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Metamaterials Using AdditiveManufacturing Technologies

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14. ABSTRACT

The purpose of this document is to provide an overview of current research being conducted in the field of additively manufactured metamaterials. This includes a historical overview of both metamaterials and additive manufacturing as well as suggestions for future research directions.

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EXECUTIVE SUMMARY

The purpose of this report is to provide an overview and summary of existing literature on the use of additive manufacturing (AM) technologies to fabricate metamaterials. The areas of both AM and metamaterials have exploded in popularity in the last two decades with each having tens of thousands of publications in that time. AM, or 3D printing, is the process of joining materials to make parts from 3D model data, usually layer upon layer, as opposed to subtractive manufacturing and formative manufacturing methodologies. The process of additive manufacturing is very flexible in both part design and material selection. Metamaterials are fabricated structures and composite materials that either mimic known material responses or qualitatively have new, physically realizable response functions that do not occur or may not be readily available in nature. Metamaterials were originally theorized and developed for electromagnetic applications but have since been extended to optical, thermal, acoustic, and mechanical applications. Metamaterials are commonly said to be "artificial" materials since the exotic properties that can be demonstrated do not depend on chemical constituents, but rather internal structure of the metamaterial. As expected, this internal structure can be complex and difficult to fabricate. The freedom derived from AM is therefor a natural choice for fabricating metamaterials.

In order to achieve the previously unachievable behaviors seen in metamaterials, the internal structure of the metamaterial manipulates wave physics at the scale of the wave length. Therefor, depending on the application, the internal structure needs to be anywhere from millimeters to nanometers in scale. Early fabrication methods were based on lithography as it was able to achieve structure resolutions at the sub-micron scale. However, lithography based methods generally suffer from a number of limitations such as difficulty in large area 3D production, the use of "negatives", and low speed. With advances in AM, it has became increasingly more advantageous to fabricate metamaterials with AM. Many common AM techniques, such as SLM, SLS, EBM, DMLS, FDM, and SLA, have already been used to successfully fabricate metamaterials. The drawback to using these methods are related mostly to precision of the build, which is generally at the upper range (sub-millimeter) of what is desired in metamaterial fabrication. To address this, a number of high precision AM methods have been developed that can achieve sub-micron and even nanometer scale resolutions. The most prominent of these methods is projection micro-stereolithography, which is based on SLA, able to achieve micron level precision, scales well to larger parts, and is a relatively simple and well understood process. The high precision AM methods developed so far have primarily been intended for non-metallic materials but some recent work has started to modify the processes to print metallic and ceramic materials. Metallic and ceramic metamaterials have the ability to offer structural stiffness in addition to the other desired properties and are an area that need further development, especially the development of high precision metallic AM methods.

The primary purpose of AM in metamaterial fabrication has not necessarily been to induce novel properties, but rather AM has functioned as an enabling technology for metamaterials. In theory, any metamaterial property can be realized without the use of AM but in practice, without AM, many metamaterials would never be able to demonstrate the theoretical properties desired. With the increasing popularity of computational simulation and topology optimization, metamaterials with extremely complex structures can be designed to exhibit more than just a single behavior, such as cloaking or negative refractive index. The metamaterial can be designed so that it demonstrates the desired property while also maintaining optimal cooling, flow, or strength for instance. Since AM is inherently a computationally driven process, it is realistically the only fabrication method available to fabricate such designs. Mechanical metamaterials are generally based on periodic lattice structures, which could be somewhat easily constructed with non-AM fabrication methods. However, the use of AM opens the design space so that disordered and/or multimaterial lattices can be fabricated. Multimaterial, mechanical metamaterials have demonstrated Poisson ratio's as low as -7, much lower than single material metamaterials have shown. In acoustic metamaterials, axisymmetric acoustic cloaks and radar absorbing metamaterials have been created using designs that can only, practically, be fabricated with the use of AM. The use of AM to construct electromagnetic, optical, and thermal metamaterials has not been widely examined, likely due to the sub-micron resolutions required in those metamaterial structure. With high precision AM methods quickly being developed, these metamaterial classes are ripe with opportunity for new developments. AM of ceramics is increasing in popularity and could enable designs of thermal metamaterials at extreme temperatures that serve as thermal cloaks for jet engines, as an example.

Another area of development in metamaterials is in 4D printing (AM of smart materials) applications. The preliminary work in this area has shown tunable and switchable metamaterials. The additional degree of freedom of time in the design space allows for metamaterial designs that can modify their behavior based on an environmental stimulus. The develop of 4D metamaterials is very new and has generally only been demonstrated in a laboratory setting and mostly with polymeric materials. The usage of metallic smart materials, like shape memory alloys (SMAs), offers even more possibilities, especially when combined with shape memory polymers (SMPs) to create "freezable" shape memory composites. Multimaterial AM methods could easily enable such materials to be fabricated. One possible design could utilize AM to embed SMAs in an SMP-based origami metamaterial. This metamaterial could be folded, unfolded, and locked into either position by using appropriate structural design and material selection, and could find application in space-based deployable structures or biomedical applications.

The use of AM to fabricate metamaterials is in many ways limited by the AM technology and not necessarily an understanding of the metamaterial. However, using AM to fabricate metamaterial makes new designs of internal metamaterial structures possible and advances the state of the art in metamaterials. The unique structure of metamaterials mean that what is best for general AM technology may not be best for metamaterial development and purposeful research should be conducted to maximize the benefits of AM for metamaterials. As one example, porosity and anisotropy in AM is generally thought to be a negative feature but in metamaterials, these defects could be used to tailor mechanical properties of the metamaterial.

METAMATERIALS USING ADDITIVE MANUFACTURING TECHNOLOGIES

1. INTRODUCTION

The topics of both metamaterials and additive manufacturing (AM) have no shortage of existing literature. As of the submission of this work, searching the terms “metamaterials” and “additive manufacturing” in the Scopus database yields over 35,000 and over 24,000 publication results, respectively. Plotting these results by publication year (Fig. 1), shows the rapid rise in popularity of both technologies in the past 20 years. While both AM and metamaterials are popular fields, the overlap of the two technologies (*i.e.* using additive manufacturing to produce metamaterials) is just starting to emerge.

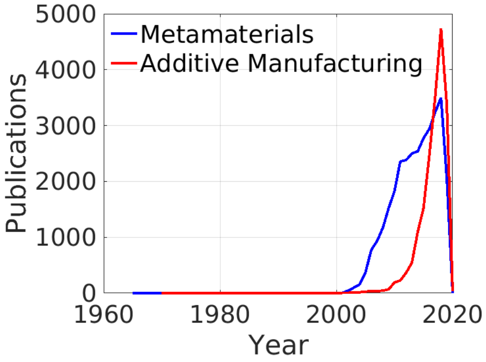

Fig. 1—Scopus publication results per year for the search terms “metamaterials” and “additive manufacturing”.

