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Dynamic DNA superstructures with emergent functions

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DNA nanotechnology enables the precise construction of intricate nanoscale structures. Over the past two decades, significant progress has been made in incorporating dynamic functionalities into these nanostructures. Concurrently, innovative strategies have emerged for their self-assembly and surface patterning into larger, more complex architectures. This review explores the convergence of these two key capabilities—reconfigurability and hierarchical assembly—to engineer DNA origami superstructures with intrinsic dynamic behavior. We begin by outlining foundational strategies in dynamic design, hierarchical assembly, and surface placement, then review recent progress in leveraging these strategies to construct dynamic superstructures with emergent behaviors. The article concludes with a roadmap of major challenges and opportunities shaping the future of this rapidly evolving field.

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1. Introduction

DNA nanotechnology is a rapidly growing field that leverages the programmability and specificity of Watson–Crick–Franklin base pairing interactions to construct precisely defined nanoscale structures.[1] A leading technique in this field is DNA origami, which uses hundreds of short single-stranded DNA (ssDNA) "staples" (typically 20–50 bases long) to fold a long piece of ssDNA "scaffold" (usually several kilobases in length) into the desired structure.[2,3] This approach, along with related methods, enables the bottom-up construction of intricate 3D architectures with nanometer precision.[4] Beyond their structural elegance, these DNA-based nanostructures serve as programmable breadboards for organizing other functional materials such as proteins, nanoparticles, and polymers, thus greatly expanding their practical applications.[5]

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Early DNA nanostructures were intentionally designed to be structurally rigid with well-defined geometries, serving as proof-of-concept demonstrations of the technology's design precision and geometric control. Over the past two decades, dynamic systems have attracted increasing attention. Inspired by the flexibility and movement of natural molecular machines like enzymes and motors, researchers have increasingly aimed to engineer dynamic DNA devices capable of conformational changes, mechanical motion, and responsive behavior.[6] This evolution has broadened the scope of DNA nanotechnology, unlocking applications in nanorobotics, targeted drug delivery, biosensing, and optoelectronic devices.[7]

In parallel, considerable efforts have focused on scaling up DNA nanostructures to achieve higher-order assemblies.[8] As individual DNA origami structures are intrinsically small—limited by scaffold length and susceptibility to assembly errors—researchers have turned to hierarchical strategies to overcome these constraints. This typically involves forming discrete origami units, then organizing these units into larger superstructures. The latter step can be achieved by interconnecting units using programmable binding motifs or by positioning units on surfaces using predefined binding sites.[9] These larger assemblies enhance both throughput and functional complexity, representing a critical step toward practical applications.

Despite the progress in dynamic functionality and hierarchical assembly, these two paradigms have largely developed independently. The prospect of combining dynamic behavior with higher-order organization promises a new class of DNA-based materials with unprecedented capabilities. Dynamic superstructures could exhibit collective emergent properties,

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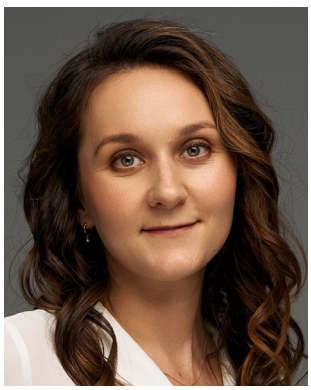

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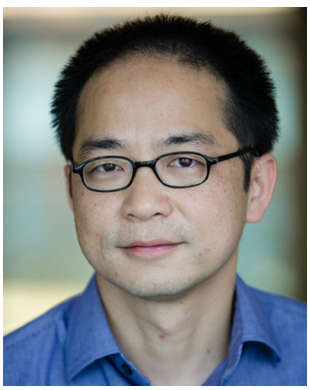

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enhance environmental responsiveness, and drive novel applications in biomedicine and optoelectronics. However, this fusion is not without challenges: while dynamic devices benefit from flexibility and disorder, robust superstructures require precision and rigidity—posing a fundamental design dilemma. Several important strides have been made toward addressing this challenge, revealing strategies for integrating dynamic function into ordered superstructures. In this article, we review these advances, discussing the strategies used to create reconfigurable higher-order assemblies, the unique emergent properties they exhibit, and the new application spaces they unlock. We also highlight remaining challenges and future opportunities in this exciting and evolving frontier.

2. Foundational work on dynamics and assembly

Before discussing dynamic higher-order assemblies of DNA nanostructures, we first review key strategies for introducing dynamic mechanisms into individual DNA origami structures and then discuss hierarchical self-assembly and surface-placement approaches for achieving large, static assemblies of origami structures.

Dynamic mechanisms

Dynamic DNA origami nanodevices leverage key features of DNA to both facilitate motion and enable actuated conformation changes. These features include: the ability to precisely control the geometry of DNA structures; the flexibility of ssDNA; and the tunable hybridization/dissociation or deformability of dsDNA helices or dsDNA bundles in response to force or other stimuli.

The most common approach to building dynamic structures leverages the precise geometric design capabilities of DNA origami to position short ssDNA segments at strategic sites to create flexible domains that connect multiple stiff dsDNA bundle components together. Short ssDNA regions (~1 to 4 nt) offer bending or rotational flexibility and are usually unpaired scaffold regions,[10] though they can also be unpaired staples connecting components.[11] Because DNA origami typically uses circular scaffolds, scaffold-based connections are often formed in pairs, while staple-based connections can form single linkages, though likely with lower assembly yield. These short ssDNA connections can provide rotational flexibility between components (Fig. 1(A)), and multiple such connections in a line can restrict motion to a single rotational axis (Fig. 1(B)).[12,13] This strategy underpins many DNA origami hinges, which is among the most commonly used dynamic designs. Longer scaffold connections when paired with short ones can tune joint flexibility,[14] while all-long ssDNA links allow for greater tethered motion between components (Fig. 1(C)).[15] Beyond flexible ssDNA joints, the design capability of DNA origami also enables constrained motion through the assembly of multi-component devices with complementary shapes. This often involves "staged" assembly to fit components together—such as inserting a cylinder into a tube[10] or

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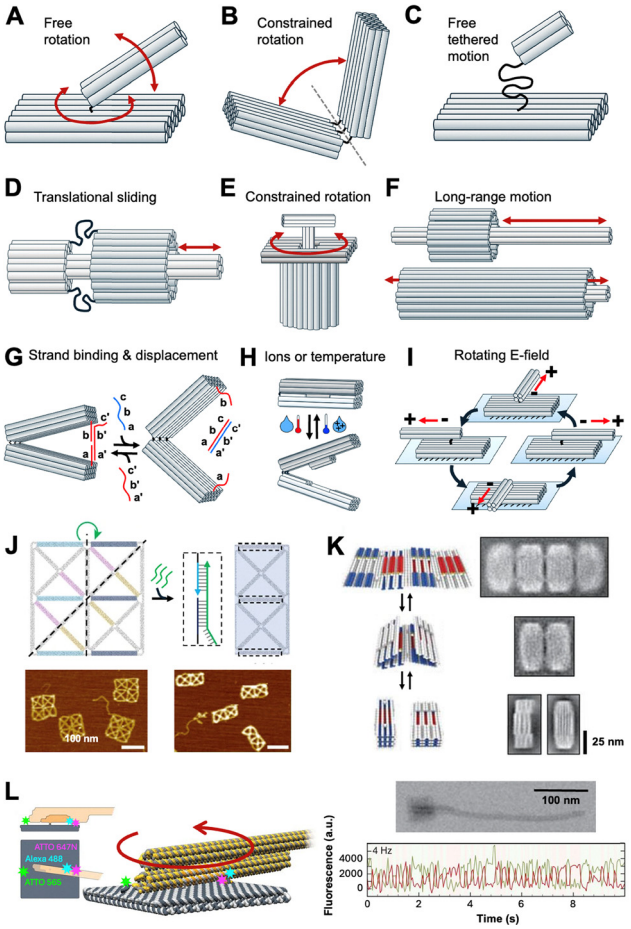

