Soft Matter

Cite this: *Soft Matter*, 2011, **7**, 3240

www.rsc.org/softmatter

HIGHLIGHT

Self-assembling DNA templates for programmed artificial biomineralization

Enrique C. Samano,*[ae] Mauricio Pilo-Pais,[b] Sarah Goldberg,[b] Briana N. Vogen,[a] Gleb Finkelstein[b] and Thomas H. LaBean*[acd]

DOI: 10.1039/c0sm01318h

Complex materials with micron-scale dimensions and nanometre-scale feature resolution created *via* engineered DNA self-assembly represent an important new class of soft matter. These assemblies are increasingly being exploited as templates for the programmed assembly of functional inorganic materials that have not conventionally lent themselves to organization by molecular recognition processes. The current challenge is to apply these bioinspired DNA templates toward the fabrication of composite materials for use in electronics, photonics, and other fields of technology. This highlight focuses on methods we consider most useful for integration of DNA templated structures into functional composite nanomaterials, particularly, organization of preformed nanoparticles and metallization procedures.

[a]*Department of Chemistry, Duke University, DurhamNC, USA. E-mail: thomas.labean@duke.edu*
[b]*Department of Physics, Duke University, DurhamNC, USA*
[c]*Department of Computer Science, Duke University, DurhamNC, USA*
[d]*Department of Biomedical Engineering, Duke University, DurhamNC, USA*
[e]*CNyN-UNAM, EnsenadaBC, México. E-mail: samano@cnyn.unam.mx*

1. Programmed assembly of DNA nanostructures

Our current ability to create complex DNA nanostructures *via* designed self-assembly owes much to the confluence of early efforts to build periodic matter for crystallography[1] with the concept of DNA-based computing.[2] Development of concepts and materials for computation by molecular self-assembly[3,4] has led to widespread success of DNA-based nanotechnology and the great diversity of DNA building blocks, architectures, and geometries described in the past decade [ref. 5–10 and other literature cited therein]. Fig. 1 gives a very brief survey of the variety of construction strategies and superstructures produced by programmed DNA assembly. It should be noted that other nucleic acids have also been applied as molecular construction materials,[11,12] but to a lesser extent than DNA. Although, other materials,

Enrique C. Samano

Enrique C. Samano earned his PhD degree in Physics from Stevens Institute of Technology in Hoboken, NJ, USA, in 1992. He is a Research Professor at Centro de Nanociencias y Nanotecnología-UNAM in Ensenada, BC, Mexico, working on thin film growth and surface science techniques. He is currently a Visiting Scholar at Duke University in the DNA Nanotechnology Group, and involved on the fabrication of nanostructures based on DNA with applications in electronics and plasmonics.

Mauricio Pilo-Pais

Mauricio Pilo-Pais received a dual BS in Physics and Chemistry from the University of Florida in 2008. He is currently a second year graduate student in Physics at Duke University where he does interdisciplinary research in Physics and Biochemistry, with focus on DNA nanotechnology. His main interest is in studying the self-assembly properties of DNA for possible uses in nanocircuits and plasmonic applications.

