Abstract
Material properties are governed by the chemical composition and spatial arrangement of constituent elements at multiple length-scales. This fundamentally limits material properties with respect to each other creating trade-offs when selecting materials for specific applications. For example, strength and density are inherently linked so that, in general, the more dense the material, the stronger it is in bulk form. We are combining advanced microstructural design, using flexure and screw theory as well as topology optimization, with advanced additive micro- and nanomanufacturing techniques to create new material systems with previously unachievable property combinations – mechanical metamaterials. The performance of these materials is fundamentally controlled by geometry at multiple length-scales rather than by chemical composition alone. We have demonstrated designer properties of these mechanical metamaterials in polymers, metals, ceramics and combinations thereof. Properties include ultra-stiff lightweight materials, negative stiffness, and negative thermal expansion. Our manufacturing techniques include Projection Microstereolithography (PµSL), Direct Ink Writing (DIW), and Electrophoretic Deposition (EPD). These tools are capable of generating the designed structures which are highly three-dimensional micro- and nanoscale architectures with multiple constituent materials in the same structure.

Introduction
Material properties can be controlled via intricate assemblies and structural organization at multiple length-scales as evidenced by naturally occurring cellular materials such as honeycombs [1], trabecular bone [2], plant parenchyma [3], and sponges [4]. It is the architecture of the material’s structure at the micro- and nanoscale, as much as the chemical composition that leads to its mechanical properties. By designing highly ordered architectures in cellular solids, we
open up the possibility of engineering the mechanical response of these materials to create so-called mechanical metamaterials [5,6].

The ability to decouple properties via micro- and nanoarchitectural control can allow for unique material performance such as ultra-lightweight, high stiffness and high strength materials [7,8], negative Poisson’s Ratio [9], negative stiffness [10], and negative thermal expansion coefficient [11]. Paramount to achieving these engineered, and often unnatural properties, is an ability to design, fabricate, and characterize these structures for the properties of interest. In fact, this methodology of choosing a unique property and engineering a material’s performance via architecture could be described as an *inverse design* problem. Normally, material properties are taken to be absolute and functional structures are then created from these materials. Mechanical metamaterials are exactly the opposite.

A classic example of architectural control and the resulting unique material performance is the octet truss stretch dominated lattice [5] shown in Figure 1. This structure which contains $b$ struts and $j$ frictionless joints satisfies Maxwell’s criterion, where $b - 3j + 6 > 0$ which defines a stretch dominated structure. Because the struts in the unit cell are designed to be in either tension or compression under applied load, as opposed to bending, the lattice is mechanically efficient with a high stiffness-to-weight ratio ($E/\rho$). In fact, it is designed to have a linear scaling relationship between stiffness and density, $E/E_s \propto (\rho/\rho_s)$ where the subscript $s$ denotes bulk properties. Most naturally occurring materials with stochastic porosity have a quadratic or even cubic relationship; for every order of magnitude decrease in density there is a corresponding 2 to 3 order of magnitude decrease in stiffness. The architected design fundamentally changes the scaling relationship of the lattice material through geometry rather than composition. This concept can be further advanced by taking advantage of nanoscale size effects. Strength to density relationships can be effectively manipulated with control at size-scales below the critical flaw and crack dimensions [12].

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**Design**

Numerous methods can be used so solve the inverse design problem for mechanical metamaterials based on architected microstructure. We have primarily been developing and utilizing two techniques; one analytical and the other computational. The analytical method is known as *Freedom, Actuation, and Constraint Topologies* (FACT) and it relies upon design of flexure and screw elements to create unit cells and lattices with prescribed properties [13,14]. The computational method we have been using is *Topology Optimization* (TO) which involves optimizing a unit cell’s layout subject to an objective function and boundary conditions.

The FACT method relies upon using a previously developed, comprehensive library of geometric shapes that define fundamental flexure and screw motion. These shapes enable the designer of a unit cell to visualize all the regions wherein various microstructural elements may be placed to achieve desired bulk material properties. As an example, consider a two dimensional unit cell design with negative thermal expansion that was derived using FACT and is shown in Figure 2 [15]. In this unit cell, two materials plus void space are required to achieve the negative property. As the unit cell heats up, the red material, which has a larger thermal expansion than the gray material, volumetrically expands more relative to its gray counterpart. Consequently, the red angled component pulls the center of the flexure element which makes up the sidewall of the unit cell inward, while simultaneously pushing the corners of the unit cell outward. When arranged in a lattice connected at the midpoint of each sidewall, the corners grow into the void space while the sidewall are pulled inward resulting in an overall contraction of the lattice and hence, negative thermal expansion. The FACT technique can be used to design other mechanical metamaterials with properties such as negative Poisson’s ratio and non-linear responses.

