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Digital strategies for structured and architected materials design

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Silvia Bonfanti, Roberto Guerra, Michael Zaiser, and Stefano Zapperi

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Silvia Bonfanti,[1] Roberto Guerra,[1] Michael Zaiser,[2,a)] and Stefano Zapperi[1,3,b)]

AFFILIATIONS

[1]Center for Complexity and Biosystems, Department of Physics, University of Milan, Via Celoria 16, 20133 Milano, Italy
[2]Institute of Materials Simulation, Department of Materials Science, Friedrich-Alexander-Universität Erlangen-Nürnberg, Dr.-Mack-Str. 77, 90762 Fürth, Germany
[3]CNR—Consiglio Nazionale delle Ricerche, Istituto di Chimica della Materia Condensata e di Tecnologie per l'Energia, Via R. Cozzi 53, 20125 Milano, Italy

[a)]**Electronic mail:** zaiser@fau.de
[b)]**Author to whom correspondence should be addressed:** stefano.zapperi@unimi.it

ABSTRACT

Designing materials with tailored structural or functional properties is a fundamental goal of materials science and engineering. A vast research activity is currently devoted to achieving metamaterials with superior properties and optimized functionalities by carefully fine tuning both the microstructure and geometry of the material. Here, we discuss the impact of digital technologies in this research field by providing fast and cost effective tools to explore a large array of possibilities for materials and metamaterials. We report on recent progress obtained by combining numerical simulations, optimization techniques, artificial intelligence, and additive manufacturing methods and highlight promising research lines. The exploration of the space of possible material microstructures and geometries is reminiscent of the process of biological evolution in which traits are explored and selected according to their fitness. Biomimetic materials have long profited from adapting features of biological systems to the design of new materials and structures. Combining biomimetic approaches with digital simulation and optimization and with high throughput fabrication and characterization techniques may provide a step change in the evolutionary development of new materials.

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I. INTRODUCTION

Discovering new materials with improved structural and functional properties has been a fundamental quest throughout centuries and millennia. The history of humankind is often characterized by new material discoveries that define entire epochs. Consider, for instance, the bronze and iron ages or, more recently, the discovery of silicon based electronics. From the traditional trial and error discovery processes used in the past, the process of materials discovery is becoming more and more systematic, thanks to large scale combinatorial approaches[1] based on high-throughput (HT) materials screening and high-speed chemical synthesis. These methods allow one to explore a wide spectrum of new materials and to overcome the limitations of the traditional path to materials discovery where new materials had to be synthesized and characterized one at a time. The material discovery process can now also rely on computational approaches, allowing for rapid calculation of electronic, mechanical, or thermodynamic properties of possible materials that can be stored into databases for further search and analysis.[2,3]

While the optimization of the chemical composition plays an important role in new materials design, both structural and functional properties of most materials strongly depend on their *microstructure* well above the atomic scale. On this level, materials properties can be optimized and new functionalities can be acquired by targeted design of "architectured" microstructures. Biological materials such as plant and animal tissues have long exploited this possibility by deriving, from a comparatively limited set of chemical building blocks, a fascinating diversity of structural and mechanical functionalities through the design of highly complex structures on the mesoscale and macroscale.

There is therefore an increasing interest in studying materials that are structured on the meso/macroscale and using

the resulting insights for the targeted design of architectured materials microstructures. One aspect of this approach involves transposing knowledge gained from the study of plant and animal tissues to design new materials microstructures and metamaterials, a class of artificial architected materials engineered to have exceptional properties (such as high elasticity, adhesion, or fracture toughness) and responses from their artificially designed internal geometry.

Metamaterials can display properties that are difficult to find in conventional materials and are typically designed considering an internal structure composed of multiple sub-elements, or cells, arranged in repeated patterns. Since these objects allow each cell to be designed in a different way, the resulting structure can display many degrees of freedom, giving rise to a variety of unusual physical properties, for instance, those described by negative constants, moduli, or indices.

The possibility to design architectured materials with tailored mechanical properties and functionalities has therefore huge implications in many research fields such as materials science or engineering, resulting in extraordinary diverse applications and societal gains.

Inspired by nature, such as honeycomb (wood, cork) and foams, materials with increasingly complex and innovative internal architectures are gradually emerging. More recent examples of structured materials include hollow materials for thermal insulation,[4] heterogeneous-structured metals showing extra strengthening and work hardening,[5] polymeric structured materials for improved strain characterization,[6] anisotropic meso-structured materials for soft robotics,[7] nanostructured ceramics with high crack-growth resistance,[8] and bimodal harmonic structured materials formed by a combination of ultra-fine and coarse grains, allowing both high strength and ductility,[9] among countless others.

The increased focus on metamaterials is partly stimulated by the recent advances in manufacturing technologies such as 3D printing and automated assembly, which enable an easier manufacture of such material structures, remove many constraints in scale and geometry imposed by conventional manufacturing processes, and allow for manufacturing of one-off structures at low cost.

The design of materials microstructure and metamaterials is ideally suited for digital techniques that can rapidly and efficiently explore a multitude of possible geometrical and structural arrangements and test them numerically. In this Perspective, we discuss recent advances and promising avenues in this direction. In Sec. II, we consider, on the one hand, the design and optimization of mechanical metamaterials (MMMs), where the emphasis is on the design and optimization of mechanical functionality through the manipulation of *geometry*. Such materials consist of a single basic material, which is manufactured into cellular assemblies that may exhibit a high degree of geometrical complexity, usually using additive manufacturing (AM) routes. On the other hand, in Sec. III, we look at the combinatorial design of *microstructures*, which combine material elements of different properties and, in general, of varying chemical composition in order to optimize properties of the three-dimensional microstructural assembly of these elements. Finally, in Sec. IV, we discuss how these different pathways to materials design may be combined in the context of bio-inspired materials.

In the Conclusions, we present some of the most important outstanding challenges in the design and practical realization of architectured materials with extreme mechanical properties and discuss the fundamental role of the digital strategies in their transposition into tailored functionalities.

II. OPTIMIZATION OF MATERIALS ARCHITECTURES: METAMATERIALS

Metamaterials are a new class of materials characterized by an internal architecture that results in properties that far exceed those of conventional materials.[10] *Mechanical* metamaterials (MMMs),[11–16] to distinguish them from other metamaterials such as electrical, optical, and acoustic metamaterials, represent a recently developed subclass of metamaterials, engineered to generate a tailored mechanical response.[17–20] Metamaterials geometries are usually produced through digital manufacturing techniques, including 3D printing and laser cutting.

