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Mini-Review

DNA Origami Colloidal Crystals: Opportunities and Challenges

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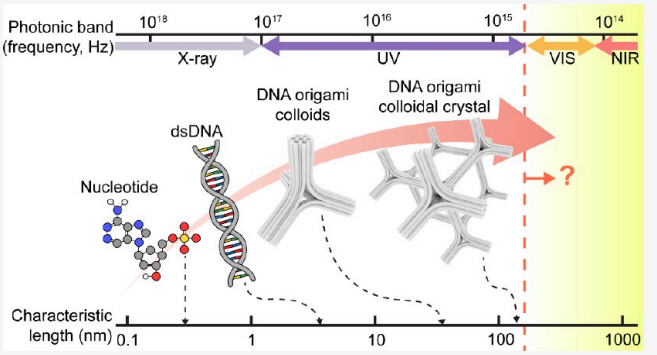
ABSTRACT: Over the last three decades, colloidal crystallization has provided an easy-to-craft platform for mesoscale engineering of photonic and phononic crystals. Nevertheless, the crystal lattices achieved thus far with commodity colloids are largely limited to symmetric and densely packed structures, restricting their functionalities. To obtain non-close-packed crystals and the resulting complexity of the available structures, directional binding between "patchy" colloids has been pursued. However, the conventional "patchy" colloids have been restricted to micrometer-scale spherical particles or clusters. In this Mini-Review, we argue that the time has come to widen the scope of the colloidal palette and include particles made using DNA origami. By benefiting from its unprecedented ability to control nanoscale shapes and patch placement and incorporate various nanomaterials, DNA origami enables novel engineering of colloidal crystallization, particularly for photonic and phononic applications. This mini-review summarizes the recent progress on using DNA origami for colloidal crystallization, together with its challenges and opportunities.



KEYWORDS: *DNA origami, colloidal crystals, self-assembly, colloids, lattice engineering*

Before the 1990s, colloids were primarily viewed as mesoscale counterparts of atoms or molecules. Their *in situ* visibility and slow Brownian diffusion enabled the use of optical microscopy to study atomic and molecular thermodynamic behaviors, such as phase transitions and crystallization kinetics.

In the late 1990s, colloids were reenvisioned as material libraries for mesoscale crystal engineering. This paradigm shift coincided with the emerging concepts of photonic crystals and photonic glasses.[1] Following E. Yablonovitch and S. John's establishment of the modern concept of photonic crystals in 1987 (notably, Lord Rayleigh has pointed out the existence of photonic bandgaps in 1D regular stacks of dielectric materials as early as 1887),[2−4] there was a strong demand for 3D crystals with mesoscale periodicities. In this context, colloidal crystallization saw a surge of interest from both colloidal and photonic societies, particularly as the field of self-assembly rapidly advanced during the 1990s and 2000s. Furthermore, in addition to photons, other types of waves—including sound, thermal, and mechanical vibrations, collectively known as phonons—can also be sculpted by their interactions with crystals with precisely controlled lattice geometries, densities, and periodicities.[5] Overall, it has been established that the diversity of lattice structures, densities, and periodicities in mesoscale colloidal crystals have been advancing toward maximizing the bandgap width of photonic and phononic crystals, to enable applications such as beam splitters, polarizers, solar absorbers, and lasers (for photonic crystals) or thermal insulators, optomechanical cavities, and acoustic rectifiers (for phononic crystals).[5,6]

Generally, colloidal crystallization is achieved by inducing an entropic or enthalpic attraction between particles. Entropy-driven packing, ligand-mediated binding, and capillary forces (e.g., dip-coating or confined assembly within emulsions) are some of the available approaches to introducing attractive interaction between colloids, leading to long-range colloidal crystallization.[7] In these cases, the lattice structure with the most efficient packing is generally obtained, as it tends to maximize the system's entropy, as in hard-sphere crystallization, or minimize its energy, as in ligand-mediated assembly.[8] As such, the available lattices of colloidal crystals assembled via such simple attractive forces are inherently limited by the shapes of the available building blocks. For example, spherical colloids typically crystallize into a face-centered cubic (FCC) lattice

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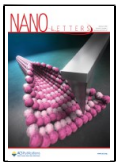