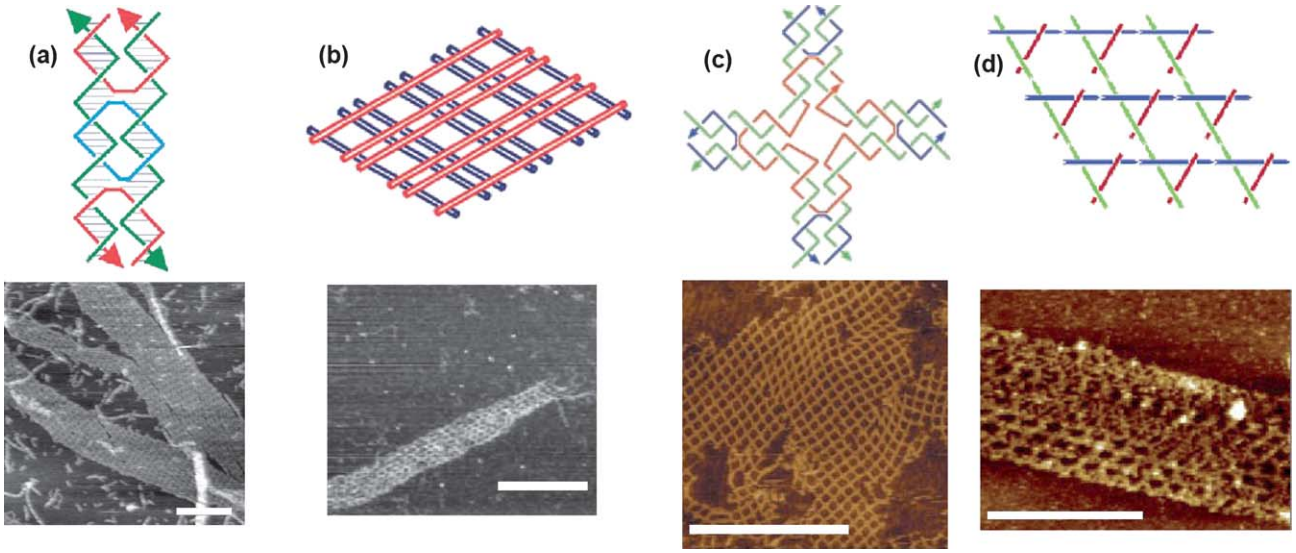

Fig. 1 Schematic drawings and AFM images of DNA nanostructure building blocks and lattices. (a) Double-crossovers forming 2D-DNA crystals,[23] (b) DNA Holliday junction arrays using rhomboid tiles,[24] (c) DNA grid assembled from 4 × 4 cross-tiles,[7] and (d) DNA triangle tiles with flexible junctions.[9] All scale bars are 300 nm.

Sarah Goldberg

Sarah Goldberg obtained her PhD in Condensed Matter Physics from the Weizmann Institute of Science in 2009. Her work focused on the long-range order of the vortex lattice in type II superconductors. Currently, she is a postdoctoral researcher at Duke University, developing nanoscale elements for electronic and plasmonic applications.

Gleb Finkelstein

Gleb Finkelstein obtained his PhD in Physics from the Weizmann Institute of Science in 1999. Following postdoctoral work at MIT, he moved to Duke University, where he is now an Associate Professor. His group works on a range of subjects in condensed matter physics and nanoscience.

Briana N. Vogen

Briana Vogen received her PhD in Chemistry from Duke University in 2010. She is currently a postdoctoral researcher at Duke in the laboratory of Prof. LaBean. Her research interests include designing and patterning surfaces for potential applications in DNA nanotechnology.

Thomas H. LaBean

Thomas H. LaBean earned his PhD at the University of Pennsylvania in 1993. He studied the folding properties of arbitrary-sequence proteins expressed by randomized, synthetic DNA libraries. He then moved to Duke University and studied de novo *protein design, and then worked on DNA-based molecular computation. He now studies self-assembling biomolecular nanostructures as a Research Professor at Duke University.*

including polypeptides, offer alternative molecular assembly systems,[13,14] our understanding of the rules of nucleic acids assembly (dominated by Watson–Crick base pairing) far outstrips our current ability to design self-assembling polypeptide systems from scratch. High-resolution structural DNA nanotechnology employs both building blocks and assembly tags made of DNA, so DNA is both "bricks and mortar" in these nanoscale assemblies;[15] by contrast, some lower-resolution nano-assemblies use DNA only as "mortar" in the directed-assembly of other nanomaterials.

This highlight will not discuss "DNA as mortar" developments[16,17] nor active, responsive DNA connectors,[18] but will focus on more complex, "smart" assemblies and their use as templates upon which other "dumb" materials (*i.e.* materials typically not associated with molecular recognition) can be organized. One family of "smart" DNA assemblies used as templates is the DNA origami structures. These are typically 2D-nanostructures based on the 7249-base, single-strand M13 bacteriophage genome that is folded in a programmed and precise manner by means of about two hundred specific, complementary "staple" strands of synthetic DNA.[19] As an extension to these 2D templates, 3D DNA nanostructures have tremendous potential to organize materials in 3D, encapsulate and release drugs, regulate the activity of encapsulated proteins, and selectively cage nanomaterials.[20–22]

The desire to fabricate functional electronic and photonic devices at the nanoscale has led to the development of two main strategies for using DNA self-assembly in the creation of metal nanostructures: metallization of DNA by chemical deposition (Section 2) and placement of preformed particles on DNA templates (Section 3). For instance, origami scaffolds contain known sequences at known locations that can be used to position DNA-binding inorganic molecules just a few nanometres apart.

2. Metallization of DNA templates

The literature reports several procedures for modifying and converting DNA templates into functional electronic nanostructures; *i.e.*, nanowires, transistors, *etc.* Nucleation and growth of metal clusters on viral DNA, by the localization of an aqueous reducing agent, is the main approach to metallization of DNA templates. Metallic nanowires templated on viral DNA frequently display a lack of crystallinity and uniformity and instead show coarse grained morphologies. DNA origami structures are quite robust and stiff and are currently being developed as scaffolds to improve the fabrication of nanowires and other optically and electronically active nanostructures.

