

1 **Design and Printing of Proprioceptive 3D Architected Robotic Metamaterials**

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1 **Summary paragraph**

2 Piezoelectricity converts electric fields to mechanical strain and vice versa. Despite their  
3 technological importance in robotics, actuators and transducers, only a small fraction of materials  
4 has been found to display piezoelectricity and the electric-field-induced strain is dictated by the  
5 available noncentrosymmetric crystal structures, limited to only normal and shear directions with  
6 low amplitude, and thus require extensive system assembly and integrations to produce complex  
7 motions with amplified amplitude. We report an inverse design and manufacturing route to create  
8 a new class of robotic metamaterials that display a wealth of previously inaccessible strain modes  
9 that bypass the limitations of natural crystals, capable of generating giant strain in an arbitrary  
10 direction from an electric field and vice versa, resulting in programmed, high-speed motions with  
11 self-sensing, feedback, as well as bidirectional ultrasonic transduction. Production of these  
12 materials are enabled by a multi-material additive manufacturing technique, capable of direct  
13 assembling piezoelectric, conductive and passive structural materials into complex micro-  
14 architectures. The resulting architected materials function as proprioceptive micro-robots capable  
15 of performing a variety of self-guided robotic tasks with feedback from self-sensed contact and  
16 remote stimuli. This work defines a possible route to create robotic metamaterials in the future,  
17 which actively sense and move comparable to a living system.

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## 1 **Introduction**

2 Recently, advances of additive manufacturing techniques have enabled the realization of novel  
3 properties that were previously inaccessible, such as ultra-high stiffness<sup>1</sup> and damage tolerance<sup>2</sup>,  
4 exotic mechanical behaviors<sup>3</sup>, negative thermal expansions<sup>4</sup>, fluid control<sup>5</sup> and wave  
5 transmissions<sup>6</sup> that can be digitally designed via placing structural elements as bits and atoms in a  
6 three-dimensional layout. The potentials of architecting electronic, multi-functional materials and  
7 their combinations are still elusive, as most additive fabrication techniques in the past decades  
8 focus on singular and structural materials, such as polymer, metal and ceramics. Architecting  
9 functional materials could offer exotic properties at the functional space, such as directional  
10 sensing<sup>7</sup>, morphing<sup>8</sup>, and reconfigurability<sup>9</sup>. However, unlike a biological system where a network  
11 of interconnected components cooperates to provide motion, force, perception, and feedback, the  
12 advances of architected materials have yet reached such system complexity. Design and  
13 manufacturing routes that move beyond structural materials to the seamless integration of  
14 electronic, functional and structural materials into complex three-dimensional (3D) micro-  
15 architectures are compelling and could potentially enable a wealth of self-regulated robotic  
16 materials that would be otherwise not possible in their native counterparts.

17 Piezoelectric materials, capable of converting electrical field into mechanical strain and vice  
18 versa<sup>10</sup>, are widely employed in precision actuators<sup>11</sup>, manipulators<sup>12</sup>, accelerometer<sup>13</sup>, tactile and  
19 ultrasonic sensors<sup>14,15</sup>, to generate robotic motions and sensing feedbacks. However, their electric-  
20 field-induced strain, as a result of asymmetric displacement of ions in crystal dimension, are  
21 derived from and thus limited by the naturally existing noncentrosymmetric crystal structures,  
22 leading to a magnitude lower than 0.5% in only normal and shear directions<sup>10</sup>. To utilize the  
23 piezoelectric materials as transducers in robotic systems, the manufacturing process must involve

1 extensive processing and assembly steps, including ceramic machining<sup>16</sup>, lamination<sup>17</sup>, attaching  
2 planar electrodes for both activating and driving<sup>18</sup> and integration with transmission mechanisms<sup>19</sup>,  
3 to amplify the piezoelectric strain and transform the strain into desired motion directions. These  
4 manufacturing routes usually only processes solid piezoelectric materials and are not able to  
5 precisely pattern the electrodes, making it impossible to reduce the weight of the actuation element  
6 and activate the bidirectional piezoelectric effects at small scales, which limits the application  
7 robotic platforms and prevent size reduction and improvement of integration density<sup>20</sup>.

8 Here we show that via 3D micro-architected metamaterials comprised of a network of  
9 interconnected piezo-active, conductive and structural phase (Fig. 1A), a two-way conversion from  
10 electric field to arbitrary mechanical strain in an arbitrary direction and vice versa can be achieved.  
11 We show, through analytical numerical and experimental validations that, without being limited  
12 by the available crystal structures<sup>21</sup>, these 3D micro-architected piezoelectric robotic  
13 metamaterials exhibit previously inaccessible piezoelectric strain constants, resulting in a wealth  
14 of electric field induced strain conversions, including tension, shear, twisting, flexure degrees of  
15 freedoms (DoF) and their combinations, as well as giant strain amplifications and logic subtraction  
16 and addition of strains. To implement these designs, we developed a charge-programmed multi-  
17 material additive manufacturing technique capable of directly assembling conductive, piezo-active  
18 and structural materials into a complex 3D micro-architecture. The design and manufacturing of  
19 multi-material 3D micro-architectures lead to the creation of robotic metamaterial, which directly  
20 serve as a micro-robot, capable of performing a wealth of robotic tasks, including locomotion,  
21 steering, stepping, two-way sound and ultrasonic transductions as well as decision making via a  
22 feedback control.

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## 1 **Rational design of robotic metamaterials with arbitrary strain modes**

2 The core concept of architected material is the free placement of materials in a 3D cellular topology  
3 that either bypass limitations in natural crystals or mimic them to achieve desired properties. We  
4 introduce a convenient and robust strategy to architect piezo-active, conductive and structural  
5 phase (Fig. 1A) in 3D space. Such multi-material metamaterials are capable of taking an input  
6 electric field and outputting a desired mode of strain including new DoFs beyond the Cauchy strain  
7 components<sup>22,23</sup> (e.g., normal and shear strain), including normal, shear, twisting, and flexure, as  
8 well as their combinations and amplifications.