Structures can be designed to exhibit a wide range of remarkable properties, such as high strength to weight ratios,[21,22] auxeticity,[23–26] negative elastic constants[27] or indices,[28,29] energy trapping,[13,30] and fracture resistance,[31,32] among many others.[18] Artificial metamaterials are composed of building blocks, which are typically arranged as unit cells so as to form repeating patterns that extend throughout the material.[14,33] The response of the material originates from the mutual interplay among the building blocks. Along these lines, recent work demonstrated the possibility to introduce an increasing degree of disorder in the metamaterial without losing effectiveness.[34–38] Remarkably, in some cases, disordered MMMs can show a superior performance, for instance, in terms of protection from defects.[39]

A. Human designed mechanical metamaterials

The traditional design of MMMs relies on heuristic methodologies or intuitive rules derived by trial-and-error approaches. Design principles of the unit cells mainly revolve around theoretical studies of slender (flexible) elements, which can describe instabilities and deformations ranging from the simpler elastic up to the fully inelastic regimes.

1. Linear elastic regime

MMMs that exhibit mechanical response in the elastic regime, where the relation between the stress and strain is linear, generally give rise to two types of mechanisms: stretching or bending.[38,52] Which of these two regimes a lattice falls into depends on the network geometry and in particular on its connectivity. When a lattice's connectivity is less than that required for rigidity in its rigid-link freely hinged analogous system, the lattice will exhibit a bending dominated behavior; otherwise, a stretching dominated behavior will be observed.[40] Typical examples of such unit cells are reported in Figs. 1(a) and 1(b).

The two classes of lattice exhibit dissimilar mechanical properties. For example, the relative stiffness of a two-dimensional lattice ($\tilde{Y}/Y$) is related to its relative density ($\tilde{\rho}/\rho$) via the expression $\frac{\tilde{Y}}{Y} \sim \left(\frac{\tilde{\rho}}{\rho}\right)^n$, where Y and ρ are Young's modulus and the density of the base material, respectively, and $\tilde{X}$ denotes the property X of the MMM. In two dimensions, $n = 1$ or 3 for stretching-dominated

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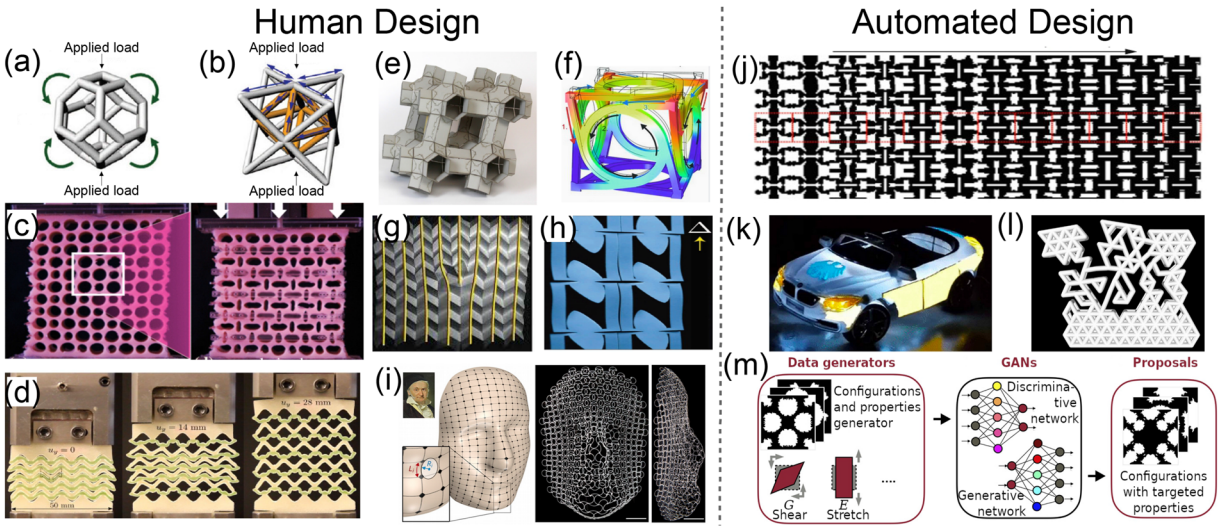

FIG. 1. Human design MMMs (left). Unit cells of MMM that (a) bend, buckle, and eventually break under load or (b) stretch or compress on reaching higher strength. Figures adapted from Ref. 40. MMM that (c) buckles[41] or (d) snaps when stretched.[42] Mechanism-based metamaterials that (e) reconfigure,[43] (f) twist,[44] (g) fold (origami),[45] (h) change shape (kirigami),[46] or (i) shape-morph.[47] Automated design MMMs (right). (j) MMMs with graded Young's modulus from 0.03 unit to 0.18 unit and constant Poisson's ratio automatically generated using topological optimization (TO) algorithms.[48] MMM actuators that (k) wrap into desired surfaces, specifically an electric toy vehicle,[49] or (l) locally respond to a prescribed input designed with generative algorithms.[50] (m) MMMs created via the ML technique (GAN) after training with TO-generated architectures. Figure adapted from Ref. 51.

and bending-dominated architectures, respectively.[53] Due to this scaling behavior, stretching-dominated lattices of low relative density are stiffer than their bending-dominated counterparts. These scaling relations are useful in the search for lightweight structures with high stiffness or fracture resistance. The structure–property relations for architectures of MMM unit cells with stretching- and bending-dominated regimes are reported (also via diagrams) and discussed in Refs. 54–58. The connectivity argument has been enriched also by theoretical studies of disordered networks that mimic the geometry of jammed particles.[59–61]

Besides auxetic metamaterials that have the unique property to contract under compression and expand under tension, another exciting class of mechanical metamaterials is that of pentamode materials, also known as meta-fluids. They have the striking feature to resist isotropic compression but very easily deform under shear behaving ideally like a fluid. Pentamode materials have been theoretically proposed in 1995[62] but could only be realized in 2012[63] due to the inability to fabricate complicated 3D structures before the advent of 3D printing.[64] Such peculiar structured materials have been used to fabricate "unfeelability" cloaks used to hide objects.[65]

2. Nonlinear metamaterials

Inelastic or nonlinear MMMs present strong geometrical deformations that lead to nonlinear stress–strain relations even for small applied forces. Slender elements in this regime show mainly two different types of mechanical responses: buckling[66] and snapping.[67,68] Notorious examples of MMMs that undergo buckling are characterized by porous lattice structures (matrix with circular voids), such as those reported in Refs. 28 and 69–72. Under uniaxial compression, the beams undergo buckling triggering a sudden switch of the initial circular holes into ellipses, up to their eventual closure. When the load is removed, the initial shape is then recovered [Fig. 1(c)]. Snapping (or bistable) MMMs instead can achieve stable geometrical transformations, snapping the beams between two configurations, maintaining the deformation after the load is removed[42,73] [Fig. 1(d)].