Braun and co-workers first constructed and tested a conductive nanowire from silver nanoparticles nucleated upon viral DNA *via* an electroless deposition method.[25] They later improved this method by employing a sequence-specific "molecular lithography" approach to selectively metallize a dsDNA template.[26] They used an aldehyde-based reducing agent on λ-DNA molecules to grow silver clusters. The silver clusters were utilized as seed particles to nucleate more silver or gold around them and the electrical properties of the resulting wires were measured. They employed the same technique to fabricate a DNA-templated carbon nanotube field-effect transistor.[27] They also demonstrated the ability to provide aldehyde derivatization of DNA in a sequence-specific manner as a means of more accurately localizing metal deposition.[28] They envisioned a multi-step self-assembly process for the preparation of nanoelectronic components and circuits using DNA templates,[29] and summarized their experience on the

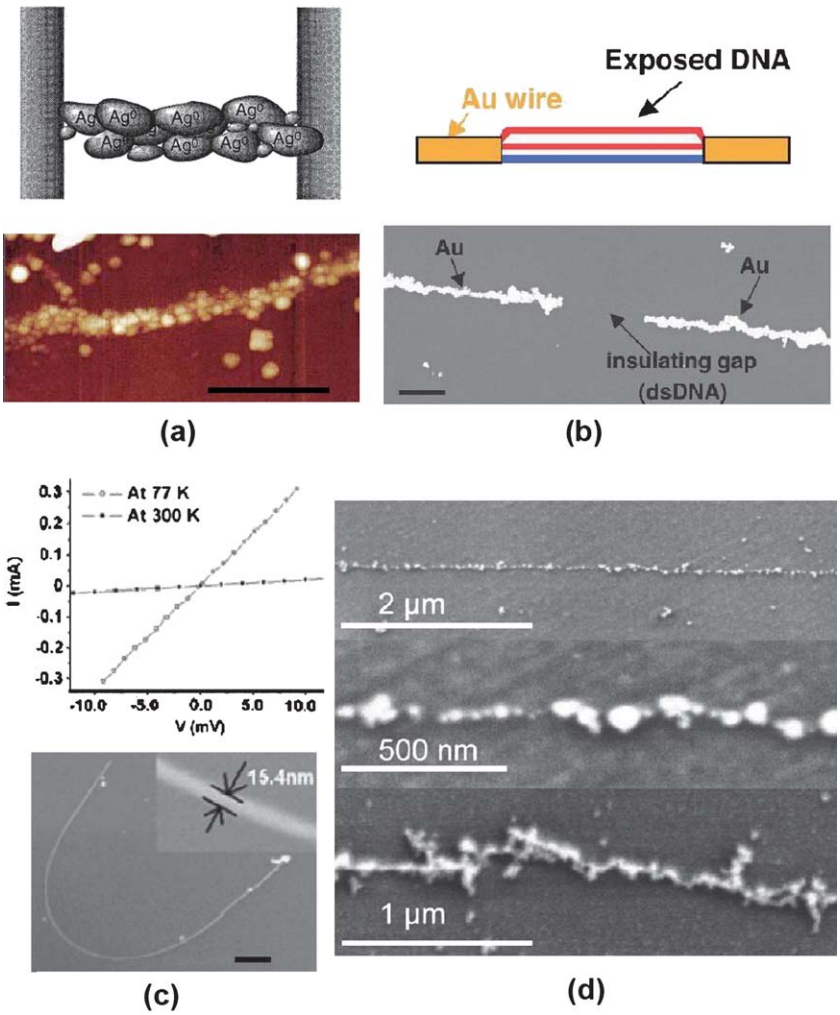

Fig. 2 Diverse DNA metallization schemes and corresponding images. Construction of silver nanowires by (a) Na^+/Ag^+ ion exchange,[25] (b) molecular lithography[26] and (c) using a $AgNO_3$ solution.[32] (d) Different stages of the metallization process by chemical deposition of palladium.[34] The DNA template is λ-DNA except (a). All scale bars are 500 nm except (d).

fabrication of electronic circuits and components based on DNA in an excellent review.[30] Recent literature also reports the production of conductive silver nanowires with controlled width using DNA nanostructures.[6,7,31,32]

Metallization of λ-DNA with other metallic particles using similar methods has also been reported, including cobalt,[33] palladium,[34] platinum.[35,36] Metallization of λ-DNA through electroless deposition of palladium to build 1D parallel or 2D crossed metallic nanowires[37] and measurements of their electrical conductivity[38] have also been reported. In all cases, ohmic behavior of the nanowires was found to be different from that of bulk metal at room temperature and under cryogenic conditions. The possibility of using copper to form DNA-templated nanowires has been investigated as well.[39,40] Fig. 2 shows the results of various methods used to synthesize nanowires by DNA metallization using chemical deposition techniques.

