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REVIEW

Origami and Kirigami Nanocomposites

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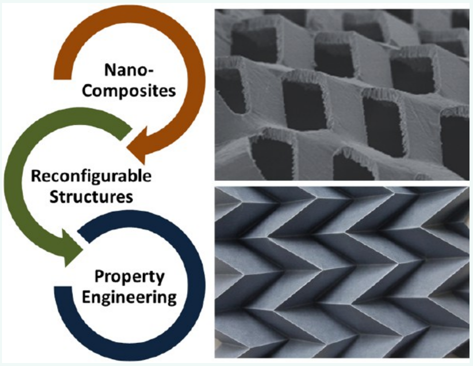
ABSTRACT: **The arts of origami and kirigami inspired numerous examples of macroscale hierarchical structures with high degree of reconfigurability and multiple functionalities. Extension of kirigami and origami patterning to micro-, meso-, and nanoscales enabled production of nanocomposites with unusual combination of properties, transitioning these art forms to the toolbox of materials design. Various subtractive and additive fabrication techniques applicable to nanocomposites and out-of-plane deformation of patterns enable a technological framework to negotiate often contradictory structural requirements for materials properties. Additionally, the long-searched possibility of patterned composites/parts with highly predictable set of properties/functions emerged. In this review, we discuss foldable/stretchable composites with designed mechanical properties, as exemplified by the negative Poisson's ratio, as well as optical and electrical properties, as exemplified by the sheet conductance, photovoltage generation, and light diffraction. Reconfiguration achieved by extrinsic forces and/or intrinsic stresses enables a wide spectrum of technological applications including miniaturized biomedical tools, soft robotics, adaptive optics, and energy systems, extending the limits of both materials engineering concepts and technological innovation.**



KEYWORDS: *kirigami materials, origami materials, nanocomposites, reconfigurable devices, 3D devices, stretchable electronics, 3D printing, implantable devices, energy harvesting and storage, sensors*

Over the past few decades, materials and structures inspired from ancient paper arts have attracted extensive research attention. Origami, a word originating from Japanese, refers to folding ("ori-") of paper ("gami-"). Kirigami, a variation of this paper art, involves cutting ("kiri-") of paper. These techniques afford fabrication of 3D objects from cutting, bending, and folding of thin paper sheets. Concepts in origami and kirigami have been recently generalized to a wide variety of scientific and engineering efforts that exploit out-of-plane deformations of 2D elements to enable large and reversible geometry changes of the devices built therefrom.

Origami and kirigami techniques as applied to composite materials can be viewed as methods of property engineering, which, arguably, lead to a new class of structures that will be defined in this article as origami and kirigami nanocomposites (OKNs). Other notations can be used as well but the preference was given to OKNs for being both general and descriptive.

OKNs have three major advantages over traditional composites engineered using primarily molecular scale approaches. First, origami and kirigami structures can be readily made with limitless variety of patterns and equally limitless range of composites whereas the choice of components for traditional composites and processing conditions are limited by miscibility, compatibility, temperature stability, *etc.* We note that the materials properties of the base nanocomposites at the scales smaller than those of the patterns do not change after patterning, but the effective properties at the scales larger than those of the patterns do.

OKNs can be considered to be close relatives of metamaterials with the distinction of reconfigurability. The typical optical and mechanical metamaterials are static, whereas the dynamic transition from two-dimensional (2D) patterns to three-dimensional (3D) constructs is an inalienable property of OKNs. The "nano" attribute of these composites is essential, too. Since most of the parts/components in our world have characteristic dimensions of 10^{-4}−10^{-1} m, the constituent materials must have patterns with characteristic dimensions ~100× smaller than the dimensions above, that is, 10^{-6}−10^{-3} m to be engineered using the effective properties of the structure and not the properties of the base materials. To afford such patterns, the base (composite) materials must have granularity in the scale ~100× smaller than that of the patterns, which is 10^{-8}−10^{-5} m. Despite the fundamental obstacle of phase separation, several nanocomposites afford uniform component distribution with adequate scale of uniformity.[1−3]

Second, current computational, simulation, and theoretical methods enable predictability of OKNs, which had a limited success for other composites and materials due to inevitable

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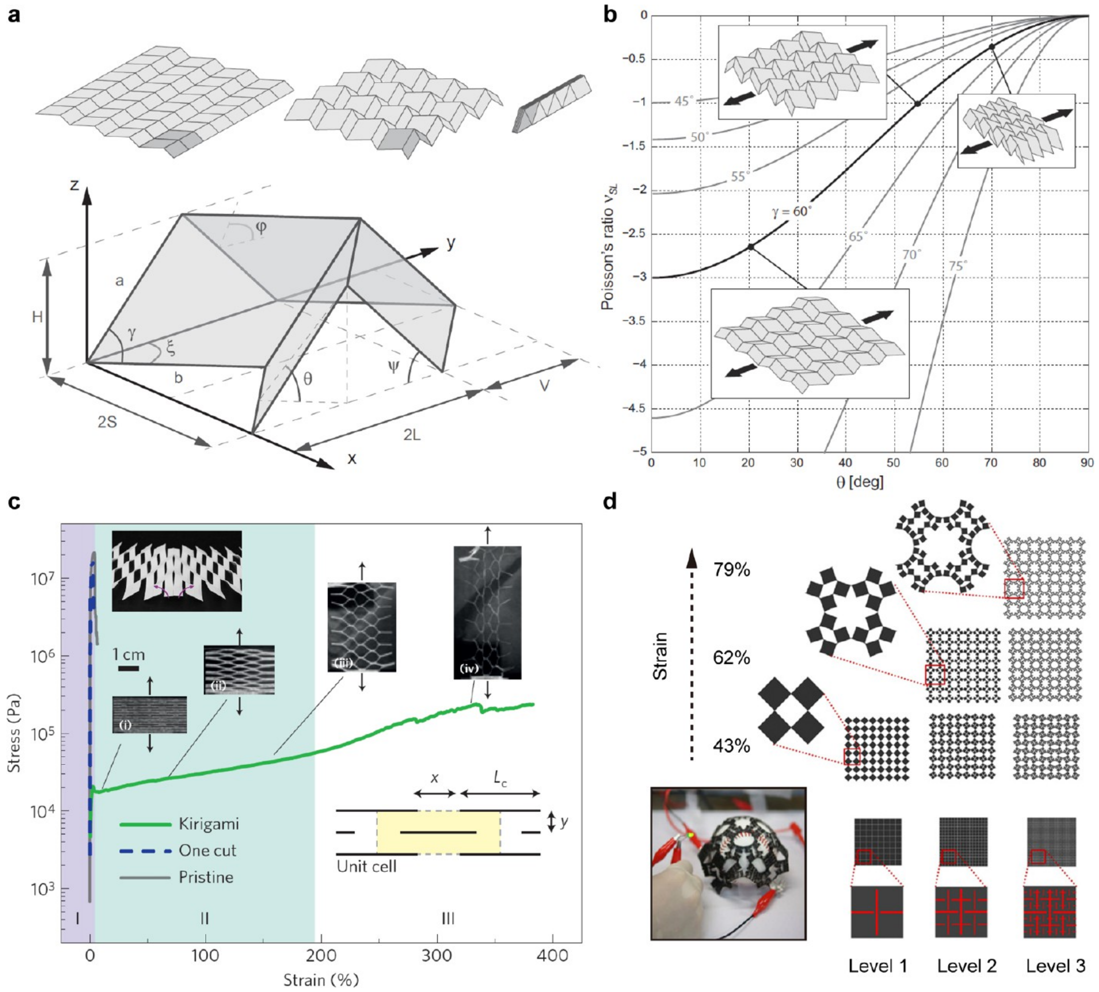