Fig. 1 Dynamic mechanisms in DNA origami structures. (A)–(F) Strategically placed ssDNA connections can produce a diverse range of motions: (A) free rotation about a point, (B) rotation constrained to a single angular degree of freedom, (C) free motion through tethers, (D) translational sliding through interlocked structures, (E) rotary motion constrained by concentric structures with an end cap on the outer structure, and (F) long-range translational sliding along tracks. (G)–(I) Various actuation methods can be used to control motion: (G) toehold-mediated strand displacement, (H) environmental triggers such as temperature or ion concentration, and (I) electric fields. (J)–(L) Experimental examples of dynamic DNA origami structures: (J) sheets that fold along different lines in response to different ssDNA trigger strands, (K) a sheet that reversibly folds into blocks using salt-dependent shape-complementary base stacking interactions, and (L) electric-field driven DNA origami rotor. Panels J, K, and L have been adapted with permission from ref. 25 (Copyright 2023, Springer Nature), ref. 22 (Copyright 2015, AAAS), and ref. 12 (Copyright 2018, AAAS), respectively.

placing a ring on a track[16]—to enable linear sliding motion (Fig. 1(D)). More complex geometries can constrain sliding while allowing rotation (Fig. 1(E)).[17] While the assembly process is more intricate, this method allows for long-range sliding translational motion, especially when integrated with higher-order assemblies (Fig. 1(F)).[16] Combining this approach with flexible ssDNA joints can further help constrain the range of sliding motion.[10]

Building on strategies to program motion in DNA origami nanodevices, various actuation methods have been developed to control their conformations. The most common of these involves ssDNA overhangs on dynamic components, which can be latched together by adding complementary DNA strands (Fig. 1(G)).[18] This latching mechanism can be designed to be reversible by incorporating an additional ssDNA domain that remains single-stranded in the latched state providing a "toe-hold" for removing the latching strand *via* strand displacement, thus releasing the structure back into the unlatched configuration. The key advantage of this approach is the relative ease of incorporating the ssDNA overhangs and the sequence specificity, which even allows for specific and sequential actuation of different strand displacement reactions on the same device.[19] However, key drawbacks include the need to introduce strands into solution either causing overall dilution or requiring solution exchange, and this approach also leads to relatively slow response times of ~1–10 minutes.[10,20] Other actuation strategies involve the use of external stimuli, such as pH, temperature, ion conditions, light or small molecule binding (Fig. 1(H)).[21] These stimuli typically operate by stabilizing or de-stabilizing base-pairing interactions, or other binding motifs such as shape-complementary base-stacking motifs.[22] These approaches can provide significant speed up in response times, down to millisecond scales.[6] Recent efforts have also explored generating rotational motion in DNA devices through the use of electric fields,[12] which steer negatively charged components, or magnetic fields,[11] which apply force to integrated magnetic particles (Fig. 1(I)). These approaches can achieve second or sub-second response times, with electric field control allowing up to several rotations per second of DNA origami rotors. Other recent advances include the design of ratchet motors[23] and the incorporation of protein motors

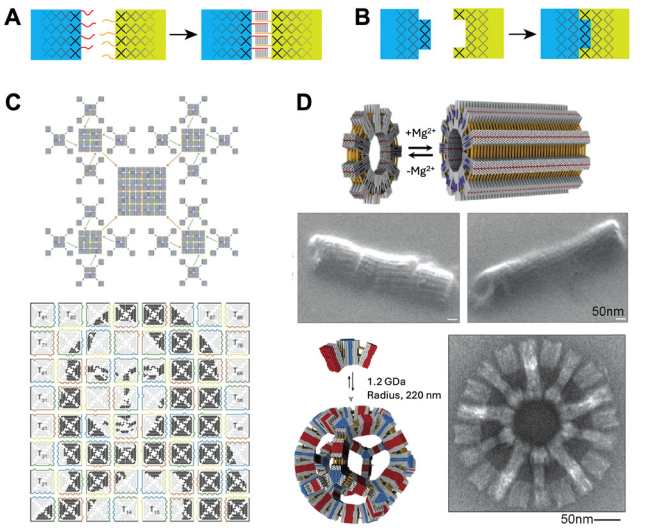

Fig. 2 Hierarchical assembly of DNA origami *via* (A) programmable sticky-end hybridization and (B) shape-matching blunt-end stacking. (C) Hierarchical assembly of large arrays, including a picture mimicking the painting of Mona Lisa, from DNA origami tiles (reproduced with permission from ref. 35; Copyright 2017, Springer Nature). (D) Hierarchal assembly of DNA-origami gigadalton structures, including a self-limiting nanotubes and 3D cages, *via* blunt-end stacking (reproduced with permission from ref. 38; Copyright 2017, Springer Nature).

that harness chemical reactions to repetitively reconfigure dynamic nanodevices.[24]

These efforts have led to powerful devices with advanced capabilities and properties including complex reconfiguration and motion (Fig. 1(J) and (K)),[25] rapid actuation,[26] mechanical transmission of forces up to ~10 pN,[27] and processive motions over large distances or angles (Fig. 1(L)).[28] Other advanced behaviors include signal propagation through cascaded conformation changes,[29] ability to apply compressive forces,[14,30] and undergo complex shape changes.[25] As a result of these features, DNA origami devices have become useful tools for biophysical measurements,[31] biosensing,[32] and actuators to control molecules and nanomaterials and their interactions.[7]

Higher-order assembly

The assembly of DNA origami superstructures is typically achieved by designing specific DNA–DNA interactions between individual building blocks. A commonly used strategy involves sticky-end hybridization (Fig. 2(A)), where complementary ssDNA overhangs form connections between the building blocks. This strategy is as old as the field itself, conceived by Seeman when he envisioned the concept of making lattices with DNA molecules.[33] It remains probably the most versatile and robust method for assembling large structures, from the earliest example of a 2D lattice of small DNA tiles,[34] to the more recent complex assemblies such as the massive DNA origami array with features mimicking the ''Mona Lisa'' painting (Fig. 2(C)).[35] Naturally, the assembly of such complex superstructures *via* sticky-end hybridization requires more sophisticated design that involves large numbers of sticky-end sequences and carefully engineered, multi-step assembly approaches. Combinations of sticky-end hybridization with other strategies continue to yield advances in hierarchical DNA origami assembly. A recent example is the formation of large microscale crisscross structures assembled from many DNA 6-helix-bundle origami ''slats''.[36] This approach used a 2D origami seed to nucleate growth and leveraged cooperative binding—where each slat can bind to the assembly only after its predecessor—to control the assembly pathway and final architecture.

A second approach to higher-order assembly takes advantage of blunt-end base-stacking—attractive π–π stacking interactions across terminal base pairs at the ends of dsDNA helices. As first observed in the seminal work on DNA origami in 2006,[2] blunt-end base-stacking can lead to unwanted aggregation of DNA origami. Later, Rothemund and Woo used non-specific blunt-end base-staking to their advantage by designing the edges of the DNA origami tiles to contain matching arrangements of helix protrusions, enabling their programmable higher-order assembly.[37] Another elegant strategy leverages stacking interactions to stabilize lock-and-key motifs introduced at the edges of DNA origami building blocks (Fig. 2(B)),[22] enabling the construction of large-scale structures, such as microscale nanotubes and gigadalton DNA cages (Fig. 2(D)).[38]

Beyond sticky-end hybridization and shape-complementary blunt-end stacking, researchers continue to explore alternative molecular interactions for hierarchical assembly. Although these alternatives have not yet matched the programmability of traditional methods, peptide–DNA conjugates are a promising strategy, as they offer the combined advantages of DNA and proteins. Peptides can be readily attached to DNA through covalent interactions while preserving peptide–peptide interactions, enabling higher-order assembly of DNA nanostructures connected through peptides.[39] In particular, coiled-coil peptides strategically attached to DNA origami have proven useful for this strategy. This emerging area has the potential for creating unique DNA–protein hybrid materials.