![Figure 2. Unit cell and lattice with negative thermal expansion designed using FACT [15].](image)

Topology optimization is a computationally driven inverse design method. In our implementation, we utilize a finite element solver as the core physics engine and an optimization
algorithm subject to an objective function and constraints to evolve the design. Again, we will use a negative thermal expansion metamaterial as an example. In a typical implementation, we begin with a unit cell with three phases randomly distributed throughout the space; a high thermal expansion constituent material, and relatively lower thermal expansion constituent material, and void space. An objective function such as a specific target thermal expansion for the unit cell is defined along with quantitative constraints such as stiffness and volume fraction bounds. Initially, the finite element solver calculates the material properties for the random distribution. The will likely be far from the target and may violate the constraints. At this point, the optimization algorithm, which in this case is a gradient-based method, will redistribute the three phases some small amount and call the finite element solver to again calculate properties. The new properties are then evaluated against the target and the previously calculated values and the optimization algorithm again redistributes material based on this information in an attempt to approach the target. This iterative process is repeated until it converges to a design which minimizes the objective function and satisfies all constraints. In some cases it may not converge due to over-constrained problems or poor initial conditions. An example of a topology optimized negative thermal expansion unit cell design is shown in Figure 3. There are significant limitations to these methods including a lack of knowledge regarding practical manufacturing constraints in the codes, a propensity to converge to a local minimum solution rather than a global one, and for more sophisticated design problems it can be computationally expensive requiring high performance computing resources.

**Fabrication**

In order to physically realize mechanical metamaterials, a suite of fabrication processes with unique capabilities is required. Due to the geometric complexity of these structures and lattices, additive manufacturing (AM) methods are particularly well suited as a fabrication technique for this application. However, there are still features and geometries in these structures that are not

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attainable with commercially available AM tools requiring us to develop our own custom processes and materials.

Projection Microstereolithography (PμSL), one of the processes we have been developing, uses a spatial light modulator — a liquid crystal on silicon (LCoS) or digital micromirror device (DMD) — as a dynamically reconfigurable digital photomask to fabricate three-dimensional materials in a layer-by-layer fashion. A three-dimensional CAD model is first sliced into a series of closely spaced horizontal planes. These two-dimensional image slices are sequentially transmitted to the reflective LCoS chip, which is illuminated with UV light from a light emitting diode array. Each image is projected through a reduction lens onto the surface of a photosensitive resin. The exposed liquid cures, forming a layer in the shape of the two-dimensional image, and the substrate on which it rests is lowered, re-flowing a thin film of liquid over the cured layer. The image projection is then repeated with the next image slice forming the subsequent layer until the desired number of layers has been fabricated to complete the 3D structure. A schematic of the basic system is shown in Figure 4 along with an example structure [16]. Additionally, we have recently developed a scanning version of this concept which enables us to rapidly fabricate structures approaching ten centimeters in size while maintaining features as small as ten microns.

Another fabrication method that we have been utilizing to fabricate these metamaterials is Direct Ink Writing (DIW). DIW is a layer-by-layer printing approach in which concentrated inks are deposited in planar and 3D layouts with lateral dimensions (minimum ~400 nm) that are at least an order of magnitude lower than those achieved by conventional extrusion-based printing methods. Paramount to this approach is the creation of concentrated inks that

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can be extruded through fine deposition nozzles as filaments which then undergo rapid solidification to maintain their shape as shown in Figure 5 [17,18]. In many cases they can even span gaps across unsupported regions [19]. Direct-write techniques, such as direct ink writing, offer an attractive alternative to conventional manufacturing technologies, due to the low-cost of the printing equipment, ease of manufacture, and flexibility in material systems and dimensions.

Finally, a third fabrication method which adds to our ability to deposit a range of materials is Electrophoretic Deposition (EPD). EPD is a bottom-up fabrication process that utilizes electric fields to deposit charged nanoparticles from a solution onto a substrate [20, 21]. EPD can be used with a wide range of nanoparticles including oxides, metals, polymers, and semiconductors. Once the particles are deposited the green body can be dried and/or sintered to adhere the particles together into a fixed structure. A schematic and fabricated nanostructure is shown in Figure 6. Traditionally EPD has been used for coating applications such as depositing ceramic materials onto metal tooling. We have expanded existing EPD technology to enable patterning of mesoscale, multimaterial structures, with micron-scale tailoring. Our modifications to traditional EPD include automated sample injection during deposition to tailor the material composition, dynamic electrodes that controllably vary the electric field profile on the deposition plane to precisely pattern geometries, and in-depth process modeling to predict the deposition parameters required to achieve a specific packing structure.