3. Mechanism-based metamaterials

Active, adaptive, and programmable MMMs[74] represent an emerging field in MMMs: Through a prudent choice of the mechanical instabilities and their combinations, MMMs can be tuned to achieve programmed structural deformation with tailored responses. Response to uniaxial compression can be programmed by lateral confinement, allowing monotonic, nonmonotonic, and hysteretic behaviors.[41,75] Shape-changing, macroscale MMMs that undergo a broad range of qualitatively different deformations [Fig. 1(e)] or self-guided, multi-step reconfiguration in response to global uniform compression have been also designed by combining nonlinear mechanical elements into a multimodal architecture that enables a sequence of topological reconfigurations caused by the formation of internal self-contacts between the elements of the MMM.[76]

Other remarkable designed smart metamaterials with successful applications are *chiral* MMMs[77] that exploit the rotation of the hinges and bending of beams or that twist when pushed[44] [Fig. 1(f)] and *crumpling* MMMs.[39]

In the mechanism-based MMM category, we can include also *origami* and *kirigami*, which use folds and cuts, respectively, to program shape changes.[45,78–81] The Miura origami [reported with an addition of pattern defects in Fig. 1(g)] is one of the most well-known examples of origami configurations and has been employed in different engineering applications, e.g., in aerospace engineering

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for satellite solar panel deployment. Origami-based MMMs[82,83] and kirigami-based MMMs[46] provide an intriguing source for the design of mechanical metamaterials [Fig. 1(h)].

Challenging so-called "4D" printed lattice shapes that encode complex and curved structures have been recently designed through the combination of multimaterial and heterogeneous unit cells, having a huge potential for the future of architectured materials. A striking example is a flat layer that is programmed to transform after stimuli into a 3D human face, precisely that one of nineteenth century mathematician Gauss[47] [Fig. 1(i)].

B. Automated design of metamaterials

An alternative strategy for the generation of MMMs is what we refer to as "automated design," as it uses robust computational algorithms that produce a rich portfolio of complex architectures, extremely difficult to realize with heuristic-driven approaches. Such kinds of techniques allow the development of high performance MMMs with tailored properties and functionalities, especially relevant for emergent fields such as soft robotics that requires localized responses to mechanical actuation. In the following, we report two major classes of automated design of MMMs: topology optimization and generative design.

1. Topological optimization design

Topology optimization (TO) is a well-known numerical method to design periodic lattice structures.[84] This technique relies on the structural optimization of the material layout within a prescribed design space for a given set of loads and constraints,[85,86] maximizing weighted objective functions with an iterative protocol. This is done by either working on the whole volume or focusing on the structure of unit cells that are then assembled inside the volume in an optimal way. Most current applications of TO use lattice structures and the finite element method (FEM) to evaluate the design performance. The method is also exploited in additive manufacturing (AM) with the purpose of realizing stiff but lightweight structures, reducing the waste of material. Many approaches to optimize lattice structures have been proposed, as explained in a comprehensive review;[48] however, the search of strategies to design complex MMMs remains an active research field.[87–90] An example of solution obtained with topological optimization (TO) algorithms requiring a metamaterial with graded Young's modulus is shown in Fig. 1(j).

Conventional TO optimization uses either gradient-based mathematical techniques, such as the optimality criteria algorithm and the method of moving asymptotes, or non-gradient-based algorithms, such as genetic algorithms.[91] The main limitation of this approach resides in the difficulty of optimizing the mechanical response of large and complex structures since most of the methods used tend to find local optimal solutions rather than global optima. The majority of reported MMMs generated with TO consist of repetitive arrays of regular unit cells where the focus is on the global mechanical response to external stimuli, hindering the development of metamaterial actuators that require a localized feedback.

2. Generative design

Generative design is considered as the next-generation method for building MMMs, overcoming the limitations of passive manual design. The underlying idea is to use computational algorithms to automatically generate complex geometries from scratch meeting custom requirements and constraints. The search for the optimal output response to an applied input generally occurs through iterative modifications of the structure. Generative design allows the exploration of innovative material architectures by growing some parts of the material rather than removing them, as done for TO methods.

Generative methods based on optimization algorithms have been employed to design allosteric materials,[92–94] fiber-reinforced actuators,[95] programmable shape-shifters[96] and kirigami patterns,[81] and buckling-driven soft cellular mechanisms[97] and to choose the optimal cell geometry in periodic MMM lattices.[98,99]

The reverse engineering computational method for conformal wrapping with origami has just recently emerged[49] [Fig. 1(k)], as well as optimized MMM actuators with efficiencies far exceeding those of human-designed counterparts[50] [Fig. 1(l)]. These generative methods can explore the topological space of the MMM in a fully flexible way so as to maximize the desired mechanical response in response to any kind of input stimulus. The optimization process is efficiently realized, coupling the optimization algorithm with discrete element simulations or with a suitably trained deep neural network.

3. Machine learning design

Recent design strategies of MMMs employ the training of machine learning methods such as deep learning with the aim of eliminating or at least reducing the need of lengthy mechanical simulations so as to speed up the design process, improving scale-up capabilities.[50,100] This field of design requires a vast amount of data to train the network for pattern recognition, for instance, to identify regions prone to buckling within the sample. The ultimate challenge is represented by machine learning methods that are able to determine, at once, the functional features of a possible structure.[50,101] Outstanding designs of MMMs with extreme properties without prior knowledge via generative adversarial networks (GAN) trained with millions of randomly TO generated architectures have just emerged[51] [Fig. 1(m)] and are definitely overcoming the designer-driven approach and thus opening the way for unlimited applications for architectured materials in the fields of aerospace, automotive, and robotics.

III. COMBINATORIAL DESIGN OF MATERIALS WITH COMPOSITE MICROSTRUCTURES

Mechanical metamaterials, as discussed in Sec. II, consist of a matrix material with constant chemical composition and mechanical properties: Their functionality derives, in essence, from geometry. An alternative viewpoint on the same class of materials is to consider them as two-phase composites consisting of the matrix material and interspersed empty space. As such, they represent an extreme case of a more general composite paradigm according to which the combination of materials with different, possibly vastly different, properties may result in materials with properties that are superior to either of the components. A quite generic problem in the design of high-performance materials is that they need to simultaneously match conflicting demands. The digital design of "architectured" microstructures can help us to meet these demands and overcome trade-offs encountered with conventional materials. We illustrate this for two important problems taken from quite different realms

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of materials science: first, the trade-off between the permittivity and breakdown strength in dielectric materials and, second, the trade-off between the strength and ductility in high-performance structural materials.