3. Particle placement on DNA templates

Placement of metallic nanoparticles on DNA templates with nanometre precision is of great importance for both photonic and electronic applications. A group of accurately positioned nanoparticles may exhibit a strong surface plasmon resonance, or may act as seeds that can later coalesce into conducting structures of any predesigned geometry. Various DNA configurations may be used as scaffolds for such metallic structures. Linearized forms of DNA have been used for electrostatic assembly of metallic wires,[41–43] as shown in Fig. 3(a). More complex nanostructures require a higher degree of control on nanoparticle placement, and therefore may utilize more complex DNA templates, such as nanogrids constructed from a repeating tile[7,23] or scaffolded DNA origami.[19]

The particles that provide the basic units of any metallic nanostructure must be functionalized with a particular biological or chemical linker that enables their specific positioning within the DNA template. Mirkin and Alivisatos independently pioneered the functionalization of gold nanoparticles using thiol-labeled oligonucleotides.[44,45] Gold nanoparticles can be self-assembled within the DNA scaffold by means of complementary binding to organize structures with nanometric accuracy. Alivisatos further demonstrated the attachment of discrete numbers of oligonucleotides to gold nanoparticles in a controlled manner utilizing either gel electrophoresis[46] or HPLC purification steps.[47] These methods have recently been extended to dithiol-capped oligonucleotides which reportedly offer superior gold–DNA attachment.[48] Although efforts have concentrated largely on gold nanoparticles, silver nanoparticles have also been functionalized.[49] Another way to biofunctionalize gold nanoparticles is *via* the self-assembly of bis-biotinylated oligonucleotides and streptavidin.[50] Semi-synthetic DNA–protein conjugates can be produced on the remarkable biomolecular recognition of biotin by the homotetrameric protein streptavidin. This approach was also used to incorporate biotin-labeled oligonucleotides into a DNA scaffold, and provide binding sites for streptavidin-coated gold nanoparticles.[51]

Using different functionalization methods, nanoparticles of assorted sizes and materials have successfully been attached to various DNA templates to build a wide variety of structures in two- and three-dimensions (for related reviews, see ref. 6 and 52). The focus in this highlight is on 2D structures of metallic nanoparticles because they are most readily utilized for initial electronic and photonic applications. Regular, large scale arrays of gold nanoparticles, in which the underlying template is a DNA nanogrid composed of a small number of DNA sequences, have been reported by a number of groups,[51,53,54] as shown in Fig. 3(b). DNA origami has recently provided a template for arbitrary metallic structures; for instance, a self-similar chain of gold nanoparticles[55] as shown in Fig. 3(c). Placement and positioning of metallic nanostructures on a useful substrate, like silicon, for electronic and photonic applications are highly important. Controlled placement of a basic metallic structure on a silicon substrate has been recently demonstrated[56] utilizing DNA origami, as shown in Fig. 3(d).

