Designing electromechanical metamaterial with full nonzero piezoelectric coefficients

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Designing topological and geometrical structures with extended unnatural parameters (negative, near-zero, ultrahigh, or tunable) and counterintuitive properties is a big challenge in the field of metamaterials, especially for relatively unexplored materials with multiphysics coupling effects. For natural piezoelectric ceramics, only five nonzero elements in the piezoelectric matrix exist, which has impeded the design and application of piezoelectric devices for decades. Here, we introduce a methodology, inspired by quasi-symmetry breaking, realizing artificial anisotropy by metamaterial design to excite all the nonzero elements in contrast to zero values in natural materials. By elaborately programming topological structures and geometrical dimensions of the unit elements, we demonstrate, theoretically and experimentally, that tunable nonzero or ultrahigh values of overall effective piezoelectric coefficients can be obtained. While this work focuses on generating piezoelectric parameters of ceramics, the design principle should be inspirational to create unnatural apparent properties of other multiphysics coupling metamaterials.

INTRODUCTION

Metamaterials are a class of engineered systems that aim at achieving exotic macroscopic physical properties by designedly organized subunits in the critical dimensions to form, most notably, equivalent parameter values beyond nature (1–8). Inspired by the theoretical prediction of left-handed materials from Maxwell’s equations (9), initial research studies focused on how to realize materials with negative permittivity (10) and permeability (11). Later experimental verification of negative refractive index enlightens scientists to craft man-made structures with subwavelength elements for manipulating wave propagation (3, 8, 12), which has been widely applied in designing invisible cloaks (13, 14) and perfect lenses (15), and in transformation optics (16, 17). Compared with photonic crystals, the local physical inhomogeneity in electromagnetic metamaterials, whose unit cell dimensions are much smaller than manipulated wavelength, can be disregarded so that physically equivalent parameters can be defined (1, 2). Besides electromagnetism, the methodology of achieving pivotal effective indices for targeting metaproperties has been the core concept extended to acoustics (4, 5), mechanics (6, 18), and thermodynamics (7). While metamaterials feature single exotic properties, the strong correlation between specific geometries and active ranges restricts tunability and real-world applications, resulting in tough problems such as narrow bandwidth and high loss in electromagnetic metamaterials (19, 20). To pave the way for practical devices, tunable parameters, and unit elements sensitive to applied external strain, electromagnetic field or other physical fields induced by multiphysics coupling effects are expected to be introduced to form smart metamaterials (21, 22).

Multifarious complex methods have been continuously developed to fabricate applicable metamaterials usually requiring topological and geometrical orderliness on several different scales (23, 24). However, until now, these artificial processing techniques still lack universality, are expensive, and are unable to imitate macroscopic properties comparable to naturally occurring materials. Perfectly manufactured metamaterials with both controllable numerous subwavelength units and applicable three-dimensional large-volume structures is an even tough challenge (19), while it is necessary for practical macro-sized devices (25). To further promote the fabrication feasibility, functional scalability, and smart tunability of new metamaterials, designing artificial unit cells with relative easily acquired topological structures, fully using intrinsic properties of natural materials, and introducing multiphysics coupling effects should be combined together for thoroughly updating traditional solely functional metamaterials and leading to apparent properties with tunable parameters (21). Guided by this methodology, we propose a systematic strategy to obtain a novel kind of piezoelectric metamaterials, whose exotic parameters are derived from structural designs, while their unit elements are macro-sized piezoelectric ceramics with naturally occurring electromechanical coupling effects.