A. Dielectric composites: Reconciling permittivity and breakdown strength

The trade-off between the permittivity (dielectric constant) and breakdown strength in dielectric materials can be easily understood even on the molecular level: In solid materials with high bandgap, and thus strongly insulating properties, electrons are tightly bound to atoms and an external electric field is unlikely to induce substantial dielectric polarization [Fig. 2(a)]. For polymeric dielectric materials, machine learning approaches can be used to push the limits of the permittivity vs breakdown strength trade-off on the molecular level. For instance, Mannodi-Kanakkithodi *et al.*[102] considered polymers with seven possible chemical building blocks (CH_2, NH, CO, C_6H_4, C_4H_2S, CS, and O) in a repeat unit of n blocks. Based on density functional theory (DFT) calculations serving as ground truth, they then train a Kernel–Ridge–Regression algorithm in order to simultaneously predict the bandgap and permittivity (dielectric constant) in a computationally efficient manner. With this, they then drive a Monte Carlo multi-objective optimization algorithm to simultaneously optimize the bandgap and permittivity.

Composite design may offer alternative pathways to combinatorial molecular design for simultaneously achieving high permittivity and acceptable breakdown strength. In dielectric composites, typically, a polymer with high breakdown strength serves as a matrix material into which conductive nanoparticles are embedded in order to enhance the permittivity. Figure 2(b) shows the permittivity and breakdown strength (relative to the pure matrix material) as a function of conductor volume fraction.[103] The increase in permittivity obtained by increasing the volume fraction of the conductive filler is concomitant with a decrease in breakdown strength triggered by local electric field concentrations [see the electric field calculations in Fig. 2(b)]. In order to resolve this conundrum, two strategies can be used—both of which can profit from machine learning approaches.

To account for the complex multi-field processes in dielectric breakdown, which include not only electrical and thermal effects but may also be affected by mechanical deformations, Shen *et al.*[104] developed a phase field model, which couples electrical, thermal, and elastic fields [Fig. 2(c)]. They used this model in high-throughput simulations in order to explore the effects of filler conductivity, dielectric constant, and elastic modulus on the breakdown behavior of dielectric composites with a P(VDF-HFP) polymer matrix and different types of nanofillers. The simulation results then serve as training data for a machine learning approach (regression analysis) to determine a semi-analytical relationship between filler properties

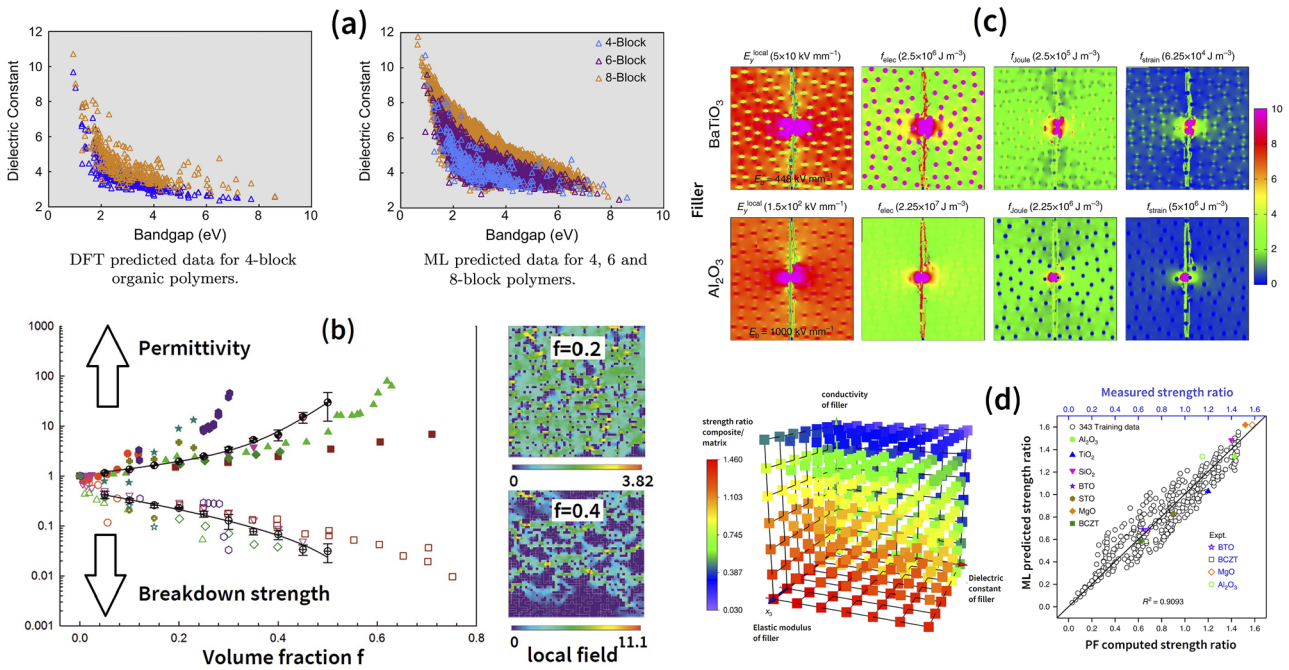

FIG. 2. Dielectric constant vs dielectric breakdown strength: (a) trade-off between the dielectric constant and bandgap (left: DFT data; right: machine learning results to achieve improved trade-off), after Mannodi-Kanakkithodi *et al.*;[102] (b) trade-off between the dielectric constant and breakdown strength in dielectric polymer-matrix composites with conducting fillers; property vs filler volume fraction f, normalized with respect to matrix data; full symbols: dielectric constant, open symbols: breakdown voltage, and color graphs: local electric field distribution for $f = 0.2$ and $f = 0.4$, after Roscow, Bowen, and Almond;[103] (c) phase-field (PF) simulations of dielectric breakdown of the polymer dielectric P(VDF-HFP) with two types of nanofillers, showing from left to right the electric field, electric energy density, heat density, and strain energy density at breakdown, after Shen *et al.*;[104] (d) high-throughput PF simulations exploring the space of nanofiller properties and comparison with machine learning predictions, after Shen *et al.*[104]