In this report, a summary of recent literature on the use of AM to fabricate metamaterials will be shown. The remainder of this report will be structured as follows: Section 2 briefly introduces the background of AM as well as metamaterials; Section 3 examines the fabrication of metamaterials and AM processes that can be applied to fabricate metamaterials; Section 4 reviews recent work on metamaterial parts that have been fabricated using AM and the unique properties and applications of those parts; finally Section 5 identifies potential opportunities for fabrication and application of AM metamaterials.

2. ADDITIVE MANUFACTURING AND METAMATERIALS

As mentioned, both AM and metamaterials are fields with a vast body of literature. Both AM and metamaterials encompass a very broad range of research and applications. In this chapter, a brief overview of AM and metamaterials is presented.

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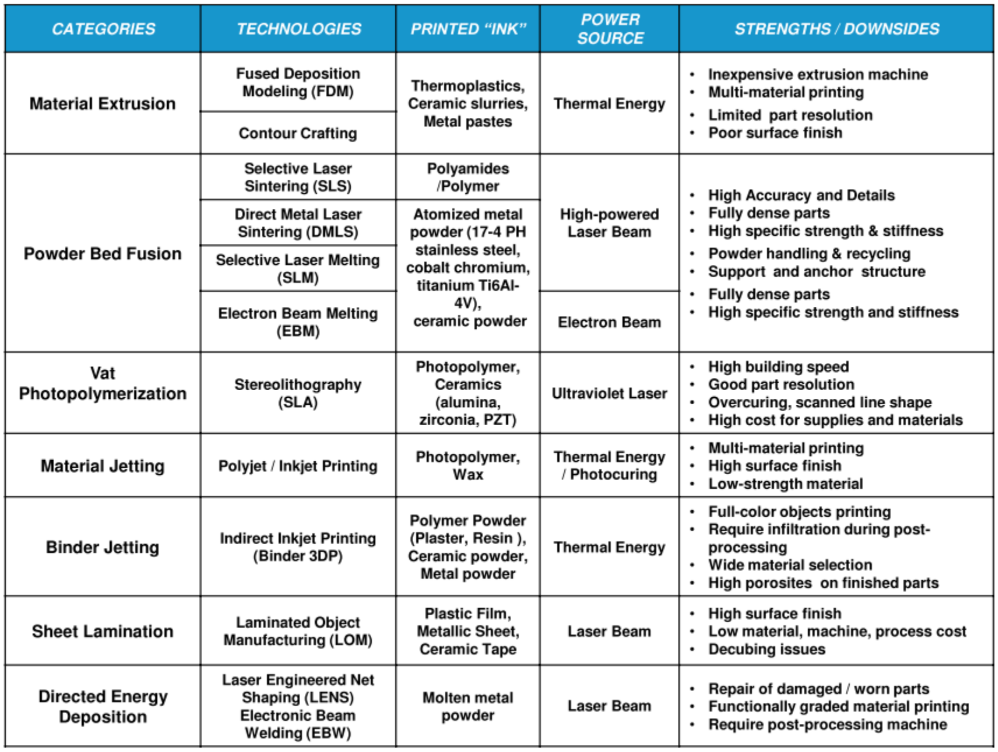
2.1 Additive Manufacturing Technologies

Much of the work in additive manufacturing (AM) has been accomplished in the past two decades but the ideas used in AM can be dated to as far back as the 1860s. [1] AM is referred to by many names in the literature including, but not limited to, 3D printing, rapid prototyping, and layer manufacturing. [1–4] The general AM process has, however, been specifically defined by ISO/ASTM standards to be the "process of joining materials to make parts from 3D model data, usually layer upon layer, as opposed to subtractive manufacturing and formative manufacturing methodologies", which gives the area of AM a very broad scope. Likewise, by the same definition standards, major AM processes can be generalized into seven categories: fused deposition modeling (FDM), inkjet printing (IJP), laminated object manufacturing (LOM), laser engineered net shaping (LENS), stereolithography (SLA), selective laser sintering (SLS), and three-dimensional printing (3DP). [2, 5] As with the naming convention for AM, the naming of the processes can vary widely [1], where, for example, technologies that all use laser processing for metallic materials can be referred to as different names depending on how the powder is fed and melted. [6–8] AM can utilize a vast range of materials from plastics to metals to ceramic to foods and other biological/polymeric materials. [4, 9–12] Typically, certain classes of materials are printed with certain method (*e.g.* metals with SLS or LENS, plastics with FDM, etc.) but this is not a strict guideline and as reserach in the area expands, materials able to be printed using a given AM method will also expand. [11, 13] A summary of the classification of AM processes can be seen in Figure 2. [14]



CATEGORIES	*TECHNOLOGIES*	*PRINTED "INK"*	*POWER SOURCE*	*STRENGTHS / DOWNSIDES*
Material Extrusion	**Fused Deposition Modeling (FDM)**	**Thermoplastics, Ceramic slurries, Metal pastes**	**Thermal Energy**	• Inexpensive extrusion machine • Multi-material printing • Limited part resolution • Poor surface finish
	Contour Crafting			
Powder Bed Fusion	**Selective Laser Sintering (SLS)**	**Polyamides /Polymer**	**High-powered Laser Beam**	• High Accuracy and Details • Fully dense parts • High specific strength & stiffness • Powder handling & recycling • Support and anchor structure • Fully dense parts • High specific strength and stiffness
	Direct Metal Laser Sintering (DMLS)	**Atomized metal powder (17-4 PH stainless steel, cobalt chromium, titanium Ti6Al-4V), ceramic powder**		
	Selective Laser Melting (SLM)			
	Electron Beam Melting (EBM)		**Electron Beam**	
Vat Photopolymerization	**Stereolithography (SLA)**	**Photopolymer, Ceramics (alumina, zirconia, PZT)**	**Ultraviolet Laser**	• High building speed • Good part resolution • Overcuring, scanned line shape • High cost for supplies and materials
Material Jetting	**Polyjet / Inkjet Printing**	**Photopolymer, Wax**	**Thermal Energy / Photocuring**	• Multi-material printing • High surface finish • Low-strength material
Binder Jetting	**Indirect Inkjet Printing (Binder 3DP)**	**Polymer Powder (Plaster, Resin), Ceramic powder, Metal powder**	**Thermal Energy**	• Full-color objects printing • Require infiltration during post-processing • Wide material selection • High porosites on finished parts
Sheet Lamination	**Laminated Object Manufacturing (LOM)**	**Plastic Film, Metallic Sheet, Ceramic Tape**	**Laser Beam**	• High surface finish • Low material, machine, process cost • Decubing issues
Directed Energy Deposition	**Laser Engineered Net Shaping (LENS) Electronic Beam Welding (EBW)**	**Molten metal powder**	**Laser Beam**	• Repair of damaged / worn parts • Functionally graded material printing • Require post-processing machine