Piezoelectric elements were initially used as the electromechanical resonators to form bandgap in locally resonant metamaterials (26, 27). Nowadays, much attention has been focused on piezoelectric metaproperties, in which the piezoelectric response arises from the orderly topological and geometrical structures rather than the materials’ chemical composition (18, 28, 29). In the field of piezoelectric metamaterials, exotic engineered piezoelectric constants are the core targeted parameters to form anomalous properties; for instance, enormous apparent piezoelectric response with an effective piezoelectric coefficient $d_{33} > 3500$ pC/N was detected in a lead-free ceramic metamaterial with designed gradient-generating structures (29). In general, piezoelectric ceramics are the most widely used electromechanical materials because of their excellent mechanical properties, high Curie temperature, easy preparation process, and economically friendly performance (30–34). However, polarized piezoelectric ceramics feature a macroscopic transversely isotropic symmetry similar to 6-mm point group belonging to the hexagonal crystal family (Fig. 1A) (35). This symmetry leads to only five naturally occurring nonzero piezoelectric strain coefficients (which quantifies the ability to transform applied electric field into strain), namely, $d_{33}$, $d_{31}$ ($d_{32}$), and $d_{15}$ ($d_{24}$), imposing restriction to the study and application of electromechanical devices for decades. Crystals with specific point groups have more practical coefficients, like $d_{36}$, but are still limited (36). Efforts have been devoted to
achieve better indices, but these few sporadic results did poorly on portability and sustainability because of a lack of clear methodology (30, 31, 37, 38). Meanwhile, the possibility of the remaining zero piezoelectric coefficients has always been ignored for decades. This work, the 13 nonzero effective piezoelectric coefficients are by metamaterial design in contrast with only five nonzero ones in natural piezoelectric ceramics.

In this study, full 13 artificial nonzero elements in the piezoelectric strain matrix and the corresponding deformation modes are unprecedentedly excited in their entirety based on 5 natural nonzero ones of ceramics by metamaterial design (see Fig. 1). Some effective piezoelectric coefficients exhibit tunable and greatly enhanced values by an order of magnitude than already existent ones, occurring even bigger than the highest natural ones in single crystals (effective $d_{11}$ of a kind of commercialized PZT-5H ceramics is designed and measured to reach 13,592 pm/V). Composed of above novel metamaterials as unit elements, arrayed electromechanical metamaterials with enhanced 20-fold displacement, first-time realized co-fired multilayer shear-mode actuators are designed. These results demonstrate that the piezoelectric metamaterials show great potential in improving traditional quasi-static or resonant transducers and enlighten brand-new piezoelectric technologies.

**RESULTS AND DISCUSSION**

**Piezoelectric metamaterial design**

Here, we show a systematic methodology to excite all nonzero effective piezoelectric coefficients in both resonant and quasi-static frequencies by an electromechanical metamaterial design (Fig. 1B). The piezoelectric matrix of $d_{ij}$ in natural ceramics is presented as

$$
[d_{ij}]^T =
\begin{bmatrix}
0 & 0 & 0 & 0 & d_{15} & 0 \\
0 & 0 & 0 & d_{24} & 0 & 0 \\
d_{31} & d_{32} & d_{33} & 0 & 0 & 0
\end{bmatrix}^T
$$

(1)

The longitudinal and transversally extensional modes ($d_{11}$, $d_{32}$, and $d_{33}$) manifest as nonzero normal strain with external electric field parallel to polarization direction, while $d_{15}$ (or $d_{34}$) is the only nonzero shear mode with orthogonal orientations. To excite other basic nonzero $d_{ij}$ values, there is no choice but to design metamaterials based on the five natural modes.

To create nonzero piezoelectric strain coefficients $d_{ij}$, our strategy starts by analyzing the requirements of applied electric field (along with $i$ direction) as well as objective strain type (normal strain with $j = 1, 2, 3$; shear strain with $j = 4, 5, 6$) to determine the adoptive basic natural modes (selecting $d_{11}$, $d_{32}$, $d_{15}$, $d_{24}$, or $d_{33}$) and topological structures (arrangement ways of meta-atoms) based on “center extrusion effect” (CEE; which is developed for normal strain; see details below) and “diagonal transformation effect” (DTE; which is developed for shear strain). It continues by dividing piezoelectric metamaterials into lattice-like subunits with programmed polarization directions and electrodes as locally deformed elements (meta-atoms) to induce anisotropic strain and, consequently, to excite desired modes. The artificial anisotropy actually achieves equivalently reduced symmetry. Finite element simulation, theoretical analyses, and experimental measurement jointly prove the validity of our methodology. The design details fall into two categories: normal-strain and shear-strain modes.