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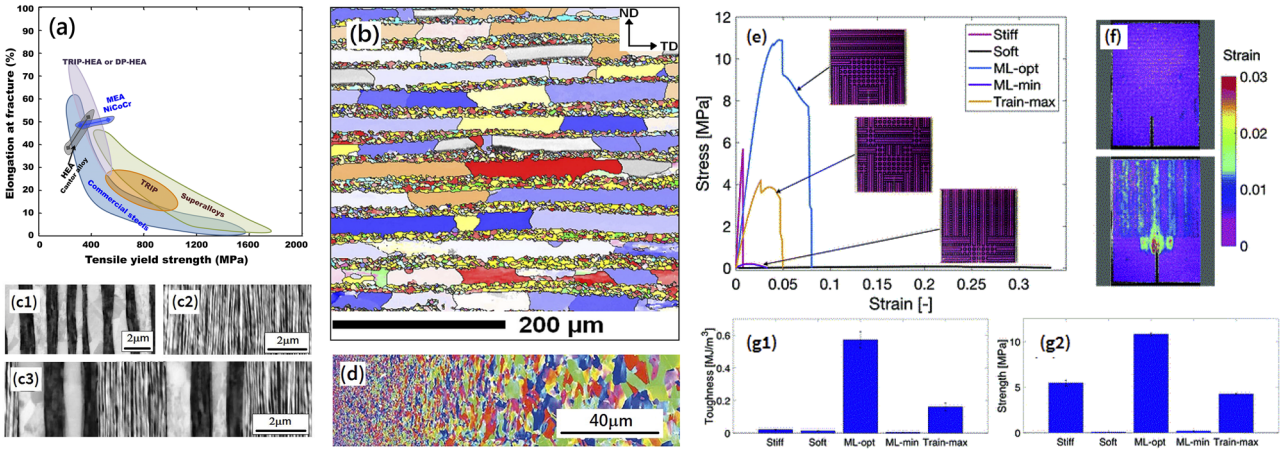

FIG. 3. (Left) The strength vs ductility problem: (a) strength–ductility trade-off in different classes of metallic materials, after Wu *et al.*;[107] (b) homogeneous nanolaminated Al composite, alternating layers of nanosized and micrometer-sized grains, after Chekhonin *et al.*;[108] and (c) heterogeneous FeCu nanolaminates produced via accumulative roll bonding with 14 passes (c1) and 10 passes (c2) (Göken, unpublished data). An architectured nanolaminate with a hierarchical combination of both structures would look as in panel (c3). Notice that the image was assembled from images (c1) and (c2) just for illustrative purposes. (d) Grain microstructure of gradient nanograined Cu, after Long *et al.*[109] (Right) Machine learning approach to enhance strength + ductility in a 2D architectured polymer composite, all graphs after Gu *et al.*;[110] (e) stress–strain curves for constituent materials (stiff/soft), the best performing sample in a training set with randomly generated microstructures (train-max), and the best performing sample generated by the trained algorithm (ML-opt); (f) strain patterns in the best and worst performing ML generated sample; and (g1) strength and (g2) toughness values of the different samples.

and composite performance. Thus, the approach of Shen *et al.*[104] aims at identifying optimal fillers in a two-phase composite.

Beyond chemical composition, one may look at effects of the shape and geometrical arrangement of filler particles. The problem of dielectric breakdown driven by electric field concentrations has close analogies with the mechanical problem of crack propagation by stress concentrations. Accordingly, computational optimization methods such as shape optimization, which were used, e.g., by Prechtel *et al.*,[105] to adjust the shape of inclusions in a composite such as to optimize crack resistance, can be used for the same purpose in order to enhance the dielectric breakdown strength. Beyond single inclusions, one may seek to optimize the spatial arrangement of fillers such as to mitigate the effect of electric field concentrations, e.g., through multi-scale hierarchical arrangements, as considered in relation to fracture by Moretti *et al.*[106] We discuss such architectured microstructures and the use of machine learning in their design in relation to the use of architectured microstructures for overcoming a dilemma that has many similarities to the dichotomy between the permittivity and breakdown strength, namely, the strength–ductility trade-off in structural materials.

B. Architectured microstructures: Overcoming the strength–ductility trade-off

The development of metallic materials has been struggling forever with an intrinsic dichotomy: In order to achieve the same structural performance at reduced volume and weight, the strength of metals should be enhanced, which implies changes in microstructures such as to reduce their ability to plastically deform at low stresses. However, plastic deformability is essential for a material's toughness, i.e., its ability to dissipate energy at crack tips and thus hinder crack propagation. Hence, there exists an intrinsic trade-off between the conflicting aims of enhancing strength and hardness on the one hand and ensuring toughness and deformability on the other hand. High-strength materials are likely to be brittle [Fig. 3(a)].

1. Tailoring microstructures to improve strength and ductility

The strength–ductility dilemma is as old as metallurgy, and so are some fundamental approaches to resolve it. First, one may use composites, where materials of different strength and ductility are combined in the hope of synergetically improving the strength–ductility ratio beyond a mere mixture of the component properties. This idea has, in recent years, received a boost through the use of nanocomposites where the size of the interspersed components is reduced to the nanometer scale. This leads to a significant fraction of extended interface regions whose properties differ from those of the bulk component materials ("interphases"). For instance, when carbon nanoparticles such as carbon nanotubes are introduced into melt-processed aluminum, a variety of synergetic mechanisms are triggered that may simultaneously enhance the strength and ductility[111] beyond the rule-of-mixtures according to which strength is increased because the ultra-strong nanotubes carry part of the load according to their volume fraction. First, well dispersed nanotubes may act as nuclei for crystallization, leading the Al matrix to develop a nanocrystalline grain structure of high intrinsic strength.[111] Second, appropriately surface processed nanotubes may not only act as crack bridges, enhancing strength, but also as toughening agents whose pull-out requires large plastic deformation and concomitant energy dissipation within an extended interface region surrounding the nanotube.[111] Third, the presence of the CNTs can modify plasticity in the Al matrix and induce an Orowan mechanism of dislocation motion,[112] which not only requires high stresses (enhancing

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strength) but also provides a significant amount of internal energy storage (enhancing toughness).

