Design, material, function, and fabrication of metamaterials

Zadpoor, Amir A.; Mirzaali, Mohammad J.; Valdevit, Lorenzo; Hopkins, Jonathan B.

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Amir A. Zadpoor,[1] Mohammad J. Mirzaali,[1,a)] Lorenzo Valdevit,[2] and Jonathan B. Hopkins[3]

AFFILIATIONS

[1] Department of Biomechanical Engineering, Faculty of Mechanical, Maritime, and Materials Engineering, Delft University of Technology, 2628 CD Delft, Netherlands
[2] University of California, Irvine, California 92697, USA
[3] Mechanical and Aerospace Engineering, University of California, Los Angeles, California 90095, USA

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[a)] **Author to whom correspondence should be addressed:** m.j.mirzaali@tudelft.nl

ABSTRACT

Metamaterials are engineered materials with unusual, unique properties and advanced functionalities that are a direct consequence of their microarchitecture. While initial properties and functionalities were limited to optics and electromagnetism, many novel categories of metamaterials that have applications in many different areas of research and practice, including acoustic, mechanics, biomaterials, and thermal engineering, have appeared in the last decade. This editorial serves as a prelude to the special issue with the same title that presents a number of selected studies in these directions. In particular, we review some of the most important developments in the design and fabrication of metamaterials with an emphasis on the more recent categories. We also suggest some directions for future research.

I. INTRODUCTION

The last decade has witnessed an explosive growth in the breadth and depth of the studies aiming to design, simulate, fabricate, and characterize metamaterials of different kinds. This unprecedented growth has primarily happened at the intersection of three major developments that have reinforced each other and have facilitated the study of metamaterials. First, the design of metamaterials that was initially limited to optical and electromagnetic properties has now expanded to mechanical (both quasi-static and elastodynamic),[1,2,183] acoustic,[3–5] biomedical,[6–10] and thermal[11,12] properties. Second, the additive manufacturing (AM) techniques, which are also referred to as 3D printing techniques, have come of age during the last decade. In particular, it is now possible to fabricate functional materials and structures at different length scales,[13–16] from different materials,[17–21] and with arbitrarily complex distributions of multiple phases with vastly different mechanical and physical properties within one single construct.[19,22–27] Third, the development and widespread availability of computational techniques, including those based on artificial intelligence (AI), as well as readily available computational capacity in the form of cloud computing,[28,29] distributed computing,[30,31] GPU (graphic processing unit) computing,[32,33] parallel computing,[34,35] and TPU (tensor processing unit),[36,37] has enabled improved canvassing of the space of possible designs and more powerful approaches to the rational design of metamaterials.

The current special issue presents a collection of selected articles from various areas of research within the broad spectrum of designer materials that are referred to as "metamaterials." It, therefore, features multiple studies employing elements from all the three above-mentioned trends. In this editorial, we try to focus on the most important recurrent themes not only in the studies published within this special issue but also in the relevant literature, in general. Electromagnetic and optical metamaterials have been extensively reviewed in other (recent) papers. Moreover, the guest editors' expertise and the topic of the many of the articles published in this special issue is non-electromagnetic metamaterials. This editorial will, therefore, focus on highlighting the most important trends seen in the current research into metamaterials that target properties and functionalities beyond optics and electromagnetism. We will particularly focus on mechanical and biomedical metamaterials.

II. DESIGN

When designing metamaterials, the principal design objective is to devise small-scale architectures that give rise to a desired set of large-scale properties. The methods applied for such a design purpose often rely on physical reasoning, analytical models, and computational models and are collectively referred to as "rational design" approaches. In this context, the term "rational" highlights the contrast with "creative" or "artistic" design approaches that rely on one's artistic, creative, and (even) intuitive design capabilities. In their purest form, rational design approaches aim at solving an inverse design problem in which the microarchitectures giving rise to a specific set of physical parameters are sought. However, solving such inverse design problems is often notoriously difficult. The vast majority of studies found in the literature, therefore, start off with a design idea that stems in physical reasoning. Such design ideas are then supported by parametric studies in which "forward" computational models are used to relate the designed microarchitectures to the large-scale properties. Starting off from a specific design idea not only is important for such hybrid approaches but also is required when trying to solve the actual inverse problem associated with any specific objective. That is because the space of all possible microarchitectural designs is too large and complex to be realistically canvassed by any viable computational method available today. It is, therefore, important to start off by limiting the space of possible designs to a specific parametrization of the possible microarchitectures. To be as minimally restrictive as possible, such parametrizations require a masterful application of physical reasoning and an intuitive understanding of the underlying physics (Fig. 1). This somewhat blurs the boundaries between "rational" and "intuitive" designs but is a worthy price to pay given the need to compensate for the inadequacy of computational hardware and software.