9 As the existing piezoelectric tensors are insufficient to describe the new DoFs, we define the  
10 generalized piezoelectric tensors  $d_{nm}$  and  $\bar{d}_{nm}$  to describe the strain conversions of the architected  
11 piezoelectric materials (Fig. 1B):

$$12 \quad \varepsilon_m = d_{nm}E_n; \varphi_m = \bar{d}_{nm}E_n \quad (1)$$

13 where  $E_n$  is the electric field along n-direction in the Cartesian coordinates (n=1-3);  $\varepsilon_m$  and  
14  $\varphi_m$  (m=1-6) are the directional strain and coupled strain tensors<sup>23</sup>, respectively;  $d_{nm}$  is the existing  
15 piezoelectric tensor; and  $\bar{d}_{nm}$  is the extended piezoelectric tensor.

16 To design a piezoelectric micro-architecture that display a desired global strain mode  $d_{nm}$  (or  
17  $\bar{d}_{nm}$ ) (Fig. 1B, see MOVIE S1 for 36 distinguished strain modes), we start by identifying the  
18 motion of a stack of virtual characteristic planes within a unit cell of the micro-architecture (Fig.  
19 1C and D) and the local strain of the piezo-active struts comprising the unit cell (Fig. 1E, MOVIE  
20 S1). The virtual characteristic planes can be considered “pinned” by the piezo-active struts to allow  
21 unconstrained motion and will display a motion representing the desired global strain. As shown  
22 in Extended Data Fig. 1a-d, the characteristic planes undergo distance change, slip, rotation and tilt

1 corresponding to strain modes including normal strain ( $\epsilon_m$ ,  $m=1-3$ ), shear ( $\epsilon_m$ ,  $m=4-6$ ), twist ( $\varphi_m$ ,  
2  $m=1-3$ ) and flexure ( $\varphi_m$ ,  $m=4-6$ ), respectively. Fig. 1D shows an example of a cube that twists  
3 upon application of electric field in the 3-direction ( $\bar{d}_{33}$  mode), which is represented by the in-  
4 plane rotation of characteristic planes (Fig. 1D). The local strain of a piezo-active strut, either  
5 expand or contract, within the unit cell is determined by the direction of the strut, the polarization  
6 and the electric field (see Methods about the details on the identification of the local strain). The  
7 electric field are generated by the conducting phase either covering the sides of the lattice topology  
8 (external electrodes, Fig. 1E①) or penetrating within the topology (localized electrodes, Fig.  
9 1E②), covering both sides of an active struts, leading to a shorter distance between electrodes  
10 and an elevated electric field.

11 The next design step is to place the piezo-active struts in a spatial layout such that their local strain  
12 will drive the virtual characteristic planes to display the motion path corresponding to the target  
13 global strain mode ( $d_{nm}$  or  $\bar{d}_{nm}$ ), as shown in MOVIE S1. Fig. 1F presents the designed piezo-  
14 active struts that generate a clockwise rotation of the characteristic planes, resulting in a new twist  
15 strain mode and corresponds to a nonzero piezoelectric twist coefficient,  $\bar{d}_{33}$ . 3D micro-  
16 architectural layout of piezo-active struts for other strain modes including expansion, shear and  
17 flexure, are summarized in Extended Data Fig. 1e-h. The full unit cell design is then completed by  
18 adding structural phase and conductive phase (Fig. 1G) in a layout that matches the symmetry of  
19 the piezo-active struts.

20 In the last step, we tessellate the unit cells into a “metacrystal” that reflect effective response of  
21 the unit cell (see SI. 1 and Extended Data Fig. 1i-l for choice of tessellation orientations). For  
22 example, to generate a twisting metamaterial, we tessellate the unit cell in cylindrical coordinates  
23 along the radial ( $r$ ), angular ( $\theta$ ) and height ( $z$ ) directions, as shown in Figs. 1H-I and MOVIE S1.

1 This aperiodic tessellation bypasses the effect of the number of unit cells when tessellated in  
2 Cartesian coordinates, where the twist strain would vanish when reduced to a Cauchy continuum  
3 as the unit cell number increases<sup>24</sup> (Extended Data Fig. 2). As such, these metamaterial concepts  
4 are scalable, where the coefficient is invariant of the number of unit cells in all directions. Designs  
5 with all strain modes shown in the extended matrix are summarized in Table S1.  
6 Additionally, the interpenetrated piezo-active, structural and conductive phase allow amplification,  
7 logic subtraction and addition of strains. Fig. 1J shows the unit cell design featuring amplified  
8 expansion via tessellating pairs of piezo-active and structural struts<sup>25</sup>. Fig. 1K demonstrates the  
9 design with localized electrodes. The localized electrode architecture that cover selected groups of  
10 struts allows for programming the polarization and electric field direction within the unit cell (Fig.  
11 1K, see Extended Data Fig. 3 for polarization and driving voltage programming), thereby  
12 achieving compound strain mode with both shear and expansion strain output or decoupled strain  
13 mode with added up shear and suppressed expansion.  
14 We implemented a computational framework that predicts and verifies the piezoelectric electric  
15 coefficients  $d_{nm}$  and  $\bar{d}_{nm}$  of the metacrystal designs. This is achieved by calculating the total  
16 induced-load contribution of the connected piezo-active struts within the cubic volume under an  
17 electric field (Methods section and SI. 2). SI. 3 and Extended Data Fig. 4 shows the finite element  
18 analysis results that verify the actuation modes.

## 19 **Additive manufacturing of robotic metamaterials**

20 To implement our design, we developed a charge programmed multi-material additive  
21 manufacturing technique capable of assembling piezo-active, structural and conducting phase into  
22 a complex 3D micro-architecture.