On larger scales, the interphase idea can be equally exploited. Thus, nanolaminated composites have been synthesized via a repeated stretching-and-folding process denoted as accumulative roll bonding (ARB).[113] In this process, stacked metal sheets are cold welded under volume reduction by cold rolling and then folded to repeat the process, resulting in a layered microstructure. Depending on the number of rolling–folding cycles such structures consisting of sheet metals of different purity (hence different propensity to recrystallization) may develop into nanolaminates where layers of nanoscale grain size (high strength, low ductility) alternate with layers of much larger grain size (low strength, high ductility), as shown in Fig. 3(b). The idea here is that plastic deformation in the large-grain-size layers is confined by the interlayer boundaries, leading to extended dislocation pile-ups and back stresses in extended "interface affected zones"[114] where plastic deformation requires increased stresses, while the dislocation pile-ups also provide an additional energy storage and enhanced hardening mechanism. At the same time, activation of additional slip systems in the large-grain-size layers provides an energy dissipation mechanism that may prevent cracks, which nucleate in the brittle nanocrystalline layers, from spreading across the microstructure. A similar mechanism can be exploited if one creates nanolaminates of chemically dissimilar materials and different strength and ductility,[115] as illustrated here for a Cu–FeSn nanolaminate in Fig. 3(c). By varying the number of ARB passes and the chemical composition of the sheets laminated together and by targeted heat treatments between ARB cycles, it is possible not only to control the layer width in the laminates [Figs. 3(c1) and 3(c2)] and the layer composition but also to simultaneously harness a wide variety of intra-layer microstructures that lend itself to combinatorial optimization [Fig. 3(c3)]. However, an evident restriction imposed by the manufacturing method is its restriction to sheet metals and layered microstructures.

A second method to address the strength–ductility problem consists in fabricating materials with spatially differentiated ("graded") properties. In toolmaking as in weaponry, ultra-high strength and hardness are required only at the cutting edge of the tool or the weapon, which may then be supported by a much softer but tough body that mitigates against brittle fracture. It is thus natural to engineer the microstructure of tools or weapons such as to optimize hardness at the cutting edge, while optimizing toughness in the supporting body of the tool or weapon. Another variant of such graded microstructures has long been used to mitigate against fatigue crack propagation: The nucleation of fatigue cracks occurs at surfaces and depends on the local loading direction and grain orientation; their early stage growth is hindered by grain boundaries. Fatigue failure can therefore be significantly delayed by surface treatments such as laser shock or severe deformation of a surface layer, which introduce ultrafine or nanograined structures within a layer beneath the surface (see, e.g., the work of Nalla *et al.*[116]).

A key feature that distinguishes graded microstructures from bulk composite microstructures resides in the possibility of spatially differentiating and locally matching materials properties and engineering demands: While microstructure design for bulk composites aims at improving the strength–ductility ratio *as such*, graded microstructures may rather aim at improving the strength and ductility (or resilience against fatigue crack propagation, or other properties) *where it matters*. In order to fully exploit the potential of such materials, discipline boundaries between materials science, process engineering, and mechanical engineering design must be overcome: Ideally speaking, the shape and external environment of a component determine local requirements of materials, which are met by spatially differentiated local properties deriving from differentiated local process conditions.

It is thus obvious that the potential of architectured microstructures can only be harnessed if highly flexible processing tools are available, which allow for detailed three-dimensional control of the chemical and physical microstructures. Conventional bulk processing routes normally fall far short of this requirement. This can be seen from the above illustrated methods for creating laminated or graded microstructures: ARB allows for heterogeneity of chemical composition, layer thickness, and grain size, but the method is restricted to sheet metals and heterogeneity can only be imposed across the sheet thickness; surface treatments can reduce the grain size underneath the surface but are restricted to near surface regions, while general three-dimensional microstructure architectures are not accessible. In these respects, additive manufacturing may prove to be a key technology that allows for the first time the design and fabrication of fairly generic, three dimensional metal microstructures.

2. Additive manufacturing: Toward 3D printing of architectured microstructures

Additive manufacturing of metals proceeds through a layer-by-layer build up process where metal powders are deposited and then processed by laser or electron beam melting (LBM/EBM) or simultaneously deposited and melt processed in direct laser deposition (DLD) processes.[117] Using these techniques, cellular structures with tailored geometry can be manufactured for a wide range of applications from acoustic metamaterials[118] over biomaterials[119] to process specific tailored catalysts.[120] Recent work extends the spatial resolution of additive manufacturing methods for metals to the nanometer range.[121]

Beyond fabrication of 3D cellular structures with specifically designed geometry, additive manufacturing of metals also provides new pathways for processing of bulk metals with tailored microstructures. For certain high-performance alloys such as γ-TiAl alloys, which are difficult to process using conventional processing routes, AM via selective electron beam melting may provide a viable processing path paving the way for their application as replacement for Ni-base superalloys in high-strength high-temperature applications.[122]

AM processes on metals provide multiple pathways toward fabrication of tailored 3D microstructures. First, the metal powder composition can be spatially graded to ensure a continuous and controlled grading of the chemical composition of manufactured parts. At the same time, process parameters of EBM and LBM processes control important microstructural parameters. On a most basic level, the layer-by-layer nature of the build-up process implies that layers that were built earlier are exposed to an environment of elevated temperature for longer times than later-built layers, an effect that led to a gradient in γ' precipitate size during AM of Ni-base superalloys.[123] On the level of the local melting-solidification process that occurs once laser or electron beams are scanned

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across the powder bed, parameters such as beam power, beam deflection speed, and beam spot width define local heating-cooling protocols that control essential parameters of the microstructure such as grain size, grain texture, and dendritic microstructure pattern.[122] A thorough understanding of the relationships between process parameters and ensuing microstructure is needed in order to map the accessible microstructures and ensure robustness of the process. However, modeling process–microstructure relationships in these AM processes is a formidable task since one is dealing with a complex multi-scale multi-physics problem involving a simultaneous description of energy deposition by the beam, heat conduction, solidification kinetics, and hydrodynamic effects within the melt pool, in conjunction with significant stochastic effects.[124–126]

3. Design and fabrication of 3D microstructures: Data based approaches and machine learning

Data driven approaches can facilitate the process of designing and fabricating designed metal microstructures via AM on two distinct levels. First, data driven approaches may be used to establish process–microstructure relationships and to optimize the manufacturing process itself. Since the process data themselves are complex and performance may be affected by parameters before and after the actual manufacturing process, data management strategies supported by digital twins[127] have been proposed. On a basic level, digital twins of the actual manufacturing process are computationally efficient and validated process models that allow us to predict for a given material system the dependency of essential microstructure parameters on process variables [see Ref. 128 for a LBM process]. On a more ambitious level, digital twin enabled data management strategies may be used in order to identify, collect, and analyze performance specific data throughout the entire life cycle of AM manufactured parts.[129]

