travel.

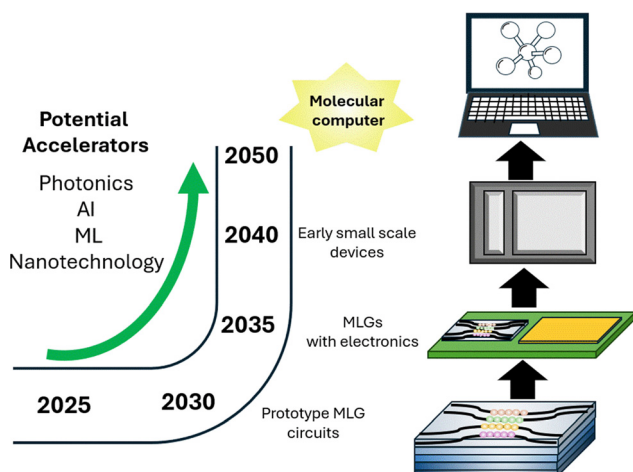


Fig. 35 A potential roadmap towards the world's first molecular computer.

opportunity to decentralize IC production, reducing our global reliance on a handful of semiconductor fabrication facilities.

Decentralizing the production of these critical components could reduce geopolitical and economic concerns. While initial molecular computing devices may be costly and limited in functionality, increased research investment and technological maturation are expected to drive down costs and enhance performance, as observed in the silicon and now photonic industries. The latest advances in AI and ML can be utilized to accelerate advancement in molecular electronics further still. While there may be further unknown obstructions on the road ahead (Fig. 35), the path is set, with molecular electronics providing the tools and resources to meet the needs of tomorrow.

## Author contributions

Alexander Ciupa authored the manuscript.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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