Normal-strain ($j = 1, 2, 3$) metamaterials exhibit contraction or extension deformation. The topological structures for apparent normal-strain modes with effective piezoelectric coefficients $d_{11}$, $d_{12}$, $d_{13}$, $d_{22}$, $d_{23}$, and $d_{33}$ are illustrated in Fig. 2 (A to C), whose detailed designs are expounded in section S1. Generally speaking, the naturally occurring $d_{15}$ (or $d_{34}$) shear mode is stimulated in each meta-atom, functioning as local elements. Head-to-head polarization or electric fields are programmed so that these subunits synergistically extrude one another to excite an objective orthogonal displacement. Typically, for nonzero $d_{12}$, $d_{13}$ coefficients, stimulation of “2” direction normal strain is quite tough because $d_{15}$ deformation is totally in perpendicular “13” plane. However, by reusing “extrusion effect,” an extruded strain along with 2 direction is created with the aid of the positive Poisson’s ratio of piezoelectric ceramics.

The topological structures of each normal-strain metamaterial behave like arm force rod under external stress, and their mechanism is named CEE (Fig. 2H, inset). The principle of center extrusion structures is illustrated in Fig. 2H, and a displacement output along the “3” axis emerges as a joint result of each meta-atom. The biggest output acquired in the metamaterial center is regarded as effective displacement $D_{eff}$ with values theoretically predicted as

$$
D_{eff} = \frac{d_{15}EI}{2t}
$$

(2)

where $l$ and $t$ are the length and thickness of a cuboid specimen, respectively, and $E$ denotes the applied electric field.

Shear strain (when $j = 4, 5, 6$) is an in-plane deformation manifesting as transformative rhombic angles. The topological designs for apparent shear modes with effective piezoelectric coefficients $d_{14}$, $d_{16}$, $d_{16}$, $d_{24}$, $d_{34}$, and $d_{36}$ are illustrated in Fig. 2 (D to G) and expounded in detail in section S2. In general, programmed meta-atoms of metamaterials jointly excite anisotropic strain along with two orthogonal diagonals (extension and contraction, respectively) and result in apparent shear-like deformation. The basic mode of meta-atoms for coefficient $d_{1j}$ (or $d_{2j}$) is the $d_{15}$ (or $d_{34}$) shear mode, while the $d_{33}$
mode is adopted for coefficient $d_{34}$ ($d_{35}$) and the $d_{31}$ mode is adopted for coefficient $d_{16}$.

The mechanism for all shear metamaterials is summarized as DTE, which imitates fork-type structures with rhombic distortion driven along diagonals (Fig. 2I, inset). In theory, combination of extension and contraction along with vertical diagonals is supposed to be equivalent to a pure shear deformation (see detailed analyses in Materials and Methods). As illustrated in Fig. 2I, for a cube-shaped specimen, the strain tensor in the prime coordinates $(1'2'3')$ is

$$S' = \begin{bmatrix} s & 0 & 0 \\ 0 & 0 & s \\ 0 & s & 0 \end{bmatrix}$$

(3)

Transformed equivalent strain tensor $S_{eff}$ in the original coordinates $(123)$ is presented as

$$S_{eff} = T S' T^{-1} = \begin{bmatrix} 0 & 0 & s \\ 0 & 0 & 0 \\ s & 0 & 0 \end{bmatrix} = S_5$$

(4)

where $T$ is the transformation matrix. The off-diagonal element $S_{13}$ is equal to $S_{31}$, indicating a pure shear strain $S_5$ stimulated equivalently. In consideration of feasibility of polarization and electrode fabrication, only a pair contractive (or extensive) strain is provided along one diagonal in some metamaterials. However, they still exhibit apparently quasi-shear distortion because another diagonal extension can arise from the positive Poisson’s ratio of the contraction, just with some extra volume contraction. These metamaterials function in both resonant and quasi-static states, with shear strain in resonance frequency being actually more ideally pure because of the equipartition of energy in this situation.