1 First, a negatively charged resin that can be selectively deposited with metals<sup>26</sup>, and a highly loaded  
2 nanoparticle colloid were printed via a multi-material 3D printing system (see details in the  
3 Methods section), as shown in Fig. 2A. The nanoparticle colloid consists of 15~50vol% lead  
4 zirconate titanate (PZT, APC855), featuring high piezoelectric strain constant (i.e.  $d_{33}=600\text{pm/V}$ )  
5 and 5~15% lead nitrate, which is used as a liquid phase sintering agent<sup>27</sup>. Then, the conductive  
6 phase was selectively deposited onto the charged resins, forming a 3D micro-architecture with  
7 electrodes (see Fig. 2B and Methods). The deposited architectures were cosintered with optimized  
8 temperature profile and gas environment (Extended Data Fig. 5) to prevent degradation of  
9 piezoelectricity of the piezoceramics and conductivity of the electrodes (Fig. 2C). Additionally,  
10 lead oxide was used to provide a liquid sealing and lead-rich environment to suppress lead  
11 evaporation of PZT at temperatures over 800°C (Fig. 2D and SI.4). The lead nitrate particles also  
12 melted and induced contraction of the PZT particles due to its surface tension, which densifies  
13 PZT and reduces porosity. Next, the architected piezoceramics are polarized by applying a high  
14 electric field through the deposited metals under elevated temperature (SI. 4 and Extended Data  
15 Fig. 5). After polarization, the regions not covered with electrodes remain inactive (unpolarized)  
16 and are employed as the structural phase (Fig. 2E). Other ceramics, such as silicon oxycarbide  
17 ( $\text{SiOC}$ )<sup>28</sup>, can also be employed herein as the structural phase to enhance the stiffness of the  
18 metamaterial. Our technique enabled the construction of 3D architectures with high-performance  
19 piezoceramics, highly conductive metals and structural structural materials, allowing the creation  
20 of multi-functional devices via additive manufacturing

21 The presented 3D fabrication approach allows the fabrication of piezo-active materials with  
22 precise, micro-scale 3D architectures and low porosity (Fig. 2F-H). Fig. 2K shows a robotic  
23 metamaterial block with embedded electrodes. The gapless bonding between the piezo-active and



1 conductive phases (Fig. 2L) leads to minimal dielectric loss during electric field application. The  
2 minimum achievable feature size of the conductive phase reaches 20  $\mu\text{m}$ , as shown in Fig. 2M,  
3 ensuring precise control of the localized electric field when applying voltages through these  
4 conductive phases. The as-printed piezoelectric solids reach a measured piezoelectric constant as  
5 high as 583 pm/V  $d_{33}$  (measured with a  $d_{33}$  meter, APC 90-2030), which outperforms all existing  
6 3D printable piezoelectric composites or ceramics (Fig. 2N)<sup>29-35</sup>. The selectively patterned  
7 conductive phases are highly conductive (SI. 5, Fig. S3), achieving  $3.7 \times 10^6$  and  $1.35 \times 10^7$  S/m  
8 after sintering, for copper (Cu) and gold (Au), respectively (Fig. 2O), making them suitable for the  
9 subsequent actuation with negligible energy loss. The high precision fabrication, highly responsive  
10 3D printed PZT and the highly conductive deposited metals enable the implement of the  
11 metamaterial designs.

## 12 **Arbitrary, amplified and programmed strain**

13 To verify the arbitrary strain modes of the robotic metamaterial, we measured the electric field  
14 activated deformation of as-fabricated and poled samples via a high-precision full-field laser  
15 vibrometer (Polytec, PSV-500) (Fig. 3A). Quasi-static driving voltage with 50V amplitude was  
16 applied through the layered external electrodes, while the activated deformation is measured by  
17 tracing the side surface of the metamaterial (as shown in Fig. 3A) and reconstructed as a surface  
18 plot for visualization. Figs. 3B-C show an optical image of a twist ( $\bar{d}_{33}$ ) mode lattice with external  
19 electrodes and its side surface deformation, showing a wrapped shape corresponding to a twisted  
20 solid (Fig. 1C). Deformation measurement for shear ( $d_{34}$ ) and flexure ( $\bar{d}_{35}$ ) mode lattices are  
21 summarized in Fig. 3D-E and Fig. 3F-G, respectively. Different strain modes can also be combined  
22 into a single element piezoelectric lattice to achieve a selection of multiple strain modes and multi-  
23 DoF metamaterial actuator with six individually actuatable architecture designs full-filling 6-DoF,

1 including the  $d_{33}$ ,  $d_{35}$ ,  $\bar{d}_{33}$ ,  $\bar{d}_{34}$ ,  $\bar{d}_{35}$  and  $d_{36}$  modes (Fig. 3H-I, MOVIE S2). This multi-DoF  
2 actuator has a density of  $0.88/\text{cm}^3$ , making it among the most compact 6-DoF actuators with high-  
3 speed and precision motions, with a density an order of magnitude lower than existing piezo-  
4 actuators with dense PZT<sup>20</sup> (Fig. S4). The multi-DoF actuator is capable of high-speed high-  
5 precision motions. As an example, we demonstrate a high-speed galvanometer actuated by two  
6 multi-DoF actuators assembled with two reflective mirrors (MOVIE S3). Extended Data Fig. 6  
7 shows a star pattern drawn by the steered laser beam (10 cycles at 20 Hz, with an RMS precision  
8 of  $50\pm 13\ \mu\text{m}$ ) by programming the input voltage that controls the tilt angle of the metamaterial  
9 (see SI. 6 for the details of the laser pattern reconstruction).