Once the crucial problem of relating process variables and ensuing local properties is resolved, automated design strategies similar to those used for metamaterials can be used to design samples with tailored 3D microstructures for manufacturing via AM. In the context of optimizing fracture resilience, several such strategies have been proposed. Gu *et al.*[110] used a convolutional neural network to predict fracture toughness (defined as the work of failure for a sample with given initial crack), assuming a hierarchical composite microstructure consisting of three types of unit cells combining a weak and ductile material with a strong and brittle material. The unit cells themselves were constructed using the domain knowledge from biomimetic materials to mimic basic traits of biological composites. The network was trained on a large number of microstructures consisting of random assembly of the three kinds of unit cells in order to be able to predict the work of failure for a given microstructure. The network output was then used in a simple stochastic optimization algorithm to evolve microstructures with optimal work of failure (see Fig. 3, right), which were produced by 3D printing from two different acrylic photopolymers and tested for validation (Fig. 3, right). This proof-of-concept study demonstrates the feasibility of automated microstructure design strategies for a comparatively simple type of problem. At the same time, it is clear that, in order to explore higher dimensional design variable spaces, more complex optimization strategies may be required.

IV. BIOLOGICALLY INSPIRED MATERIALS AND METAMATERIALS DESIGN

An interesting source of inspiration for the design of novel materials and structure comes from biological systems where the engineer can find ready-made solutions to a variety of problems.[135] Through the course of evolution, nature was able to produce remarkable structures whose design principles can now be exploited for different technological applications.[136] Consider, for instance, the adhesive feet of geckos[137] whose structural features [see Fig. 4(a)] have provided inspiration for micro-fabricated adhesives[130,138] or the self-cleaning surfaces of lotus leaves [see Fig. 4(c)] that found direct application for the design of super-hydrophobic surfaces.[139]

The applications of natural materials span different technological areas, ranging from optics, as in the case of light interference of a beetle exoskeleton,[140] to mechanics, if we consider all the mechanically intriguing structures found in biological organisms. Biological structures have been able to reconcile strength and toughness, two often mutually exclusive properties.[141] Consider, for instance, the exceptional mechanical properties of bones[142] and nacre[132] [see Fig. 4(b)], a composite material produced by some molluscs for their inner shell,[143] or the damage tolerant structure of the dactyl club of the marine crustaceans (stomatopods).[144] For instance, nacre is composed of hexagonal platelets of aragonite separated by sheets of an organic matrix forming a characteristic brick and mortar structure illustrated in Fig. 4(b). This combination of soft and hard constituents makes the material exceptionally tough and resilient. High-resolution scanning/transmission electron microscopy experiments showed that nacre displays nanograin deformation and locking across the organic interface, leading to the joining of the platelets.[145] Locked platelets respond to deformation, but crack propagation is prevented by the organic boundaries.[145]

Nature provides direct inspiration to engineers not only in terms of new possible structures but also in the general design principles. Most biological structures are composed of polymers or polymer-ceramic composites,[146] which in most cases would not represent the standard choice for an engineer. Yet these cheap and simple materials allowed nature to achieve remarkable properties focusing on geometrical and structural properties. It is thus possible to identify broad structural design principles underlying the mechanical properties of many natural materials[147] such as the hierarchical structural organization, which is responsible for the extraordinary mechanical properties of bones,[142] sea sponge ridges [see Fig. 4(d)],[133] or corals.[143] In particular, bones display a hierarchical structure in which collagen molecules are arranged into nanoscale fibrils, which are then assembled together into larger scale arrays. The observed macroscopic toughness then stems from the cooperation of distinct plasticity mechanisms operating at these different scales.[148]

Bioinspired material design strategies follow either a top-down or bottom-up approach. The top-down strategy aims at simply reproducing the micro- or meso-structure of known biomaterials and structures. For example, microelectromechanical sensor systems that mimic the structure of hair cells have been made using photolithography.[135] While this approach has led to successful development of devices, it requires a precise reproduction of the natural structures and could therefore suffer from drawbacks in terms of

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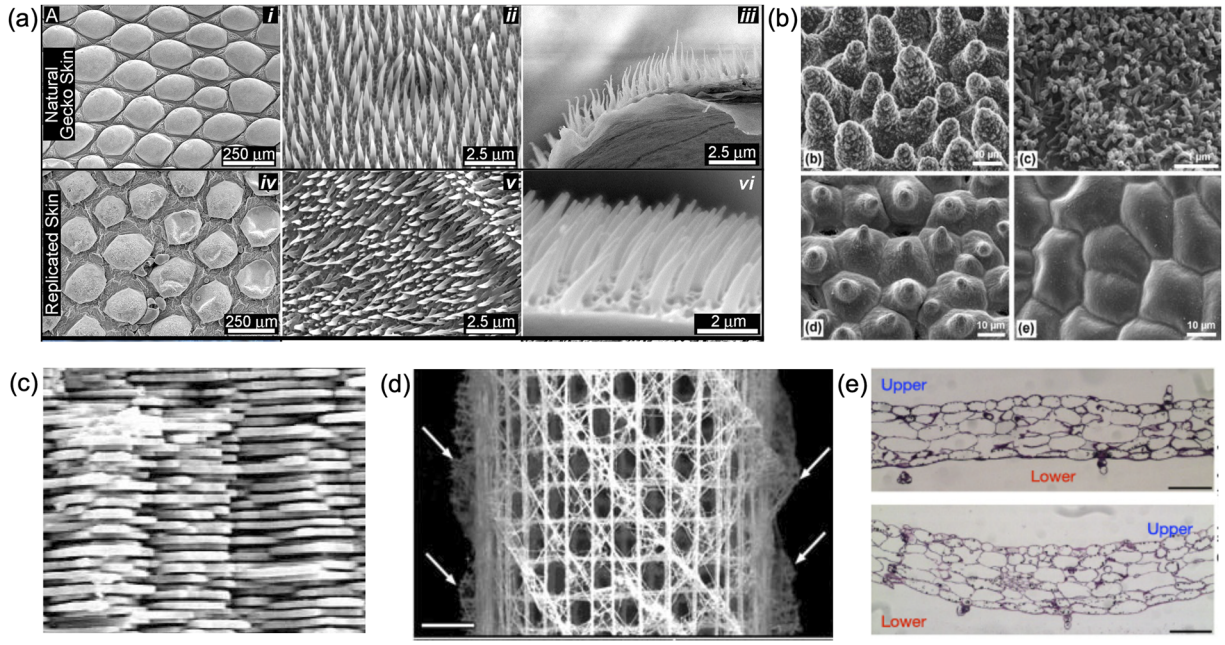