Figure 1. Physical behaviors of origami and kirigami structures. (a) Miura-ori and the geometrical parameters of its unit cell. Reproduced with permission from ref 23. Copyright 2013 National Academy of Sciences. (b) In-plane expansion coefficient, or Poisson's ratio, as a function of parameters γ and θ specified in (a). Reproduced with permission from ref 23. Copyright 2013 National Academy of Sciences. (c) Stress−strain curve for a kirigami sheet (green), compared with unnotched (gray) and single-notched (blue) counterparts. Insets show the pattern of cuts and photographs of kirigami sheets under different strain states. Reproduced with permission from ref 6. Copyright 2015 Macmillan Publishers Limited. (d) Finite element simulation of kirigami sheets with 2D fractal cuts. The maximum lateral strain for fractal iteration levels 1, 2, and 3 are 43, 62, and 79%, respectively. Inset shows a fractal kirigami sheet conforming to a spherical surface. Reproduced with permission from ref 35. Copyright 2014 National Academy of Sciences.

heterogeneity, defects, and interfaces; all of these property-determining factors are associated with tremendous computational complexity.[4,5] The molecular composition and the notch/crease patterns are used in the case of OKNs as input parameters; mechanical, optical, and electrical properties in macroscale can be predicted with high accuracy and wide dynamic range.[6,7]

Third, OKNs provide macroscopic deformability far beyond the strain limit of the constituent materials, best performing elastomers, foams, *etc.* Simultaneously, they offer reproducible manufacturability using established additive or subtractive patterning techniques. It results in the possibility to combine properties that are typically contradictive to each other, for instance, high conductivity and stretchability. Also, high macroscopic deformability and high accuracy of patterning make possible programmable and complex response to strain for deformable systems. Consequently, OKNs are likely to find utility in a wide spectrum of applications, including biomedical microdevices, human-centered electronics, wearable energy harvesters, adaptive optics, and many others. While a number of review articles related to this area have been focused on specific structures,[8−12] processing techniques,[13,14] or applications,[15,16] herein, we provide our analyses of origami and kirigami materials, with an emphasis on the three distinguishing advantages outlined above. The discussion starts with a fundamental exploration of OKNs, including their general physical behaviors, applicable materials, and fabrication techniques, followed by various schemes for achieving reconfigurable structures. We then highlight the broad range of technological applications associated with OKNs and conclude with an outlook of emerging opportunities for future research.

Fundamental Explorations of OKNs. An early example of applying the concepts of paper art to the creation of engineering systems was reported by Miura.[17] In this work, a pseudocylindrical concave polyhedral shell was developed as a deployable device. The folding patterns in the thin shell structure, which are similar to those in paper art, enable high deformability of the system. This class of folding patterns, since named "Miura-ori" (Miura folding), received extensive

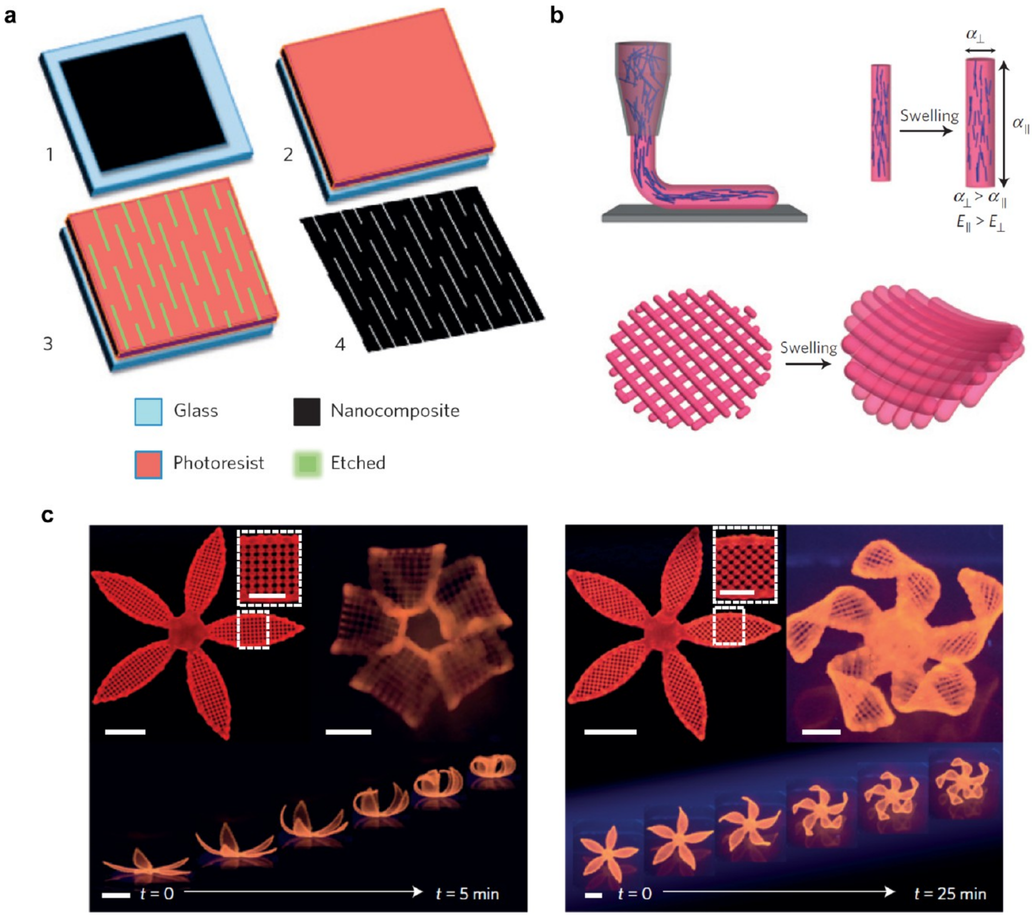

Figure 2. Fabrication techniques for OKNs. (a) Schematics of planar patterning techniques for kirigami of LbL-assembled nanocomposites. Reproduced with permission from ref 6. Copyright 2015 Macmillan Publishers Limited. (b) Schematics of a 3D printing process for hydrogel-cellulose fibrils composites. Shear-induced alignment of the cellulose fibrils enables anisotropic swelling behaviors. E and α denote stiffness and swelling strain, respectively. Reproduced with permission from ref 43, Macmillan Publishers Limited. (c) "4D-printed" origami flowers with different self-folding schemes determined by orientations of the filaments. Scale bars: 5 mm (2.5 mm for the insets). Reproduced with permission from ref 43. Copyright 2016 Macmillan Publishers Limited.