Surface placement

In addition to the solution-phase assembly methods described above, an alternative strategy for organizing DNA nanostructures across large length scales involves placing them on surfaces. Surface-based approaches can be broadly divided into two main categories. The first is ''self-assembly'', where structures self-organize into a higher-order pattern on a uniform substrate (Fig. 3(A)). This method is simple and cost-effective, as it does not require precise patterning of surfaces with binding sites, but it does not offer control over the exact position and orientation of each structure relative to a fixed frame of reference. The second is ''precision-placement'', which involves positioning structures at predefined locations and orientations on a patterned substrate (Fig. 3(B)). This approach would allow structures to interface effectively with fixed external components, such as circuits imprinted on a surface.

In surface self-assembly, DNA nanostructures can either be assembled directly on an attractive substrate or first self-assembled in solution and then deposited onto the surface. In the first case, the substrate must not be overly attractive to DNA, as some surface mobility is needed for assembly with less defects. Researchers have found that materials like mica[40,41] and supported lipid membranes[42] are well-suited for this purpose. Most such assemblies on surfaces have been driven by base stacking interactions between dsDNA blunt ends at the edges of the nanostructures (Fig. 3(C)). Even without such energetic connections, structures can entropically self-organize on substrates at high packing densities, potentially yielding larger and more efficient assemblies (Fig. 3(D)).[40,43] Additionally, top-down methods can be used to guide the localization of structures to predefined sticky regions on the substrate.[44–46] In the second case, the structures must be interconnected through hybridization or base stacking interactions before deposition. Careful handling is required during deposition to ensure the sheet-like assemblies flatten correctly on the surface, which can constrain the maximum pattern size that can be achieved.[34,47]

In precision placement, nano-lithographic methods such as e-beam lithography are used to create attractive nanoscale patches on passivated substrates.[48–50] The number of DNA structures that bind per patch depends on its size: large patches allow multiple structures to bind, while small ones favor single structures. However, for true precision, the patches should closely match the size and shape of the DNA structure; this encourages the structures to align and fully overlap with the

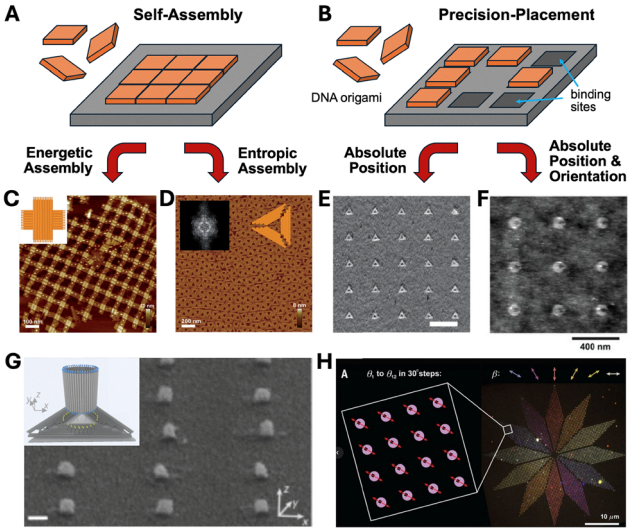

Fig. 3 Approaches for organizing DNA origami structures on surfaces: (A) self-assembly, which yields relative order without fixed absolute positioning, and (B) precision-placement, which uses prepatterned substrates to enforce greater control. Within self-assembly, organization can be achieved (C) energetically *via* attractive connections, to assemble triangular origami into densely packed arrays (reproduced with permission from ref. 40; Copyright 2014, Wiley-VCH), or (D) entropically *via* packing effects, to assemble cross-shaped DNA origami into 2D lattices (reproduced with permission from ref. 40; Copyright 2014, Wiley-VCH). Within precision-placement, organization can be achieved with (E) absolute position control of triangular origami *via* lithographically defined binding sites (reproduced with permission from ref. 51; Copyright 2014, American Chemical Society), and (F) absolute position and orientation control *via* shape-complementary binding domains (reproduced with permission from ref. 53; Copyright 2021 AAAS). (G) Vertical placement of barrel-shaped DNA origami on top of triangular origami positioned on triangular binding patches (reproduced with permission from ref. 52; Copyright 2023, Springer Nature). (H) Micrometers-sized polarimeter device created from arrays of precisely positioned and oriented circular DNA origami on silica substrate (reproduced with permission from ref. 53; Copyright 2021 AAAS).

patches to maximize binding energy. This method has successfully enabled the precise placement of simple symmetric origami shapes, such as equilateral triangles (Fig. 3(E)),[49,51] which possess "concave" binding-energy landscapes, *i.e.*, no metastable traps that might otherwise cause misalignments. Moreover, once correctly placed, these base-layer structures can support the docking of additional layers of 3D DNA origami (Fig. 3(G)).[52] However, due to the rotational symmetry of these shapes, achieving absolute orientation remains challenging; for instance, a well-aligned equilateral triangle can still deposit in any of the three orientations defined by the patch's vertices. Anisotropic shapes, even simple ones like scalene triangles, fail to resolve alignment issues due to metastable states in their binding energy landscapes, which cause structures to become trapped in misaligned orientations. A recent solution employed circular DNA origami with off-centered holes, creating a metastability-free energy landscape that enables precise placement and orientation control of anisotropic structures (Fig. 3(F)).[53] Using this method, researchers patterned arrays of DNA origami embedded with light-sensitive dyes in defined orientations, constructing a ~40 μm device capable of detecting the direction of light polarization (Fig. 3(H)).[53]

3. Dynamic higher-order assemblies

While remarkable progress has been made in both dynamic nanodevices and static higher-order assemblies, efforts to unify these themes to produce large, dynamic assemblies have only just begun to take shape. Yet, recent years have seen a surge of innovation, with many developments already achieved and more on the horizon. These emerging systems can be broadly classified into three categories: reconfigurable lattices that retain overall structural connectivity, static lattices that incorporate dynamic components, and lattice systems that undergo controlled assembly and disassembly.

Reconfigurable lattices

Much of the work on dynamic DNA superstructures has centered around systems assembled from dynamic building blocks, where the structure and mechanical properties of the entire lattice can be changed by reconfiguring its building blocks (Fig. 4(A)). These adaptable lattices have been realized in a variety of geometries, including linear arrays, 2D grids, and 3D crystals. Among these, linear systems are the most straightforward to implement and thus often serve as model systems for exploring different forms of dynamics. For instance, researchers have assembled tubular structures by linking DNA origami rings into extended cylinders, a design concept that has found multiple implementations. In one example, adjustable struts were embedded within the rings to control the tube's cross-sectional area in response to trigger strands.[54] In another example, the rings encapsulated liposomes, enabling tunable control over the liposome size and spatial organization by modifying the connections between the rings.[55] Linear assemblies have also been used to dynamically modulate the chirality of gold nanoparticle arrangements, achieving nanoscale optical control through strand displacement reactions (Fig. 4(C)).[56] Beyond traditional DNA connections, alternative linking strategies have been introduced, such as coiled-coil peptides that enable orthogonal and modular assembly of DNA origami chains with configurable mechanical properties, such as persistence length.[57] In another application, researchers achieved rapid, reversible shape changes using thermally responsive nanoparticle–DNA assemblies, pushing the boundaries of functional dynamics in linear origami lattices (Fig. 4(D)).[58]

Moving beyond 1D systems, dynamic 2D superstructures have also been explored. These lattices typically feature a grid of DNA origami units with struts spanning the joints to regulate the grid shape. Stimuli-responsive behavior is encoded by designing the struts to dissociate or change length in response to environmental changes. A prominent example involves the use of triplex-forming DNA motifs, which are sensitive to pH changes; in basic conditions, these motifs associate and lock the lattice geometry (Fig. 4(B)).[59] In another study, researchers expanded on