Fig. 2—Classification of AM processes by ASTM International. [14]

The adaption and expansion of AM can in part be attributed to the reducing cost of programmable controllers, lasers, and computer-aided design (CAD) software. [14] The process of creating a part to be built by AM begins with 3D solid modeling as a CAD file which is then sliced into many discrete layers that are used as inputs to the AM machine to build the part. The largely digital workflow of the AM process allows for significant design and geometric flexibility with high dimensional accuracy. The AM process is also cost efficient as there is no need for assembly of parts and there is very little waste material. [13, 14] Additionally, AM processes can be scaled to print parts that range from the nano/micrometer scale [15–17] to the scale of meters. [18, 19]

2.1.1 4D Printing

4D printing is a subset of 3D printing (or AM) that is applied when smart materials or multi-material systems are used in the AM process with the capability to transform over time. [20, 21] The transformation of the material system in time is a result of an applied stimulus in the form of thermal, mechanical, electrical, magnetic, chemical, and/or many other forms or combinations of stimuli. [22–24] The bulk of research in 4D printing is centered around shape memory alloys [25], polymers [26, 27] and their composites [28] but recent advances have begun to extend this to multi-material systems and metamaterials. [29] Applications of 4D printing are growing as well with 4D parts being built for applications in biomedicine, textiles, soft robotics, oragami, adaptive joints/structures, and aerospace. [24, 27]

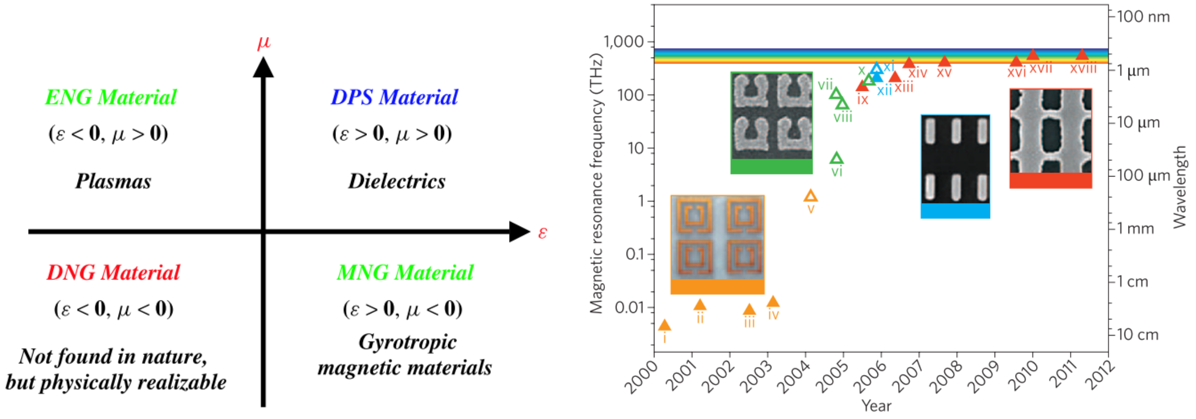
2.2 Metamaterials

As with AM, metamaterials have exploded in popularity in the past 20 years but the origins of these materials can be traced back to the early nineteenth century. [30] Metamaterials are fabricated structures and composite materials that either mimic known material responses or qualitatively have new, physically realizable response functions that do not occur or may not be readily available in nature. [31] These "artificial" materials can have exotic properties, which are not primarily dependent on the intrinsic properties of the chemical constituents, but rather on the internal, specific structures of metamaterials. [32] The origins of modern metamaterials are derived from the theoretical work of Veselago [33] then, later, Pendry [34] and Smith *et al.* [35]. However, these works were purely theoretical and it was the work of Shelby *et al.* [36] that first fabricated such a material and demonstrated a negative refraction index.

The underlying scheme of metamaterials is that the structures manipulate wave physics to achieve unusual or previously unachievable behaviors. [37] As mentioned previously, the earliest works in metamaterials were in electromagnetics to realize the theoretical double negative (DNG) material (Fig. 3a) of Veselago's early work. The work in artificial magnetism of Pendry *et al.* in the 1990's was the first to demonstrate a magnetic material with negative permeability (μ) and positive permittivity (ε). [38] This work was key to the development of an artifical structure known as a split-ring resonator (SRR), which first demonstrated a realizable DNG. [34, 36, 39] Initially, these metamaterials were limited to microwave ranges (Fig. 3b) [40] but research quickly expanded the area to encompass frequencies in the terahertz and optical range. [41, 42] The structures originally proposed and built to create and demonstrate these metamaterials were single functional layers but Valentine *et al.* [43] realized a 3D structure by appropriately stacking several individual layers. [44, 45] The success of metamaterials in electromagnetics/optics led to a natural extension of metamaterials that were designed to operate at much larger wavelengths and length scales such as those in acoustics, thermodynamics, and mechanics. [46, 47]

2.2.1 Thermal Metamaterials

Thermal metamaterials are governed by equations similar to those used in electromagnetic metamaterials [48] and are referred to as transformation thermodynamics. [49, 50] Thermal metamaterials are perhaps the



(a) Classification of metamaterials by permittivity and permeability. [31] DPS: double positive, DNG: double negative, MNG: μ negative, ENG: ε negative.

(b) Recent progress in electromagnetic/optical metamaterials. [45]

Fig. 3—Classification and progress in electromagnetic/optical metamaterials.

least studied class of metamaterials due to the relatively late derivation of transformation thermodynamics. [51] However, thermal metamaterials are quickly being advanced for applications in thermal cloaking, protection, and management. [52, 53]

2.2.2 *Acoustic Metamaterials*

Acoustic and mechanical metamaterials are similar and in many cases grouped together since acoustics depend on the bulk modulus in a similar manner as classical continuum mechanics. [54] The first acoustic metamaterials (AMMs) developed by Liu *et al.* [55] used rubber-coated spheres to create locally resonant and deeply subwavelength structures that responded to incident acoustic waves. Since then many reserachers have investigated AMMs for their ability to control the attenuation of acoustic waves, invisibility cloaking capabilities, and acoustic wavefront engineering, such as focusing via manipulating the acoustic impedance of metamaterials. [56] AMMs are able to control wave propagation by changing the bulk modulus (κ) and/or the mass density (ρ) in much the same way that is done with electromagnetic metamaterials (Fig. 4). As such, it can be shown that κ^{-1} corresponds to ε and ρ to μ in the acoustic and electromagnetic governing equations. [47] The literature is vast in the area of AMMs and there are a number of very thorough reviews on the topic of AMMs which discuss acoustic hyperlens' [57], mechanisms of absorption [58], tunable AMMs [56], and other aspects and applications such as cloaking and active AMMs. [59–61].