With naturally occurring ceramics programmed as unit elements, piezoelectricity is introduced as coupling effects, and a new class of metamaterials, piezoelectric metamaterials, is obtained. To clarify the basic science principles and design methodology, we simplify metamaterials to several meta-atoms. Metamaterials composed of multiple unit elements can also be easily designed (see section S3). However, more meta-atoms usually bring problems such as complex fabrication processes and worse device reliability. Our methodology contributes to full piezoelectric coefficients and practical applications in the situation of only several subunits.

Fig. 2. Schematic designs of piezoelectric metamaterials. With programmed polarization and applied electric field of subunits, the metamaterials realize all effective normal or shear-strain modes in both quasi-static and resonant frequencies. (A to G) Schematic metamaterial designs and deformation states by finite element simulation in both resonant (with ▲ label) and quasi-static (without ▲ label) states of $d_{11}$ ($d_{22}$) mode (A), $d_{13}$ ($d_{33}$) mode (B), $d_{12}$ ($d_{21}$) mode (C), $d_{14}$ ($d_{41}$) mode (D), $d_{16}$ ($d_{61}$) mode (E), $d_{34}$ ($d_{43}$) mode (F), and $d_{36}$ mode (G). (H and I) Diagrams of two kinds of fundamental design mechanism learned from natural structures we established, namely, CEE for effective normal strain (H) and DTE for shear strain (I).
Investigations on effective values of piezoelectric coefficients

The effective values of artificial coefficients mainly depend on two factors: (i) topological structures and geometrical sizes of metamaterials and (ii) the intrinsic electromechanical coupling strength of piezoelectric ceramics. Moreover, the measured or simulated values are also related to electric or mechanical boundary conditions. To verify nonzero piezoelectric coefficients, we adopted finite element method (FEM; by COMSOL Multiphysics) with theoretical analyses and experimental results to investigate their magnitudes and variation tendency along with geometric dimensions. A kind of relaxor ferroelectrics, PNN-PT (31), and commercialized PZT-5H and PZT-8 are selected as sample ceramics, and the simulation results are illustrated in Fig. 3. The definition formulas, boundary conditions, and computational details of all artificial coefficients are expatiated in Materials and Methods, section S4, and table S1. It is obvious that metamaterials feature not only overall nonzero but also some tunable and ultrahigh piezoelectric coefficients relevant to geometrical dimensions.

Effective piezoelectric coefficients of several normal-strain metamaterials are theoretically predicted as

\[ d_{ij}^{\text{eff}} = \frac{d_{ij}}{2l} \]  \( ij = 11, 12, 13, 23 \)  \( (5) \)

which indicates feasibly tunable values in wide range by changing LTR (length-to-thickness ratio) of specimens (Fig. 3B). Attributed to the uniform deformation of their topological structures, simulated values match theoretical predictions well. To our best knowledge, the largest piezoelectric strain coefficient of single crystals is reported to be 7500 pm/V (37), while these electromechanical metamaterials can be easily tuned and feature much bigger apparent values (above 10,000 pm/V) without any additional amplification mechanism. Their remarkable outputs show great potential for high-performance actuators.

For the \( d_{12} (d_{31}) \) mode, simulation results denote a positive nonlinear correlation between the nominal coefficients and LTR (Fig. 3C). The precisely analytic solution is hard to be obtained because of the anisotropic strain state from extrusion effects. Coefficients of shear mode \( d_{ij}^{\text{eff}} \) \( ij = 14, 25, 16, 26 \) increase with the thickness-to-length ratio (TLR) with similar nonlinearity from extrusion deformation (Fig. 3D). The \( d_{34} (d_{35}) \) shear mode shows steady effective coefficients (Fig. 3E), and \( d_{36} \) has relatively linear increasing values proportional to TLR in a wide range (Fig. 3F). Compared with other shear modes, these unique ones derived by subunits of the \( d_{33} \) or \( d_{31} \) mode feature profound advantages.

Further design will demonstrate that these technical improvements are of great importance in co-fired electromechanical devices.