attention thereafter, especially in the development of space solar panels.[18] In the past few decades, academic research inspired by origami and kirigami has expanded rapidly.[19−22] Among these works, the fundamental studies are mainly focused on two aspects: (1) physical behaviors of origami and kirigami as they relate to scientific and engineering interests; and (2) materials and processing techniques that enable origami and kirigami structures.

Miura-ori is one of the most extensively studied origami structures. Schenk and Guest systematically studied the geometry and kinematics of Miura-ori (Figure 1a,b).[23] A negative Poisson's ratio is characteristic for in-plane deformation of this structure and is closely connected to the excellent foldability and deployability of such structures. In this work, the geometrical parameters of a unit cell and its tessellations are quantitatively analyzed as the deterministic factors for the physical behaviors, as exemplified by the tunable Poisson's ratio for in-plane deformation. In addition, out-of-plane deformation and 3D stacking of Miura-ori are also analyzed in this and other reports.[24−27] Cohen *et al.* noted that the mechanics of Miura-ori-based metamaterials are highly sensitive to defects, which allow for broad tuning of a material's stiffness by introduction of local geometrical modifications.[28] Nojima *et al.* created classes of folding schemes that extend the achievable structures through origami.[29] In addition to Miura-ori and its variations, other foldable structures, such as folded polyhedral[30,31] and rings with overcurvature,[32] have been theoretically modeled and experimentally realized, expanding the library of origami schemes. Beyond specific structures, general mathematical analyses of flat-foldability problems provide useful guidelines for origami-based designs.[33]

While typical origami mostly involves the folding of thin planar sheets, introduction of additional cuts might further enhance the deformability of the resulting structures. The class of methods that involve cutting, referred to as kirigami, has recently attracted wide attention. A simple yet typical example of kirigami consists of a pattern of parallel slits (Figure 1c).[6] These cuts enable out-of-plane deformation of the membrane structures in response to in-plane stretching, accommodating large macroscopic strains which would otherwise cause fracture of an un-notched counterpart. Here, the stark contrast between bending stiffness and in-plane stiffness of a thin plate favors out-of-plane deformations in order to minimize the strain energy. Mechanical simulation based on finite element methods accurately predict the deformation patterns, revealing that the local strain in the kirigami materials can be more than 2 orders of magnitude lower than the macroscopic strain exerted on the entire devices, resulting in only elastic deformation of the constituent nonstretchable materials even under a macroscopic tensile strain of 100%.[7] Such kirigami techniques enable