2.2.3 *Mechanical Metamaterials*

General mechanical metamaterials are perhaps the most complex metamaterials to design due to the number of possible parameters involved in describing a continuum material (*i.e.* anisotropic elasticity tensor, thermo-elasticity, elasto-plasticity, *etc.*). [48] Mechanical metamaterials are typically engineered to yield properties such as zero or negative Poisson ratio (auxetic) [62–65], very low mass or negative mass density (light-weight), vanishing shear modulus (pentamode or anti-auxetic) [66, 67], negative stiffness or compressibility (negative parameter) [68, 69], origami structures, and/or negative coefficient of thermal expansion [70, 71]. Examples of each of these classes of materials can be seen in Figure 5.

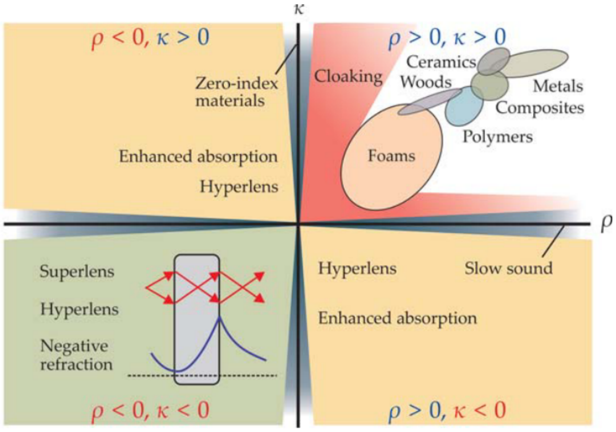


Fig. 4—Classification of acoustic metamaterials by mass density, ρ, and bulk modulus, κ, in the same manner as Figure 3a. [60]

While most mechanical metamaterials are metallic, research in the area of polymers [72] and ceramics [73] have also been studied. As with AMMs, research in the area of mechanical metamaterials is vast and for further information, the reader is directed to some recent reviews of the field. [54, 74–76]

3. FABRICATION AND ADDITIVE MANUFACTURING OF METAMATERIAL

As has been discussed in the previous section, metamaterials behavior is dependent on the internal structure of the metamaterial. This structure is in general, a complex, small repeating structures. As one can imagine this structure is extremely suitable to be manufactured by AM technologies due to the many unique advantages it offers, such as the ability to print geometries from the macro to micro scale and being free from space limitations. [77, 78] This chapter will address general metamaterial fabrication briefly then review recent work on fabrication of metamaterials with AM. Specific properties and applications of AM fabricated metamaterials will be discussed in Section 4.

3.1 Metamaterial Fabrication

Since the first metamaterials were demonstrated in 2001, enhanced fabrication techniques for metamaterials in both 2D and 3D have been of interest. The single, largest, current barrier to metamaterial widespread adaption is the difficulty in scaling from 2D to 3D, and doing so in a manner that maintains sufficient resolution of the desired unit cell while fabricating parts at the macro scale.

The most common class of methods for 2D metamaterial fabrication is lithography. Lithography for metamaterials generally takes the form of photolithography, shadow mask lithography, soft lithography, electron beam lithography, x-ray lithography, or two photon lithography. [79] Photolithography was among the earliest method of fabricating metamaterials. Generally speaking, lithography methods use a stamping or transfer printing process requiring a “negative” of the design to be printed and are exposed to an energy source such as UV light or an electron beam. These methods can produce features at the sub-micron and nano scales, and can be scaled to 3D by stacking multiple layers. [80] However, lithography methods are typically slow, expensive, and lack scalability to large areas. [81] Stereolithography is a method commonly used in AM but is not widely used for metamaterial fabrication due to insufficient resolution. However,

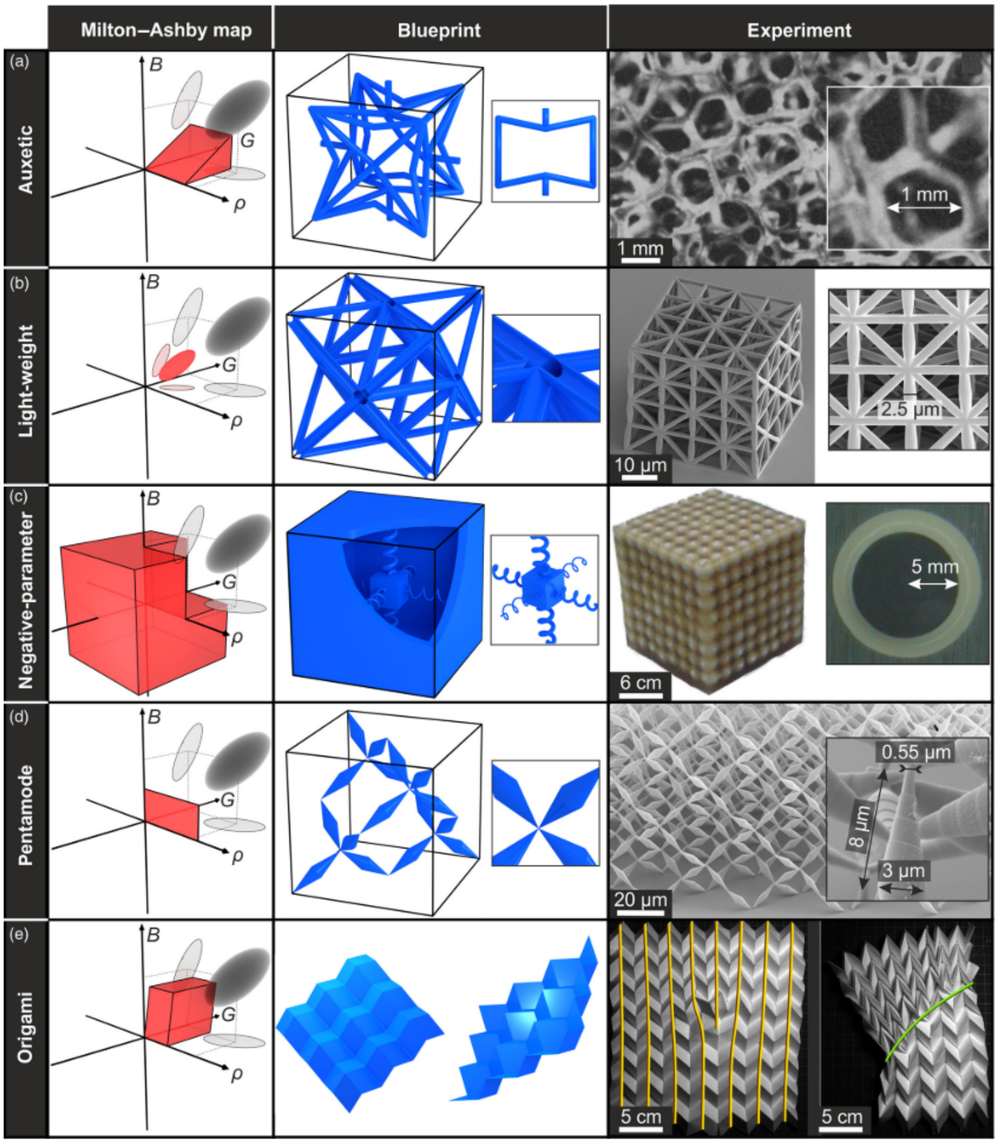


Fig. 5—Overview of mechanical metamaterials by Christensen *et al.* [54]. B: bulk modulus, ρ: mass density, and G: shear modulus. In the first column, red represents the metamaterial regime while black represents traditional materials.

recent advances in microstereolithography have resolved this issue. [82] Further discussion on this topic is delayed to the next section.