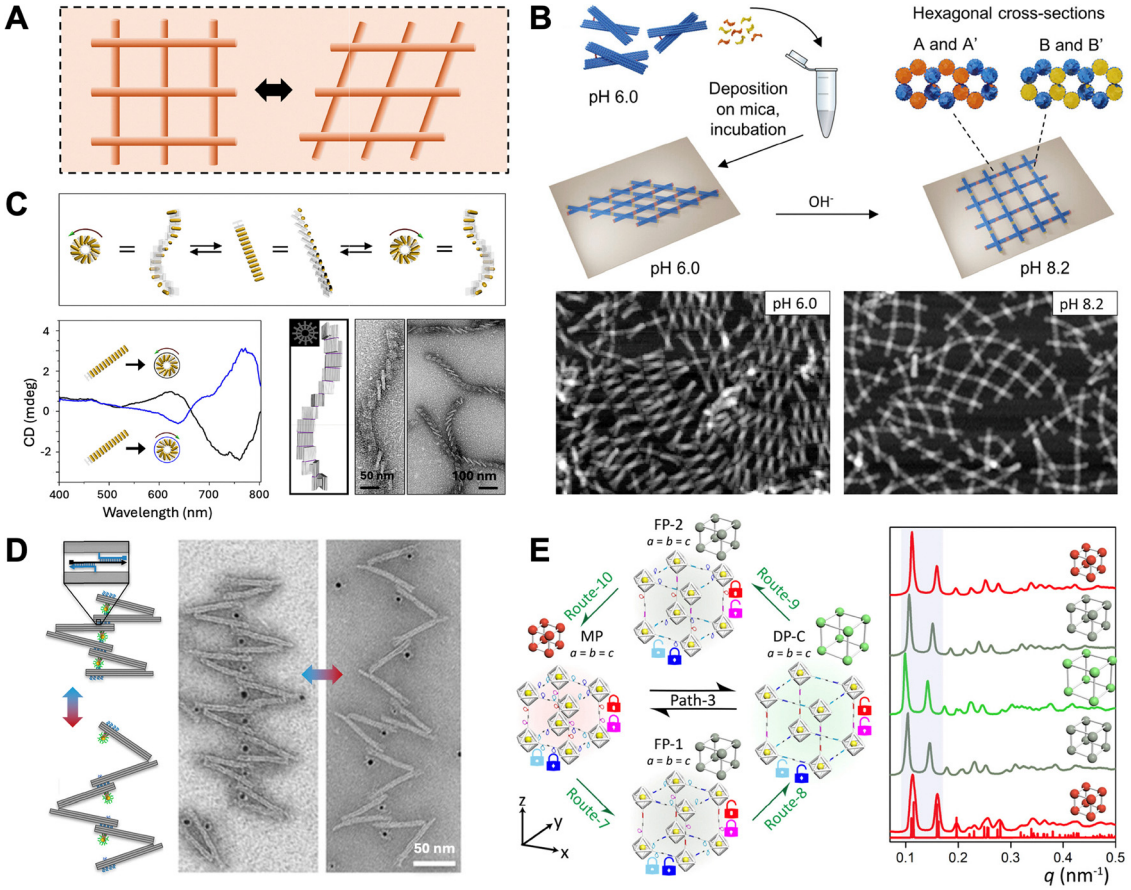

Fig. 4 Reconfigurable superstructures assembled from dynamic origami building blocks. (A) Schematic of a reconfigurable lattice. (B) Accordion-like reconfiguration of an origami assembly in response to changes in pH (reproduced with permission from ref. 59; Copyright 2023 American Chemical Society). (C) Linear array of origami-nanoparticle assemblies that switch chirality after addition of trigger strands (reproduced with permission from ref. 56; Copyright 2018 American Chemical Society). (D) Origami-nanoparticle chains that can rapidly cycle between two states at different temperatures (reproduced with permission from ref. 58; Copyright 2019 American Chemical Society). (E) Crystals of nanoparticle-carrying origami with changeable lattice parameters and the corresponding SAXS spectra for each lattice type (reproduced with permission from ref. 63; Copyright 2023 American Chemical Society).

this approach by incorporating trigger and suppressor strands that allow programmable control over which regions of the lattice respond to pH. Furthermore, salt-sensitive motifs were introduced, creating a versatile multi-responsive platform for sensing and adaptive materials.[60,61] Another method for creating multi-responsive assemblies involves decorating the edges of block-shaped origami with shape-complimentary motifs that bind with other origami through base stacking.[22] By adding trigger strands that disrupt the shape-complementarity of these motifs, or by changing the temperature or salt concentration which alter the base-stacking interactions, the assemblies could be transformed between open and closed states. In another innovative approach, a 2D lattice composed of identical Holliday junctions was designed to undergo a signal-induced wave of conformational change. The array initially adopts a state that favors extensive base stacking, but the introduction of a trigger strand imposes a constraint that initiates a sequential reconfiguration that propagates across the lattice.[29] This cascade mechanism opens up new avenues for cooperative behavior and long-range information transmission within DNA materials.

In recent years, researchers have started extending these design principles of reconfigurability to 3D lattices. These lattices typically rely on adjustable connectors between DNA origami units that allow the lattice parameters to change in response to external cues. In one example, connectors incorporating C-quadruplex-forming sequences enabled pH-responsive contraction of the crystal, as the quadruplexes dissociate under acidic conditions.[62] More elaborate designs have introduced connectors with multiple independently controlled domains, allowing the same crystal framework to switch between different lattice types when triggered by specific strands (Fig. 4(E)).[63] However, a recurring challenge in reconfigurable 3D systems is the trade-off between dynamic behavior and structural integrity: the flexibility required for reconfiguration often leads to softer, defect-prone crystals, and in some cases, significant aggregation during the assembly process. To address this, researchers have developed stiffer connectors with multiple contacts that preserve mechanical stability while maintaining reconfigurability.[64] One final form of reconfigurable DNA materials are constitutional dynamic network (CDN)

hydrogels, where reversible strand displacement reactions enable continuous structural and functional adaptation within a dynamic framework. In one particularly relevant study, the addition of trigger strands to a CDN increased the degree of crosslinking within the network, resulting in a hydrogel that could reversibly switch between three levels of stiffness.[65]

Static lattices with dynamic components

In this class of DNA superstructures, each structural unit contains two domains: a rigid one that self-assembles and provides order to the system, and a dynamic one that can respond to external signals (Fig. 5(A)). This ensures that the dynamic components are uniformly arranged in the desired pattern. Researchers have started exploring this idea to achieve intriguing phenomena such as communication, collective behavior, and cargo manipulation.

For example, motivated by protein allostery, researchers exploited steric interactions between the dynamic components to build a communication device. This principle has been demonstrated in a DNA origami system composed of a rigid base and two hinged arms, where both experimental results and coarse-grained simulations confirmed that the closure of one arm shifted the conformational distribution of the other arm (Fig. 5(C)).[66] This demonstrates the potential of such devices for transmitting signals along longer chains of such devices through a cascade of conformational changes. Beyond signaling, researchers have explored how arrays of rotatable elements could be used to enact order–disorder transitions, inducing macroscale structural changes through simple stimuli such as temperature and salt concentration (Fig. 5(D)).[67] This concept is embodied through so-called rotor lattices, where rotatable DNA origami components embedded in a static 2D framework interact with their neighbors in an orientation-dependent manner. Monte Carlo simulations of such systems have shown that they exhibit temperature-dependent order–disorder transitions, like the Ising lattice and beyond. Experimental realization of one such system used ssDNA to connect the rotors to the base lattice and enable free rotation.

In addition to dynamic components that rotate, these lattices can incorporate translatable components. In one study, dynamic transport was pursued through rotaxane-like structures, where a movable ring mechanically interlocks around a long fiber.[16] These systems allow for the controlled positioning and release of the ring, offering a platform for programmable transport mechanisms over mesoscopic distances (Fig. 5(E)). Finally, static lattices can also contain domains that store and release cargo. Recently, researchers have created DNA crystals with reconfigurable elements that release gold nanoparticles in response to specific molecular inputs.[68] When coupled with signal-amplifying transcriptional circuits, such crystals demonstrated the ability to sense and react to subtle environmental changes, laying the groundwork for innovations in dynamic photonic materials and catalytic systems (Fig. 5(B)).