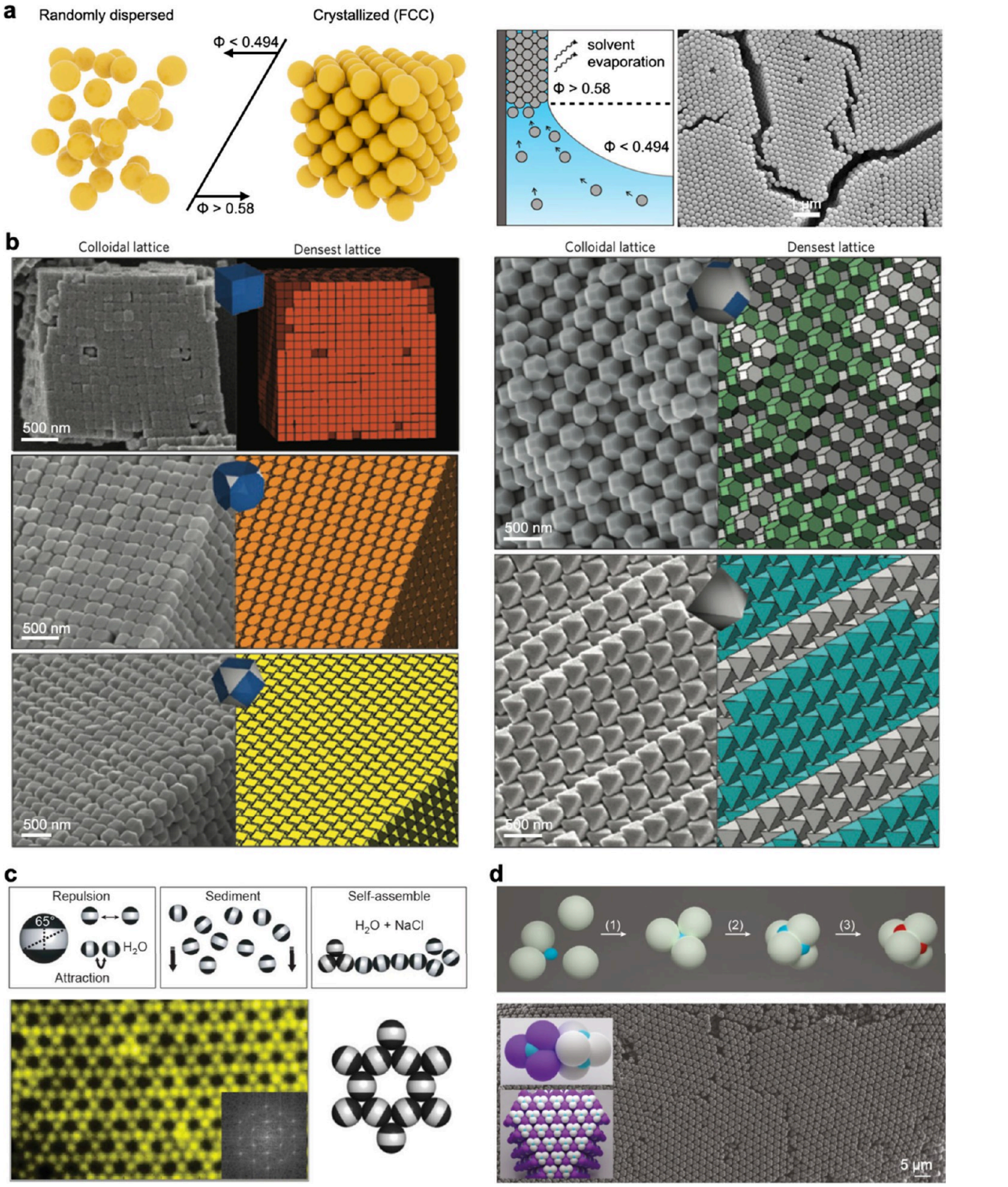

Figure 1. Crystallization of commodity colloids. (a) Entropy-driven packing of the spherical colloids into a face-centered cubic (FCC) lattice through condensation of colloids (left panel) and capillary-assisted self-assembly (right panel).[7] Adapted from ref 7. Available under a CC-BY 4.0. Copyright 2010 National Academy of Sciences. (b) Entropy-driven self-assembly of polyhedral noble-metal colloids into densest polyhedron lattices. (Left panels) Scanning electron microscopy (SEM) micrographs of the assembled colloidal lattice. (Right panels) Corresponding schematics of their known lattice under densest packing.[11] Adapted from ref 11. Copyright 2012 Springer Nature. (c) (Top panels) Schematics illustrating the self-assembly process of triblock Janus spheres with hydrophobic patches (black) and charged equator section (white). (Bottom panel) Fluorescence microscope images of the Kagome lattice assembled by directional assembly of the patch colloids. The corresponding fast Fourier transform (FFT) pattern is also shown in the bottom right corner.[15] Adapted from ref 15. Copyright 2011 Springer Nature. (d) Crystallization of diamond cubic crystals using tetrahedral patch clusters. (Top panel) Schematics illustrating the synthesis of compressed tetrahedral patchy clusters. White, blue, and red regions represent polystyrene particles, oil droplet, and DNA patches, respectively. (Bottom panel) A SEM image of the 111 plane of colloidal diamond crystals. Schematics on the left represent staggered assembly of the patch clusters and resultant diamond lattice.[16] Adapted from ref 16. Copyright 2020 Springer Nature.

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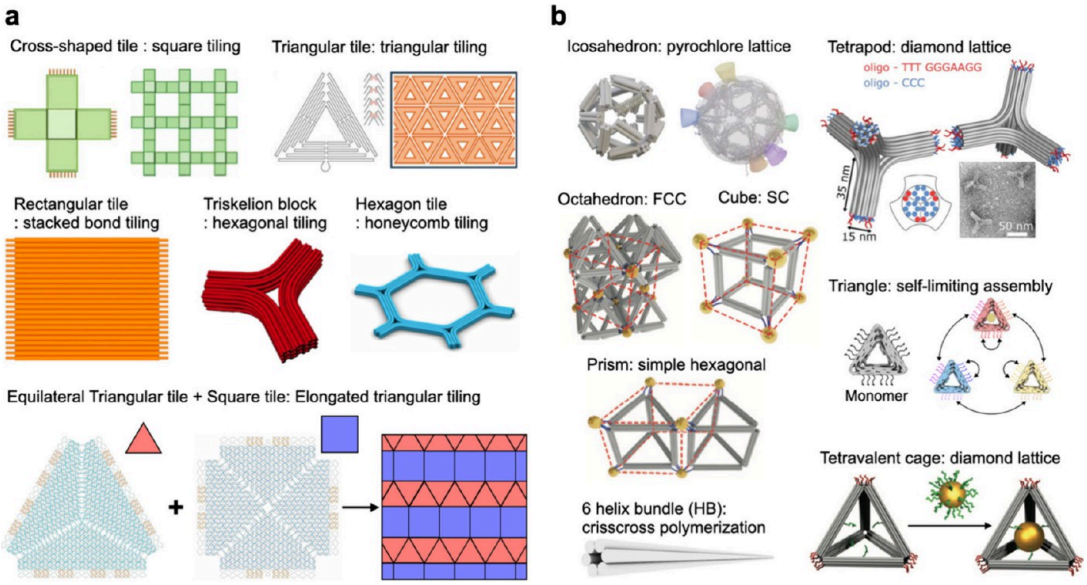

Figure 2. Representative DNA origami motifs used for colloidal crystallization and lattice engineering. (a) DNA origami motifs with on-plane directional patches for 2-dimensional (2D) lattice formation (i.e., tilling).[25−30] Adapted from refs 18, 25−27, 29, 30. Copyright 2006 Springer Nature; Copyright 2023 American Chemical Society; Available under a CC-BY 4.0. Copyright 2015 American Chemical Society. Available under a CC-BY 4.0. Copyright 2015 Springer Nature; Copyright 2014 Wiley-VCH GmbH. Copyright 2016 American Chemical Society. (b) DNA origami designs with out-of-plane directional patches for 3D crystallization.[19−24] Adapted with permission from refs 19−24. Copyright 2016 Springer Nature; Available under a CC-BY 4.0. Copyright 2024 American Association for the Advancement of Science. Copyright 2016 American Association for the Advancement of Science. Copyright 2024 American Association for the Advancement of Science. Copyright 2024 American Association for the Advancement of Science. Copyright 2023 Springer Nature.

(also referred to as opals) with a packing ratio of approximately 74 vol % and a coordination number of 12 rather than into other Bravais lattices (Figure 1a).

During the early stages of colloidal science and engineering (1960s−1980s), synthetic routes were developed for polymeric (such as polystyrene (PS) and poly(methyl methacrylate) (PMMA)) and ceramic (including silica and titanium dioxide (TiO_2)) colloids, henceforth collectively referred to as commodity colloids.[9] These commodity colloids generally exhibit a spherical shape, thus limiting the accessible crystal lattices to primarily opals, in accordance with the densest packing rule.

In the early 2000s, there was significant progress in the synthesis of noble-metal colloids, such as gold (Au) and silver (Ag).[10] This advancement was promoted by precise control over the reduction of metallic ions and the subsequent crystallization of these reduced atoms, benefiting from developments in surface nanochemistry (e.g., ligand chemistry, exchange, and more). As a result, various polyhedral colloids, such as cubes, octahedra, and concave dodecahedra, could be synthesized. This diversity in shapes led to an expanded diversity of crystal structures. Indeed, simple cubic, body centered cubic, Minkowski, and face-centered tetragonal crystals, which are challenging to achieve with spherical colloids, were successfully assembled with polyhedral metallic colloids (Figure 1b).[11]

However, the colloidal lattices discussed thus far are still limited to symmetric and densely packed structures, which, in turn, limit the range of practical applications in photonics and phononics. For instance, opals tend to exhibit a relatively narrow photonic bandgap due to their high volume fraction and lattice geometry.[12] Although inverse opals can be fabricated through infiltrating the interstitial space and etching out the original opal colloids to achieve low volume fraction, their high symmetry points W and U in the Brillouin zone make it difficult to open up a wide photonic bandgap. In contrast, lower-density dielectric lattices with complex symmetry and connectivity can achieve a much wider photonic bandgap, even with lower refractive index contrasts. Due to these considerations, the diamond and related structures are considered among the most desirable lattices for photonic crystals.[12] The Kagome lattice is another desirable structure because it enables topologically protected wave propagation.[13] Both of these lattices are generally constructed with a lower volume fraction (less than 40 vol %) and coordination numbers of four or fewer. While closely packed lattices of polyhedral metallic colloids are useful in their own right for achieving exotic properties, like near-field enhancement and unnatural refractive indices,[14] methods for assembling low-density two-dimensional (2D) and three-dimensional (3D) structures are still needed.