10 Next, we fabricated micro-architectures with embedded electrodes and demonstrate the strain  
11 amplification, strain compounding, strain addition and subtraction enabled by the localized  
12 electrodes and the micro-architecture design. Fig. 3J, L&N presents as-fabricated lattice lattices  
13 with localized electrodes, featuring amplified expansion ( $d_{33}$ , Fig. 1J), amplified twist ( $\bar{d}_{33}$ ,  
14 Extended Data Fig. 3) and amplified shear ( $d_{34}$ , Fig. 1K), respectively. The displacement versus  
15 the driving voltage of lattices is captured with the laser vibrometer and plotted in Fig. 3K, M&O,  
16 which were used to derive the corresponding strain coefficients. Compared to the strain  
17 coefficients of lattices with layered external electrodes, the localized electrodes achieve giant  
18 piezoelectric coefficients (i.e.  $d_{33}=10050\text{pm/V}$ ), which is two orders of magnitude higher than  
19 that of their native material ( $583\text{pm/V}$ ). Fig. 3P-Q demonstrate the compound expansion-shear  
20 strain and the decoupled pure shear strain using designs from Fig. 1K, via programming the local  
21 polarization and driving electric field within the embedded electrode architectures.

## 1 **Robotic metamaterials as proprioceptive microrobots**

2 These piezoceramics architectures possess bidirectional electrical-to-mechanical energy  
3 conversion, namely its direct and inverse piezoelectric effect. We show that these metamaterials  
4 with their strain amplification, multi-DoF strain modes as well as the direct piezoelectric effects,  
5 can be directly exploited as a proprioceptive micro-robotic system with feedback control without  
6 the need to combine transmissions and external sensors. Here, within a single piece of piezoelectric  
7 architecture, selected regions serve as actuation limbs utilizing the inverse piezoelectric effect,  
8 while other regions utilizing the direct and bidirectional piezoelectric effect serve as strain sensor  
9 and pulse-echo element, respectively (Fig. 4A).

10 To demonstrate this concept, we fabricated one piece of metamaterial with amplified  $d_{33}$  mode  
11 (Fig. 1J) as a multimodal microrobot (Fig. 4B), within which selected regions serve as robotic  
12 limbs capable of actuation, load carrying, as well as sensing of both contact forces and remote  
13 objects. The microrobot is controlled by a closed-loop system via its sensing components  
14 (Extended Data Fig. 7). The microrobot consists of three individually actuatable regions serving  
15 as the body, the rear leg and the front leg (Fig. 4B). The coordinated motion of these three regions  
16 enables three distinguishable modes, including walking, turning and jumping modes (see Figs. 4B,  
17 Methods, Extended Data Fig. 8a-b for motion planning based on the three limbs). The 3D localized  
18 electrode interconnects all piezo-active elements within one region and minimizes the number of  
19 connection leads, enabling a compact design without the need for additional circuit boards or  
20 wiring leads to individual piezo-active element, making the microrobot lightweight (740 mg).

21 In contrast to current actuation mechanism where a leveraging/transmission mechanism is  
22 employed to amplified displacement at the cost of reduced blocking force, a unique feature of the  
23 actuation performance of the robotic metamaterial is its simultaneously increased blocking force

1 and displacement via its skeletal actuator layout. An actuator connected with a transmission system  
2 for robotic motions typically leads to a trade-off between blocking force and stroke due to the  
3 compliant levering mechanism involved<sup>36,37</sup>. We show that these architected piezoelectric robotic  
4 limbs (Fig. 4A), comprised of a network of microscale actuation units distributed throughout the  
5 robot body in a fashion similar to skeletal muscles, breaks this trade-off and simultaneously  
6 produce high stroke, high blocking force with high system stiffness. A comparison between the  
7 skeletal actuation architecture and actuator-transmission mechanism is described in Extended Data  
8 Fig. 9 and SI. 7. While a transmission system effectively amplifies the displacement, its additional  
9 levering inevitably reduces system stiffness and thereby blocking force (Fig. 4C), resulting  
10 decreased payload of the robot at driven at higher speed. The interconnected micro-actuator and  
11 embedded electrode micro-architecture allows driving voltage to be locally applied at each element,  
12 enabling a higher driving electric field thus amplified displacement, while simultaneously  
13 maintaining the overall stiffness through the interconnections of the unit cells without compliant  
14 transmissions. These unique features enhance performances of the microrobot, including high  
15 speed, high payload and high resonance.

16 The large stroke and high resonance of the metamaterial limb enable a maximum speed of 128  
17 mm/s in the walking mode and a turning rate of  $90^\circ/\text{s}$  (Fig. 4D, MOVIE S4, see Extended Data  
18 Fig. 8c for the frequency response of the limb), making this robot one of the fastest and lightest  
19 small-scale piezoelectric locomotion robots (Method, Extended Data Fig. 8d). With its broadband  
20 response (Extended Data Fig. 8c), the robotic metamaterial achieves short rising time to impulse  
21 voltage inputs<sup>38</sup>. This enables the immediate release of actuation energy within 1ms, lifting the  
22 front leg up into the air and subsequently lifting the body and rear leg, thus allowing the microrobot  
23 to step up from the ground (MOVIE S4). The jumping modes enable the microrobot to climb over

1 rough terrains (Fig. 4E, MOVIE S4), which is not achievable in piezoelectric microrobots with  
2 small strokes<sup>39,40</sup>. The high stiffness of the piezoelectric metamaterial elements enables a high  
3 system payload. Various weights (1g~5.6g) are loaded on the microrobot and its speed is captured  
4 by digital image correlation. As shown in Fig. 4F, over 80% of the original speed is retained for a  
5 500% payload, compared to an over 80% speed reduction with less than a 200% payload for  
6 microrobotic systems utilizing a compliant mechanism<sup>36,41</sup>. The high payload capacity allows  
7 integration of onboard devices, opening possibilities for onboard power sources and control within  
8 the microrobot.