Ideally, large-scale 3D metamaterials will be achieved through some low cost, high throughput process but, at present, most fabrication methods fall into either 3D or large-scale with some techniques beginning to bridge the gap. One of the first 3D fabricated metamaterials was by Deubel *et al.* [83] using direct laser writing, but this method suffers from low throughput and material selection issues. [80] Other advanced 3D fabrication techniques such as imprint lithography, interference lithography, multi-photon polymerization [17], and focused ion beam milling have been studied but generally suffer from the same issues of low throughput, lack of resolution, lack of scalability beyond the micron scale, and/or limited material selection. [79, 84]

Additional 3D methods derived from traditional manufacturing processes such as ceramic casting [85] and injection molding [86] have also been adapted to metamaterial fabrication but these have not been widely adapted due to issues with mold creation and required sintering for final part formation. The most promising methods for fabricating large-scale 3D metamaterials are nanoimprint lithography [87], pattern transfer [88], self-assembly [89, 90], and AM. Deatils of all but AM are omitted here but the reader is referred to Yoon *et al.* [80] and Bishop-Moser *et al.* [81] for an overview of these methods.

3.2 Additive Manufacturing of Metamaterials

AM has been used to build light-weight, high stiffness structures through topology optimization since its inception and its use has only become more widespread. [14, 91] While many examples of AM-built topology optimization are truly freeform structures, many are based on patterned unit cells. [92] Thus, it is natural to pair AM and metamaterials. However, most common AM techniques operate at the micron to meter scale while metamaterials typically use structures in the millimeter to nanometer scale, especially in electromagnetic and optical applications where the substructure of the metamaterial needs to be of the same size as the wave length. In order to accommodate this scale difference, many new high precision AM techniques have been developed/implemented for metamaterial fabrication.

There are a number of works that use a modified high precision version of stereolithography (SLA) called projection microstereolithography (PμSLA). [78] This method is capable of producing features with 1 μm resolution in a nearly identical fashion as traditional SLA. PμSLA is perhaps one of the most common metamaterial fabrication techniques used in the literature. [82, 93, 94] This is in part due to the fact that it is a relatively simple process that is well understood, has a throughput, and has been scaled well to large build planes. As with traditional SLA, PμSLA is typically limited to photopolymers but some work in ceramics [95] and metals [96] has been conducted.

Two other high precision methods of metamaterial fabrication by AM, which are both somewhat common, are direct laser writing and direct ink writing. The former shares some similarities with SLA while the latter more closely resembles binder jetting or material extrusion methods. [97] Within direct laser writing is a method known as two-photon polymerization (TPP), which uses a femtosecond-pulsed laser beam to initiate the photopolymerization process. This method is extremely accurate with resolutions under 100 nm and can operate in a truly 3D space (*i.e.* no support structure is required and photopolymerization happens on a continuous 3D laser path rather than a layer by layer approach). [98] TPP has a very small material selection and while extremely precise, does not scale to large parts well. In direct ink writing, colloidal inks are used which solidify based on pressure and not phase change as traditional extrusion methods. [99, 100] Direct ink writing can reach sub-micron resolutions and has a wide selection of materials but suffers from slow printing processes. [78, 101]

Before continuing, two other methods of high precision metamaterial fabrication are noted. Electrophoretic deposition [102] and electrohydrodynamic printing [103] both use electrostatic fields to transport material to the desired location. Both processes are very promising as they are low cost, reliable, high precision methods. However, the processes suffer from a number of drawbacks including low rate deposition. [78] Additionally, while both methods are additive, neither fits in the standard classification of AM processes (Fig. 2) and as such will not be discussed further.

While the techniques discussed above are high precision AM methods, there are still a number of applications in metamaterial fabrication that make use of traditional AM methods, particularly in mechanical and acoustic metamaterials where metamaterial structures need not be micro/nanoscale. To avoid repetition, only a brief overview of some traditional AM methods that have been used for metamaterial fabrication are given.

SLA has been used to produce mechanical octet lattices for energy absorption [104] and metamaterial embedded geometric optics [105]. Powder bed fusion methods such as SLS [106–109], DMLS [110], SLM [15], EBM [111] have all been extensively implemented, particularly for mechanical metamaterials where the strength of metallic structures is desired. FDM has also been used in 4D metamaterials [112] and other polymer applications [113]. Other methods such as binder jetting [114] have been utilized but their usage is sparse. While not directly related to directed energy deposition (DED), aerosol jet printing [115] shows some similarities DED as well as aerosol deposition (AD) [116] and cold spraying (CS) [117] but with much more precision.

Many of the AM processes reported in Figure 2 are seen in the above overview. Methods such as sheet lamination and DED seem to be the least common methods of fabricating metamaterials. Sheet lamination is most likely not used since it does not have a mechanism to produce the structural features required for metamaterials. DED generally does has a lower resolution than other comparable sintering or melting methods for metallic materials, which likely explains it's lower uptake in the metamaterial fabrication process. However, DED has the ability to operate with multi-axis milling machines thus has more freedom spatially than other AM processes. This freedom could be beneficial in creating complex metamaterial structures and doing so may aid in the creation of isotropic metamaterials, a highly desirable feature. DED also has the ability to create functionally graded and multi-material parts, which other methods, especially metallic AM methods, cannot do.

3.2.1 4D Metamaterials

As previously mentioned, another emerging class of 4D materials are 4D metamaterials. Due to the relatively recent uptake of 4D printing, work in 4D metamaterials is limited but provides the prospect of creating materials that function as multi-state metamaterials. [118] The most common methods of 4D metamaterial printing are FDM and SLA/PμSLA for shape memory polymers (SMPs) but other methods such as SLS and inkjet pritning for SMP fabrication and SLM for shape memory alloys (SMAs) have been successful. [24, 119–121] Other works in 4D printing have started to examine functionally graded printing and multi-material printing but these techniques have not been used to demonstrate conventional metamaterial behaviors. [122]

4. ADDITIVELY MANUFACTURED METAMATERIALS

In this section, properties achieved by AM fabricated metamaterials and some applications will be discussed. As one would expect, AM fabricated metamaterials exhibit many of the same properties and non-AM metamaterials but effort to discern the two classes will be made. It should also be mentioned that a recent review by Wu *et al.* [123] provides an excellent overview of additively manufactured metamaterials and their applications/properties. The current work differentiates itself by giving a broader overview of applications and properties rather than many examples of prominent metamaterial usages.