To achieve non-close-packed structures and the resulting complexity of crystal lattices, directional interactions between colloidal building blocks have been extensively exploited using patchy colloids. For instance, glancing angle deposition onto preassembled monolayers of dielectric colloids has enabled the creation of patches on opposite poles of spherical particles, which can then be assembled into the Kagome lattice (Figure 1c).[15] Additionally, chemical synthesis methods have been developed to create patchy colloidal particles. The method introduced by Pine et al. is one of the most widely known approach for synthesizing patchy colloids.[16] By clustering spherical colloids into the symmetric clusters and subsequently applying partial masking (colloidal fusion), they synthesized colloids with tetravalent and symmetric patches (Figure 1d). These clusters can then be hierarchically assembled into complex crystal structures, like the diamond lattice, although the lattice was not independently stabilized (Figure 1d).[16]

While such commodity colloids with attractive patches have successfully circumvented the densest packing problem, it

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remains to be seen whether several critical limitations can be addressed to make photonic and phononic materials this way. To date, patchy colloids used in the assembly of non-close-packed crystals have been mainly restricted to micrometer-scale, spherical particles, or clusters of spheres. This limited range of applied shapes still constrains the variety of colloidal crystal lattices that can be formed. Although anisotropically shaped patchy particles were occasionally produced for shape-directed self-assembly, they did not assemble into large lattices with regular periodicity.[17] The relatively large periodicity of colloidal crystals is unsuitable for engineering photonic or phononic properties, as they require much smaller feature sizes. For example, thermal phononic waves have wavelengths on the nanometer scale. Furthermore, the patch geometries on the colloids are simply symmetric and thus far limited to just 2−4 patches per particle. To overcome these technical challenges and to advance both photonic and phononic evolutions, the time has come to widen our scope of what is considered to be a "colloidal particle" and to develop new approaches to crystallization.

WHY DNA ORIGAMI AS PATCHY COLLOIDS?

Scaffolded DNA origami, pioneered by Rothemund in 2006, presents a new method to synthesize arbitrarily shaped, 2D and 3D nanoscale particles, composed entirely of DNA duplexes.[18] By utilizing a user-programmed set of oligonucleotide "staple" strands, a long single-stranded DNA (ssDNA) can be folded exactly into any desired shape, resulting, in the ideal case, in a one-to-one transformation of each scaffold ssDNA into a well-defined nanoscale particle. Once folded, DNA origami remains stably dispersed in a salty, aqueous solution. Thus, DNA origami can be considered as a distinct class of colloids capable of self-assembly, including crystallization. Nevertheless, research on DNA origami and traditional colloids has largely been pursued separately over the last two decades, and DNA origami has not been widely regarded as a colloidal particle.

In this Mini-Review, we propose that DNA origami should be equally viewed as a patchy colloidal particle and seamlessly integrated with traditional colloidal systems. Figure 2 summarizes representative DNA origami motifs used in colloidal crystallization to date.[19−30] From these studies, we argue that DNA origami enables colloidal engineering that would otherwise be impossible with traditional colloid-synthesis methods and that its integration into colloid science can help to push the field in new and exciting directions.

DNA origami presents an attractive array of features for colloid synthesis. First, unlike the commodity colloids mentioned earlier, DNA origami can be used to design colloids with nearly arbitrary shapes with a resolution of a few nanometers (Figure 2).[18] Various computational tools with user-friendly interfaces, such as caDNAno, canDO, and oxDNA, enable rapid and deterministic designs of any desired DNA origami shape for both experts and nonexperts alike.[31]

Second, these intricately shaped particles can be synthesized through a one-pot, single-step thermal annealing process. By simply mixing the scaffold and staple ssDNAs and heating and cooling them in the same batch, nanomolar concentrations of DNA origami can be obtained, although purification is generally required to remove unincorporated ssDNAs.

Third, the surface of DNA origami can be precisely decorated with "sticky end" ssDNA sequences of your choice, which can act as handles for hierarchical assembly with neighboring DNA origami. This "molecular glue" can be positioned precisely at any location on the surface. Direct complementary binding between handle and antihandle ssDNA has been the gold standard for "sticky ends". Blunt-end interactions[31] and dynamic strand displacement[32] can be also effective. These ssDNA-enabled programmable interparticle interactions can be precisely controlled by adjusting the temperature and salinity. If the temperature increases and/or the salinity decreases, the binding affinity decreases, which may prevent the crystallization of DNA origami. On the other hand, if the temperature decreases and/or the salinity increases, the binding affinity strengthens, which may lead to nonspecific binding. The unprecedented control over shape and patch placement makes DNA origami a versatile colloidal platform, expanding the range of colloidal geometries and patch numbers.

Fourth, DNA origami enables colloidal crystallization across multiple scales. The size of individual 2D or 3D DNA origami objects spans from a few tens to hundreds of nanometers, making it adaptable for engineering photonic crystals working at the ultraviolet (UV) and visible regimes and thermal phononic crystals with customizable periodicities.

Lastly, DNA can be conjugated to a range of functional nanomaterials such as quantum dots (QDs), molecular quantum emitters, and metallic nanoparticles (NPs).[33] In addition, the surface of DNA origami can also be coated with inorganic materials to enhance structural reliability and provide additional functionalities.[31] Therefore, DNA origami colloidal crystals can be further functionalized with optically active materials, for instance, enabling the seamless integration of quantum emitters into photonic crystal cavities.

HISTORICAL BENCHMARKS OF DNA ORIGAMI COLLOIDAL CRYSTALS

When Ned Seeman published his visionary paper on nucleic acid junctions and lattices,[34] he could not have foreseen the development of the field of DNA nanotechnology with its multiple branches and its hundreds of laboratories around the world. Fulfilling his original proposal from 1982 to make "three-dimensional networks of nucleic acids which are periodic in connectivity and perhaps in space", in 2009, his team published single crystals assembled purely from DNA that exhibit edge lengths of ∼200 μm and that refract with 4 Å resolution.[35] Along this journey, multiple crystalline assemblies from DNA oligonucleotides have been generated, leading up to "DNA brick assemblies" that enabled the addressable assembly of a large 3D molecular canvas.[36] These 2D/3D DNA crystals inherently retain extremely small periodicities (on the scale of individual oligonucleotides, i.e., the order of a few nanometers), limiting their applicability in photonics and phononics, as well as their ability to accommodate larger nanomaterials such as QDs and metallic NPs.