Assembly-disassembly lattices

The third category of dynamic DNA superstructures includes those that undergo controlled assembly and disassembly in response to specific stimuli (Fig. 6(A)). For instance, tiles

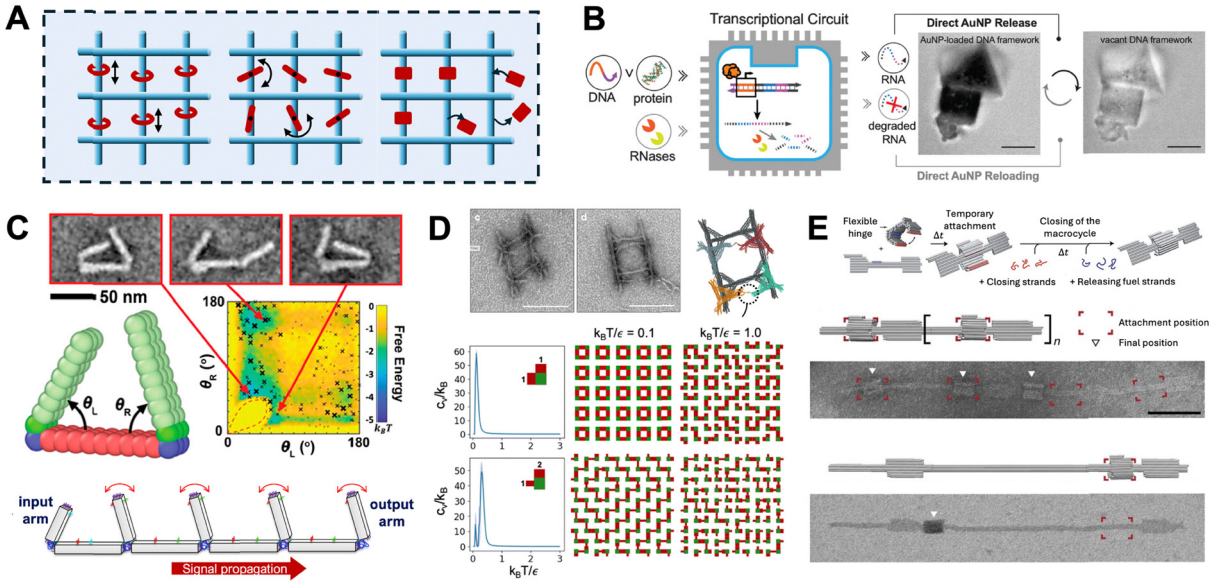

Fig. 5 Static superstructures housing dynamic DNA origami subunits. (A) Schematic of such lattices with translatable, rotatable, and dissociable components. (B) Crystal that loads and unloads gold nanoparticles in response to the output of a transcriptional circuit (reproduced with permission from ref. 68; Copyright 2024, American Chemical Society). (C) TEM images and angle distributions from coarse-grained simulations of minimal unit of a sterically communicating DNA origami array (reproduced with permission from ref. 66; Copyright 2023, American Chemical Society). (D) TEM images of 2 × 2 rotor lattice and order–disorder transitions of 10 × 10 lattices as predicted by Monte Carlo simulations (reproduced with permission from ref. 67; Copyright 2023, Royal Society of Chemistry). (E) Method for assembling mechanically interlocked rotaxane-like structures, as well as TEM images showing movement of the dynamic component (reproduced with permission from ref. 16; Copyright 2016, Springer Nature).

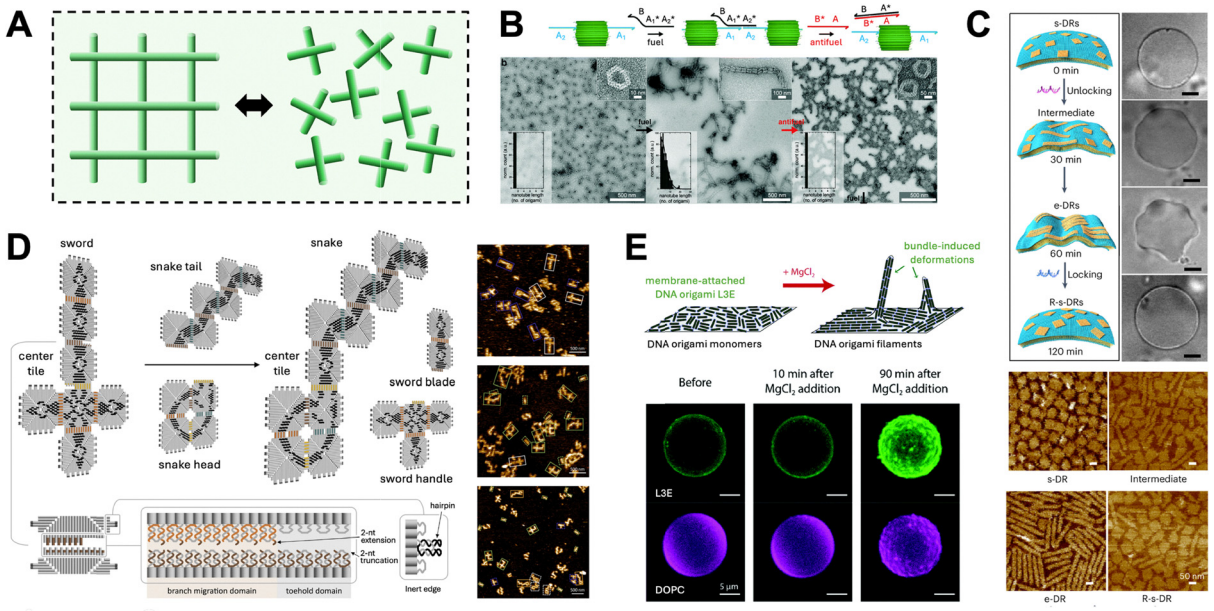

Fig. 6 Dynamic origami lattices capable of assembly and disassembly. (A) Schematic of one such lattice. (B) Barrels and interlocking strands that reversibly bind them into tubules, as shown in TEM images (reproduced with permission from ref. 71; Copyright 2020, Royal Society of Chemistry). (C) Origami clusters on a vesicle that reconfigure to modulate membrane shape, mimicking cellular behavior (reproduced with permission from ref. 76; Copyright 2025, Nature publishing group). (D) Tile displacement systems that reconfigure between distinct shapes, as seen in AFM images (reproduced with permission from ref. 70; Copyright 2023, AAAS). (E) Salt-induced origami filament bundling that form spiky membrane features (reproduced with permission from ref. 74; Copyright 2021, Royal Society of Chemistry).

equipped with triplex-forming overhangs can be programmed to assemble into different shapes depending on the pH, using overhangs with varied pH sensitivities to direct shape selection.[69] In another approach, sticky ends and hairpin loops could be arranged on the edges of tiles to mimic the logic of toehold-mediated strand displacement, enabling dynamic replacement of one tile assembly with another. This was demonstrated in a system that transitioned from a "sword" to a "snake" configuration upon addition of new tiles (Fig. 6(D)).[70] Beyond finite-sized assemblies, other systems form polymer-like filaments of indefinite length. These are typically constructed by attaching single-stranded overhangs to DNA origami units and introducing fuel strands that bridge these overhangs. Incorporating a toehold into the fuel strand allows for subsequent disassembly *via* the addition of anti-fuel strands. This mechanism has been shown using hollow cylindrical origami that polymerize into tubules, with tunable kinetics and chain properties based on the overhang and origami design (Fig. 6(B)).[71] In another application, six-helix bundle (6HB) filaments co-assembled with photo-responsive lipids into dense fibrous networks that disintegrate upon UV exposure. Notably, the lipids shield the DNA origami from nuclease digestion when assembled, offering a promising direction for therapeutic delivery systems that activate upon light-triggered disassembly.[72]