FIG. 4. (a) Scanning electron microscopy (SEM) image of the natural gecko skin with increasing scale (i–iii) and the polystyrene replicate of the same gecko skin (iv–vi).[130] (b) SEM view of the lotus leaf showing the hierarchical structure.[131] (c) SEM image showing the cross section of nacre as the "brick and mortar" structure.[132] (d) Skeletal structure of *Euplectella* showing a square-grid lattice and diagonal elements organized in a chessboard fashion,[133] (e) cellular architecture and growth inducing leaf bending for open (top) and closed (bottom) *Drosera capensis* leaves.[134]

robustness. Unlike typical artificial manufacturing processes focusing on the fabrication of identical components, evolution shapes structures through biologically controlled self-assembly. Hence, the top-down approach overlooks one of the key features of biomaterials in nature.[146]

The alternative to top-down engineering is thus bottom-up self-assembly, which more closely mimics biological growth processes. In the context of tissue engineering, top-down engineering has struggled to recreate the microstructures of biological systems, a drawback that bottom-up self-assembly addresses with success.[149] Beyond the self-assembly of identical passive particles into precise structures, new challenges open up if we take inspiration from the flexible and adaptive approach of living systems. For example, the self-assembly of carefully synthesized block copolymers has been studied for decades, and more recently, considerable attention has been given to the self-assembly of random copolymers, in which the units to be assembled are random structures of monomers. The assembled structures can be controlled by the solvent used, that is, these systems are responsive to their environment.[150] Another exciting possibility is the self-assembly of active particles, such as motile bacteria. For example, in the self-assembly of mixtures that include active bacteria, phase separation is suppressed[151] and a novel behavior such as spontaneous rotation of aggregates is observed.[151,152] Such systems open new avenues of inquiry, the exploration of which will both deepen our understanding of known biomaterials as well as potentially open new possibilities that have not yet been observed in nature.

The peculiar design philosophy of biological evolution means that we can learn from natural structures and materials especially since biological solutions are often quite different. In the context of plants, the distinction between natural and artificial processes is even more remarkable since plants exhibit developmental plasticity[153] so that the overall form and size of genetically identical plants depend on their environmental conditions. As an extreme example, consider *Pinus parviflora* that grows to 15 m–25 m in height in the wild but can also be used for bonsai.[154] An interesting playground to explore the combination of geometry and mechanics to direct motion is provided by carnivorous plants such as *Dionaea muscipula*[155,156] or *Drosera capensis*.[134] The snap through instability used by the leaf of *Dionaea muscipula* has been implemented in soft fluid actuators[157] and in tunable mechanical metamaterials.[158] The closure mechanism of the *Drosera capensis* leaf was shown to arise from the asymmetric cellular structure [see Fig. 4(d)], which is able to convert a symmetric stimulus into a bending motion.[134] This observation inspired the design of a bending mechanical metamaterial with a similar asymmetric geometry.[134]

V. CONCLUSIONS

In this article, we have addressed the general long-standing issue of designing new materials and highlighted the increasingly important role played by digital strategies. The search for materials and structures with improved functional properties has been going on not only throughout all human history but also across natural history as it is illustrated by the remarkable optimized properties found in biological materials. These naturally occurring structures are now routinely used as templates to develop novel artificial bio-inspired materials and metamaterials.

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The advent of powerful computational tools is now reshaping the traditional materials discovery process, which was based on trials and errors, into a more systematic exploration of a vast realm of possibilities for new materials. Algorithms can now be used to predict the most suitable chemical composition, choose the optimal microstructure, and select the most effective geometry needed to achieve a given functional property. Digital strategies can also help us overcome typical hurdles found in materials design, as when we try to maximize two mutually exclusive properties, such as strength and toughness.

We are now facing an era where human imagination, the most important limitation to the design of architected materials, is gradually being replaced by the ever increasing digital strategies assisted by artificial intelligence. In the near future, it is expected that the contribution to machine-designed structured materials will be of major importance to solve efficiently many of the open design challenges and unexplored research directions that we face today. Some of the most promising routes that need further investigation would be the following:

- Explore the role of disorder in the geometry. Architected materials are generally designed with a strict periodicity, e.g., by repeating unit cells' patterns in a regular arrangement. Such well ordered architectures are in contrast with the disordered structures (or structures with defects) present in nature, which are realized by growth processes in response to environmental conditions and are generally observed to provide robustness to modifications due to the geometrical frustration.
- Enable architectured metastructures that morph into multishapes with the goal to achieve multifunctions. The aid of digital strategies in this context is of primary importance to identify novel architectures with innovative properties or hierarchical responses reducing the time to find a suitable solution.
- Unify the categories of different metamaterials to give rise to hybrid metamaterials with remarkable properties, such as electro-mechanical, thermo-mechanical, or even bi-materials mechanical. An outstanding example of such interdisciplinary interaction is that of the recent micrometer *walking* robots that can move, thanks to origami-inspired foldable legs activated by little voltages.[159]
- Miniaturization of architected material properties down to small scales such as graphene-origami machines,[159] opening the way for the use of these materials on a wide range of length scales with important medical implications.

At the same time, the field of AM has also made unprecedented progress. Its advent is considered a breakthrough in the material design process since it made feasible the large-scale production of materials, especially with highly complex internal geometries. Additive manufacturing nowadays plays a central role in multiple markets, which will continue to grow. The employment of additive manufacturing techniques allows us to rapidly transform computer generated models into actual objects that can be tested in the lab.

There are practical constraints that still limit AM and need to overcome: for instance, the presence of defects, such as surface imperfections, that arise in the production process due to the granular structure of the constituents. Technological progress has shown that the vapor deposition process with aluminum can improve the resistance of the overall structure. Besides experimental AM challenges, digital strategies might also play a central role in the AM design process when passing from the computer model to the realistic object. The biggest challenge lies in the ability of 3D printing techniques (or manufacturing, in general) to produce more and more precise (resolution) and diversified structures (combination of different base materials) in order to make the structuring more and more controllable and effective.

Ultimately, we could think about the extreme case to submit to the computer analysis also the problem of exploring the landscape of all the possible applications with the goal of creating completely innovative materials and solutions from scratch. The starting data pool for the learning could be a merge of the data obtained from both natural and human systems, and from those designed by machines themselves, up to what we define here the "meta-machine learning." Recently, some interesting research along this direction is ongoing.[160] The great challenge of the future should focus on further optimizing materials processes and protocols in order to achieve the best results in the most efficient way.

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DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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