To experimentally verify the actual performance of piezoelectric metamaterials, we fabricated and investigated all seven types of piezoelectric metamaterials composed of commercialized PZT-5H ceramics, which are discussed in detail in Materials and Methods and section S5.
The preparation processes mainly consist of bonding meta-atoms with prepolaryization by structural adhesives and coating electrodes by screen printing, and photographs of all metamaterials are shown in fig. S3. Table 1 lists all specimen sizes. Under the condition of steady mechanical boundary, the displacement responses of metamaterials to applied AC voltages are characterized by a high-precision laser feedback interferometer and then automatically recorded by a data acquisition system (LabVIEW myDAQ) so that the corresponding nominal piezoelectric coefficients can be calculated (see Fig. 4A). The results of two typical metamaterials with the $d_{11}$ mode (on behalf of normal strain and CEE) and the $d_{36}$ mode (on behalf of shear strain and DTE) are shown in Fig. 4. Figure 4B presents the measured displacement outputs of the $d_{11}$ mode metamaterial (with LTR = 40) under different driving voltages, where the responses exhibit low noise, high controllability, and stable repeatability. It is obvious from Fig. 4C that the measured displacements are comparable to simulation ones in the voltage range of 0 to 250 V/mm. The reasonable disparity between experimental and analytical data is assigned to technological gaps such as full polarization, bonding junction loss, and ideal boundary conditions. Moreover, when electric field turns stronger than 250 V/mm, the specimen shows unprecedented giant outputs, which are even higher than simulation ones and reach around ±4.5 μm under an electric field of ±400 V/mm. This interesting phenomenon may attribute to the additional out-plane shear-bending amplification effects (39), leading to further enhanced displacement. The experimental outputs below 250 V/mm are fitted, and piezoelectric coefficient $d_{11}$ is calculated to be 13,592 pm/V, which is close to the theoretical prediction of 16,000 pm/V and nearly 20-fold bigger than natural $d_{13}$ values of PZT-5H (700 pm/V).

Figure 4 (D and E) shows the transverse displacement responses of a side vertex of the $d_{36}$ shear-mode metamaterial. This accordant effective output by simulation and experiments demonstrates the viability of our fresh methodology to generate shear-like deformation based on normal-strain modes. In practical device applications, the nonlinear extrusion effect of $d_{36}$ or other coefficients may lead to some stress concentration in metamaterial bodies especially at interfaces. However, this phenomenon can be diminished or avoided effectively by creating stress release holes [like multilayer structures in classical piezoelectric technologies (40)], using ductile glues or modifying topological configurations.

The simulated and experimental displacement responses of all other kinds of metamaterials are shown in fig. S4, and their corresponding effective strain and CEE) and the $d_{36}$ mode (on behalf of normal strain and CEE) and arrangement of generalized deformation (section S6).

### Potential applications of piezoelectric metamaterials

To take full advantage of piezoelectric materials for electromechanical applications such as actuators, ultrasonic motors, sensors, or energy harvesters, scientists used proper modes to form other diversiform configurations. The novel metamaterials bring unprecedented full nonzero elements to the piezoelectric matrix; moreover, their tunable or ultra-high nature promises to broaden designs of devices, improve their properties, and enlighten brand-new piezoelectric technologies. Here, we show two potential applications.

### Arrayed and enhanced electromechanical metamaterials

Using metamaterials as unit elements, interesting arrayed electromechanical metamaterials can be further constructed through our basic arrangement methodologies (CEE and DTE) and exhibit enhanced and compounded extension, contraction, shear, twist, or other programmed deformation (section S6).