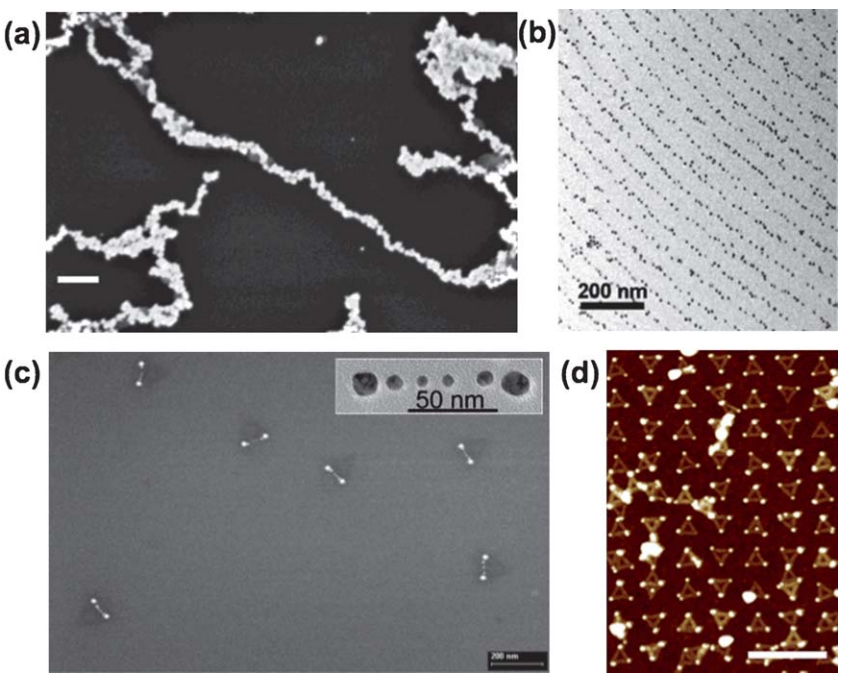

Fig. 3 Organization of preformed metallic nanoparticles templated on DNA. (a) Calf-thymus DNA as a template for Au nanowires.[42] Scale bar = 300 nm. (b) Placement of Au nanoparticles on a two-dimensional DNA nanogrid.[53] (c) A chain of different sized Au nanoparticles on DNA origami.[55] Scale bar = 200 nm. (d) Controlled positioning of metallic nanostructures on a silicon substrate using DNA origami.[56] Scale bar = 500 nm.

4. Vision for future artificial biomineralization

The future of biomolecular assembly for the fabrication of functional nanoelectronic and photonic devices is very

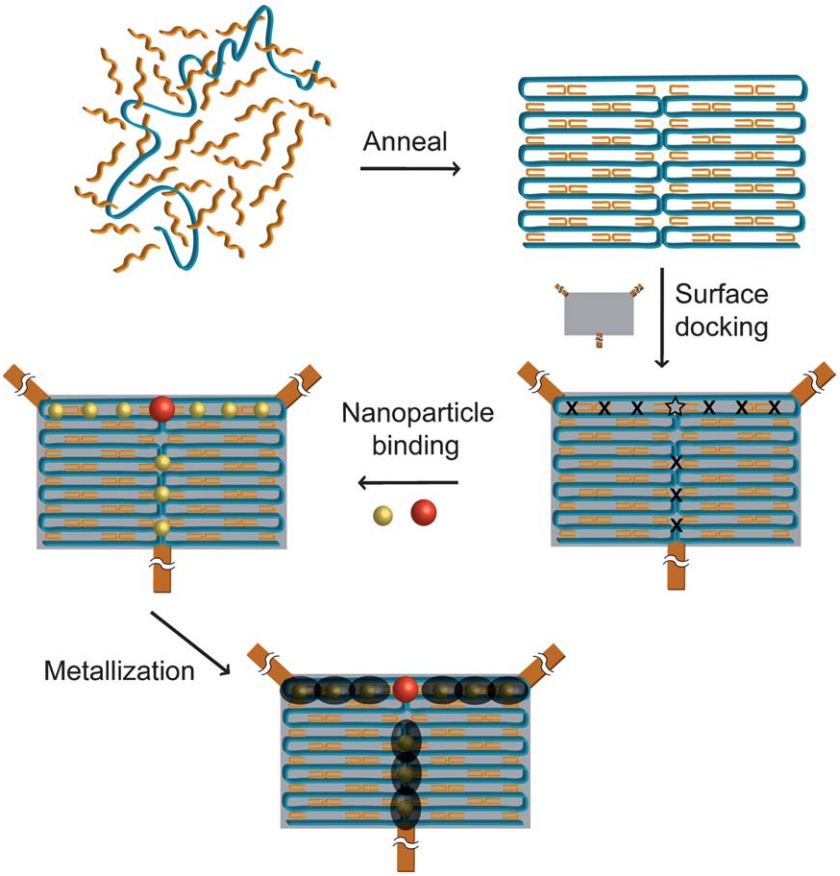

Fig. 4 A complete process for DNA-directed device formation. (1) Anneal of ssDNA scaffold strand (blue) with ssDNA staple strands (orange) into finite size DNA origami. (2) Surface docking of DNA origami with displayed binding sites (star and X′s) in a specific orientation on a surface containing electrodes (brown). (3) Nanoparticles (red and yellow) specifically bind to their complementary sites on the origami. (4) Metallization occurs on the metallic nanoparticles (yellow), fusing the nanoparticles together into nanowires also joined to the microscale electrodes. The central island (red) is assembled at the gap.

bright. Development of construction methods incorporating the placement of preformed nanomaterials with the chemical deposition of addition inorganics, as outlined in the sections above, offers high probability of creating significant advances in massively parallel fabrication of useful devices and circuits. Combining these techniques with location specific docking of the DNA templates on silicon substrates, as demonstrated in ref. 56, will serve to bridge the gap between bottom-up and top-down fabrication methods, and allow bionano-assembly steps to be fully integrated with conventional microfabrication processes. Future research will also focus on expanding the repertoire of inorganic materials available for incorporation, including semiconductors and dielectrics, as well as moving to the use of 3D DNA templates for patterning.