9 By assigning sections of the piezo-active phases as the sensing element utilizing its direct  
10 piezoelectric effects via the 3D embedded electrodes, we demonstrate that the robotic metamaterial  
11 is capable of self-sensing its strain change and responding to external stimuli, which allows closed-  
12 loop control and quick reacts to external stimuli. As shown in Fig. 4B, the selectively deposited  
13 electrodes separate a strain sensing section from a piezo-active element with a 100  $\mu\text{m}$  isolation  
14 gap, which eliminates electrical coupling. This section is then integrated with a micro-controller  
15 forming a close-loop control system that receives the sensing voltage and sending control voltage  
16 to the actuation regions. The self-sensing voltage signal updates as the actuator element deforms,  
17 allowing real-time monitoring of the gait status and stimuli-responses (see Extended Data Fig. 7  
18 and SI. 8 for control loop design). Figs. 4G demonstrates the gait monitoring signal, the detection  
19 of the external impact and programmed reaction within 0.1ms upon a drop weight impact during  
20 locomotion. MOVIE S5 shows other programmed reactions including stop or making a turn upon  
21 detecting an impact.

22 The robotic metamaterial is also capable of sensing and reacting to non-contact stimuli via the  
23 pulse-echo element functioning with bidirectional piezoelectric effects. As shown in Fig, 4B, an

1 element on the frontal leg is capable of emitting and receiving focused ultrasonic waves with a  
2 semispherical acoustic lens printed with the structural phase. The echo signal (Extended Data Fig.  
3 10 and Methods) from the remote object is processed by the closed-loop control system that  
4 updates the driving voltage to navigate through an “S” shaped path and avoid remote obstacles  
5 (see Fig. 4H-I and Movie S6). The high integration density makes the metamaterial one of the  
6 smallest robots to date capable of multimodal locomotion, proprioception and closed-loop obstacle  
7 avoidance.

## 8 **Discussion and outlook**

9 We introduced a strategy to design and additive manufacture a class of robotic metamaterials that  
10 incorporate electronic, structural and conducting micro-scale strut elements in a 3D architecture.  
11 These multi-functional metamaterials uncovered a myriad of new strain modes including twisting,  
12 flexure, compound, decoupled and amplified strain without combing any leveraging or  
13 transmission system at a fraction of the weight. This bypasses the limitations of natural  
14 piezoelectric crystals where the piezoelectric strain relies on the available natural crystalline  
15 structures, of which only a fraction of the tensor has been discovered and measured thereby limited  
16 to only normal and shear directions with low amplitude. The design strategy can be further  
17 combined with a topology optimization algorithm to generate arbitrary piezoelectric tensors.

18 While current additive manufacturing either produce soft polymers, or ceramics and metals alone,  
19 no techniques allow the co-printing of functional, conducting and structural materials. The multi-  
20 material additive manufacturing technique reported herein patterns piezoceramic, metallic and  
21 structural materials into a complex 3D architecture. The resulting metamaterial blocks with milli-  
22 to-centimeter dimensions are capable of outputting multi-DoF motions with high blocking force  
23 as well as sensing contact and remote stimuli, completely free from external sensors, transmissions.

1 With continuing development in colloidal material synthesis, and emerging multi-material 3D  
2 printing techniques to rapidly pattern multiple materials, these robotic metamaterials may involve  
3 other electronic functionalities, such as millimeter wave transmission and untethered operations.  
4 Moreover, as the rapid advancement of electric converter design pushed the envelope of power  
5 miniaturizations, embedded low power battery and a voltage converter to drive the material will  
6 expand these intelligent materials to be completely autonomous with a feedback control. The  
7 reported design framework and manufacturing methods have direct implications for miniaturized  
8 robots, transducers, and robotic materials in the future, where desired motions and decision making  
9 can be realized via simplistic artificial material.

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1 **Figure legends**

2 **Fig. 1 Rational designs of robotic metamaterials with arbitrary strain modes** A) Schematic of  
3 the piezoelectric metamaterial consisting of piezo-active, conductive and structural phases. B)  
4 Schematic of various strain modes and piezoelectric strain matrix with extended tensors (red), as  
5 enabled by metamaterial design in contrast to only 18 tensors in natural piezoelectric ceramics  
6 (blue). C-I) Schematic of the design rationale of the piezoelectric metamaterials. The example  
7 shown here is for the  $\bar{d}_{33}$  (twisting) mode. J) Designs of piezoelectric robotic metamaterial with  
8 amplified expansion. K) Schematic of the compound and decoupled expansion-shear mode with  
9 strain amplification.

10 **Fig. 2 Multi-material fabrication platform** A-B) Schematic illustration of the custom 3D  
11 printing system and multimaterial composite lattice with programmed surface charges and  
12 selective deposition of 3D electrodes. C-D) Schematic of the sintering process of the highly loaded  
13 piezoelectric ceramics with localized electrodes. E) Schematic of the activation (polarization)  
14 process of the piezo-active phase. F-I) Optical image of piezoelectric thin mesh, spiral, bridge and  
15 coil. J) Scanning electron microscope (SEM) image of the cross-section of the sintered PZT. K)  
16 Optical image of a piezoelectric metamaterial block with 3D electrodes. L) SEM image of the  
17 boundary between the PZT and Au electrode. M) Elemental mapping of a selectively deposited  
18 thin wire, showing the minimum deposition width, W. N) Comparison of piezoelectric charge  
19 coefficients between piezoceramics presented in this study and existing 3D-printed piezoelectric  
20 materials. O) Conductivity of Cu and Au conductive phases before and after sintering.