Recently, the application of machine learning techniques has enabled two other approaches to the design of metamaterials [Figs. 2(a) and 2(d)]. First, it has become now possible to solve the inverse design problems with the help of deep learning and other AI tools.[38–44] Second, generative models, such as generative adversarial networks (GANs)[45,46] and variational autoencoders (VAEs),[47] can now take over some parts of the rational design process by generating designs that correspond to some given sets of target properties.[48]

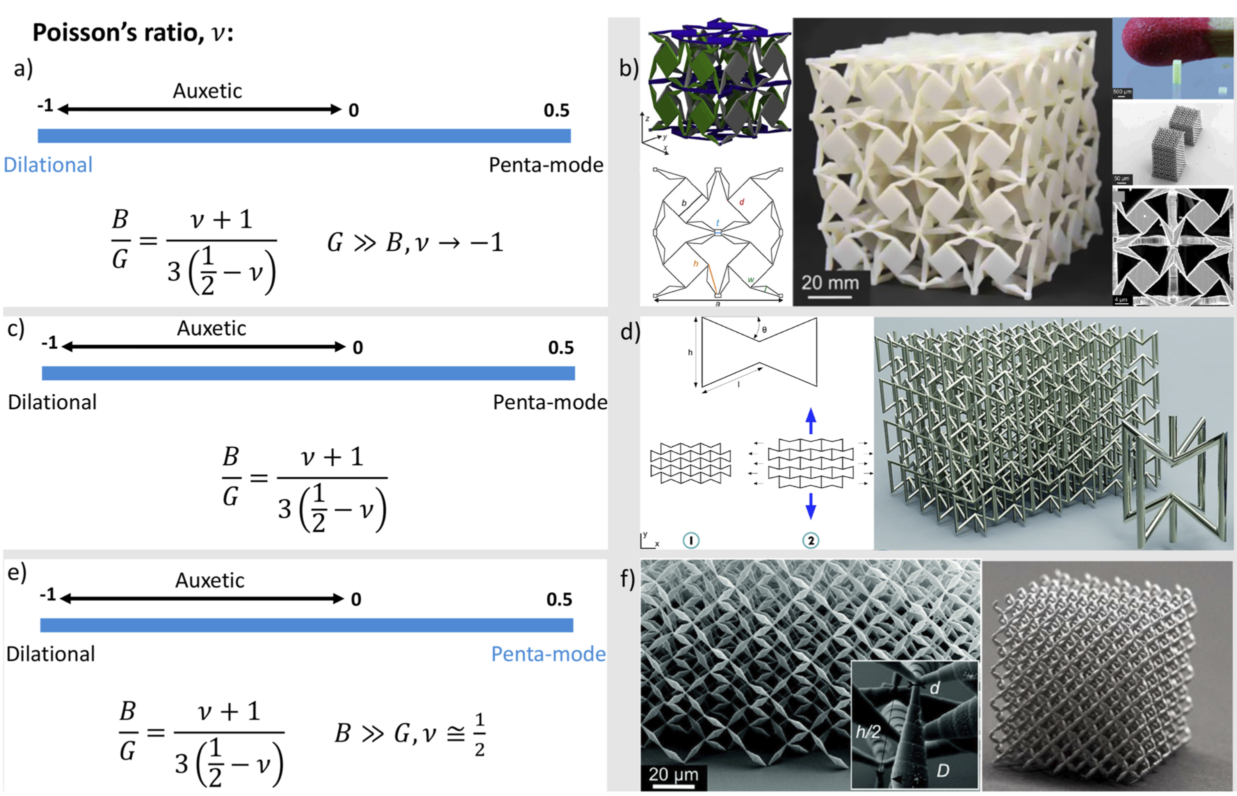

FIG. 1. Mechanical metamaterials can show unusual properties. As an example, three categories of metamaterials with different values of Poisson's ratio, ν, are shown here. This includes dilational behavior with $\nu = -1$ [(a) and (b)], auxetic behavior with $\nu < 0$ [(c) and (d)], and penta-mode properties with $\nu = 0.5$ [(e) and (f)]. Subfigure (b) is reprinted with permission from Bückmann *et al.*, "On three-dimensional dilational elastic metamaterials," New J. Phys. **16**, 033032 (2014). Copyright 2023 IOP Publishing. Subfigure (d) is reprinted with permission from Kolken and Zadpoor, "Auxetic mechanical metamaterials," RSC Adv. **7**, 5111–5129 (2017). Copyright 2017 The Royal Society of Chemistry. Sub-figures (f)-left and (f)-right are, respectively, reprinted with permission from Kadic et al., "On the practicability of pentamode mechanical metamaterials," Appl. Phys. Lett. **100**, 191901 (2012), and Hedayati *et al.