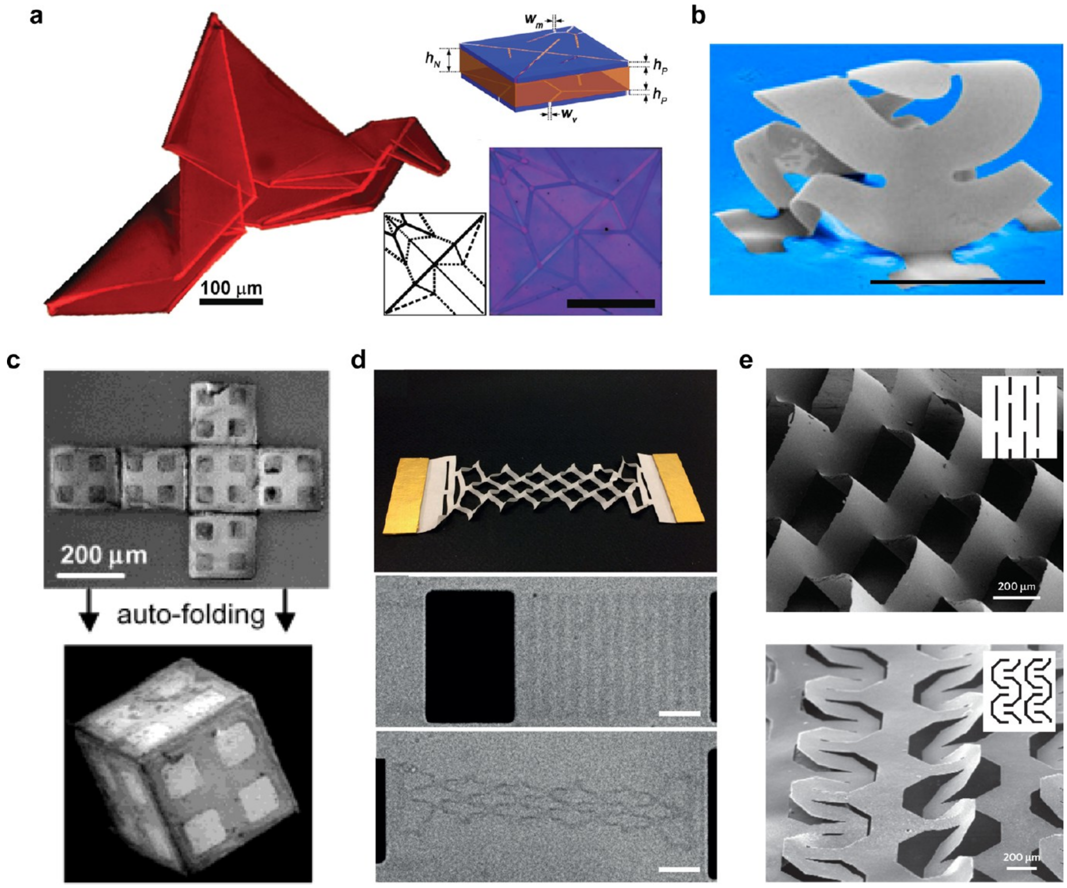

Figure 3. Examples of constituent materials for OKNs. (a) Left: Fluorescent image of an origami bird made from self-folded polymeric composites. Upper right: schematics of the trilayer structure with differential patterning on the top and the bottom surfaces. Lower right: crease patterns for self-folding. Scale bar: 400 μm. Reproduced with permission from ref 46. Copyright 2015 Wiley-VCH. (b) Scanning electron microscopy (SEM) image of a kirigami 3D structure made of silicon nanomembrane/polymer bilayer. Scale bar: 200 μm. Reproduced with permission from ref 48. Copyright 2015 National Academy of Sciences. (c) SEM images of a self-folded metallic microcube. Reproduced with permission from ref 50. Copyright 2002 Wiley-VCH. (d) Top: Photograph of a macroscale kirigami structure similar to that applicable to graphene. Bottom: Optical microscope images of kirigami-patterned graphene, enabling a stretchable transistor. Scale bar: 10 μm. Reproduced with permission from ref 52. Copyright 2015 Macmillan Publishers Limited. (e) SEM images of kirigami sheets based on graphene oxide/poly(vinyl alcohol) composites. Reproduced with permission from ref 6. Copyright 2015 Macmillan Publishers Limited.

unusual engineering structures with high tolerance to extreme physical deformation.[34] More sophisticated kirigami that involves 2D fractal iterations affords biaxial expansion, allowing a 2D membrane to conform to 3D surfaces (Figure 1d).[35] Higher orders of fractal iterations result in greater deformability of the kirigami sheets. In addition, assembly of complex 3D objects from lattice kirigami can also be theoretically modeled and practically achieved.[36]

Importantly, the deformation of origami and kirigami-based structures is mostly determined by the kinematics associated with the geometrical parameters and are scale-independent. This behavior allows for the construction of a diverse range of materials with multiscale fabrication routes. Realization of origami/kirigami structures and devices often involves composite materials that can be finely tailored in terms of geometry and functionality. The expansive toolbox of both subtractive and additive fabrication techniques can be readily applied to OKN manufacturing. For example, carbon-based nanocomposites prepared using layer-by-layer (LbL or LBL) assembly can be kirigami-patterned by standard photolithography and etching techniques (Figure 2a).[6,7] The LbL approaches, which involve sequential deposition of alternating nanoscale components, can generate a large variety of thin-film composites with tailored properties.[37,38] As exemplified by a carbon nanotube (CNT)/poly(vinyl alcohol) (PVA) LbL composite, a conductivity of ~4.15 × 10^4 S/m, Young's modulus of ~12 GPa, mechanical strength of ~220 MPa, and optical transmittance of ~80% are combined in the thin-film structure.[39] Such a combination of parameters is rarely found in traditional polymeric composites or electrical conductors but is possible with LbL-engineered materials with nanoscale control of chemical composition and structural iteration. To generate kirigami patterns, planar fabrication approaches adapted from conventional semiconductor technologies are particularly useful. Photolithography can be applied to transfer the digitally designed patterns from photomasks to the photoresist (PR) layer with nanometer-scale fidelity. Etching techniques including wet chemical treatment or plasma etching can selectively remove unwanted materials not covered by PR and/or release the thin film from the handling substrates, generating free-standing kirigami composites. In addition to the traditional planar fabrication approaches, emerging classes of 2D patterning methods, such as imprint lithography, are also useful.[40]

Recently developed techniques in additive fabrication, referred to as 3D printing, can be applied to generate OKNs