The hierarchical assembly of DNA origami into larger-scale structures has been reported since the early stages of DNA origami development. Both 2D and 3D DNA origami were decorated at their edges with ssDNA handles or blunt-ended duplexes and programmed to bind with neighboring structures. Aside from the formation of colloidal crystals, rigid 3D DNA origami have also been used to form self-limiting finite assemblies,[20] but these types of assemblies are outside of the scope of this mini-review and lack the kind of long-range order required for many photonic and phononic applications.

Over the past decade, two-dimensional crystals have been assembled from DNA origami, including origami crystals decorated with metal nanoparticles. The earliest demonstrations used surface diffusion-assisted assembly to create long-range

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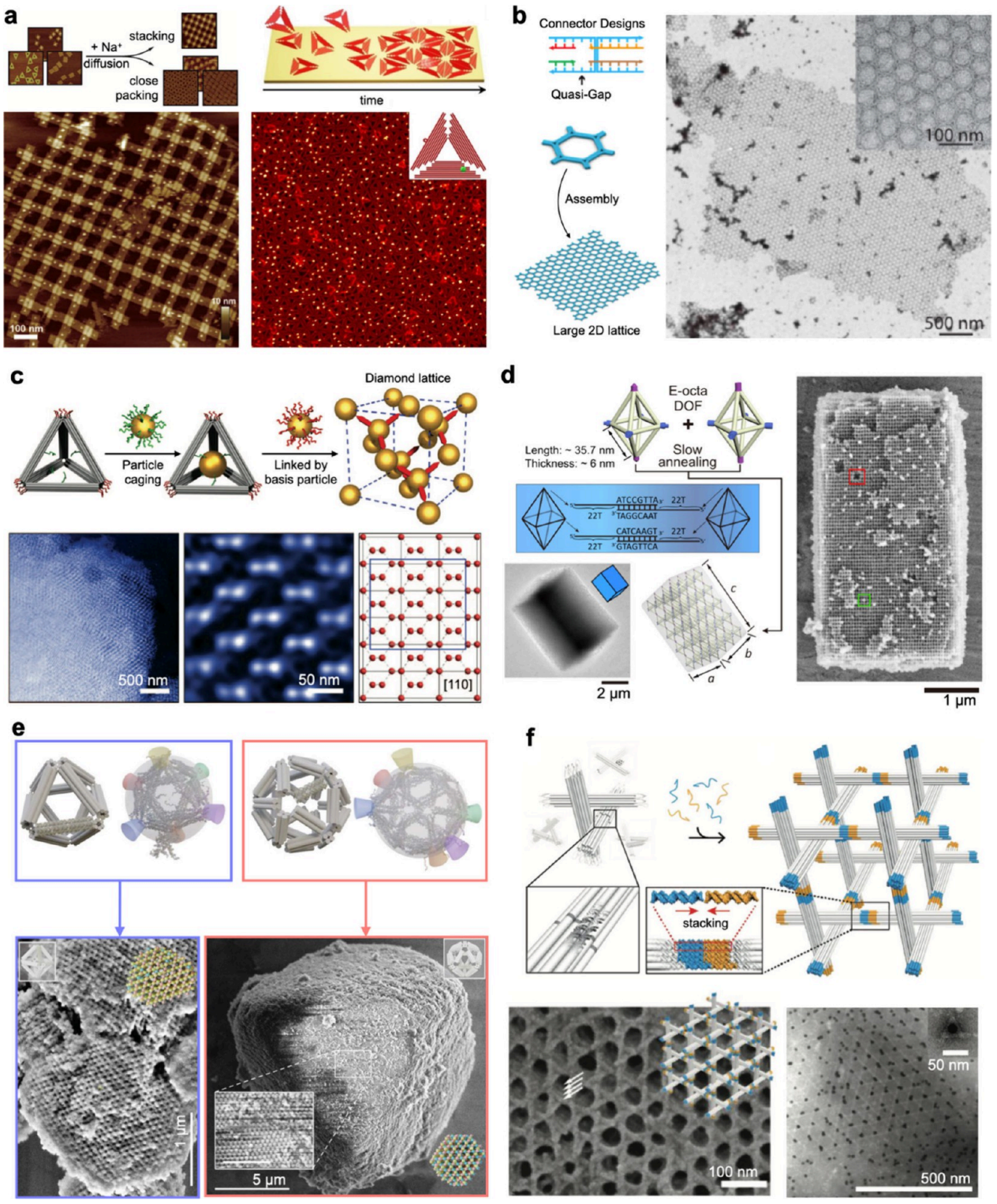

Figure 3. Hierarchical assembly of DNA origami into long-range lattices. (a) Representative atomic force microscopy (AFM) images showing surface diffusion-assisted ordering of DNA origami tiles. (Left panel) 2D lattice of cross-shaped DNA origami assembled through DNA base-stacking on the edge of DNA origami.[29] (Right panel) Hierarchically assembled triangle DNA origami through surface diffusion, which facilitates entropy-driven packing.[37] Adapted from refs 29 and 37. Copyright 2014 Wiley-VCH GmbH. Available under a CC-BY 4.0. Copyright 2021 Wiley-VCH GmbH. (b) 2D Honeycomb lattice assembled from hexagon DNA origami tile (HT). (Left panel) Schematics illustrating the assembly of HT via DNA sticky-end on the 6-fold patches. (Right panel) Representative transmission electron microscopy (TEM) images of the assembled honeycomb lattice.[30] Adapted from ref 30. Copyright 2016 American Chemical Society. (c) Cubic diamond superlattice assembled through the cocrystallization of gold nanoparticles (Au NPs) and tetravalent DNA origami cage. (Top panel) Schematics illustrating the coassembly of Au NPs and DNA origami cage. (Bottom panel) Cryo-scanning TEM (cryo-STEM) images and schematic projection of the assembled crystal along the [110] zone axis.[21] Adapted from ref 21. Copyright 2016 American Association for the Advancement of Science. (d) Wulff-shaped single crystals of elongated octahedral DNA origami frames. (Left panel) Schematics illustrating the single-crystal growth of pure 3D DNA origami and the corresponding TEM image of a bare microcrystal.

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Figure 3. continued

(Right panel) A SEM image of the silica-encapsulated microcrystal. Crystal vacancy and adatom are highlighted with red and green boxes, respectively.[38] Adapted from ref 38. Available under a CC-BY 4.0. Copyright 2021 Springer Nature. (e) Pyrochlore lattice of DNA origami realized through trap-free self-assembly pathways designed by multiscale modeling and algorithm optimization. (Top panels) Schematics of octahedral and icosahedral DNA origami used for assembly of pyrochlore lattice. (Bottom panel) Representative SEM images of the assembled pyrochlore lattice.[22] Adapted from ref 22. Copyright 2024 American Association for the Advancement of Science. (f) Rhombohedral lattice assembled from DNA origami tensegrity triangle structures. (Top panel) A schematic illustration of tensegrity triangle building blocks and their assembly through blunt-ended DNA stacking interactions. (Bottom left panel) A magnified SEM image of the assembled crystals revealing multilayered lattices. (Bottom right panel) A representative TEM image of the origami lattice with hosted Au NPs.[39] Adapted from ref 39. Copyright 2018 Wiley-VCH GmbH.

packing of 2D DNA origami, as shown in Figure 3a.[29,37] The DNA origami in these examples was limited to 2D single sheets, typically triangles and squares. Importantly, the key enabling features of DNA origami as patchy colloids, such as molecular programmability and patch addressability, were not fully utilized in these early studies, limiting the range of available crystal lattices, periodicities, and densities.