Higher effective piezoelectric constants contribute to bigger apparent displacement and lower drive voltage, bringing in benefit to miniaturization and integration of MEMS (micro-electromechanical systems) driving modules. As illustrated in Fig. 5 (A to C), on the basis of the giant $d_{11}$ normal-strain mode (CEE) and arrangement of generalized DTE, a novel multilayer metamaterial is designed, which exhibits remarkable effective strain (0.4%) and very large apparent displacement (more than 40 μm) under very small applied electric field (200 V/mm for each layer) (see Materials and Methods and section S6 for FEM calculation details). These values are 20-fold bigger than traditional $d_{33}$-derived multilayer piezoelectric actuators of the same geometrical size and driving voltage. The apparent performance can be mediated as required or further optimized by increasing LTR of each layer.

| Neotype modes | Specimen sizes (length × width × thickness) | Simulation values (pm/V) | Experiment values (pm/V) |
|---------------|---------------------------------------------|--------------------------|--------------------------|
| $d_{11}$ (LTR = 40) | 20 mm × 7 mm × 0.5 mm | 16,000 | 13,592 |
| $d_{12}$ (LTR = 14) | 14 mm × 7 mm × 0.5 mm | 11,200 | 6,984 |
| $d_{14}$ (LTR = 0.1) | 10 mm × 14 mm × 1 mm | 200 | 2,205 |
| $d_{16}$ (LTR = 0.1) | 10 mm × 10 mm × 1 mm | 28 | 64 |
| $d_{14}$ (LTR = 0.1) | 10 mm × 10 mm × 1 mm | 22 | 57 |
| $d_{16}$ (LTR = 0.1) | 10 mm × 10 mm × 1 mm | 531 | 714 |
| $d_{16}$ (LTR = 1/4) | 14 mm × 14 mm × 1 mm | 372 | 259 |

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Co-firing multilayer shear-mode actuators

The co-fired multilayer structure is an efficient method to manufacture integrated and multifunctional devices such as actuators and multilayer ceramics capacitors. Since tape casting and co-fired technologies were invented decades ago, shear-mode multilayer actuators have never been successfully fabricated using these methods because the polarization and applied electric field are in orthogonal directions, and electrodes require polishing and repreparation after polarization. However, our piezoelectric metamaterials promise to first solve the puzzling problems by $d_{34}$ ($d_{35}$) or $d_{36}$ normal-strain-derived shear modes. Figure 5 (D to F) and section S7 demonstrate multilayer $d_{36}$ shear-mode ceramic actuators. Surfaces of every other layer are partitioned into four squares, with their electrodes coated; the remaining surfaces function as grounding electrodes, forming interdigital structures. After squares in each layer polarized along the same direction, one diagonal positive and another diagonal negative driving voltages will stimulate in-plane $d_{36}$ shear deformation and will eventually obtain shear-mode multilayer actuators. Conventional $d_{36}$ face-shear actuators (35) usually require an expensive crystal resonator, while the designed novel co-firing ones based on ceramic metamaterials feature lower operating voltage, higher mechanical strength, lower cost, and still comparable precise positioning, promising to motivate research in brand-new miniaturized actuating devices such as...
as nanoshear actuators, piezoelectrically walked step motors, and ultrasonic motors in the microelectronics industry.

Besides inspiring brand-new piezoelectric inventions, electromechanical metamaterials with full nonzero coefficients will hopefully renovate and enhance traditional piezoelectric devices. Previously nondispersive shear horizontal waves used in damage detection are generated by the $d_{15}$ mode, while novel artificial modes with milder voltage may be more competitive (35). A kind of shear-bending actuators reported by us before can also be classified into the application examples of $d_{11}$ mode metamaterials (39). Serving as additive tunable elements with abundant deformation modes, electromechanical metamaterials are able to introduce piezoelectric tunability to traditional smart metadevices (25).

CONCLUSION AND DISCUSSION

We implement a metamaterial design to piezoelectric ceramics and report a novel class of electromechanical metamaterials with fully unnatural nonzero piezoelectric coefficients, which promises to profoundly promote the fundamental research in piezoelectric materials and technologies. The methodology of CEE and DTE arises from basic concepts of condensed matter physics, including symmetry principle, finite element idea, and multiphysics coupling effects.

The broken symmetry, an extensive mechanism in physics, leads to many charming phenomena such as antiferromagnetism and superconductivity. Usually, novel broken symmetry is pretty tough to controllably build in the atomic level or micro-level, while our results demonstrate that some equivalent effects may be architected through macroscopic metamaterial design, in which the finite meta-atoms function as locally equivalent elements to introduce artificial anisotropy.