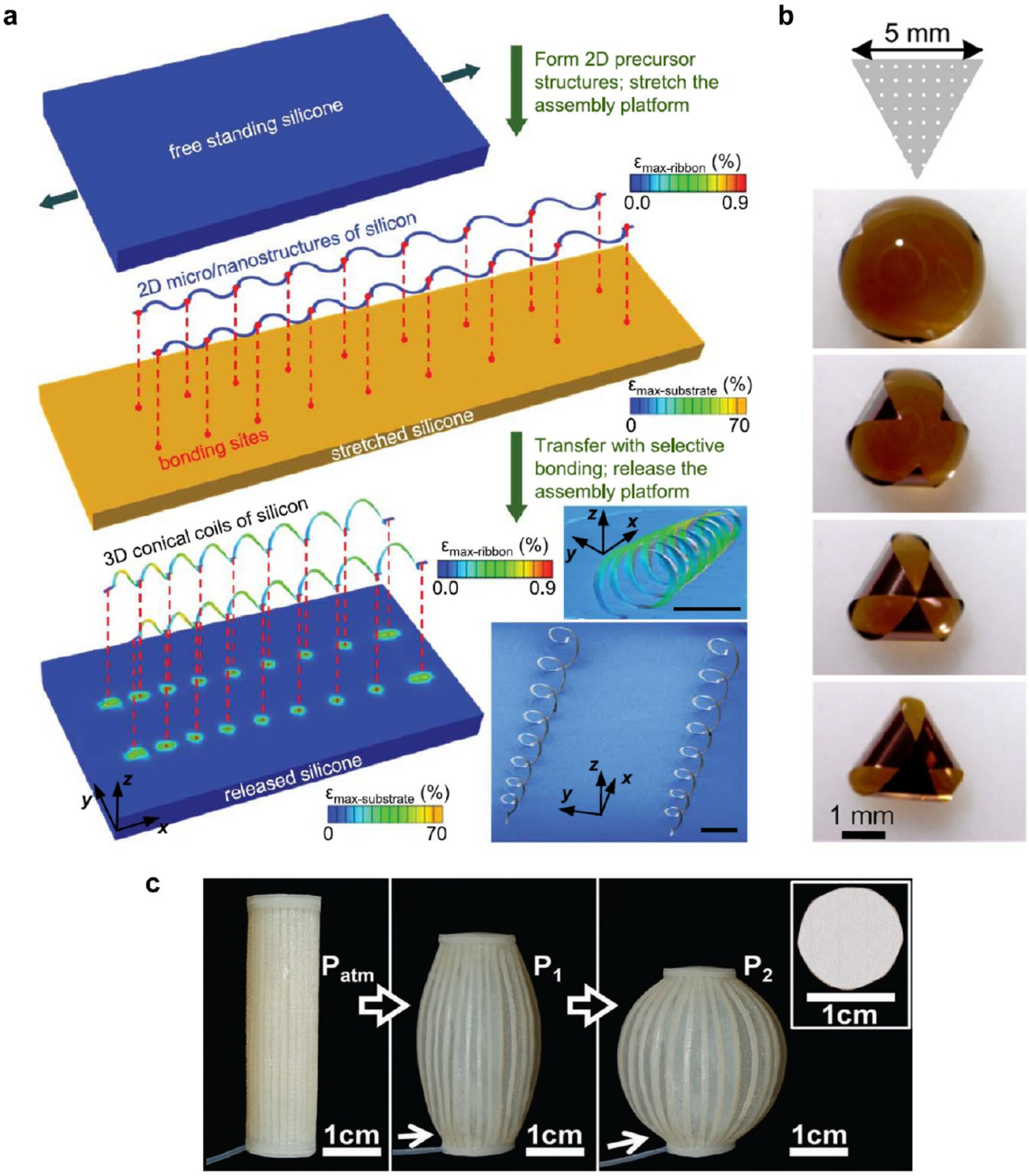

Figure 4. Reconfigurable OKNs actuated with extrinsic forces. (a) Finite element analyses on strain distribution associated with the 3D assembly processes for a silicon nanoribbon selectively bonded to an elastomer slab. Lower right shows SEM images of experimental results. Scale bars: 400 μm. Reproduced with permission from ref 61. Copyright 2015 AAAS. (b) Spontaneous folding of a single-crystalline silicon membrane of a 1.25 μm thickness, as a result of evaporation of an applied water droplet. Reproduced with permission from ref 63. Copyright 2009 National Academy of Sciences. (c) Photographs of pneumatic origami of paper-elastomer composites: P_1 = 80 mbar, P_2 = 200 mbar. Reproduced with permission from ref 64. Copyright 2012 Wiley-VCH.

on the micro- or macroscale. A typical route for generating 3D-printed OKNs is reported by Lewis *et al.*[41] Here, engineering of the viscosity of complex fluids affords direct writing of various materials with custom-designed 3D geometries.[42] Folding can be naturally incorporated in these 3D-printed composites. For example, hydrogels with shear-aligned cellulose fibrils can be directly written in 3D space (Figure 2b).[43] The anisotropic swelling behavior of the composites with preferentially oriented cellulose fibrils encodes stimuli-responsive self-folding of the printed structures. Upon exposure to appropriate solvents or other physical/chemical triggers, time-dynamic and deterministic geometry changes can be realized through autonomous folding, which is also referred to as "4D printing".[43−45] A variety of complex 3D structures can be achieved with different inverse-engineered folding routes dictated by the spatial arrangement of the printed materials (Figure 2c).

Using the expansive toolbox of subtractive (*e.g.*, photolithography and etching) and/or additive (*e.g.*, 3D printing) fabrication techniques, a variety of material components, including polymers, metals, inorganic semiconductors and emerging 2D nanomaterials, are demonstrated for utilization as OKNs. Hayward and co-workers reported origami from thermally responsive hydrogels sandwiched by photopatternable, stiff polymer layers (Figure 3a).[46] The creases are differentially defined at the top and bottom surfaces by selective removal of the stiff polymers. In this scheme, thermally triggered expansion of the hydrogel is guided in different patterns by the top and bottom surfaces, enabling origami of complex structures. Rogers and co-workers developed routes for origami and kirigami of semiconductor nanomembranes.[47,48] In contrast to the rigid and brittle format of inorganic semiconductors wafers, nanomembranes can be

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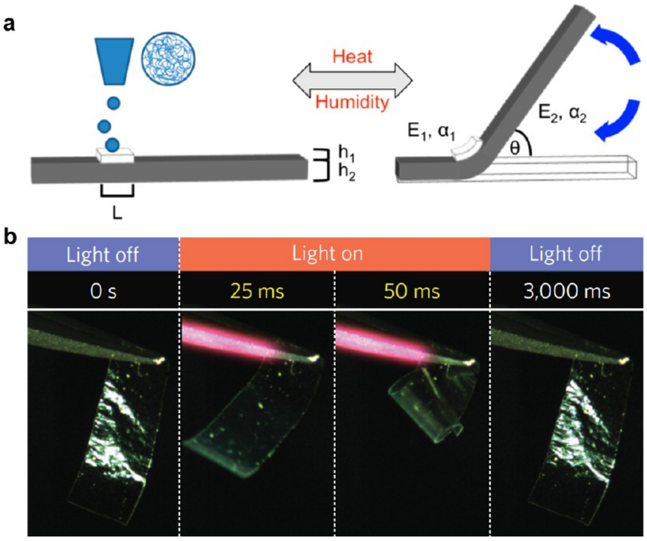

Figure 5. Reconfigurable OKNs actuated with intrinsic stress. (a) Schematic of a humidity-sensitive nanocomposites bimorph (white) deposited on an arm of conductive, humidity "inert" nanocomposite (gray). The bimorph bends in response to exposure to humidity and temperature stimuli. The bending angle θ depends on the thickness (h), stiffness (E), and expansion coefficient (α) of each layer, along with the total length of the bimorph (L). Reproduced with permission from ref 82. Copyright 2014 American Chemical Society. (b) High-speed snapshots of the motions of a humidity- and light-sensitive carbon nitride polymer film in response to ultraviolet (365 nm) irradiation. Reproduced with permission from ref 98. Copyright 2016 Macmillan Publishers Limited.

extremely flexible due to the favorable bending mechanics associated with the reduction of thickness.[49] Therefore, origami and kirigami become achievable with engineered out-of-plane deformation of these nanomembranes. For example, a cutting pattern can be made in a Si membrane with thickness of ~100 nm. When a compressive mechanical load is applied to the nanomembrane that is selectively bonded to a soft substrate, a complex 3D assembly can be achieved through buckling of the 2D elements (Figure 3b).[48] Gracias and co-workers developed techniques to fold metallic composites at the microscale.[50] In this technique, a photopatterned Cr/Au/Ni composite with deformable hinges can be folded through the surface tension of melted solders, allowing for generation of patterned cubes from 2D sheets (Figure 3c). Although further reconfiguration might be difficult for this particular structure, the concept of surface-tension-driven assembly represents a powerful tool for building reconfigurable OKNs. Emerging classes of 2D nanomaterials, as exemplified by graphene and MoS_2, have also been explored (Figure 3d).[51−55] The atomic-scale thickness and strong intraplane bonding afford extreme deformation of kirigami structures made from 2D nanomaterials. Composites based on graphene and/or graphene oxide can be patterned with kirigami motifs (Figure 3e), enabling stretchable electrodes with relevance to tunable plasma generation.[6] In addition, plasmonic nanoparticle superlattice sheets are also demonstrated for generating origami structures.[56]

Interestingly, the principles of origami and kirigami are also found in natural biological materials. For example, structures similar to Miura-ori can be seen in hornbeam leaves during the blooming process.[57] The self-guided folding is associated with