In terms of applications, a particularly compelling platform of dynamic assembly and disassembly are lipid membranes, especially giant unilamellar vesicles (GUVs). In these applications, DNA origami structures are confined to the two-dimensional membrane surface but retain lateral mobility. This setup enables programmable organization and reconfiguration on the membrane, enabling feature formation and membrane deformation. As before, these origami can be connected through sticky ends that assemble upon the addition of trigger strands. Researchers have utilized this approach with cross-shaped origami to assemble grid-like arrays on membranes that noticeably deform the membrane.[73] In another approach, origami filaments were designed to self-assemble *via* blunt-end stacking.[74] As a result, the filaments remain dispersed in low salinity, but in high-salt environments they associate into spiky surface projections, transforming the membrane topology (Fig. 6(E)). Recently, researchers engineered triangular building blocks that can assemble into lipid-coated polyhedral shells when in contact with lipid vesicles.[75] By placing cholesterol on either the inside or outside of the triangles, the directionality of the budding process could be controlled, enabling the generation of vesicles with either DNA exoskeletons or endoskeletons. In another recent advancement, researchers designed DNA origami bundles to cluster on vesicle surfaces to induce vesicle deflation, mimicking cytoskeletal contraction.[76] Upon the addition of biogenic pores, the origami reorganized into synthetic, sealable channels and the vesicle re-inflated, effectively reconstituting a synthetic cell-like structure (Fig. 6(C)). Such membrane-associated assembly systems,[77] not only provide a route to programmable shape control but also represent a powerful toolkit for creating responsive, bioinspired devices capable of transporting macromolecules and mimicking fundamental cellular processes.

4. Outlook

As reconfigurable DNA-based superstructures become increasingly sophisticated, they present a host of new application

opportunities, as well as significant challenges. In this section, we discuss some of the core challenges that must be addressed to realize robust, higher-order dynamic architectures and highlight several potential applications in optoelectronics and biomedicine.

Assembly yield

One of the central challenges in realizing dynamic DNA origami superstructures is improving the fidelity (yield) of self-assembly when flexible connecting elements are involved. The flexible elements typically required for dynamic mechanisms inherently introduce a high degree of configurational entropy. While some flexibility is beneficial for tolerating structural imperfections during assembly, excessive softness increases the number of pathways that lead to defects or aggregation and reduces the thermodynamic driving force for assembly by exacerbating the entropic cost of ordering. Strategies to solve this issue could include programmable mechanical constraints, for instance, temporarily rigidifying flexible elements during assembly. Another potential solution, specifically for 1D and 2D systems, would be to divide the building blocks into rigid and dynamic components, assembling the static ones first, then attaching the dynamic components to the pre-formed static lattice. This pathway should promote larger and more ordered systems.

Energy landscapes

Another important direction is the ability to design tunable free energy landscapes for dynamic connections, such as hinges and rotors. Depending on the intended application, rotating joints may need to be entirely free (*e.g.*, for molecular rotors), exhibit spring-like harmonic potentials (*e.g.*, for sensing or resonance devices), or possess bistable potentials (*e.g.*, for switchable actuators). While some experimental and computational work has characterized the free-energy profiles of DNA hinges as a function of geometric and sequence-based parameters,[78,79] it remains difficult to design arbitrary energy landscapes with precision. Creating systematic design rules or predictive models remains an open problem, and advances here could help to optimize the balance of structural stability required for assembly while maintaining some dynamic function. Emerging efforts include machine learning approaches that could help correlate structural features—such as ssDNA segment length, crossover density, or junction topology—with energy landscape properties like barrier height and well depth.

Collective phenomena

There is growing interest in shifting from studying the dynamics of individual structures to exploring how complex, collective behaviors can emerge from interactions among many dynamic structures. This review has highlighted early examples such as rotor lattices, cascading conformational waves, and steric communication chains, which begin to demonstrate the potential of ''many-body'' dynamics in DNA structures. Looking ahead, more complex collective phenomena like swarming behavior, resonance, pattern formation, adaptation, and topological solitons could be explored. These directions would require a careful mapping of the conditions and material properties underlying the desired phenomena onto those achievable with DNA. Furthermore, to facilitate achieving these goals, new mechanisms may need to be developed to control these collective responses, such as electrical, magnetic, or optical fields. Such stimuli are non-invasive and could offer precise, rapid, and reversible control over system-wide transitions. This is especially important for order–disorder transitions in rotor lattices, where thermal actuation is constrained by DNA's narrow stability range. At the same time, theoretical models must evolve to better reflect the continuous, thermally fluctuating nature of these nanoscale systems. For instance, in rotor lattices, current models often rely on rigid or discrete-state approximations that fail to capture the inherent flexibility and interactions of DNA origami components.

Orthogonal stimuli

Another persistent challenge in the development of dynamic DNA origami systems is the lack of specificity and orthogonality in the stimuli. Most current designs rely on a small set of physical and chemical triggers—namely pH, temperature, ionic strength, and strand displacement reactions. While these inputs are effective, they tend to overlap in their effects and are not easily decoupled from each other, leading to undesired crosstalk in complex or multi-component environments. Moreover, the global nature of many of these stimuli limits their spatial and temporal resolution, making it difficult to actuate one component in a structure without affecting others nearby. Addressing this issue requires the development of more orthogonal and localized control mechanisms. One avenue is the incorporation of molecular inputs that are chemically and functionally distinct, such as photo-responsive moieties, redox-active groups, aptamer–ligand pairs, or restriction enzymes. From a systems perspective, the use of multiplexed logic circuits—where multiple stimuli must be satisfied for activation—could dramatically increase the selectivity of responses. Integrating such systems into higher-order origami assemblies would require careful design of overlapping input conditions and could benefit from computational tools that optimize trigger combinations to minimize undesired cross-activation.

Optoelectronic applications

Higher-order DNA origami structures have already enabled the fabrication of advanced optical and electronic materials, including DNA-templated photonic crystals,[80] chip-integrated polarimeter,[53] prototypical photodetectors,[46] chiral metamaterials,[81] and plasmonic metasurfaces.[82] Introducing dynamic behavior into these superstructures would add a powerful new dimension, allowing for reconfigurable, stimuli-responsive architectures that move beyond static templating. Such systems could lead to tunable photonic crystals, chiral metamaterials, and metasurfaces capable of real-time optical modulation—promising for applications in sensing, communications, and adaptive optics. For instance, DNA-based photonic crystals with adjustable lattice spacing could shift their bandgaps in response to external stimuli like heat or light, mimicking biological systems such as chameleon skin.[83] Likewise, arrays

that undergo sharp order–disorder transitions could be functionalized with optoelectronic components, such as gold nanoparticles, to induce abrupt changes in optical or electronic properties. However, realizing these applications often requires operation in dry air, which means DNA structures must be converted into more robust templates—for example, by silica coating followed by deposition of high–refractive index materials such as silicon or TiO_2.[46,80] Ensuring that functional activity is preserved after coating and achieving reliable performance in non-aqueous environments remain critical challenges. Another key hurdle is the precise and multiplexed integration of different species of origami structures onto surfaces, as current placement techniques typically accommodate only a single origami species at a time.

Biomedical applications

Similarly, dynamic DNA superstructures could offer powerful new capabilities for biomedical applications by enabling responsive, amplified, and programmable behaviors. For example, in the context of smart delivery systems, dynamic assemblies could provide means for amplified delivery responses where a single stimulus can be rapidly communicated throughout an assembly, leading to an amplified response and release of many drug molecules or a combination of drug molecules. In biosensing, dynamic superstructures could amplify signals or perform multi-input logic operations to detect complex biomarker patterns with high sensitivity. Higher-order arrays also enable interfacing with microscale systems—from microelectronic devices to live cells—potentially allowing direct modulation of processes like cell division or endocytosis. Furthermore, these dynamic assemblies are uniquely suited to serve as cytoskeletal-like elements in synthetic cells,[76,84] enabling controllable mechanical and biochemical responses. Despite these exciting possibilities, realizing such systems remains challenging due to the need for robust and coordinated inter-device communication, improved stability in physiological environments, and smoother integration with living or engineered biological platforms.