*, "Additively manufactured metallic pentamode meta-materials," Appl. Phys. Lett. **110**, 091905 (2017) with the permission of AIP Publishing LLC.

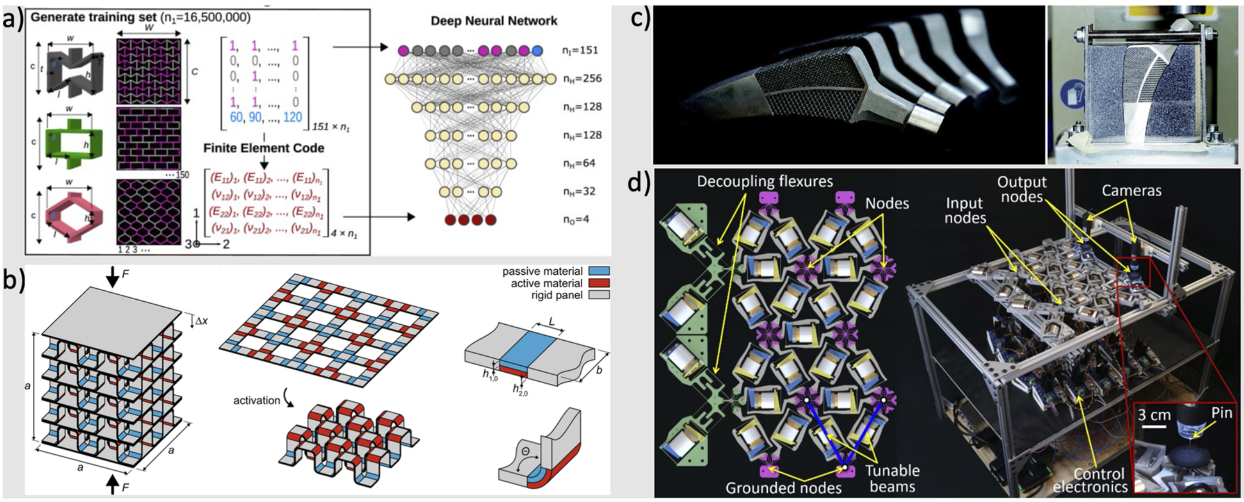

FIG. 2. An example of physics-informed deep learning models (a) that can be used for the rational design of the microarchitectures of mechanical metamaterials.[38] An example of self-folding lattices composed of passive and active materials (b), reprinted with permission from van Manen *et al.*, "Theoretical stiffness limits of 4D printed self-folding metamaterials," Commun. Mater. **3**, 43 (2022). Copyright 2023 Springer Nature Limited. An example of the applications of mechanical metamaterials in biomedical engineering for creating meta-biomaterials (c), reprinted with permission from Kolken *et al.*, "Rationally designed meta-implants: A combination of auxetic and conventional meta-biomaterials," Mater. Horiz. **5**, 28–35 (2018). Copyright 2023 Royal Society of Chemistry. An example of mechanical neural networks (d) that demonstrates the unique features for learning various mechanical behaviors simultaneously. Sub-figure (d) is reprinted with permission from Lee *et al.*, "Mechanical neural networks: Architected materials that learn behaviors," Sci. Rob. **7**, eabq7278 (2022) with the permission of AAAS.