More recently, new strategies for making rigid DNA origami subunits further opened the door to the robust assembly of large-area two-dimensional crystals. In particular, Ke et al. developed a two-layer or four-layer 3D DNA origami hexagon frame—referred to as a hexagon tile (HT) (Figure 3b).[30] The design of the ∼106 nm (end-to-end) HT has a 6-fold symmetry with six patches for connecting to neighboring tiles. Through systematic experimental testing and simulation, this work revealed the importance of subtle differences in the HTs' structural rigidity and patch connecting strength in achieving low-defect, long-range crystallization. Moreover, the HT design featured a relatively large cavity with a diameter of ∼60 nm, which enabled the incorporation of sizable nanomaterials, such as ∼30 nm Au NPs. These Au NPs were arrayed over a large area within the 2D HT crystals, functioning as electric metamaterials. However, these HT crystal structures were limited to 2D, because the HTs are designed to be flat tiles with patches located in a 2D plane.

Around the same time, 3D crystals were also assembled from DNA origami. In 2016, Gang et al. first reported the successful assembly of DNA origami and its subsequent 3D long-range crystallization using valence-limited, specific interactions.[21] In particular, they first used a tetrahedron DNA origami cage, consisting of a 10 helix bundle (HB) approximately 36 nm in length for each edge, as the main motif to encapsulate ∼14.5 nm Au NPs via coassembly (see Figure 3c). Then, DNA-functionalized Au NP were used to bind to each vertex of Au NP containing DNA origami cage through complementary DNA hybridization, enabling the cocrystallization of DNA origami cages and Au NPs into a diamond lattice with a relatively low volume fraction. This represented the first materialization of a colloidal diamond structure.

Following this breakthrough, Gang and co-workers expanded the library of DNA origami cages to include cubes, octahedra, elongated square bipyramids, prisms, and triangular bipyramids. This diversification allowed the formation of previously inaccessible 3D crystal lattices, varying in terms of symmetry and space groups.[19] They also demonstrated that single crystals could be assembled from DNA origami frames alone—without the assistance of Au NP—as evidenced by the formation of single crystals with distinct crystal habits (Figure 3d).[38]

The crystallization of DNA origami cages has since been further advanced by the development of new inverse design strategies.[22] For example, Sulc et al. used satisfiability problem (SAT) assembly (i.e., a constrained optimization-based design algorithm for patch-particle interactions) to inversely design the assembly of patchy particles into pyrochlore lattices, which have the potential to exhibit a large and omnidirectional photonic bandgap. They demonstrated that precise control over the connectivity of tetravalent patches on each particle enabled the self-assembly of cubic diamond crystals, specifically in the form of pyrochlore lattices. For this purpose, octahedral and icosahedral DNA origami cages, ∼50 nm in size, were chosen as an experimental model system (Figure 3e). These cages were successfully crystallized into the designed pyrochloric lattice, forming long-range, well-ordered structures. However, verifying the photonic bandgap of these DNA origami diamond crystals has yet to be exploited.

3D crystallization of DNA origami is not limited to the periodic assembly of polyhedral cages. In 2018, Liedl et al. made another breakthrough in 3D DNA origami crystallization.[39] Inspired by the work on the DNA tensegrity triangle-based crystals of Ned Seeman,[35] they used a DNA origami tensegrity triangle structure for their crystal design. In tensegrity, or tensional integrity, structural motifs are formed that are balancing forces between tension and compression as exemplified in DNA origami multihelical bundles bearing compressive forces that are fixed in space by ssDNA bearing tension. The DNA origami triangles for crystal growth were composed of three struts of 14 HB, with a diameter of ∼12.5 nm (see Figure 3f). The intrinsic triangulation of the tensegrity triangles endowed with enough rigidity to robustly and programmably connect the ends of each strut, enabling the crystallization of the tensegrity triangles into rhombohedral lattices via blunt end stacking interactions. Each 14 HB strut was 67 nm in length, double the length of Gang's cage structure, allowing the crystals of tensegrity triangles to form larger cavities compared to those of the DNA origami cages. However, despite this advancement, the nanomaterials that were hosted within these DNA origami crystals were still relatively small but enabled the placement of 20 nm Au NPs. It was also the first DNA origami lattice design comprising controlled orientation of the building blocks with respect to each other via the staggered interconnection of the 14-helix bundles.

TOWARD LARGER LATTICE PERIODS OF DNA ORIGAMI CRYSTALS

The success over the past decade corroborates a key principle: for DNA origami to crystallize into long-range structures, it must exhibit sufficient structural rigidity.[40] The importance of rigidity is one reason why smaller 3D DNA origami sheets, rather than 2D sheets, have predominantly been used for colloidal crystallization. In solution, origami designs composed of single layers of parallelly connected duplexes (similar to the original designs by Rothemund in 2006)[18] are typically too floppy to sustain long-range order. In contrast, the use of multihelical bundles has proven effective in reinforcing the rigidity of DNA origami structures, whether in the form of frames or struts.