21 **Fig. 3 Experimental verification of robotic metamaterial designs** A) Schematic of the test set  
22 up with laser vibrometer to capture the deformation of the side surface of the lattice. B-G) Optical  
23 image of the twist, shear and flexure mode lattice and their side surface deformation. H) Optical

1 image of a modularized 6-DoF piezoelectric actuator. I) Deformation of the side surface of the 6-  
2 DoF piezoelectric actuator working in three representative working modes ( $\bar{d}_{35}$ ,  $\bar{d}_{33}$  and  $d_{36}$   
3 modes). J) Optical image of a piezoelectric metamaterial with amplified expansion. K)  
4 Displacement of the metamaterial with localized electrodes and solid PZT material with external  
5 electrodes as a function of the input voltage. L) Optical image of the piezoelectric metamaterial  
6 with amplified twist and suppressed expansion. M) Twist angle of the metamaterial with external  
7 and localized electrodes as a function of input voltage. N) Optical image of the piezoelectric  
8 metamaterial with amplified shear. O) Shear displacement of the metamaterial with external  
9 electrodes and localized electrodes as a function of input voltage. P-Q) Displacement with  
10 compound and decoupled expansion-shear modes with sinusoidal voltage input.

11 **Fig. 4 Stimuli-responsive multimodal locomotion micro-robot** A) Schematic of the  
12 microarchitected piezoelectric metamaterial building block with an embedded actuation element,  
13 an embedded self-sensing element and a contactless detection element. B) Optical image of the  
14 locomotion robot. C) Working curves of a unimorph, a unimorph assembled with a transmission  
15 mechanism and the fabricated piezoactuator with multiple unit cells. D) Locomotion speed as a  
16 function of driving frequency. E) Optical image of the microrobot climbing over a 1 mm height  
17 obstacle in jumping mode. F) Normalized locomotion speed of the microrobot under various  
18 payload conditions. G) Sensing voltage and driving voltage as a function of time during the impact  
19 test. Inset: optical image of drop weight impact test of the microrobot with a basket. H) Locomotion  
20 trajectory of the microrobot navigating through an “S” shaped path. I) Locomotion trajectory of  
21 the microrobot with and without obstacles.

22

# 1 **Methods**

## 2 *Identification of the local strain*

3 We identify the deformation mode of spatially oriented piezo-active strut within the building block  
4 based on the direction of polarization (3-direction), electric field (denoted by  $n$ ) and the angle  
5 between the electric field and the strut ( $\vartheta$ ). Due to the large permittivity difference between the  
6 piezo-active material and the air (or polarization liquid), upon application of the driving (or  
7 polarization) voltage, the electric field within the building block is mainly distributed along the  
8 struts that are not perpendicular to the electric field<sup>42</sup>. When  $\vartheta=90^\circ$ , the strain of the strut is  
9 negligible. When  $n=1$  or  $2$ , the strut expands if  $\vartheta<90^\circ$  and shrinks if  $\vartheta>90^\circ$ . When  $n=3$ , the strut  
10 expands if  $\vartheta>90^\circ$  and shrinks if  $\vartheta<90^\circ$ .

## 11 *Theoretical prediction of the piezoelectric strain constant*

12 The inverse piezoelectric effect, characterizing the mechanical strain experienced by a  
13 piezoelectric material per unit of electric field applied, is quantified via the constituent equation<sup>38</sup>:

$$14 \quad S = d^t E$$

15 where  $S$  is the strain field of the piezoelectric material induced by external electric field  $E$ . We  
16 define the generalized piezoelectric coefficients,  $d_{nm}$  and  $\bar{d}_{nm}$  (as shown in Equation. 1), to  
17 describe the strain response of piezoelectric metamaterials.

18 We classify the working mode and evaluate the generalized piezoelectric coefficients  $d_{nm}$  and  
19  $\bar{d}_{nm}$  of a unit cell under an electric field by calculating the effective force  $\mathbf{F}_p^t$  and moment  $\mathbf{H}_p^t$  of  
20 each piezoactive strut member  $\mathbf{L}^t$  as follows:

$$21 \quad \begin{cases} \mathbf{F}_p = \sum_{t=1}^N \mathbf{F}_p^t = \sum_{t=1}^N A^t C \mathbf{E}_t \mathbf{d}_{ij}^t \mathbf{N}_{ip}^t \\ \mathbf{H}_p = \sum_{t=1}^N \mathbf{H}_p^t = \sum_{t=1}^N A^t C \mathbf{E}_t \mathbf{d}_{ij}^t \mathbf{N}_{ip}^t \mathbf{r}_q^t \end{cases} \quad (i, j, p, q = 1, 2, 3)$$

1 where  $A^t$  is the cross-sectional area of the t-th strut,  $C$  and  $\mathbf{d}_{ij}^t$  are the elastic modulus and  
 2 piezoelectric coefficient matrix of the constituent materials, respectively,  $\mathbf{E}_i$  is the electric field  
 3 applied on the piezoactive struts, and  $\mathbf{r}_q^t$  is the distance vector that connects the origin of the global  
 4 coordinate system (center of the unit cell) and the starting point of the t-th strut. Herein, we  
 5 introduce a local coordinate system x-y-z and correlate the stress of the t-th strut member in local  
 6 and global coordinate systems 1-2-3 via the stress-transformation matrix  $\mathbf{N}_{ip}^t$  as follows:

$$7 \quad \mathbf{N}_{ip}^t = \begin{bmatrix} \cos\theta_2 & 0 & \sin\theta_2 \\ 0 & 1 & 0 \\ \sin\theta_2 & 0 & -\cos\theta_2 \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\theta_1 & \sin\theta_1 \\ 0 & \sin\theta_1 & -\cos\theta_1 \end{bmatrix} \begin{bmatrix} \cos\theta_3 & \sin\theta_3 & 0 \\ \sin\theta_3 & -\cos\theta_3 & 0 \\ 0 & 0 & 1 \end{bmatrix}$$

8 The deformation mode is identified by evaluating the nonzero components of the effective force  
 9  $\mathbf{F}_p$  and moment  $\mathbf{H}_p$  of the metamaterial. For instance, a nonzero  $\mathbf{F}_5$  term represents the shear mode  
 10 in the 5-direction (13-direction in Einstein notation), and a nonzero  $\mathbf{H}_3$  term indicates a twist mode  
 11 along the 3-direction<sup>43</sup>.