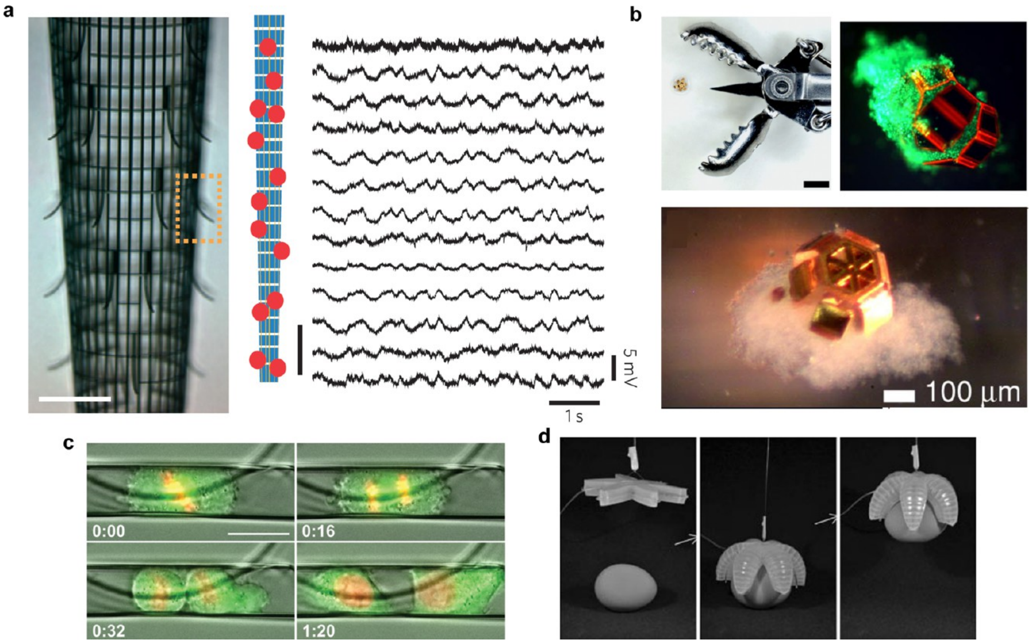

Figure 6. Applications in biomedicine and soft robotics. (a) Left: Micrograph of self-folded, macroporous nanoelectronic 3D neural sensors, with a representative probe outlined by the orange dashed box. Right: Sensor positions and the local field potentials in mouse somatosensory cortex measured through these probes. Scale bars: 200 μm. Reproduced with permission from ref 99. Copyright 2015 Macmillan Publishers Limited. (b) Origami micrograppers with utilities in biopsy. Top left: photograph of a microgripper compared with conventional biopsy forceps. Scale bar: 1 mm. Reproduced with permission from ref 107. Copyright 2013 Elsevier. Top right and bottom: micrograph of viable cells and tissues captured by a microgripper. Reproduced with permission from ref 109. Copyright 2009 National Academy of Sciences. (c) Fluorescent and phase-contrast time-lapse images of a dividing HeLa cell entrapped in a self-rolled-up microtube. Scale bars: 15 μm. Time in hour/min format. Reproduced with permission from ref 114. Copyright 2014 American Chemical Society. (d) Photographs of a pneumatically actuated soft gripper handling an uncooked chicken egg. Reproduced with permission from ref 120. Copyright 2011 Wiley-VCH.

compressive buckling of layered materials with stiffness mismatch.[58] In another example, hydro-actuated self-folding is characteristic in ice plant seed capsules.[59] Similar concepts have led to artificial origami materials actuated with capillary/Laplace pressure.[60] These studies illuminate some of the thermodynamic driving forces involved and provide inspiration for engineering origami and kirigami systems.

Reconfigurable Materials and Structures. The ability to dynamically and reversibly change geometry is characteristic of many origami and kirigami systems and is crucial for potential engineering applications of OKNs. Here, the driving force for actuation is critically important. While manual folding is normally exercised in paper art, engineering origami and kirigami require alternative actuation schemes as device miniaturization and/or geometrical complexity restrict direct manipulation. The various research efforts made thus far can be categorized into two general actuation strategies. The first utilizes extrinsic forces applied on the OKNs. For example, compressive loading can be applied for guiding the 3D assembly of nanomembranes.[61,62] A soft elastomer substrate can be prestretched and selectively bonded to a Si membrane through covalent bonding sites patterned by photolithography (Figure 4a). With controlled release of the stretch, the substrate tends to recover to the original geometry and exerts a compression force on the Si membrane through the bonded interfaces. Due to the nanoscale thickness (∼100 nm) and serpentine design (∼10 μm in line width and ∼100 μm in radius), the Si structures experience out-of-plane buckling to minimize strain energy associated with the compression. Consequently, 3D conical coils, for example, can be assembled from a 2D serpentine strips. In another example of kirigami composites, tensile loading can be directly applied through the periphery of the structures to actuate the reconfiguration.[6,35] In both of these cases, theoretical modeling plays a central role of guiding the design of 3D assemblies, wherein the deformation patterns are readily predictable based on the inputs of the mechanical parameters of the base materials as well as the geometry of the structures. At reduced dimensions, the surface tensions of liquids become a powerful source for actuation,[50,63] as they scale linearly with characteristic length, whereas gravitational and elastic forces scale with the length cubed. For example, evaporation of a water droplet can effectively fold a patterned Si membrane with millimeter scale in planar dimensions.[63] With the tapered corners in the membrane geometry, minimization of the water−Si−air interface energy will compete with the elastic bending energy as the water evaporates (Figure 4b). In addition to water, other liquids such as melted solders can also be utilized for surface-tension-actuated origami.[50]

An emerging class of soft elastomer origami exploits pressurized or depressurized fluid for actuation. Such pneumatic or hydraulic devices involve composite chambers with non-homogeneous mechanical compliance.[64] A typical structure can be made from paper sheets selectively embedded into soft silicone elastomer (Figure 4c). When pressurizing the chamber, the relatively stiff paper restricts elongation of its structure and leads to anisotropic deformation that primarily expands the soft elastomer membrane. As a result, complex 3D deformation can be generated with as few as one pneumatic input. Additional channels can provide a higher degree of freedom for actuation.[65] The 3D printing techniques facilitate custom-designed geometry desirable for complex systems.[66] In addition to expansive actuation, depressurizing soft composite chambers results in contractive deformation, which enables similar functions to those in skeleton muscles.[67] Other forces including electrostatic[68] and magnetic[69,70] interactions can also be effective for actuating OKNs.