Naturally nonexistent effective physical parameters may be induced by programmed quasi-symmetry breaking not only in polycrystalline materials but also in amorphous materials or crystals. Like amorphous metallic glass, a kind of magnetostriction material of 6-mm symmetry, overall nonzero 18 magnetostriction coefficients are supposed to be designed and generated, despite the hard control of the space-confined magnetic field.

Previous metamaterials aim at single exotic function, while the combined effects derived from topological structures and intrinsic nature of metamolecules have been paid little attention, such as the novel electromechanical metamaterial in this work. Further exploration should aim at introducing multiphysical coupling effects to form smart metamaterials, which hopefully feature properties such as tunability by external physical field or self-adaptability to external environment (for example, pyroelectric metamaterials with controllable current and Kerr effect with tunable oriented refractive index).

MATERIALS AND METHODS

Metamaterial design methodology

To wholly create nonzero piezoelectric coefficients $d_{ij}$, we developed two basic topological deformation or arrangement methods, including CEE (for normal strain) and DTE (for shear strain), which contributes to artificial anisotropy and changes the apparent symmetry of 6-mm point group. The normal-strain modes can be excited through extrusion of meta-atoms, while for shear-strain modes, artificial anisotropic strain along different diagonals is the basic requirement for shear-like deformation. For DTE, the combination of extension and contraction strains along with diagonals can be demonstrated to be equivalent to a pure shear strain in theory. As illustrated in Fig. 2I, for a square-shaped...
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specimen, tensile strain $s$ and compressive strain $-s$ are applied simultaneously along the (101) and (101) orientations, respectively. When we take $1'$ and $3'$ axes as the prime coordinates, the strain tensor is

$$
\begin{bmatrix}
s & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & -s
\end{bmatrix}
$$

(6)

Using the transformation matrix (where parameter $\theta = 45^\circ$)

$$
T = \begin{bmatrix}
\cos \theta & 0 & \sin \theta \\
0 & 1 & 0 \\
-\sin \theta & 0 & \cos \theta
\end{bmatrix}
$$

(7)

The transformed equivalent strain tensor $S_{\text{eff}}$ in the original coordinates (123) can be converted into

$$
S_{\text{eff}} = T^S T^{-1} = \begin{bmatrix}
0 & 0 & s \\
0 & 0 & 0 \\
0 & 0 & s
\end{bmatrix} = S_5
$$

(8)

The off-diagonal components $S_{13}$ and $S_{31}$ have the same values $s$, which means that a pure shear strain $S_5$ is generated equivalently.

In the situation where only compressive strain $3'$ is produced in some metamaterial modes, the transformed equivalent strain tensor $S_{\text{eff}}$ is

$$
S_{\text{eff}} = T^S T^{-1} = T = \begin{bmatrix}
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & -s
\end{bmatrix} T^{-1} = \frac{1}{2} \begin{bmatrix}
s & 0 & s \\
0 & 0 & 0 \\
s & 0 & s
\end{bmatrix}
$$

(9)

The strain tensor in the original coordinates (123) is composed of two items: The first item is equivalent shear strain, and the second item denotes some extra volume contraction, which will not influence the application of metamaterials. The results prove that shear-like deformation was successfully excited. To satisfy the preparation feasibility and convenient real-world application, we usually adopted the design of only contraction (or extension) along with one diagonal, and the other diagonal deformation was automatically generated because of the positive Poisson’s effect.