12 The generalized piezoelectric coefficients  $d_{nm}$  and  $\bar{d}_{nm}$  are quantified in Equation. 2.  $\mathbf{C}_{pm}$  and  
 13  $\mathbf{I}_{pm}$  are the effective stiffness and rigidity of the unit cell, respectively, which are estimated  
 14 through the homogenization method as follows<sup>44</sup>:

$$15 \quad \mathbf{C}_{pm} = C \sum_{t=1}^N [\iint (r^t)^2 dA] / A_{unit}; \quad \mathbf{I}_{pm} = C \sum_{t=1}^N \int_{\mathbf{L}^t} [\iint (r^t)^2 dA] / V_{unit}$$

16 where  $r^t$  is the radius of the t-th strut,  $A_{unit}$  and  $V_{unit}$  are the cross-sectional area and volume of  
 17 the unit cell, respectively.

18 The piezoelectric electric coefficients  $d_{nm}$  and  $\bar{d}_{nm}$  of the design can be calculated by the ratio  
 19 between the total induced-load contribution of the connected piezo-active struts within the cubic  
 20 volume and the driving electric field  $E_n$ :

$$21 \quad d_{nm} = \frac{\sum_{t=1}^N \mathbf{F}_p^t \mathbf{C}_{pm}^{-1}}{E_n}; \quad \bar{d}_{nm} = \frac{\sum_{t=1}^N \mathbf{H}_p^t \mathbf{I}_{pm}^{-1}}{E_n} \quad (n = 1 - 3; p, m = 1 - 6)$$

22 where  $\mathbf{C}_{pm}$  and  $\mathbf{I}_{pm}$  are the effective stiffness and rigidity of the unit cell (see details in SI. 2), and  
 23  $\mathbf{F}_p^t$  and  $\mathbf{H}_p^t$  are the effective force and moment induced by the electric field.

## 1 ***Multi-material additive manufacturing***

2 The printing process began with the preparation of negatively charged resin, highly loaded  
3 piezoelectric nanoparticle colloids and SiOC preceramic monomers. The negatively charged resin  
4 was made by combining 5 g 2-carboxyethyl acrylate (CEA), 5 g polyethylene glycol diacrylate  
5 Mn~250 (PEG250), 0.2 g phenylbis(2,4,6-trimethylbenzoyl)phosphine oxide (Irg819), and 0.015  
6 g Sudan I in an opaque bottle and gently stirring the mixture upon heating at 60 °C. The PZT  
7 particles (APC 855) were mixed with a photosensitive resin (PEG250 mixed with 2wt%  
8 Phenylbis(2,4,6-trimethylbenzoyl)phosphine oxide) for 3D printing and lead nitrate particles in a  
9 high-energy ball mill (Retsch Emax) for half an hour to form uniform UV-sensitive colloids. The  
10 SiOC synthesis procedure can be found in our previous work<sup>28</sup>.

11 A self-developed 3D printer was used to print the lattices. During printing, the 3D lattice designs  
12 shown in Fig. 1 were first sliced into a series of closely spaced two-dimensional (2D) patterns with  
13 a commercial software (Netfabb 2021, Autodesk). These 2D patterns were sequentially transmitted  
14 to a UV light engine (Wintech PRO4500 with 405nm wavelength UV) to illuminate the UV-  
15 sensitive colloid or polymer, forming a solid layer in the shape of the 2D pattern. During the  
16 illumination, the exposure time varies with the particle loading to ensure proper curing depth. The  
17 projection was repeated with different materials within one layer; after each projection, ethanol  
18 was sprayed onto the layer and it was subsequently dried with compressed air. After each layer  
19 was exposed, the substrate on which the printed part rested was lifted up and the area below was  
20 refilled with a thin film of the UV-sensitive colloid or polymer. The exposure and material  
21 switching process was repeated for each layer with different exposure patterns to form 3D  
22 architectures with multiple materials layer-by-layer.

## 23 ***Selective deposition of the piezoelectric architecture***



1 After printing, the lattice designs were selectively deposited with metals to form the 3D embedded  
2 electrodes. Tetraaminepalladium(II) chloride monohydrate (Pd<sup>+</sup>) and borane dimethylamine  
3 complex (DMAB) were purchased from Millipore-Sigma. Electroless copper (Cu), electroless  
4 nickel-phosphorus (Ni-P), copper electroplating, and gold electroplating setups and solutions were  
5 purchased from Caswell Inc. A catalyst solution was made by vortexing 0.014 g Pd<sup>+</sup> in 10 g  
6 deionized (DI) water. A DMAB solution was made by combining 0.0118 g DMAB in 10 g DI  
7 water. The multimaterial 3D-printed parts with negatively charged resin were first rinsed in DI  
8 water and wicked dry with a paper towel. To load electroless catalyst, the parts were placed in Pd<sup>+</sup>  
9 solution for 5 minutes and wicked dry, placed in DMAB solution for 2 minutes to reduce Pd<sup>+</sup> to  
10 Pd metal, and wicked dry, and then placed in electroless copper solution for 10 to 15 minutes, as  
11 required to grow a visible deposit of copper. Then, the parts were rinsed three times in DI water  
12 and wicked dry. Copper electroplating was then performed by placing the parts in the copper  
13 electroplating solution with a copper metal counter electrode. During the electroplating, a  
14 stainless-steel needle was then contacted with the copper deposit under 0.2 A and 2 V, and contact  
15 was maintained for 1-2 minutes. The gold electroplating was done through electroless Ni-P  
16 deposition. For Ni-P deposition, the deposition solution was first heated to 80 °C. After soaking in  
17 Pd<sup>+</sup> and DMAB solutions as previously described, the parts were placed in this solution and plated  
18 for 2-5 minutes until a visible silver-metallic deposit of Ni-P formed. The parts were then allowed  
19 to cool to room temperature, rinsed with DI water several times and wicked dry. Gold  
20 electroplating was performed using a stainless-steel counter electrode in a gold electroplating  
21 solution heated to 80 °C. A stainless steel needle was contacted with the Ni-P deposit under 0.01  
22 A and 2 V for 5-10 minutes until a visible deposit of gold was achieved. With the selective  
23 deposition process, the as-printed multi-material parts are selectively deposited with highly

1 conductive embedded 3D electrode based on the digital designs, which is ready for the sintering  
2 process.