The second actuation scheme exploits intrinsic stress built into the composites. Mismatch stress is typically utilized and can be induced, for example, by chemical or thermal effects, leading to self-folded OKNs. Established theories on stress-induced bending in thin films formulate the underlying mechanics.[71] An example of incorporating intrinsic stress for microscale origami is reported by Smela *et al.*[72] Here, conducting polymers (*e.g.*, polypyrrole doped with dodecylbenzenesulfonate) are bonded to metal thin films to form bilayer composites. Upon electrochemical insertion of cations, the conducting polymer layer expands dramatically while the metals experience minimal expansion. Therefore, a bending moment will occur in response to the mismatch of membrane stress between conductive polymers and the metals, generating self-folded structures. A similar actuation scheme with differential expansion can be induced also by the swelling of polymers[73,74] or redox reactions.[75] In an ambient environment, self-folded origami can be actuated using humidity- and/or temperature-sensitive materials. For example, thermally responsive hydrogels such as poly(*N*-isopropylacrylamide) enable origami structures modulated by heating and cooling.[76,77] Shape memory alloys or polymers can be used for origami devices with sharp temperature responses and large deformation ranges;[78−80] however, the engineering of two-way actuation might require additional considerations.[81] Humidity-absorbing components, such as LbL-assembled poly(diallyldimethylammonium)chloride/poly(sodium-4-styrenesulfonate),[82] poly(3,4-ethylenedioxythiophene):poly(sodium-4-styrenesulfonate),[83,84] poly(acrylic acid)/poly(allylamine hydrochloride),[85] and polypyrrole,[86,87] are applicable as active materials for vapor-based actuation. Here, the expansion or contraction can be modulated with exposure to moisture or thermal evaporation of water, respectively (Figure 5a). In addition, stress associated with vacuum deposition of metal thin films can be another source of actuation.[88,89] In a typical configuration, a Cr thin film is deposited on Cu (or other metals) supported by a rigid planar substrate. Once patterned and released from the handling substrate, the multilayer composites experience self-folding in order to release the internal tensile stress built up in the Cr film. The folding angles and 3D morphology can be engineered by optimizing the thickness of metal stacks and the geometry of the hinges, *etc.*[90,91] Combination with magnetic components allows for remote manipulation of these micro-origami devices.[92] Incorporation of light-sensitive materials, exemplified by CNTs,[93] graphene,[94] and other polymeric components,[95−97] affords optical actuation through photothermal or photochemical effects. While typical intrinsic-stress-induced origami actuates with an approximate time scale of seconds, recent advances in chemistry enable much faster response. A class of layered carbon nitride thin films can be surprisingly sensitive to humidity and ultraviolet irradiation, leading to photothermal actuation times as low as ∼50 ms (Figure 5b).[98] The various actuation schemes are mutually complementary, creating versatile embodiments for reconfigurable engineering systems.

Applications. The unusual combination of OKN properties and their predictability in the wide range of conditions are attractive for a long list of traditional and advanced technologies, as exemplified by automotive industry, robotics,

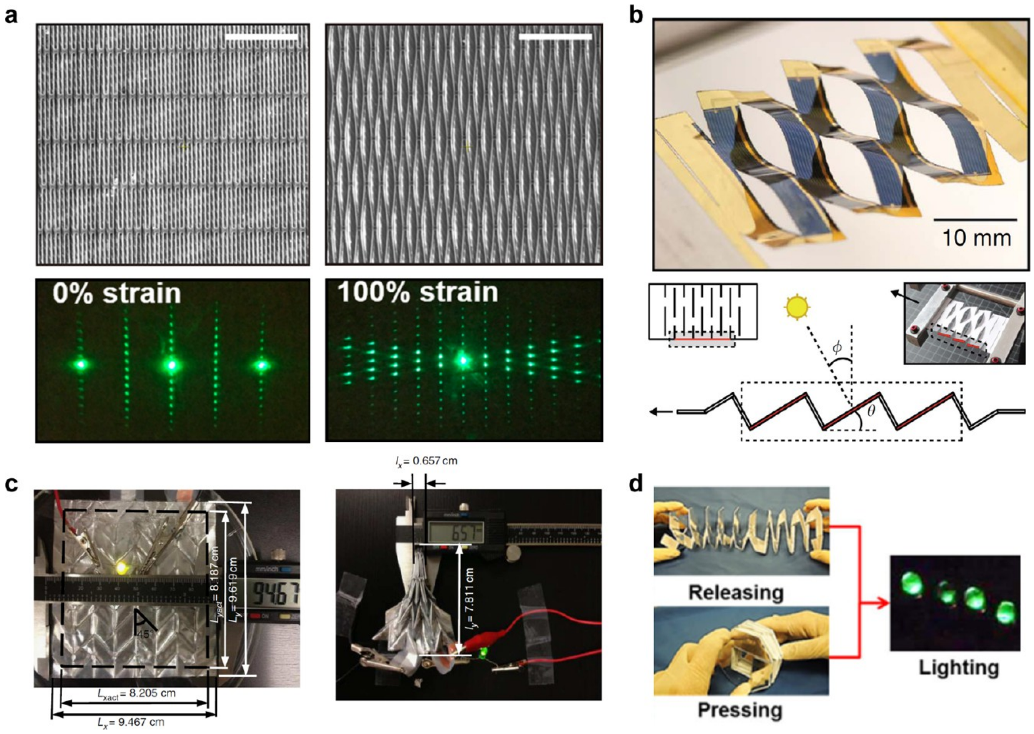

Figure 7. Applications in optics, electronics, and energy harvesting/storage devices. (a) SEM images (top) and laser diffraction patterns (bottom) of a nanocomposite kirigami grating under 0% (left) and 100% strain (right). Scale bars: 50 μm. Reproduced with permission from ref 7. Copyright 2016 American Chemical Society. (b) Photograph and schematic of a kirigami solar cell for dynamic sunlight tracking. Reproduced with permission from ref 125. Copyright 2015 Macmillan Publishers Limited. (c) Photographs of a foldable lithium ion battery based on Miura-ori structures, powering a light-emitting diode. Reproduced with permission from ref 127. Copyright 2014 Macmillan Publishers Limited. (d) Photographs of a slinky-format triboelectric nanogenerator based on the concepts of origami. Reproduced with permission from ref 129. Copyright 2015 American Chemical Society.

biomedical devices, tunable electronics and optics, and energy harvesting and storage systems.