4.1 Properties

As in Section 2.2, electromagnetic and optical metamaterial properties are discussed first, followed by thermal, then acoustic, and finally mechanical. Few authors purely discuss properties of electromagnetic or optical metamaterials that have been fabricate by AM. Rather, most authors take lesson learned from electromagnetic materials and design a metamaterial for a specific application then use AM to print the part. Intuitively, this makes sense as electromagnetic metamaterials have been extensively studied in 2D and their properties are generally well understood so the use of AM to fabricate the part is purely an application driven method. Additionally, the small scales required for electromagnetic metamaterials make AM of even simple structures a challenge. However, some authors do discuss properties that can only be achieved by AM. For instance, Garcia [124] describes the use of FDM to build a part based on unit cells that yielded an anisotropic dielectric tensor. Along the same lines, Isakov *et al.* [113] used high and low dielectric permittivity materials, layered appropriately, to achieve artificial anisotropic dielectric permittivity.

4.1.1 Thermal Metamaterials

As with AM electromagnetic materials, there is only a small body of literature in the design of tailored properties for thermal metamaterials by AM without a specific application in mind. Wu *et al.* [125] used multimaterial inkjet printing to design and fabricate antichiral structures that exhibited a negative thermal expansion. Similarly, Wang *et al.* [70] used multimaterial SLA to design a lattice structure that had a negative thermal expansion. Specific work on thermal cloaking, negative thermal conductivity, *etc.* that was enabled by AM does not appear to be an active area of research and thus has many opportunities for new studies such as designing metamaterial substructures that could only be produced by AM.

4.1.2 Acoustic Metamaterials

Locally resonant metamaterials are designed to prevent waves from propagating in certain frequency regions (bandgaps) for acoustic insulation or cloaking purposes. In 2013, Sanchis *et al.* [126] used AM to develop a prototype of an axisymmetric cloak, which was able to suppress 90% of the waves scattered by a sphere at a certain frequency. In acoustic cloaking, it is particularly important for a cloak to be isotropic and omni-directional. The techniques demonstrated by Sanchis *et al.* could be extended in such a way that an omni-directional cloak could be designed and enabled by AM. Raza [127] later made locally resonant metamaterials for energy absorption similar to Sanchis *et al.* , but Raza used a novel multi-material printing method to fabricate the part. The method used was a hybrid droplet direct write printing and continuous direct writing printing method with 3 different materials, 2 polymers and an adhesive. This technology could be significant if slightly modified to expand the range of materials used. For instance, Matlack *et al.* [128] designed a composite structure of polycarbonate and steel cubes for vibration absorption. The polycarbonate used in the study was 3D printed but paused intermittently to place the steel cubes. If the technology developed by Raza were modified properly, it is conceivable that a complete composite structure could be made without interruption. In another study, Wormser *et al.* [111] used EBM AM with titanium to design an LRM based on an optimized 3D lattice. Multiple designs were found, fabricated, then tested and showed different phononic band gaps and a slightly auxetic behavior. The dual purpose nature of the designed structure leads one to the conclusion that it may be possible to design metamaterials with both structural and acoustic or thermal purposes.

4.1.3 Mechanical Metamaterials

In comparison to other metamaterials, mechanical metamaterials produced by AM have a large body of literature. This can most likely be contributed to the fact that the AM process is a thermomechanical and materials driven process, which has obvious strong ties to structural mechanics and therefor, mechanical metamaterial design. Additionally as mentioned earlier, mechanical metamaterials do not require the same resolution as electromagnetic and optical metamaterials thus making them easier to fabricate and more readily available to study.

One of the earlier uses of AM methods to build mechanical metamaterials was Kashdan *et al.* [106], where a negative stiffness element was fabricated out of Nylon by SLS. The element fabricated was intended for vibration isolation and was a prototype which used a spring to control the buckling behavior of a beam. This prototype was a unique design for a metamaterial but does share some similar design elements as the negative-parameter design in Figure 5. SLS has been used in other works to construct disordered lattices for mechanical constant tuning [109] and fabrication of an auxetic structure, which used polyamide impregnated with carbon nanotubes (CNTs) [108]. The disordered lattices created by Mirzaali *et al.* [109] were unique since most metamaterial substructures are based on symmetric octet, re-entrant, or similar periodic lattices. Since the disordered lattices were complex and AM was required in order to fabricate them. These disordered structures were able to achieve a much broader range of elastic modulus-Poisson ratio duos than typical lattice restricted networks. AM was also a necessity to fabricate the CNT-modified polyamide auxetic bucklicrystals of Yuan *et al.* [108]. This work was able to show how energy absorption characteristics could be modified as a function of the design of the structures. Additionally, the use of non-metallic materials modified by CNTs were able to avoid processing defects inherent in most metallic AM. The use of CNTs in this work could be extended to give the auxetic structure more unique electrical/thermal properties that are commonly displayed by CNT-based materials.

Other AM methods, such as EBM, SLM, and SLA, have also been used to fabricate mechanical metamaterials. Amendola *et al.* [129] used EBM to fabricate a variety of titanium based pentamode metamaterial structures to study how the micro- and macroscopic features affect the structure, intended for vibration isolation applications. Some cyclic tests were completed but it would be interesting to examine if long term fatigue under low vibration loads is significantly effected by the metamaterial structure. Similar to the previous work, Hedayati *et al.* [130] used SLM to fabricate titanium pentamode materials. The novelty of this work was the variable energy density used to produce different strut shapes in the design of a pentamode metamaterial. In another study, Tancogne-Dejean *et al.* [15] used SLM to manufacture 316L stainless steel pentamode metamaterials where the strut dimensions were varied to produce different behaviors in the macroscopic structure. Additionally, this work showed results of the actual metallic microstructure where defects in manufacturing were analyzed. These defects are well known in the study of the AM process and more work in this area for metamaterials could lead to tailored microstructures that further enhance mechanical properties or possibilities beyond what is currently being studied.