One possible process for integrating the described methods is outlined in Fig. 4. The particular structure shown would serve as a single-electron transistor[57] and could be used in properly designed circuits to represent computational bits (1 or 0) by gating one electron at a time. Such structures would vastly reduce energy requirements and heat buildup compared to conventional micron-scale, lithographically defined devices. Besides single-electron transistors, the described scheme could be adapted to the fabrications of other electronic devices, resonators, metamaterials, waveguides, and sensors, among others. Whether or not this particular envisioned biofabrication process ends up being used in the manufacture of computing and communication devices, harnessing some aspects of artificial, programmable biomineralization appears to be a promising path toward the production of complex, functional nanostructures.

Acknowledgements

The authors acknowledge support from the National Science Foundation (CCF-0829749) and the Office of Naval Research (N000140910249). E. C. Samano is thankful to DGAPA-UNAM for financial support.

References

1 N. C. Seeman, Nucleic acid junctions and lattices, *J. Theor. Biol.*, 1982, **99**, 237–247.
2 L. M. Adleman, Molecular computation of solutions to combinatorial problem, *Science*, 1994, **266**, 1021–1024.
3 E. Winfree, On the Computational Power of DNA Annealing and Ligation, in *DNA Based Computers*, ed. R. J. Lipton and E. B. Baum, American Mathematical Society, Providence, RI, 1996, vol. 27, pp. 199–219.
4 P. W. K. Rothemund, N. Papadakis and E. Winfree, Two computational primitives for algorithmic self-assembly: copying and counting, *Nano Lett.*, 2005, **5**, 2586–2592.
5 N. C. Seeman, An overview of structural DNA nanotechnology, *Mol. Biotechnol.*, 2007, **37**, 246–257.
6 H. Li, J. D. Carter and T. H. LaBean, Nanofabrication by DNA self-assembly, *Mater. Today (Oxford, U. K.)*, 2009, **12**(5), 20–28.
7 H. Yan, S. H. Park, G. Finkelstein, J. H. Reif and T. H. LaBean, DNA-templated self-assembly of protein arrays and highly conductive nanowires, *Science*, 2003, **301**, 1882–1884.
8 D. Liu, S. H. Park, J. H. Reif and T. H. LaBean, DNA nanotubes self-assembled from triple-crossover tiles as templates for conductive nanowires, *Proc. Natl. Acad. Sci. U. S. A.*, 2004, **101**, 717–722.
9 D. Liu, M. Wang, Z. Deng, R. Walulu and C. Mao, Tensegrity: construction of rigid DNA triangles with flexible four-arm DNA junctions, *J. Am. Chem. Soc.*, 2004, **126**, 2324–2325.
10 H. Yang, C. K. McLaughlin, G. Hamblin, F. Aldaye, A. Rys and H. F. Sleiman, Metal-nucleic acid cages, *Nat. Chem.*, 2009, **1**, 390–396.
11 P. S. Lukeman, A. C. Mittal and N. C. Seeman, Two dimensional PNA/DNA arrays: estimating the helicity of unusual acid polymers, *Chem. Commun.*, 2004, 1694–1695.
12 A. Chworos, I. Severcan, A. Y. Koyfman, P. Weinkam, E. Oroudjev, H. G. Hansma and L. Jaeger, Building programmable jigsaw puzzles with RNA, *Science*, 2004, **306**, 2068–2072.
13 K. Channon and C. E. MacPhee, Possibilities for 'smart' materials exploiting the self-assembly of polypeptides into fibrils, *Soft Matter*, 2008, **4**, 647–652.
14 M. Sarikaya, C. Tamerler, A. K. Y. Jen, K. Schulten and F. Baneyx, Molecular