![Figure 6. Schematic of the EPD process and a fabricated structure.](image)

In addition to these fabrication processes, there are many available post-processing techniques that can help to expand the useable materials palette and/or improve final component properties. Thermal treatments such as sintering and hot isostatic pressing are commonly used. Also, using polymer structures as templates for other materials is another common method for accessing an

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expanded material set. For example, a polymer structure could be fabricated with any one of these methods and subsequently coated via electroless plating, atomic layer deposition (ALD), or CVD processes. The polymer in these hybrid structures could then be thermally or chemically removed converting the structure to pure metallic or ceramic hollow structures. Finally, nanoparticles can be suspended in the base feedstocks such as liquid monomers, again resulting in an interesting hybrid structure with particles distributed throughout polymer. Thermal processing can be used to remove the polymer and densify the particles leaving a relatively pure, non-polymeric structure.

**Performance**

Mechanical metamaterials are now becoming a reality due to advanced fabrication and design methods. Two examples of simple lattice-based materials with unique properties include ultralight, ultrastiff microlattices and elastomeric cellular architectures with negative stiffness. This structures and their performance highlight many of the key concepts already discussed.

Figure 7 shows an example of octet truss stretch dominated lattices fabricated at the microscale using PμSL. In the first column of the figure, a basic polymeric lattice made from HDDA (hexanediol diacrylate) with solid microscale struts can be seen with 11% relative density. In the next column, the same polymer structure was electrolessly plated with Ni-P and the polymer core was subsequently removed via thermal processing resulting in a lattice with 0.5% relative density. A hollow tube ceramic lattice, shown in the third column, was formed via ALD and similar polymer removal. This structure represents the lightest fabricated material in this test series with a relative density of 0.025% and wall thickness below 50 nm. Finally, we fabricated a lattice with alumina nanoparticles suspended in the polymer and conducted sintering procedures to remove the polymer and densify the ceramic. A solid ceramic lattice was obtained with 8% relative density [22].

The performance of these mechanical metamaterials is highlighted in Figure 8 where non-dimensional stiffness is plotted versus relative density. Note the measured scaling relationship between these two parameters is approximately linear across all constituent material types, all relative density regimes, and regardless of hollow tube or solid strut configurations clearly

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demonstrating the impact of the stretch dominated architecture. For comparison, a bend dominated Kelvin foam architecture was fabricated and tested displaying the classic quadratic relationship. Furthermore, in absolute terms, the hollow tube alumina lattices have densities approaching aerogels (known for being some of the lightest materials in the world) but with 4 to 5 more orders of magnitude in stiffness due to the architected structure [22].

Another interesting mechanical metamaterial is shown in Figure 9. These woodpile structures were fabricated out of silicone with the DIW process. By simply varying the architecture between a “simple cubic” (SC) type structure and a “face centered tetragonal” (FCT) configuration, different bulk-scale mechanical properties can be achieved. From the cross sections shown in the figure, it becomes clear that the two structures will have different compressive

Figure 7. Octet truss-based mechanical metamaterials of varying materials and configurations [22].

Figure 8. Non-dimensional performance of ultralight octet truss, stretch dominated lattices[22].
behavior with the SC layout being stiffer than the FCT structured material due to the alignment of the nodes [23].

However, what is not as obvious is the difference in shear response of the two materials. Figure 10 elucidates this difference which manifests itself as “negative stiffness” and can be seen in the negative slope of the stress-strain response in the SC material. This is a unique “snap through” property which can now be engineered into the material. Not only can we control the compressive response, but we can somewhat independently design and control the shear response, possibly even to have this uncommon negative property in this elastomeric mechanical metamaterial [23].

**Future Directions**

By combining the concept of mechanical metamaterials with inverse design methods and custom micro- and nano- additive manufacturing techniques, we have been able to demonstrate unique properties not previously attainable in known materials. However, this is just the beginning of a powerful new methodology for approaching material design and realization.

There are many potential future directions that one could take to advance the state of the art including continued exploration of size-scale effects and pushing the boundaries of multimaterial design and fabrication. However, one interesting direction that we have begun to explore is the concept of multifunctional metamaterials – combining normally disparate physics into lattice type architectures. An initial example of this can be seen in Figure 11 where we show a printed...
graphene aerogel structure [24]. This simple woodpile lattice has enhanced mechanical properties but due to the nature of graphene, there can be significant electrical and/or chemical functionality as well. Super-capacitors with high compressibility and durability for example, may become possible.

References


Figure 11. Image of printed graphene aerogel along with mechanical and electrical performance data [24].

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