Pentamode metamaterials have also been studied with polymeric materials printed by SLA. The printed lattice structures in the metamaterials have been varied [131] and shown high energy absorption characteristics with large recoverable strains [104]. These phenomena have also been shown at smaller scales from pentamode metamaterials produced by PμSLA. [82, 94] The metamaterials have shown very light weight structures with near linear mass-stiffness scaling in contrast to standard materials, which follow a square or cube relationship. PμSLA has also been used to print multimaterial lattices with tunable mechanical constants including Poisson ratio's that can range from nearly 0 to -7. [132] The range and control of elastic constants in the demonstrated multimaterial lattices show the possible range of the multimaterial design space enabled by AM. Combined with topology optimization and computational simulations, this design space could be exploited to architect metamaterials with behaviors even more extreme than those seen in the previous work. In addition to PμSLA, direct ink writing has been used to produce structures exhibiting the same near linear mass-stiffness scaling and negative stiffness. [101]

Another emerging field in mechanical metamaterials is heirarchical structural design. Hierarchical design is enabled by the ability to create structures at the nano/microscale (by using such methods as self assembly, TPP, or PμSLA) and incorporate those features into much large macroscale structures. [107] Among the first groups to study this type of structure was Meza, Zheng *et al.* [96, 133, 134]. Starting at the nanolattice, the group was able to use large area PμSLA to construct nanoscale substructures that could take advantage of the material size effect and propagate this all the way up to a part on the centimeter scale. Parts constructed at using this hierarchical approach could yield extreme mechanical properties, tailored to any desired application, while maintaining extremely low densities. Additional, due to the scale of the base nanolattice, it may be possible to use this technique to construct metamaterials other than mechanical metamaterials, such as optical and electromagnetic or thermal metamaterials.

4.1.4 4D Properties

As discussed previously, 4D printing enables unique metamaterial properties such as materials that act as switches or can freeze in specified positions. [112, 120] This type of metamaterial could be designed as an auxetic metammaterial and has the potential be used in confined applications such as deployable space structures. The metamaterial could be stored in a compacted state then released and expanded to a much larger form using only environmental stimulii. Additionally, the transforming nature of the material could enable tunable band gaps that can interact with an entire range of frequencies rather than a single frequency as demonstrated by previous works. [135] The majority of 4D metamaterial printing has been conducted on SMPs, but SMA are an emergening area. [136] These materials have not been specifically constructed as mechanical metamaterials so far, but the lesson learned from the current work can be easily extended to metamaterial design. For instance, Saedi *et al.* [137] showed that porous NiTi could be manufactured by SLM and based on the porosity, the mechanical properties could be tuned. Detailed simulation and optimization could extend the ideas demonstrated by Saedi *et al.* to construct mechanical, thermal, and/or acoustic metamaterials which are tunable depending on the application.

4.2 Applications

Additive manufacturing has not only enables unique properties in metamaterials, unattainable by conventional processing methods, but has also enabled the construction of parts for unique applications. The multimaterial system developed by Raza [127] was used to fabricate different LRM designs, which were used for energy harvesting. Likewise, Yin *et al.* [138] developed and demonstrated a radar absorbing structure fabricated by SLA. This application used alternating "woodpile" unit structures and patterned them in such a way that they become more dense near the structure to be protected from radar. This type of design can only be enabled by the freeform fabrication abilities of AM.

As previously mentioned, Sadeqi *et al.* [105] used SLA to partially develop a metamaterial embedded geometric optic. Through the use of AM, Sadeqi *et al.* were able to cover an optic in microwave absorbing metamaterials which absorbed a designed frequency. The authors noted that with increasing resolution and area, THz and optical metamaterials could be embedded in the optic. It is postulated that a system such as the one developed by Meza, Zheng *et al.* [96, 133, 134] combined with SMP printing could enable high resolution printing over the large desired area and also give a frequency tunable optic from the SMP actuation.

A class of mechanical metamaterials not yet discussed are those involved in origami structures as shown in Figure 5. This class of metamaterial uses periodic arrangements of folds to generate design with high flexibility, potential for deployability, and low weight. A number of applications are discussed in Zadpoor [74] but one of the most interesting of these are the use of 4D printing to construct the origami. This enables self folding and unfolding of the origami, greatly increasing its applicability. Liu *et al.* [139] used FDM to create 4D Miura-ori origami using SMPs, which showed 300% reversible volume change through only a thermal stimulus. Han *et al.* [140] have recently developed a multimaterial PμSLA system and suggest that it could be used for smart material printing. Han *et al.* used this machine to demonstrate a multi-responsive hydrogel beam through thermal and electrical inputs. Active origami is a field which uses SMAs embedded in elastomers to generate self-folding origami structures. [141] In this field, shape memory composites using SMA wires embedded in SMPs has been suggested as a mechanism to create self-folding origami that is able to stay locked into its actuated, folded position. [142] The utilization of a system like the one developed by Han *et al.* could expanded the possible design space of shape memory composite origami by enabling the simultaneous printing of SMAs and SMPs. The SMAs would not necessarily have to be embedded as wires but could be printed as a planar grid within an SMP that was printed similarly to the one printed by Liu *et al.*

The discussed applications are in no way exhaustive and AM can be used to fabricate almost any metamaterial for any application. There are, of course, restrictions such as part size to be built and material selection but these types of hurdles would need to be overcome with traditional fabrication as well. Some additional applications not discussed can be found in review articles of metamaterials and AM usages such as [13, 78, 97]. Some applications for AM fabricated metamaterials suggested in these works are supercapacitors, battery electrodes, and biomedical applications, which will be discussed briefly in the coming section.

4.2.1 Biological Applications

AM use for biological/biomedical applications is an innovation that makes use of the relatively low throughput of AM and the flexibility in design to create patient-specific structures and medical phantoms. These type of personalized parts could be for anything from dentures to hearing aids to prosthetics and recent work has begun to make use of metamaterials in biomedical applications. [143] For instance, a metamaterial can be fabricated using AM to exhibit auxetic behavior and this can be used to control local cell density for cell growth/regeneration.

Biological tissue is among the most mechanically complex materials to characterize and can exhibit hyperelasticity, viscoelasticity, incompressibility, and anisotropy with some materials exhibiting all these behaviors simultaneously. Additionally, biological materials (*e.g.* bone) can contain hierarchical structures or complex microstructures such as those seen in trabecular or cancellous bone. These features make metamaterials, which are based on lattice structures and can exhibit a near infinite range of mechanical properties, ideal for bio-mimicking materials. Johnson *et al.* [144] have done exactly that and used AM to fabricate metamaterials which were able to mimic a range of tissue behaviors. The use of metamaterials allowed them to use well understood elastic bio-compatible materials to create the lattices needed to mimic the bio-material and AM made the lattice construction possible. Similarly, Wang *et al.* [145–147] examined multimaterial AM to create tissue-mimicking phantoms. This multimaterial approach even more closely approximates the tissues and with further advances, it is conceivable that AM could be one day used to create entire limbs, which would aid in understanding of injurious events. In another work, EBM and titanium was used to fabricate auxetic metamaterials combined with traditional materials to create "meta-implants", which were able to reduce the risk of bone-implant interface failure. [148] The use of both metamaterials and conventional materials combined with the patient specific nature of the product could only be enabled by AM. Beyond just tissue-mimicking phantoms that capture the biomechanical response, AM fabricated metamaterials are able to grow/regenerate tissue using implanted cells. Raman *et al.* [93] used mulitmaterial PμSLA to create a cell and biomaterial pattern which replicated neurovasculature. The use of AM here was crucial to create the proper patterning which most closely mimicked live tissue so that the cells would be as close to *in-vivo* as possible. It is suggested that this type of multimaterial AM could be used in a wide range of applications, beyond the neurovasculature, including high-throughput drug testing and *in-vitro* disease development studies.