biomimetics: nanotechnology through biology, *Nat. Mater.*, 2003, **2**, 577–585.
15 N. C. Seeman and P. S. Lukeman, Nucleic acid nanostructures: bottom-up control of geometry on the nanoscale, *Rep. Prog. Phys.*, 2005, **68**, 237–270.
16 D. Nykypanchuk, M. M. Maye, D. van der Lelie and O. Gang, DNA-guided crystallization of colloidal nanoparticles, *Nature*, 2008, **451**, 549–552.
17 S. Y. Park, A. K. R. Lytton-Jean, B. Lee, S. Weigand, G. C. Schatz and C. A. Mirkin, DNA-programmable nanoparticle crystallization, *Nature*, 2008, **451**, 553–556.
18 D. Sebba, J. J. Mock, D. R. Smith, T. H. LaBean and A. A. Lazarides, Reconfigurable core-satellite nanoassemblies as molecularly-driven plasmonic Switches, *Nano Lett.*, 2008, **8**, 1803–1808.
19 P. W. K. Rothemund, Folding DNA to create nanoscale shapes and patterns, *Nature*, 2006, **440**, 297–302.
20 S. M. Douglas, H. Dietz, T. Liedl, B. Högberg, F. Graf and W. M. Shih, Self-assembly of DNA into nanoscale three-dimensional shapes, *Nature*, 2009, **459**, 414–418.
21 E. S. Andersen, M. Dong, M. M. Nielsen, K. Jahn, R. Subramani, W. Mamdouh, M. M. Golas, B. Sander, H. Stark, C. L. P. Oliveira, J. S. Pedersen, V. Birkedal, F. Besenbacher, K. V. Gothelf and J. Kjems, Self-assembly of a nanoscale DNA box with a controllable lid, *Nature*, 2009, **459**, 73–77.
22 Y. Ke, S. M. Douglas, M. Liu, J. Sharma, A. Cheng, A. Leung, Y. Liu, W. M. Shih and H. Yan, Multilayer DNA origami packed on a square lattice, *J. Am. Chem. Soc.*, 2009, **131**, 15903–15908.
23 E. Winfree, F. Liu, L. A. Wenzler and N. C. Seeman, Design and self-assembly of two-dimensional DNA crystals, *Nature*, 1998, **394**, 539–544.
24 C. Mao, W. Sun and N. C. Seeman, Designed two-dimensional DNA holliday junction arrays visualized by atomic force microscopy, *J. Am. Chem. Soc.*, 1999, **121**, 5437–5443.
25 E. Braun, Y. Eichen, U. Sivan and G. Ben-Yoseph, DNA-templated assembly and electrode attachment of a conducting silver wire, *Nature*, 1998, **391**, 775–778.
26 K. Keren, M. Krueger, R. Gilad, G. Ben-Yoseph, U. Sivan and E. Braun, Sequence-specific molecular lithography on single DNA molecules, *Science*, 2002, **297**, 72–75.
27 K. Keren, R. S. Berman, E. Buchstab, U. Sivan and E. Braun, DNA-templated carbon notube field-effect transistor, *Science*, 2003, **302**, 1380–1382.
28 K. Keren, R. S. Berman and E. Braun, Patterned DNA metallization by sequence-specific localization of a reducing agent, *Nano Lett.*, 2004, **4**, 323–326.
29 Y. Eichen, E. Braun, U. Sivan and G. Ben-Yoseph, Self-assembly of nanoelectronic components and circuits using biological templates, *Acta Polym.*, 1998, **49**, 663–670.
30 E. Braun and K. Keren, From DNA to transistors, *Adv. Phys.*, 2004, **53**, 441–496.
31 S. H. Park, H. Yan, J. H. Reif, T. H. LaBean and G. Finkelstein, Electronic nanostructures template on self-assembled DNA scaffolds, *Nanotechnology*, 2004, **15**, S525–S527.
32 S. H. Park, M. W. Prior, T. H. LaBean and G. Finkelstein, Optimized fabrication and electrical analysis of silver nanowires templated on DNA molecules, *Appl. Phys. Lett.*, 2006, **89**, 033901.
33 Q. Gu, C. Cheng and D. T. Haynie, Cobalt metallization of DNA: toward magnetic nanowires, *Nanotechnology*, 2005, **16**, 1358–1363.
34 J. Richter, M. Mertig, W. Pompe, I. Mönch and H. K. Schackert, Construction of highly conductive nanowires on a DNA template, *Appl. Phys. Lett.*, 2001, **78**, 536–538.
35 R. Seidel, L. C. Ciacchi, M. Weigel, W. Pompe and M. Mertig, Synthesis of platinum cluster chains on DNA templates: conditions for a template-controlled cluster growth, *J. Phys. Chem.*, 2004, **108**, 10801–10811.
36 W. E. Ford, O. Harnack, A. Yasuda and J. M. Wessels, Platinated DNA as precursors to template chains of metal nanoparticles, *Adv. Mater.*, 2001, **13**, 1793–1797.