**Finite element simulation**

**Basic electromechanical metamaterials**

The deformation of the basic 13 electromechanical metamaterials or the specific values and variation tendency of nonzero piezoelectric coefficients in Fig. 3 were simulated by the COMSOL code using the piezoelectric device module. The geometrical models of all modes were built by parameterized coordinates and divided into unit elements according to the topological designs as shown in Fig. 2 and table S1. The ceramic materials PZT-5H and PZT-8 are the default materials in the material libraries, and PNN-PZT is a user-defined new material, the full set of parameters of which is derived from our previous work (31). The different unit cells were polarized along with head-to-head or back-to-back directions by establishing rotated coordinate systems and applied voltages to electrode surfaces by 200 V/mm (note that for some shear modes, the electric field is average values; see section S4). With boundary conditions of partially fixed constraints and very detailed mesh generation controlled by physical fields, the displacements and effective coefficients were simulated in the steady state. To calculate the effective values of piezoelectric coefficients, centers of normal-strain modes and vertices of shear-strain modes were regarded as motion points. The length of shear-strain specimens for calculating effective piezoelectric coefficients was assumed to be the original value without regard to contraction. Parameterization function was used to investigate their variation tendency with TLR or LTR. As a comparison to experimental results as shown in Fig. 4 and fig. S4, the simulated results were based on default PZT-5H in the material libraries of COMSOL, while its $d_{33}$, $d_{31}$, and $d_{15}$ values were altered to 700, −320, and 800 pm/V according to the commercialized PZT-5H we used.

**Arrayed and enhanced electromechanical metamaterials**

The arrayed and enhanced electromechanical metamaterials are the multilayer structures of the $d_{11}$ mode. The geometrical model contains 20 PNN-PZT ceramic layers, and each layer was connected by polymethyl methacrylate (with thickness and length being both $1/10$ of thickness of each PNN-PZT layer) in the material libraries. With side surfaces of the bottom layer fixed, the displacement output was calculated.

**Co-firing multilayer shear-mode actuators**

The simulation for shear-type multilayer actuators was similar with arrayed normal-strain metamaterials. Notably, the interdigital electrodes and ceramic layers with various polarization directions should be carefully set up. With one side surface fixed, the shear-type deformation and stress distribution were simulated.

**Experimental verification**

**Metamaterial preparation**

A kind of commercialized PZT-5H (with $d_{33}$ coefficients being 650 to 700 pm/V, and $d_{31}$ and $d_{15}$ coefficients being around −320 and 800 pm/V, respectively; Suzhou PANT Piezoelectric Technology Corporation) plate was cut into meta-atoms with desired geometric sizes using a precision cutting machine (CNC-400, MTI Corporation). After silver paste electrodes were screen-printed onto surfaces with a postfire at 650°C for 0.5 hour, these meta-atoms were polarized in silicone oil bath under a DC electric field of 10 kV cm$^{-1}$ in 120°C for 15 min. Later on, the initial electrodes were removed (not necessary for all modes), and meta-atoms were arranged and bonded together by structural adhesives according to the specific topological structure design. Newly designed electrodes were printed to apply the drive voltage, and wires were bonded onto the electrodes by low-temperature silver paste without postfire procedures for further measurement.

**Parameter measurement**

The piezoelectric strain coefficients of all seven kinds of metamaterial specimens were measured using electric field–induced displacement method. The specimens were installed onto the three-dimensional printed fixture according to the desired boundary conditions. A sinusoidal driving voltage was produced using a signal generator (Tektronix AFG3022B) and amplified using a high-voltage amplifier (PINEK HA-405 or Kepco BOP-1000M). The driving voltages were monitored using an oscilloscope (Keysight MSOX4024A), and the metamaterial displacement responses were measured using a high-precision laser feedback interferometer (LeiCe LY1000) with nanometer resolution.
The data were automatically recorded using a data acquisition system (LabVIEW myDAQ).

SUPPLEMENTAL MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/11/eaax1782/DC1

Section S1. Designing piezoelectric metamaterials with normal-strain mode
Section S2. Designing piezoelectric metamaterials with shear-strain mode
Section S3. Piezoelectric metamaterials composed of multiple meta-atoms
Section S4. FEM simulation details
Section S5. Experimental verification details
Section S6. Arrayed metamaterials with enhanced performance
Section S7. Design of neotype co-firing multilayer shear-mode actuators

Table S1. Schematic diagrams and definition formulas for calculating all effective piezoelectric coefficients.

Fig. S4. Displacement responses to electric field of other five kinds of metamaterials.
Fig. S5. Arrayed shear-mode metamaterials with in-plane arrangement way.
Fig. S6. Preparation processes of neotype co-firing shear-mode multilayer actuator.

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