### 3 ***Measurement of the surface deformation and $d$ coefficients of the metamaterials***

4 During testing, a signal generator generated a 5V peak-to-peak square wave with a 5Hz frequency  
5 (nonresonance) that was fed into a power amplifier. The voltage magnitude was increased to 50V  
6 after amplification. The voltage was used to actuate the piezoelectric metamaterial samples, and  
7 an oscilloscope was used to monitor the voltage input. A full-field laser vibrometer (Polytec PSV-  
8 500) was used to capture the motion of the side surface and top surface of the sample. The  
9 deformation of the side surface was reconstructed as a function of the voltage input into the sample  
10 using the software provided by Polytec Inc. and then normalized by the largest displacement. The  
11 piezoelectric strain coefficients of the lattice were calculated from the ratio between the induced  
12 strain captured by the laser vibrometer and the applied electric field.

### 13 ***Working principle of locomotion gaits***

14 The driving voltages ( $V_f$ ,  $V_b$ ,  $V_r$ ) of three actuation elements were applied on the corresponding  
15 terminals of the 3D electrodes, as shown in Fig. 4B and Extended Data Fig. 7. The driving voltages  
16 were independent pulse width modulation (PWM) signals with two amplitude levels (“1” – activate,  
17 200V amplitude voltage, “0” – deactivate, no voltage supply) and variable frequency and duty  
18 cycle (Extended Data Fig. 8a), enabling three distinguishable locomotion modes. Four anisotropic  
19 frictional feet are attached to the bottom of the robot to create unidirectional forward movement.  
20 During the walking gait, the driving voltage activates the body and rear leg, pushing the front leg  
21 forward with released front feet and locked rear feet (Extended Data Fig. 8b), after which the body  
22 and rear leg contract, and the front feet lock onto the ground, which completes one full walking

1 stroke cycle. The motion cycle repeats to generate continuous forward motion of the microrobot.  
2 The locomotion speed and weight of the micro-robot is benchmarked with other compact robotic  
3 systems (Extended Data Fig. 8d)<sup>39-41,45-54</sup>. As the operation frequency increases over 220 Hz, the  
4 deformation of the body actuator changes to a bending resonance mode (Extended Data Fig. 8b),  
5 initiating the turning mode of the microrobot through an unbalanced forward motion of the two  
6 front feet. During the jumping gait, the front leg is first activated by an impulse voltage with a  
7 pulse width of 5ms, which lifts the front leg up into the air. Before the front leg lands, the same  
8 impulse voltage is applied to the body and rear leg simultaneously, which activates the whole robot  
9 to jump upward and forward (Extended Data Fig. 8b). Three gait modes are enabled by  
10 programmed actuation of three regions of the micro-robot, making it possible for the robot to  
11 achieve fast, steerable locomotion and locomotes through rough terrain.

### 12 *Characterization of the contactless sensing element*

13 Our 3D printed micro-robot is capable of sensing and avoiding remote obstacles via 3D printed  
14 ultrasonic contactless sensing element and feedback control. The contactless sensing element is  
15 printed with the actuation element with the same piezo-active and structural material and is rigidly  
16 connected to the frontal leg. As shown in Extended Data Fig. 7a, the control system triggers the  
17 ultrasonic pulser-receiver chip and the chip send pulse signals to the contactless sensing element,  
18 which emit focused acoustic waves. The focused acoustic waves hit potential targets and the  
19 reflected echo waves are then received by the ultrasonic transducer which generate voltages that  
20 are then collected by the ultrasonic chip and the control system. The distance between the robot  
21 and the object can then be calculated by the echo time (Extended Data Fig. 10a-b). The working  
22 range of the contactless sensing element was also characterized with a 8mm diameter cylinder, as  
23 shown in Extended Data Fig. 10c.

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1 **Acknowledgements**

2 We acknowledge the support of Deepam Maurya, Prashant Kuma on the operation of laser  
3 vibrometer and power amplifier. We acknowledge the support of Ariel Calderon , Salar Chamanian,  
4 Petrick Mercier on the development of the control and powering system of the micro-robot.

5 **Funding:** We acknowledges support from XXX.

6 **Author contributions**

7 X. Z. proposed the research, developed the metamaterial design concept and directed the overall  
8 experiments. H. C. designed the experiments, created the metamaterial design method and  
9 generated architecture designs. D. Y. developed the extended piezoelectric tensor matrix and  
10 verified the architecture designs analytically and numerically. H. C., Z. X., H. L. R. H. and Z. W.  
11 developed the multi-material fabrication platform and fabricated all testing samples. H. C. and S.  
12 D. carried out the verification experiments. H. C. and H. L. developed and characterized the micro-  
13 robotic systems. All authors discussed the results and commented on the manuscript.

14 **Data and materials availability**

15 All data is available in the manuscript or the supplementary material.

16 **Competing interests**

17 The authors certify that they have no affiliations with or involvement in any organization or entity  
18 with any financial interest, or non-financial interest in the subject matter or materials discussed in  
19 this manuscript.

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- 1 **Additional information**
- 2 **Supplementary information** is available for this paper.
- 3 **Correspondence and requests for materials** should be addressed to X. Z.