37 Z. Deng and C. Mao, DNA-templated fabrication of 1D parallel and 2D crossed metallic nanowire, *Nano Lett.*, 2003, **3**, 1545–1548.
38 J. Lund, J. Dong, Z. Deng, C. Mao and B. A. Parviz, Electrical conduction in 7 nm wires constructed on λ-DNA, *Nanotechnology*, 2006, **17**, 2752–2757.
39 C. F. Monson and A. T. Woolley, DNA-templated construction of copper nanowires, *Nano Lett.*, 2003, **3**, 359–363.
40 H. Kudo and M. Fujihara, DNA-templated copper nanowire fabrication by a two-step process involving electroless metallization, *IEEE Trans. Nanotechnol.*, 2006, **5**, 90–92.
41 F. Patolsky, Y. Weizmann, O. Lioubashevski and I. Willner, Au-nanoparticle nanowires based on DNA and polylysine templates, *Angew. Chem., Int. Ed.*, 2002, **41**, 2323–2327.
42 O. Harnack, W. E. Ford, A. Yasuda and J. M. Wessels, Tris(hydroxymethyl)-phosphine-capped gold particles templated by DNA as nanowire precursors, *Nano Lett.*, 2002, **2**, 919–923.
43 G. Braun, K. Inagaki, R. A. Estabrook, D. K. Wood, E. Levy, A. N. Cleland, G. F. Strouse and N. O. Reich, Gold nanoparticle decoration of DNA on silicon, *Langmuir*, 2005, **21**, 10699–10701.
44 C. A. Mirkin, R. L. Letsinger, R. C. Mucic and J. J. Storhoff, DNA-based method for rationally assembling nanoparticles into macroscopic materials, *Nature*, 1996, **382**, 607–609.
45 P. A. Alivisatos, K. P. Johnson, X. Peng, T. E. Wilson, C. J. Loweth, M. P. Bruchez and P. G. Schultz, Organization of 'nanocrystal molecules' using DNA, *Nature*, 1996, **382**, 609–611.
46 D. Zanchet, C. M. Micheel, W. J. Parak, D. Gerion and P. A. Alivisatos, Electrophoretic isolation of discrete Au nanocrystal/DNA conjugates, *Nano Lett.*, 2001, **1**, 32–35.
47 S. A. Claridge, H. W. Liang, S. R. Basu, J. M. J. Fréchet and P. A. Alivisatos, Isolation of discrete nanoparticle-DNA conjugates for plasmonic applications, *Nano Lett.*, 2008, **8**, 1202–1206.
48 J. Sharma, R. Chhabra, C. S. Andersen, K. V. Gothelf, H. Yan and Y. Liu, Toward reliable gold nanoparticle patterning on self-assembled DNA nanoscaffold, *J. Am. Chem. Soc.*, 2008, **130**, 7820–7821.
49 S. Pal, Z. Deng, B. Ding, H. Yan and Y. Liu, DNA-origami-directed self-assembly of discrete silver-nanoparticle architectures, *Angew. Chem., Int. Ed.*, 2010, **49**, 2700–2704.
50 C. M. Niemeyer, The developments of semisynthetic DNA-protein conjugates, *Trends Biotechnol.*, 2002, **20**, 395–401.
51 H. Li, S. H. Park, J. H. Reif, T. H. LaBean and H. Yan, DNA-templated self-assembly of protein and nanoparticle linear arrays, *J. Am. Chem. Soc.*, 2004, **126**, 418–419.
52 A. Kuzuya and M. Komiyama, DNA origami: fold, stick, and beyond, *Nanoscale*, 2010, **2**, 310–322.
53 J. D. Le, Y. Pinto, N. C. Seeman, K. Musier-Forsyth, T. A. Taton and R. A. Kiehl, DNA-templated self-assembly of metallic nanocomponent arrays on a surface, *Nano Lett.*, 2004, **4**, 2343–2347.
54 J. Zhang, Y. Liu, Y. Ke and H. Yan, Periodic square-like gold nanoparticle arrays templated by self-assembled 2d DNA nanogrids on a surface, *Nano Lett.*, 2006, **6**, 248–251.
55 B. Ding, Z. Deng, H. Yan, S. Cabrini, R. N. Zuckermann and J. Bokor, Gold nanoparticle self-similar chain structure organized by DNA origami, *J. Am. Chem. Soc.*, 2010, **132**, 3248–3249.
56 A. M. Hung, C. M. Micheel, L. D. Bozano, L. W. Osterbur, G. M. Wallraff and J. N. Cha, Large-area spatially ordered arrays of gold nanoparticles directed by lithographically confined DNA origami, *Nat. Nanotechnol.*, 2010, **5**, 121–126.
57 U. Coskun, H. Mebrahtu, P. Huang, J. Huang, A. Biasco, A. Makarovski, A. Lazarides, T. LaBean and G. Finkelstein, Single-electron transistors made by chemical patterning of silicon dioxide substrates and selective deposition of gold nanoparticles, *Appl. Phys. Lett.*, 2008, **93**, 123101.