5. SUMMARY AND FUTURE WORK

This work has presented an overview of additively manufactured metamaterials including their unique properties and applications. A brief overview of AM and metamaterials was shown followed by a review of metamaterial fabrication methods. It was shown that many common methods used to fabricate metamaterials are similar to those used in AM and that the extension of metamaterial fabrication to AM was a natural and inevitable one. A review of AM process currently used for metamaterials fabrication was then shown and found that many AM technologies have already been used to fabricate metamaterials. However, many of these processes needed to be modified in order to be applicable to the small (nano/micro) scale required in many metamaterial designs, such as those in electromagnetic and optical applications. In the previous chapter, unique properties and applications of metamaterials fabricated by AM technologies were shown. Throughout the report, a number of knowledge gaps and opportunities for future developments have been made, a few of these points will be elaborated on now.

AM Fabrication Method Opportunity — As discussed, directed energy deposition (DED) is not an AM method that has been used for fabrication of metamaterials. The abilities of DED methods should be explored further since they can be applied to metallic materials and additionally have the capability to produce multimaterial and functionally graded parts, which powder bed based method cannot do. It is postulated that the reason for the lack of adoption in metamaterial fabrication is due to the relatively low dimensional accuracy in the method. However, the DED process is comparable to the aerosol jet printing method of Jahn *et al.* [115] with the exception that DED uses a laser to sinter the material as it is sprayed. High precision lasers are available such as those used in two-photon polymerization and combined with aerosol jet, could in essence result in a high precision DED method. Such a development could generate possibilities for unique metallic metamaterials with thermal, acoustic, or electromagnetic/optical properties in addition to high structural strength. This type of development could also be combined with functional grading of the material and topology optimization to yield structures that maintain their strength, decrease density, and could still function as metamaterial cloaks. Another, more easily accessible avenue for DED implementation to fabricate metamaterials is the use of a hybrid-AM system. This system utilizes both additive and subtractive techniques to produce high quality, high resolution parts. CNC subtractive processes can be as precise as a single micron, which could provide sufficient accuracy for many metamaterial applications.

AM Technique Development — Another possible development in AM technologies that could enhance the current state of art metamaterial fabrication is multi-resolution laser processing. In this theorized system, there would essentially be a macroscale laser that would shape the overall part and fabricate a large portion of the volume, then there would a more focused laser that would sinter the fine details in the metamaterial structure. Alternatively, the system could use a single laser that is able to be focused to different scales on the fly. The envisioned system would likely be implemented in a powder bed method for metallic materials. A major hurdle of this process would be powder processing and control as the powder particle would need to be fine enough to produce the required detail in either scenario. A system such as this, that could produce high resolution metallic parts, could aid in expansion of electromagnetic, optical, and thermal metamaterials fabricated by AM, thus giving researchers/engineers more design freedom while being able to maintain structural rigidity.

Microstructure Refinement and Mechanical Properties — In printing metallic materials with AM, it is well known that the microstructure is unique and very far from traditionally manufactured parts of the same material, typically exhibiting anisotropy and heterogeneity (*i.e.* porosity). It is also known that this unique microstructure is dependent on the AM process parameters and affects the mechanical behavior of the printed part greatly. In addition to the microstructure, residual stresses from the thermal processing are commonly developed and can warp finished parts, especially finer structures. The understanding of how residual stress and the unique microstructure are developed, and how they effect the final mechanical properties is highly studied in the AM community (see process-structure-property, PSP, linkages in the literature). This type of work needs futher development in the area of AM fabricated metamaterials, especially those which require high resolution structures that are particullarly susceptible to warping. The understanding of residual stresses and microstructure anisotropy and heterogeneity could lead to potential developments where the microstructure could be tailored to further enhance the metamaterial properties, *e.g.* utilizing/manipulating the anisotropic stiffness tensor throughout the structure to achieve even larger negative Poisson ratios than have been realized while maintaining axial stiffness. This sort of behavior has already been explored somewhat by Saedi *et al.* [137] with porous SMAs. In a related point, many studies on mechanical metamaterial examine the elastic, shear, and bulk properties of a given metamaterial but few do this with any cyclic behavior. Fatigue life is an important concern in any part that is to be implemented in a real application and this area needs further development in metamaterials, particularly for AM fabricates metamaterials where defects can serve as nucleation sites for crack growth.

Smart Materials and Composites — As already discussed smart composites have been proposed, which incorporate SMA wires in an SMP matrix. The SMP has the ability to lock the composite in its deformed state and thus actuation in applications such as origami can be maintained until subsequent activation/deactivation. The usage of AM to print origami metamaterials from smart materials has not been attempted yet but could offer much more flexibility in the composite than previously achievable. In addition to smart/4D composites, tunable 4D metamaterials need to be further examined. Some authors have started to recognize the ability to tune 4D metamaterials to specific frequency ranges, rather than single frequencies, but this has only been accomplished using an SMP for band gap tuning. Applications far beyond this are possible and should be examined. For instance, an SMA-based auxetc metamaterial could be design and printed so that, depending on the thermal stimulus, it has a different volume. The thermal stimulus could be an environmental condition and the volume change could help to facilitate cooling and maintain an optimal temperature.

Multipurpose Metamaterials — A final knowledge gap in the literature identified by this work is the use of metamaterials for multiple purposes *i.e.* acoustic-thermal or mechanical-thermal metamaterials. It is very easy for one to conceive of an application where both acoustic and thermal absorption/cloaking are both simultaneously needed (*e.g.* jet or UAV engines) or an application where a mechanical metamaterial property is desired but proper thermal conductivity or mass flow through the structure is also required (*e.g.* biomedical applications). There are no physical principle that prevents such a metamaterial from being built and with computational abilities continuing to increase, the design of such a metamaterial is feasible. It is likely that the more requirements that are placed on the design of the metamaterial, the more complex the structure would become. Thus, AM would likely be required to fabricate the designed metamaterial.

As a final concluding remark, it should be noted that in many ways, metamaterial development using AM is limited by the AM tech and not necessarily an understanding of the metamaterial structure. AM is a complex process that will take time to fully understand and exploit but with its development, metamaterials can become more complex and the bounds of possible properties can continue to be pushed.

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