It is important to understand what constitutes a microarchitecture. Partially motivated by the unavailability of free-form multi-material (additive) manufacturing technologies, the first microarchitectural designs of metamaterials were focused on geometry. Even in such single-material constructs, there has usually been a second phase that constitutes the voids often seen in the design of architected materials. In such material–void composites,[49–52] the design problem reduces to that of devising a small-scale geometry that gives rise to the desired properties. Multi-material 3D printing techniques have, however, become increasingly available during the last 5–10 years.[53–55] It is, therefore, possible nowadays to combine arbitrarily complex geometries with an arbitrary spatial distribution of materials with different properties and functionalities. The space of possible designs has, thus, greatly expanded and now includes not only the topology and geometry of the individual repetitive unit cells making up the design but also the exact mechanical and physical properties of each voxel within the construct[19,38,56,57] [Fig. 2(b)]. Computational methods, such as topology optimization, can be used to design the microarchitecture of both single- and multi-material metamaterials.[53,58,59] However, there are multiple challenges that need to be addressed to enable the efficient application of such techniques. For example, it is not always feasible to find differentiable objective functions that can be combined with the available gradient descent-based topology optimization techniques. Future research should, therefore, address the above-mentioned challenges to enable more objective design approaches and the discovery of metamaterial concepts that can hardly be conceived through intuition and physical reasoning alone.

III. MATERIAL

While the properties and functions of metamaterials are, to a large extent, determined by their microarchitecture, the bulk material from which they are made also plays an important role in determining the properties of the metamaterial. In particular, the bulk material properties may define the boundaries of the envelope of (absolute) properties that can be achieved through various microarchitectural designs.

Metamaterials made from various material categories, including metals,[60–64] polymers,[65–69] and ceramics,[51,70–73] have been reported in the literature. As the number and complexity of the materials that can be processed with advanced manufacturing techniques, such as AM, increases, more examples of architected materials with exotic properties appear in the literature. An interesting application of AM techniques to produce metamaterials with exceptional constituent properties is the fabrication of polymeric structures with nanoscale resolution via two-photon polymerization Direct Laser Writing (2pp-DLW) followed by pyrolysis,[51,72,74,75] or ALD coating and polymer removal by plasma etching.[76] The result is an architected ceramic material with local dimensions at the submicron scale. At this scale, the intrinsic cracks are too small to grow by brittle fracture and the material locally approaches its theoretical strength (approximately one tenth of its Young's modulus).[77] These size effects can be combined with near optimally stiff and strong unit-cell architectures to achieve metamaterials with specific strengths higher than diamond.[51]

In some cases, the role of the bulk material properties goes beyond defining the boundaries of what is possible. In fact, some