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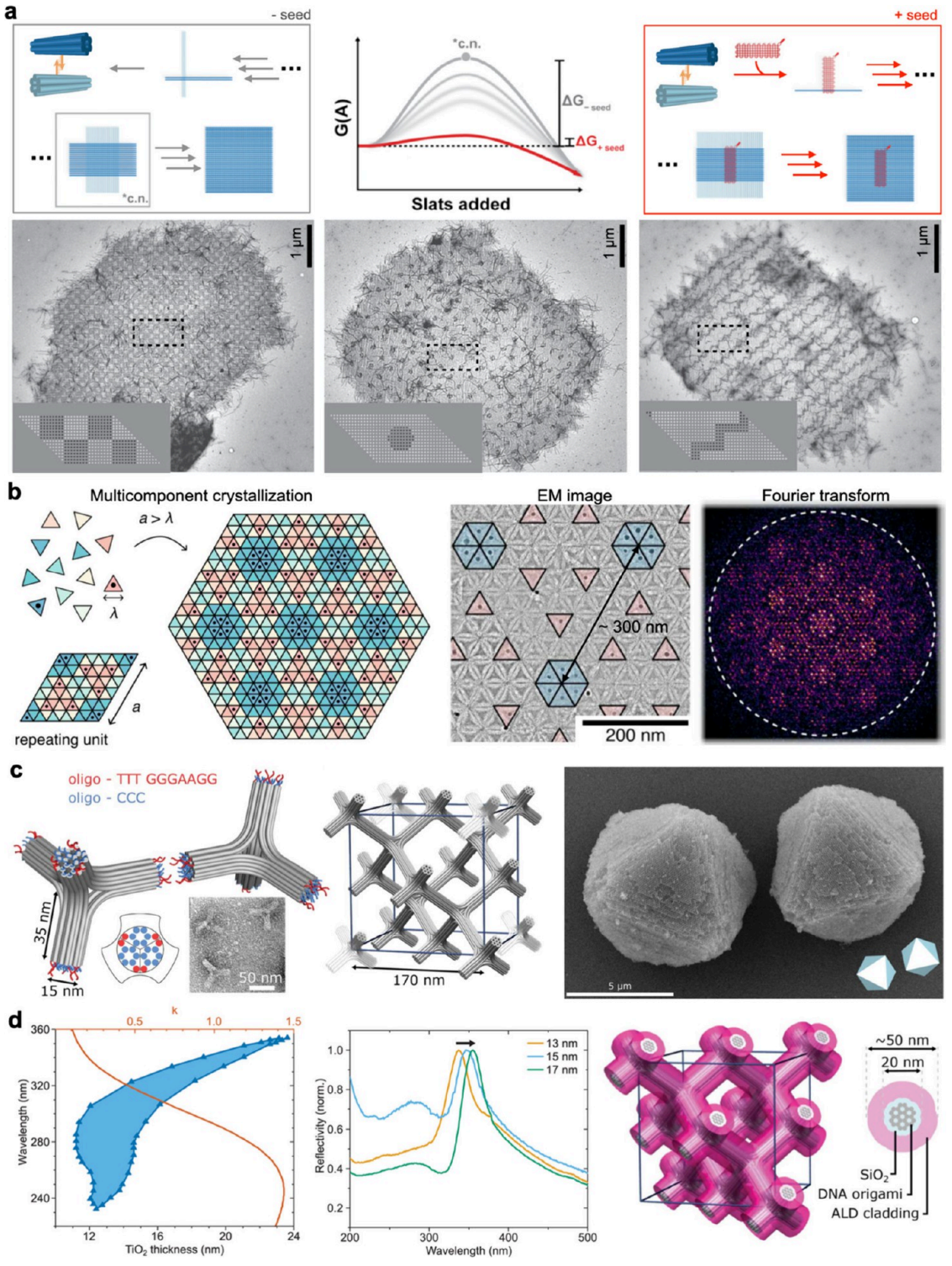

Figure 4. Advances in DNA origami crystals toward large lattice periodicity. (a) Crisscross polymerization of DNA origami slats into long-range crystals.[24] (Top panel) Schematics depicting the qualitative energy landscape in crisscross assembly with and without the addition of a seed that strongly organizes the slats during the initial stage of assembly. *c.n. indicates critical nucleus. (Bottom panel) Representative TEM images of the periodic crisscross megastructures with prescribed integration of guest materials (i.e., DNA nanocubes). Adapted from ref 24. Copyright 2023 Springer Nature. (b) Assembly of 12 distinct DNA origami triangle species into a 2D superlattice with large periodicities.[43] User-defined multicomponent unit cells, distinct from the underlying triangular lattice, can be achieved by utilizing the allowed 2D symmetries, as depicted in the schematic illustration (left panel), with corresponding EM (middle panel) and Fourier transform images (right panel). Adapted from ref 43. Copyright 2024 American Chemical Society. (c) Cubic diamond lattice photonic crystals assembled by DNA origami tetrapod. (Left panel) A schematic illustration of two tetrapods assembling in a staggered configuration and the designed unit cell of the cubic diamond lattice. (Right panel) A representative SEM image of the assembled diamond lattice.[23] Adapted from ref 23. Copyright 2024 American Association for the Advancement of Science. (d) Numerically calculated (left panel) and measured (middle panel) photonic band properties of the DNA origami photonic crystals with SiO_2 and TiO_2 coating (right panel).[23] Adapted from ref 23. Copyright 2024 American Association for the Advancement of Science.

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However, HBs are generally formed by corrugating and hierarchically stacking 2D DNA origami sheets into hexagonal or square lattices. This stacking inherently limits the accessible size of the DNA origami building blocks, which usually remains smaller than 100 nm. Note that the synthesis of DNA origami relies on folding a long ssDNA scaffold (~8,500 bases) into NPs in a one-to-one fashion, which further restricts the scale of the final product. Consequently, despite the promising prospects, the relatively small periodicities of DNA origami colloidal crystals, generally less than 100 nm, have limited their potential applications especially in photonics.

Despite their small size, DNA origami crystals still hold significant potential for phononic applications, including thermal phononic crystals and mechanical metamaterials.[41,42] However, for DNA origami to be effectively used in photonic crystals, the crystal periodicities need to be extended to match the wavelengths of visible or UV light, typically ranging from 150 to 300 nm. Also, plasmonic applications, such as metamolecules, metasurfaces, and metamaterials, require noble-metal NPs larger than 40−50 nm in size. This is because NPs smaller than 30 nm tend to exhibit considerable absorption with absorption cross sections exceeding that of resonant scattering. As a result, DNA origami colloidal crystals have been underutilized toolsets for photonic applications, particularly compared with their commodity colloidal counterparts, which have more established utility in these areas. Despite these limitations, the field continues to evolve, and DNA origami presents untapped potential for more advanced photonic applications if the scale and properties of its assemblies can be further optimized.

In just the last two years, DNA origami crystallization has made significant progress toward achieving larger periodicities. One breakthrough was the hierarchical assembly of size-limited 3D DNA origami structures into larger, yet rigid, building blocks, which were then grown into long-range crystals. In 2023, Shih et al. experimentally generalized the concept of crisscross molecular polymerization from ssDNA to DNA origami slats (6 HB or 12 HB, each ~450 and 225 nm in length, by engineering sufficiently weak binding handles) (Figure 4a).[24] The weak nature of the individual binding handles allowed for the enforcement of a strict seed dependence. In the case of 6 HB slat monomers that can each make up to 32 bonds, the formation of a "critical nucleus" where each monomer is stably bound required paying for the entropic cost of colocalizing up to 32 monomers, with only small enthalpic gains along the assembly pathway, rendering spurious nucleation highly improbable. However, the addition of a seed that preorganizes the first set of bonds yielded rapid assembly of the desired structures by bypassing the kinetic barrier, as each monomer could immediately make up to 16 bonds. Such robustness to spurious nucleation enabled crisscross assembly with yields significantly higher than those attained with other hierarchical tile assembly approaches.

DNA origami slats were used to generate micrometer-scale finite addressable assemblies from over 1,000 unique slats reaching lateral dimensions over 2 μm, as well as periodic assemblies of over 10,000 origami components with lateral dimensions over 10 μm. These structures are addressable down to 14 nm pixel spacing with the attachment of guest molecules through complementary DNA handles (e.g., DNA nanocubes decorated on the assembled slats in Figure 4a). Crucially, the multicomponent nature of the 2D origami-slat crystals provides an opportunity to make 2D crystals of metallic NPs with lattice constants much larger than those of a single DNA origami particle by functionalizing only a subset of the slats. Indeed, multiple lattices with different periodicities and crystal symmetries can be made by labeling different subsets. However, the practical applications of crisscross in photonics and phononics have yet to be established, and modifications such as the use of multiple slat layers and attachment points per guest molecule might be necessary for additional levels of rigidification.