5. Conclusions

In this Minireview, we explored the growing interest at the intersection of dynamic mechanisms and hierarchical assembly within the field of DNA nanotechnology. Before discussing how these two subfields can be combined to create dynamic higher-order DNA architectures, we first reviewed the foundational approaches for actuating individual DNA origami devices and the assembly techniques for forming large arrays of DNA nanostructures. We proceeded to analyze the current work on dynamic higher-order assemblies, grouping it into three overarching strategies: lattices composed of dynamic constitutive subunits, static frameworks housing responsive elements, and assemblies that undergo triggered dissociation and reassociation. Next, we highlighted some of the remaining fundamental challenges in achieving high assembly yield, tuning the energy landscapes that control motion, harnessing collective phenomena, and utilizing orthogonal stimuli to enable complex responses. Finally, we looked ahead to some of the promising applications of dynamic superstructures, particularly in optoelectronics and biomedicine.

Author contributions

Conceptualization: I. V. M., Y. K., C. E. C., G. A. Investigation: D. D., S. S., A. A., T. T., I. V. M., Y. K., C. E. C., G. A. Writing – original draft: D. D., S.S., I. V. M., Y. K., C. E. C., G. A. Writing – review and editing: all the authors. Supervision: G. A. Visualization: D. D., A. A., Y. K., C. E. C., G. A.

Conflicts of interest

There are no conflicts to declare.

Data availability

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

References

1 N. C. Seeman and H. F. Sleiman, *Nat. Rev. Mater.*, 2018, **3**, 17068.
2 P. W. K. Rothemund, *Nature*, 2006, **440**, 297–302.
3 M. DeLuca, D. Duke, T. Ye, M. Poirier, Y. Ke, C. Castro and G. Arya, *Nat. Commun.*, 2024, **15**, 3015.
4 S. Dey, C. Fan, K. V. Gothelf, J. Li, C. Lin, L. Liu, N. Liu, M. A. Nijenhuis, B. Saccà and F. C. Simmel, *Nat. Rev. Methods Primers*, 2021, **1**, 13.
5 O. I. Wilner and I. Willner, *Chem. Rev.*, 2012, **112**, 2528–2556.
6 M. DeLuca, Z. Shi, C. E. Castro and G. Arya, *Nanoscale Horiz.*, 2020, **5**, 182–201.
7 P. Zhan, A. Peil, Q. Jiang, D. Wang, S. Mousavi, Q. Xiong, Q. Shen, Y. Shang, B. Ding, C. Lin, Y. Ke and N. Liu, *Chem. Rev.*, 2023, **123**, 3976–4050.
8 A. E. Marras, *MRS Commun.*, 2022, **12**, 543–551.
9 I. V. Martynenko, V. Ruider, M. Dass, T. Liedl and P. C. Nickels, *ACS Nano*, 2021, **15**, 10769–10774.
10 A. E. Marras, L. Zhou, H. J. Su and C. E. Castro, *Proc. Natl. Acad. Sci. U. S. A.*, 2015, **112**, 713–718.
11 S. Lauback, K. R. Mattioli, A. E. Marras, M. Armstrong, T. P. Rudibaugh, R. Sooryakumar and C. E. Castro, *Nat. Commun.*, 2018, **9**, 1446.
12 E. Kopperger, J. List, S. Madhira, F. Rothfischer, D. C. Lamb and F. C. Simmel, *Science*, 2018, **359**, 296–301.
13 A. Kuzyk, R. Schreiber, H. Zhang, A. O. Govorov, T. Liedl and N. Liu, *Nat. Mater.*, 2014, **13**, 862–866.
14 Y. Wang, J. V. Le, K. Crocker, M. A. Darcy, P. D. Halley, D. Zhao, N. Andrioff, C. Croy, M. G. Poirier, R. Bundschuh and C. E. Castro, *Nucleic Acids Res.*, 2021, **49**, 8987–8999.
15 M. Schickinger, M. Zacharias and H. Dietz, *Proc. Natl. Acad. Sci. U. S. A.*, 2018, **115**, E7512–E7521.