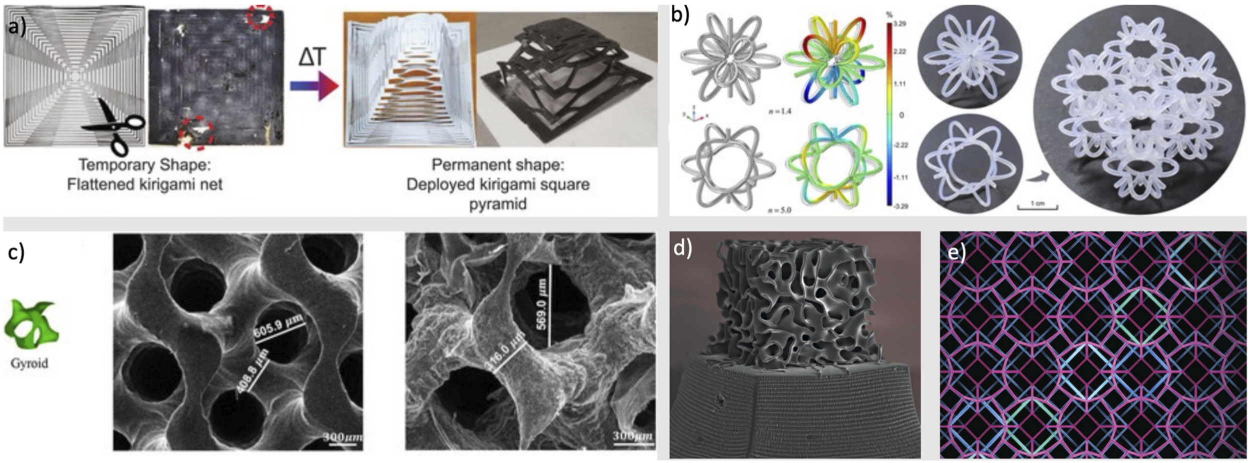

FIG. 3. Examples of programmable morphing using active (a) and passive (b) materials. Even more complex geometries can be considered in the design of such active metamaterials (c). An example of nano-architected ceramics with ultrahigh energy absorption (d). An example of a tensegrity metamaterial with failure-resistant property (e). Subfigures (a), (b), and (c) are, respectively, reprinted with permission from Lai-Iskandar *et al.*, "Programmable morphing, electroactive porous shape memory polymer composites with battery-voltage Joule heating stimulated recovery," APL Mater. **10**, 071109 (2022); Zhang and Krushynska, "Programmable shape-morphing of rose-shaped mechanical metamaterials," APL Mater. **10**, 080701 (2022); and Ashraf *et al.*, "On the computational modeling, additive manufacturing, and testing of tube-networks TPMS-based graphene lattices and characterizing their multifunctional properties," APL Mater. **10**, 121107 (2022) with the permission of AIP Publishing. Subfigures (d) and (e) are, respectively, reprinted with permission from Guell Izard *et al.*, "Ultrahigh energy absorption multifunctional spinodal nanoarchitectures," Small **15**, 1903834 (2019), and Bauer *et al.*, "Tensegrity metamaterials: Toward failure-resistant engineering systems through delocalized deformation," Adv. Mater. **33**, 2005647 (2021). Copyright 2023 John Wiley and Sons, Inc.

metamaterial functionalities may be impossible to realize without very specific bulk properties. For example, many designs of shape-shifting metamaterials, such as self-folding origami,[78–81] are dependent on the shape memory behavior found in some polymers[82,83] and metallic alloys[84,85] to program the underlying shape transformation behavior. Another example is metallic meta-biomaterials[86–88] [Fig. 2(c)] that require a specific set of biomedical requirements, such as biocompatibility, bioactivity, and biodegradability.[89–91] Bioactivity and biocompatibility are relatively less challenging to address. That is because there are metals (e.g., tantalum[92]) that are intrinsically highly biocompatible. Moreover, it has been possible to use traditional surface treatment techniques, such as anodizing[93–95] and plasma electrolytic oxidation,[96–98] to enhance the bioactivity of metals and their alloys. Biodegradability is, however, a relatively new addition to the possibilities offered by metallic meta-biomaterials.[99,100] Most reports related to metal biodegradability are limited to Mg,[101,102] Zn,[103] Fe,[104,105] and their alloys. The first reports of architected meta-biomaterials made from biodegradable metals have only recently appeared in the literature.[87,100,102] This has to do with the difficulty of processing some biodegradable metals with currently available AM processes. For example, Mg is highly inflammable and creates safety concerns, while Zn has a relatively low evaporation temperature that makes it difficult to process with direct metal printing techniques.

A final example concerns the integration of electronics into architected materials such that the structural properties can be combined with other functionalities, such as sensing, actuation, and processing.[106] The incorporation of electronics into architected materials requires the ability to print the main structural material while also distributing the other materials needed for the electronics, such as conductors and semiconductors. Simultaneous 3D printing of structural, conductive, and semiconductive materials into a coherent architected construct with arbitrarily complex geometries remains a major challenge that needs to be addressed in the coming years.