Following a similar conceptual path, Hayakawa, Videbaek et al. developed a simple inverse-design approach to assembling 2D crystals of DNA origami with user-specified multicomponent unit cells (Figure 4b).[43] Learning from the lessons introduced above, namely, the importance of the subunit rigidity, they used rigid triangular subunits that self-assemble via sticky-end cohesion. Then, by exploiting the allowed 2D symmetries, they designed large intersubunit interaction matrices that programmed the assembly of 2D crystals with periodicities much larger than the individual subunits themselves. In particular, they demonstrated the assembly of large-area single crystals from each of the four available Wallpaper groups, including metal-NP-labeled crystals with lattice constants of up to 300 nm (i.e., comparable to the wavelength of visible light). Importantly, they also identified a series of rules for choosing economical designs that enable the assembly of crystals with the largest possible lattice parameters by using the minimal number of DNA origami subunit species. Critical next steps will be to extend these and related ideas to 3D crystals as well as crystals assembled from a larger diversity of subunit shapes.

Another advance has come from efforts to increase the size of the subunits and target low volume-fraction structures. In 2020, Lee et al. numerically suggested a hierarchical assembly of DNA origami tetrapod-based structures, forming a direct rod-connected diamond lattice capable of exhibiting a complete photonic bandgap in the visible regime.[44] In 2024, Liedl et al. achieved a significant experimental breakthrough by developing a diamond cubic lattice assembled from DNA origami tetrapods with arms measuring 35 nm in length and a width of 15 nm (Figure 4c).[23] The four arms of the tetrapod were designed with programmable patches at their ends capable of retaining a torsional potential. These patches provided orientation-dependent bonding potentials between neighboring DNA origami monomers, favoring a 60° rotation between adjacent DNA origami tetrapods, that is needed to achieve robust assembly of diamond cubic lattices.

The assembly of these DNA origami tetrapods led to reliable formation of a diamond cubic lattice with a periodicity of ~170 nm. This achievement represents the largest periodicity reported for DNA origami-based colloidal diamonds, pushing the boundaries of structural control and scalability. Most importantly, this advancement enabled the first experimental demonstration of a photonic bandgap in a diamond photonic crystal at near-UV frequencies (Figure 4d), a milestone that had remained elusive due to the difficulty of forming a diamond lattice and the smaller periodicities of previous DNA origami crystals. A crucial step in achieving the photonic bandgap comprised achieving a large mismatch of the refractive indices between the crystal's material and its surrounding medium. The typically aqueous buffers where DNA-assembled crystals are grown have a refractive index of 1.3−1.4 in the visible and the near-UV. DNA, on the other hand, has an apparent refractive index around 1.6 in this spectral range, resulting in a relatively small mismatch. Taking the crystals into the air or a vacuum already decreases the refractive index of the surrounding

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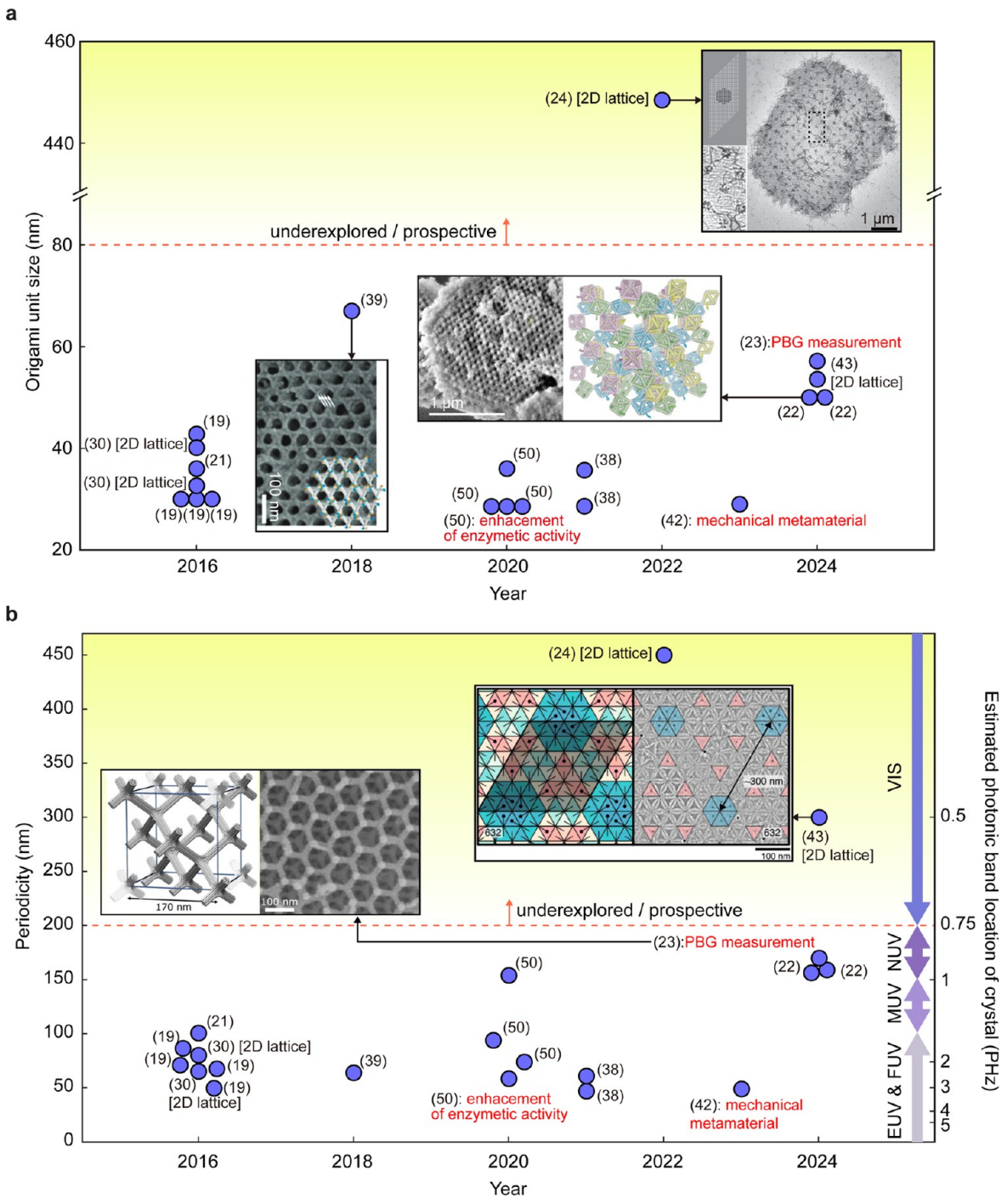

Figure 5. Historical benchmarks in colloidal crystallization of 3D DNA origami motifs.[19,21−24,30,38,39,42,43,50] (a, b) Advancement in the DNA origami unit size (a) and the periodicity of their assemblies (b) over time. The estimated photonic band location of the DNA origami crystals in (b) was determined based on the assumption that a photonic band appears when the relative phase difference between light waves scattered from one structure and its neighboring periodic structure is an integer multiple of 2π. The numbers in parentheses correspond to the reference numbers of the DNA origami motifs used. Adapted from refs 22−24, 39, and 43. Copyright 2024 American Association for the Advancement of Science. Copyright 2024 American Association for the Advancement of Science, Copyright 2023 Springer Nature. Copyright 2018 Wiley-VCH GmbH. Copyright 2024 American Chemical Society.