16 J. List, E. Falgenhauer, E. Kopperger, G. Pardatscher and F. C. Simmel, *Nat. Commun.*, 2016, **7**, 12414.
17 P. Ketterer, E. M. Willner and H. Dietz, *Sci. Adv.*, 2016, **2**, e1501209.
18 B. Yurke, A. J. Turberfield, A. P. Mills, Jr., F. C. Simmel and J. L. Neumann, *Nature*, 2000, **406**, 605–608.
19 C. Zhou, X. Duan and N. Liu, *Nat. Commun.*, 2015, **6**, 8102.
20 E. S. Andersen, M. Dong, M. M. Nielsen, K. Jahn, R. Subramani, W. Mamdouh, M. M. Golas, B. Sander, H. Stark, C. L. Oliveira, J. S. Pedersen, V. Birkedal, F. Besenbacher, K. V. Gothelf and J. Kjems, *Nature*, 2009, **459**, 73–76.
21 S. Nummelin, B. Shen, P. Piskunen, Q. Liu, M. A. Kostiainen and V. Linko, *ACS Synth. Biol.*, 2020, **9**, 1923–1940.
22 T. Gerling, K. F. Wagenbauer, A. M. Neuner and H. Dietz, *Science*, 2015, **347**, 1446–1452.
23 A. K. Pumm, W. Engelen, E. Kopperger, J. Isensee, M. Vogt, V. Kozina, M. Kube, M. N. Honemann, E. Bertosin, M. Langecker, R. Golestanian, F. C. Simmel and H. Dietz, *Nature*, 2022, **607**, 492–498.
24 M. Centola, E. Poppleton, S. Ray, M. Centola, R. Welty, J. Valero, N. G. Walter, P. Sulc and M. Famulok, *Nat. Nanotechnol.*, 2024, **19**, 226–236.
25 M. Kim, C. Lee, K. Jeon, J. Y. Lee, Y. J. Kim, J. G. Lee, H. Kim, M. Cho and D. N. Kim, *Nature*, 2023, **619**, 78–86.
26 A. E. Marras, Z. Shi, M. G. Lindell, 3rd, R. A. Patton, C. M. Huang, L. Zhou, H. J. Su, G. Arya and C. E. Castro, *ACS Nano*, 2018, **12**, 9484–9494.
27 M. Darcy, K. Crocker, Y. Wang, J. V. Le, G. Mohammadiroozbahani, M. A. S. Abdelhamid, T. D. Craggs, C. E. Castro, R. Bundschuh and M. G. Poirier, *ACS Nano*, 2022, **16**, 5682–5695.
28 P. Stommer, H. Kiefer, E. Kopperger, M. N. Honemann, M. Kube, F. C. Simmel, R. R. Netz and H. Dietz, *Nat. Commun.*, 2021, **12**, 4393.
29 J. Song, Z. Li, P. Wang, T. Meyer, C. Mao and Y. Ke, *Science*, 2017, **357**, eaan3377.
30 C. Xie, K. Chen, Z. Chen, Y. Hu and L. Pan, *Small*, 2024, **20**, e2401396.
31 G. A. Knappe, E. C. Wamhoff and M. Bathe, *Nat. Rev. Mater.*, 2023, **8**, 123–138.
32 S. Wang, Z. Zhou, N. Ma, S. Yang, K. Li, C. Teng, Y. Ke and Y. Tian, *Sensors*, 2020, **20**, 6899.
33 N. C. Seeman, *J. Theor. Biol.*, 1982, **99**, 237–247.
34 E. Winfree, F. Liu, L. A. Wenzler and N. C. Seeman, *Nature*, 1998, **394**, 539–544.
35 G. Tikhomirov, P. Petersen and L. Qian, *Nature*, 2017, **552**, 67–71.
36 C. M. Wintersinger, D. Minev, A. Ershova, H. M. Sasaki, G. Gowri, J. F. Berengut, F. E. Corea-Dilbert, P. Yin and W. M. Shih, *Nat. Nanotechnol.*, 2023, **18**, 281–289.
37 S. Woo and P. W. Rothemund, *Nat. Chem.*, 2011, **3**, 620–627.
38 K. F. Wagenbauer, C. Sigl and H. Dietz, *Nature*, 2017, **552**, 78–83.
39 J. Jin, E. G. Baker, C. W. Wood, J. Bath, D. N. Woolfson and A. J. Turberfield, *ACS Nano*, 2019, **13**, 9927–9935.
40 A. Aghebat Rafat, T. Pirzer, M. B. Scheible, A. Kostina and F. C. Simmel, *Angew. Chem., Int. Ed.*, 2014, **53**, 7665–7668.
41 S. Woo and P. W. Rothemund, *Nat. Commun.*, 2014, **5**, 4889.
42 Y. Suzuki, M. Endo and H. Sugiyama, *Nat. Commun.*, 2015, **6**, 8052.
43 C. Kielar, S. Ramakrishnan, S. Fricke, G. Grundmeier and A. Keller, *ACS Appl. Mater. Interfaces*, 2018, **10**, 44844–44853.
44 J. M. Yun, K. N. Kim, J. Y. Kim, D. O. Shin, W. J. Lee, S. H. Lee, M. Lieberman and S. O. Kim, *Angew. Chem., Int. Ed.*, 2012, **51**, 912.
45 R. Meyer, B. Saccà and C. M. Niemeyer, *Angew. Chem., Int. Ed.*, 2015, **127**, 12207–12211.
46 A. Michelson, L. Shani, J. S. Kahn, D. C. Redeker, W. I. Lee, K. R. DeOlivares, K. Kisslinger, N. Tiwale, H. Yan, A. Pattammattel, C. Y. Nam, V. S. Pribiag and O. Gang, *Sci. Adv.*, 2025, **11**, eadt5620.
47 A. Rajendran, M. Endo, Y. Katsuda, K. Hidaka and H. Sugiyama, *ACS Nano*, 2011, **5**, 665–671.
48 A. Gopinath, E. Miyazono, A. Faraon and P. W. Rothemund, *Nature*, 2016, **535**, 401–405.
49 A. M. Hung, C. M. Micheel, L. D. Bozano, L. W. Osterbur, G. M. Wallraff and J. N. Cha, *Nat. Nanotechnol.*, 2010, **5**, 121–126.
50 R. J. Kershner, L. D. Bozano, C. M. Micheel, A. M. Hung, A. R. Fornof, J. N. Cha, C. T. Rettner, M. Bersani, J. Frommer, P. W. K. Rothemund and G. M. Wallraff, *Nat. Nanotechnol.*, 2009, **4**, 557–561.
51 A. Gopinath and P. W. Rothemund, *ACS Nano*, 2014, **8**, 12030–12040.
52 I. V. Martynenko, E. Erber, V. Ruider, M. Dass, G. Posnjak, X. Yin, P. Altpeter and T. Liedl, *Nat. Nanotechnol.*, 2023, **18**, 1456–1462.
53 A. Gopinath, C. Thachuk, A. Mitskovets, H. A. Atwater, D. Kirkpatrick and P. W. K. Rothemund, *Science*, 2021, **371**, eabd6179.
54 A. Kucinic, C. M. Huang, J. Wang, H. J. Su and C. E. Castro, *Nanoscale*, 2023, **15**, 562–572.
55 Z. Zhang, Y. Yang, F. Pincet, M. C. Llaguno and C. Lin, *Nat. Chem.*, 2017, **9**, 653–659.
56 X. Lan, T. Liu, Z. Wang, A. O. Govorov, H. Yan and Y. Liu, *J. Am. Chem. Soc.*, 2018, **140**, 11763–11770.
57 T. Teng, J. Bernal-Chanchavac, N. Stephanopoulos and C. E. Castro, *Adv. Sci.*, 2024, **11**, e2307257.
58 J. A. Johnson, A. Dehankar, J. O. Winter and C. E. Castro, *Nano Lett.*, 2019, **19**, 8469–8475.
59 S. Julin, V. Linko and M. A. Kostiainen, *ACS Nano*, 2023, **17**, 11014–11022.
60 Y. Kobayashi, R. Toho and Y. Suzuki, *ACS Appl. Nano Mater.*, 2025, **8**, 8106–8112.
61 K. Watanabe, I. Kawamata, S. Murata and Y. Suzuki, *JACS Au*, 2023, **3**, 1435–1442.
62 Y. Wang, X. Yan, Z. Zhou, N. Ma and Y. Tian, *Angew. Chem., Int. Ed.*, 2022, **61**, e202208290.
63 X. Yan, Y. Wang, N. Ma, Y. Yu, L. Dai and Y. Tian, *J. Am. Chem. Soc.*, 2023, 3978–3986.
64 N. Ma, L. Dai, Z. Chen, M. Ji, Y. Wang and Y. Tian, *Nano Lett.*, 2021, **21**, 3581–3587.

65 L. Yue, S. Wang, V. Wulf and I. Willner, *Nat. Commun.*, 2019, **10**, 4774.
66 Y. C. Wang, S. Sensale, M. Pedrozo, C. M. Huang, M. G. Poirier, G. Arya and C. E. Castro, *ACS Nano*, 2023, **17**, 8271–8280.
67 M. DeLuca, W. G. Pfeifer, B. Randoing, C. M. Huang, M. G. Poirier, C. E. Castro and G. Arya, *Nanoscale*, 2023, **15**, 8356–8365.
68 Y. Xiong, C. Yancey, H. J. Lee, D. G. Lee, E. Helm, B. Kang, A. Grinthal, D. McKeen, O. Gang and R. Schulman, *J. Am. Chem. Soc.*, 2024, **146**, 30802–30812.
69 S. Yang, W. Liu and R. Wang, *Nanoscale*, 2019, **11**, 18026–18030.
70 N. Sarraf, K. R. Rodriguez and L. Qian, *Sci. Robot.*, 2023, **8**, eadf1511.
71 S. Groeer and A. Walther, *Nanoscale*, 2020, **12**, 16995–17004.
72 S. Julin, N. Best, E. Anaya-Plaza, E. Enlund, V. Linko and M. A. Kostiainen, *Chem. Commun.*, 2023, **59**, 14701–14704.
73 J. Yang, K. Jahnke, L. Xin, X. Jing, P. Zhan, A. Peil, A. Griffo, M. Skugor, D. Yang, S. Fan, K. Gopfrich, H. Yan, P. Wang and N. Liu, *Nano Lett.*, 2023, **23**, 6330–6336.
74 H. G. Franquelim, H. Dietz and P. Schwille, *Soft Matter*, 2021, **17**, 276–287.
75 M. T. Pinner and H. Dietz, *bioRxiv*, 2024, preprint, DOI: **10.1101/2024.01.18.576181**.
76 S. Fan, S. Wang, L. Ding, T. Speck, H. Yan, S. Nussberger and N. Liu, *Nat. Mater.*, 2025, **24**, 278–286.
77 P. Zhan, J. Yang, L. Ding, X. Jing, K. Hipp, S. Nussberger, H. Yan and N. Liu, *Sci. Adv.*, 2024, **10**, eadn8903.
78 Z. Shi and G. Arya, *Nucleic Acids Res.*, 2020, **48**, 548–560.
79 C. K. Wong and J. P. Doye, *Appl. Sci.*, 2022, **12**, 5875.
80 G. Posnjak, X. Yin, P. Butler, O. Bienek, M. Dass, S. Lee, I. D. Sharp and T. Liedl, *Science*, 2024, **384**, 781–785.
81 B. Dong, X. Xu, R. Guan, S. Jiang, L. Ma, H. Hu, Y. Ke, N. Liu and X. Lan, *Nano Lett.*, 2025, **25**, 5705–5712.
82 M. Dass, L. Raab, C. Pauer, C. Sikeler, L. Heinze, J. Tavacoli, I. V. Martynenko, U. Rührmair, G. Posnjak and T. Liedl, *Adv. Opt. Mater.*, 2025, 2500496.
83 J. Teyssier, S. V. Saenko, D. van der Marel and M. C. Milinkovitch, *Nat. Commun.*, 2015, **6**, 6368.
84 P. Zhan, K. Jahnke, N. Liu and K. Gopfrich, *Nat. Chem.*, 2022, **14**, 958–963.