Regardless of the type of the properties pursued in the design of metamaterials, a recurrent theme is the need to incorporate differential material response into a single construct because many advanced functionalities are dependent on the co-existence of highly different material properties next to each other and within the fabric of a single metamaterial construct. Examples include conductive vs non-conductive vs semiconductive materials for electronics applications,[107,108] magnetic vs non-magnetic properties for magnetic applications,[109–112] soft vs hard materials for creating simultaneously tough and stiff materials,[113–115] and shape-shifting vs delayed shape-shifting vs passive materials for programming complex (e.g., sequential) shape transformations[116–124] (Fig. 3) and phase transitions.[125] Creating this type of differential responses remains one of the major challenges of AM techniques to be tackled in the coming years.

IV. FUNCTION

Depending on the type of the metamaterial, the design objective may be different. Indeed, there has been a gradual shift over the years from a primarily property-driven approach to a functionality-driven one. In this context, property refers to the effective properties of the metamaterial at the macroscale when the size of the metamaterial specimen is large enough as compared to its microarchitecture. The "design for property" approaches generally aim at the creation of metamaterials with unusual properties that are not found in ordinary engineering materials, including the dilational behavior [Fig. 1(a)],[126–128] negative Poisson's ratios [i.e., auxetic behavior, Fig. 1(b)],[2,124,129–131] negative stiffness,[132–134] negative thermal expansion,[67,71] ultra-high stiffness,[15,63,135] directional compliance,[136] and penta-mode [i.e., fluid-like, Fig. 1(c)] properties.[62,127,137,138]

On the other hand of the property–functionality spectrum, one finds the "design for functionality" paradigm where the designed metamaterial exhibits functionalities that are generally observed in devices. The boundary between the material and device is thereby somewhat blurred. This has given rise to terms such as "machine matter,"[139–142] where the material is the machine. Examples of such functionalities include shape-morphing behaviors,[143–150] self-folding origami,[78,151,152] information storage (i.e., memory metamaterials),[120,153] power transmission and motion conversion,[154] and digital logic in the format of mechanical logic gates.[155,156]

There are also design concepts that take an intermediate position in the spectrum from a material to a device. An example of such intermediate concepts from mechanical metamaterials is strain rate-dependent switching in the properties (e.g., from auxetic to conventional or the other way around) and functionality (e.g., from clockwise to counterclockwise rotation)[23,139,141] of a metamaterial. Another example from meta-biomaterials is the hybrid auxetic–non-auxetic meta-implants,[6] where a rational distribution of the Poisson's ratio is used to enhance the longevity of orthopedic implants.

The move from property-driven design approaches to functionality-driven ones is a welcome change in the direction of this research area because the scope of possible designs is much broader when dealing with functionalities as opposed to properties, which are limited both in number and in their possible ranges due to, among other factors, thermodynamics constraints. Indeed, there are well-defined theoretical limits for the range of various properties that could be achieved through the microarchitectural design of metamaterials. For example, the Poisson's ratio of isotropic materials is limited to the specific range [−1, 0.5],[157] while the possible ranges of elastic modulus and bulk modulus of metamaterials are coupled and limited by the Hashin–Shtrikman bounds.[158] As a result of the latter theoretical bound, it is, for example, theoretically impossible to design metamaterials that are simultaneously highly auxetic and highly stiff. The envelope of functionalities that can be realized with metamaterials is, on the other hand, only dependent on the availability of suitable materials and (additive) manufacturing techniques. For example, the availability of AM techniques that could process both stress-worthy materials (e.g., hard polymers, metals, or composites) and (semi)conductors would enable the development of metamaterials with both structural and (distributed) electronic functionalities. Given the ever-expanding range of materials that can be processed with (multi-material) AM techniques, it is expected that we will see many novel functionalities appearing in the literature in the coming years.

V. FABRICATION

The fabrication of metamaterials can be performed using several techniques, of which AM is the most important one. That is because the form-freedom offered by AM techniques is essential for the creation of the often highly complex microarchitectures that result from rational design processes and are required for the realization of unusual properties and advanced functionalities. AM techniques have been under development for more than three decades, initially under the names "rapid prototyping" and "3D printing" and later under the umbrella of "additive manufacturing technologies," which, according to the American Society for Testing and Materials (ASTM) classification, consists of seven different categories.[159] While the first attempts at "rapid prototyping" were primarily focused on the fabrication of physical models without necessarily requiring the use of industrial-grade, stress-worthy materials, the recent research since the turn of the century and particularly in the last decade has been focused on the processing of stress-worthy materials to create fully functional parts with complex geometries and high fidelities that are on a par with industrially made parts.