Probing the complex 3D structures of the human body and its various tissues has long been a technical challenge. Conventional medical tools based on bulk metals and plastics or electronic devices based on semiconductor chips provide useful functionalities, but their performance is compromised by the limited physical interface between devices and soft biological tissues. Emerging OKNs can enable miniaturized 3D devices to address this challenge. For example, planar-fabricated microelectrode arrays can be self-folded into 3D geometries that naturally integrate with brain tissue.[99,100] A 2D-patterned electrode mesh self-folds into a cylindrical geometry with active recording sites bending outward (Figure 6a).[99] The resulting structure facilitates minimally invasive insertion into the brain and allows for high-fidelity measurements of surrounding neuronal activities (Figure 6a). These porous devices feature cellular-scale dimensions (~10 μm in line width) and ultracompliant mechanics (~0.64×10^{-15} Nm^2 in bending stiffness), desirable for integration with brain tissues. Histology studies on animal models demonstrate that their utilization significantly reduces tissue damage and unwanted immune responses compared with traditional tools, indicating possibilities for development of reliable and long-term brain-machine interfaces. Similar forms of foldable electronic mesh enable other applications including syringe-injectable electronics[101] and electronic tissue engineering scaffolds.[102,103] In another example, ultrastretchable electronics built with kirigami structures can conform to the 3D, time-dynamic surfaces of tissues and are capable of measuring physiological activities.[104]

In addition to microbioelectronics, OKNs from electro-inactive materials can also enable smart devices for medical intervention. Gracias and co-workers developed origami microgrippers for biopsy applications.[105−109] These micrometer-sized devices can effectively enter organs of interest and self-fold to sample biological tissues (Figure 6b). Remote control and navigation of the devices are achieved with magnetic components.[106] Compared with traditional biopsy forceps, these microgrippers have dramatically smaller dimensions, which is beneficial for reducing side effects and the operational difficulties associated with the physical mismatch between medical tools and the soft tissues.[107] Self-folded structures made from OKNs can also be useful for other application scenarios including microdrillers[110] or drug-delivery capsules.[108,111] In addition, Miura-ori motifs can be used to form reconfigurable stents that are useful in aortal or esophageal implants.[112]

OKNs enable 3D devices for tissue engineering platform and microfluidics, creating schemes for regenerative medicine or laboratory biomedical research. For example, self-folded microtubes or microcubes allow for guided growth and precise characterization of single cells (Figure 6c).[113−115] Paper-based origami composites lead to facile constructs for *in vitro* biomineralization[116] or *in vivo* trachea regeneration.[117] Complex 3D microfluidic networks can be generated from self-folded elastomer[118] or paper composites[119] customized with 2D patterning techniques. In most of these cases, the microscale 3D objects are difficult to fabricate directly with top-down approaches but are readily achievable through origami and kirigami engineering using self-folding 2D precursors. In addition to devices that directly interface with the human body, tissues and cells, biomimetic soft-robotics also benefit from OKNs. Pneumatically actuated elastomer origami represents a core element in these systems. The soft robots can be utilized as, among other examples, soft grippers for food handling[120] (Figure 6d) or deep-reef biological sampling.[121] These soft robots complement traditional industrial hard robots and allow for cooperative interaction with humans and various soft or fragile objects.

Highly tunable electronic and optical devices are achievable through OKNs. For example, microscale kirigami structures with periodic slit patterns can be utilized as a tunable optical diffraction gratings (Figure 7a).[7] Long-standing problems related to the limited strain tolerance of bulk optical materials have hindered the development of tunable optical devices. Utilization of kirigami concepts bypasses these problems and generates gratings with 100% reversible stretchability even from traditionally stiff metals and carbon materials. Such devices enable wide-angle steering of optical beams and simplify engineering embodiments, providing advanced building blocks for light detection and ranging (LIDAR), spectroscopy, and other technologies. Other electronic or optical devices, including tunable inductors[61] and antennas,[122] optical metamaterials,[123] and 3D wiring of electronic components,[124] are also achievable though origami and kirigami routes.

Application in energy harvesting and storage is another growing area of OKN research. For example, photovoltaics capable of dynamic solar tracking are possible with kirigami constructs.[125] Here, thin-film gallium arsenide modules can be tilted in response to the direction of incident sunlight, maximizing solar power generation (Figure 7b). Their geometrical configuration is finely controlled by out-of-plane buckling of kirigami flaps. While conventional solar tracking systems rely on the sophisticated assembly of mechanical components, the use of kirigami composites simplifies the actuation scheme and significantly reduces the weight and cost of the systems. Jiang and co-workers developed a class of foldable solar cells[126] and lithium ion batteries[127,128] based on various origami and kirigami schemes. These devices show stable and reliable performance under cycles of large mechanical deformation, suggesting their potential utilization in mobile platforms (Figure 7c). A recently emerged class of triboelectric nanogenerators also benefit from origami and kirigami concepts (Figure 7d). Starting from paper-based composites, various device embodiments, including slinky- and doodlebug-shaped[129] or interlocking kirigami[130] structures, can be made for harvesting ambient mechanical energies. Their high deformability couples with the stretching, pressing and twisting motions that are frequently encountered in daily life, providing promising energy solutions for self-powered wearable systems.

CONCLUDING REMARKS

The science and engineering of OKNs enable 3D-reconfigurable structures with the advantages of universality, predictability, deformability, tunability, and simplicity. The expansive library of available materials and actuation schemes afford devices spanning a wide spectrum of physical dimensions, response times, and functionalities. The systems built therefrom complement—or even revolutionize—existing technologies in an expansive range of applications. Research into origami and kirigami materials is highly interdisciplinary and incorporates inputs from nearly all of the aspects of art, mathematics, science, and engineering.

There are promising opportunities lying ahead. Investigation into the mathematics and physics of origami and kirigami structures will continue to uncover surprising behaviors, including additional degrees of freedom for reconfiguration[131−133] or foldability of unconventional thick plates.[134] Pushing the limits of applicable materials and dimensions will enable molecular-scale origami structures such as those made from DNA molecules.[135,136] Technological development translates simple OKN elements into sophisticated engineering systems for untethered robots[137,138] and other practical applications. Collaborative efforts from all disciplines, including fine arts, mathematics, physics, chemistry, materials science, biomedicine, engineering, and many others, will move this area forward in a way that promotes positive feedback between the disciplines. Current advances suggest continuous growth of origami and kirigami materials research in the future, based on a broad expansion of both fundamental understanding and technological innovation.

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VOCABULARY

origami, a word of Japanese origin, referring to the folding (*ori-*) of papers (*-gami*); it has been generalized to a variety of engineering structures and fabrication techniques inspired by the traditional art of paper-folding; **kirigami**, a variation of origami that involves cutting (*kiri-*) of paper (*-gami*); **reconfigurable structures**, structures that are capable of deterministically changing geometrical configuration upon appropriate stimuli; **composite materials**, materials that involve two or more constituents to provide properties usually unachievable with any single constituent alone; **layer-by-layer assembly (LbL/LBL)**, a thin-film fabrication technique that involve repeated and alternating adsorption of two or more types of nanoscale components on a given substrate

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