medium to 1.0. This can be achieved by silica coating of the DNA structures rendering the crystals mechanically and chemically stable.[31] At the same time, silica has a refractive index around 1.5 in the visible, leading to a mismatch of only ~0.5, still too small to open a photonic bandgap in the diamond lattice. So far, it was possible to achieve the required mismatch only via atomic layer deposition (ALD)-deposition of a material like TiO_2 (refractive index between 2.5 and 3 in the visible and near UV).[23]

FUTURE DIRECTIONS

By harvesting the precise programmability of DNA origami, researchers have been able to control not only the crystallization kinetics but also the specific interactions between DNA origami

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units, thus enabling the hierarchical assembly of complex 3D lattices with unprecedented periodicities. This breakthrough in DNA origami-driven colloidal crystallization is particularly significant for the development of advanced photonic/phononic devices, metamaterials, and other applications requiring precise control of material properties at the mesoscale and nanoscale.

Figure 5 provides a comprehensive summary of the historical benchmarks in DNA origami colloidal crystallization categorized by the size of crystallized DNA origami units, periodicities, and their applications. Over the past decade, the achievable size of crystallized DNA origami units has expanded significantly—from a few tens of nanometers to well over hundreds of nanometers. This dramatic increase in the structural complexity and periodicity of DNA origami crystals highlights a pivotal shift, allowing for the exploration of a new horizon of material science. Most notably, diamond photonic crystals, once considered a "pipedream" during the 1990s and 2010s, have now become a reality through DNA origami colloidal crystallization, offering the first experimental evidence of complete photonic bandgaps at UV−visible frequencies. This achievement highlights the unique ability of DNA origami to enable self-assembly of 3D crystals that were previously unattainable using conventional colloidal systems.

In addition to advances in photonics, DNA origami colloidal crystals have been shown to host various nanomaterials, including metallic NPs and QDs, which can further enhance the functionality of these crystals by introducing optical, electronic, mechanical, and catalytic properties. This versatility, coupled with the ongoing progress in DNA origami colloidal crystallization techniques, makes these materials promising candidates for next-generation photonic, phononic, and plasmonic devices. As the field continues to evolve, future research will likely focus on further expanding the size and periodicity of DNA origami crystals, exploring new nanomaterials for hosting and pushing the boundaries of what can be achieved in both optical and mechanical metamaterials.

Despite the impressive success in DNA origami colloidal crystallization, several limitations need to be addressed to enhance their utility for a diverse range of applications.

1. Defects and random orientation in self-assembly: The self-assembly process inherently leads to defects and random orientations within the lattice, which can compromise the photonic bandgap and its application in devices such as waveguides and color pigments. This challenge limits the size of the assembled crystal and hinders an in-depth analysis of its photonic bandgap structure. While simple reflection or transmission measurements (e.g., conventional UV/vis/NIR spectrometer measurements) on small (few μm of lateral dimension) colloidal crystal ensembles can provide qualitative information about the presence of a photonic bandgap,[23] a quantitative analysis of the full photonic band structure requires angle-resolved transmission measurements along all high-symmetry directions in the Brillouin zone defined by the wave vector.[45] Such angle-resolved transmission measurements necessitate relatively large colloidal crystals with high lattice fidelity and lateral dimensions exceeding hundreds of micrometers, which have not yet been achieved through DNA origami-based colloidal crystallization. This challenge could potentially be addressed through site-specific growth of DNA origami colloidal crystals on prepatterned surfaces. Inspired by epitaxial growth techniques, the deterministic placement of seed DNA origami over a large area could facilitate controlled growth, thereby effectively addressing defect issues. Similar approaches have been successfully used with Au NPs, demonstrating their potential applicability to DNA origami systems.[46]
2. Limited flexibility in lattice control: Currently, the flexibility in controlling lattice geometry, density, and lattice constants is quite low, directly restricting the width of photonic and phononic bandgaps. The largest periodicity reported for DNA origami colloidal crystals with 3D lattices is still relatively small (~170 nm, achieved with 50 nm sized, tetrapod DNA origami), with the photonic bandgap primarily working in the near UV region;[23] the visible spectrum remains largely unexplored. To achieve a photonic bandgap in the full visible regime, DNA origami building blocks to be crystallized (e.g., tetrapods) need to be roughly doubled in size (e.g., from 50 to 100 nm) to achieve periodicities around 300 nm. Furthermore, the existing libraries of successfully crystallized DNA origami motifs are relatively narrow, which restricts the possible range of lattice geometries and the resultant band structures. A shift from empirical trial-and-error approaches to more deterministic strategies is needed, utilizing recent advancements in inverse design methods and all-atom simulation tools. Additionally, upon assembling larger periodic structures, the incorporation of Au NPs larger than 40−50 nm will be essential for significant scattering, along with the types of nanomaterials hosted. For instance, the integration of silicon (Si),[47] or other high-index NPs can introduce unnaturally extreme electromagnetic resonances, such as optical magnetism. Recently, Acuna et al. have developed a method for decorating Si NPs with ssDNA and their subsequent assembly on DNA origami, providing a promising platform for future applications.[47]

   It is also noteworthy that combining commodity colloids with DNA origami offers another promising approach for creating complex and large-scale lattices. This synergy, initially proposed by Rogers, Shih, and Manoharan,[48] was demonstrated experimentally by Chaikin et al., showing colloidal clustering with chiral geometries.[49] DNA origami assemblies, referred to as DNA origami belt[49] or stick frame,[48] could fully wrap around the 200−300 nm sized commodity colloids. The directional bonding between these DNA origami-patched colloids could offer a versatile assembly of a large-periodicity diamond lattice, which is needed for a complete photonic bandgap in the visible range.
3. Low refractive index of DNA origami: The refractive index of DNA origami is relatively low, which may limit their effectiveness in certain photonic applications. Enhancing the refractive index through material modifications or composite strategies could be beneficial. Indeed, the silicification of DNA origami crystals and the subsequent deposition of TiO_2 by ALD were reported for enhancing the refractive index of DNA origami crystals.[23] Exploring the incorporation of larger refractive index materials within the DNA origami colloidal crystals (e.g., Si, germanium, and gallium arsenide) or utilizing alternative designs may offer pathways to diversify the applications of DNA origami crystals. For example, the patching of 200−300 nm sized, high-index Si[47] or other

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high-index colloids with a DNA origami belt or stick frame could enable the direct self-assembly of a diamond lattice with a complete photonic band gap in the visible regime—eliminating the need for postcoating of high index materials (e.g., ALD of TiO_2) or structural inversion (e.g., inverse pyrochlore lattice).

By addressing these challenges, researchers can unlock the full potential of DNA origami colloidal crystals, enhancing their functionality and broadening their applicability in advanced photonic and phononic devices and metamaterials/metasurfaces. Continued interdisciplinary collaboration and innovation will be critical for overcoming these challenges and realizing the next generation of DNA origami colloidal crystals.

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Notes

The authors declare no competing financial interest.

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