The recent developments of AM have expanded the length scales, types, and number of co-printed materials. As far as the length scales are concerned, it is currently possible to additively manufacture materials with a few nanometer resolutions using electron beam induced deposition,[160,161] with submicron resolutions using two photon polymerization,[1,162,163] with a few micrometer resolutions using variants of stereolithography,[71,164,165] with sub-100 micron resolutions using polyjetting[57,166] as well as with microselective laser melting,[167,168] and with submillimeter resolutions using a variety of techniques (e.g., selective laser melting[169] and electron beam melting[170] for metals; fused deposition modeling[171] and selective laser sintering[49] for polymers).

Even though printing with very fine resolutions has become possible, there are two major obstacles that need to be tackled in future studies. First, the additive nature of printing processes means that the fabrication of objects with dimensions that are a few orders of magnitude larger than the printing resolution takes a formidably long time. To date, this limitation has been primarily addressed through the use of indirect AM techniques where molds,[9,17,140,172] (lithography) masks,[173,174] or (imprinting) stamps[175–177] are created using AM and are then applied to scale up the manufacturing of the target devices both in number and in dimensions [Figures 3(d) and 3(e)]. An emerging approach to design and fabricate scalable nano-architected materials is the use of self-assembly approaches (e.g., spinodal decomposition[178,179]). While the unit cell topology is somewhat limited by the natural process, recent studies have shown that spinodal shell-based metamaterials [Fig. 3(d)] have exceptional mechanical[180,181] and biomechanical[182] properties.

The second limitation concerns the limited number of materials that can be processed with small-scale AM techniques. As a rule of thumb, AM techniques working with the finest resolutions can only process a limited number of materials with a relatively limited range of (mechanical) properties. Once more, indirect AM may be used to address this limitation to some extent. However, indirect AM techniques have their own limitations, including a lower degree of design freedom as compared to direct AM techniques. It is, therefore, important to address both the above-mentioned challenges more directly and through the development of AM machines that are specifically designed for scalable manufacturing of metamaterials (e.g., machines with many laser sources) as well as through the development of novel, bespoke materials that could be processed using ultrahigh resolution AM techniques.

While we discussed the materials used for the fabrication of metamaterials in Sec. III, the focus of that section as well as that of much of recent research has been on the application of AM for the processing of already existing materials. The optimal conditions for the processing of metamaterials are, however, only achieved when new materials are developed for the specific AM technique at hand. Future research should, therefore, focus on the mutual optimization of AM processes and functional materials to enable a high resolution, high fidelity, and scalable fabrication of multi-functional metamaterials, meta-structures, and meta-devices.

VI. CONCLUSIONS

In summary, the research into metamaterials has been growing in both breadth and depth over the last decade and it currently constitutes an important, thriving area of research with a large community of researchers attracted from different disciplines and areas of the world. This diversity of topics, research groups, and researchers is also reflected in the current special issue where a selected number of studies are presented that cover various types of properties/functionalities, design techniques, and fabrication methods. The recent developments in rational design processes, particularly advanced computational methods (e.g., machine learning and multi-objective topology optimization), as well as the ever-increasing availability of highly functional materials for AM and the coming-of-age of AM techniques themselves are expected to enable the development of novel types of metamaterials. More specifically, there is a move from engineering properties to creating multiple advanced functionalities where the boundary between materials and devices is blurred.

AUTHOR DECLARATIONS

Author Contributions

Amir A. Zadpoor: Conceptualization (lead); Investigation (lead); Project administration (lead); Supervision (lead); Visualization (supporting); Writing – original draft (lead); Writing – review & editing (lead). **Mohammad J. Mirzaali**: Conceptualization (supporting); Investigation (supporting); Project administration (supporting); Supervision (supporting); Visualization (lead); Writing – original draft (supporting); Writing – review & editing (supporting). **Lorenzo Valdevit**: Conceptualization (supporting); Investigation (supporting); Visualization (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Jonathan B. Hopkins**: Conceptualization (supporting); Investigation (supporting); Visualization (supporting); Writing – original draft (supporting); Writing – review & editing (supporting).

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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