Supplementary Materials for

Deployable mechanical metamaterials with multistep programmable transformation

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Materials and Methods

1. Fabrication of bistable units by laser cutting

The bistable units are made of the widely available elastic thermoplastic polyurethane (TPU) with good ductility to improve structural design capabilities. First, we use an additive manufacturing method – the fused deposition modeling (FDM) technology to fabricate TPU sheets with a thickness of 1.5 mm by Ultimaker 3 (Fig. S1A and B). Second, we cut the TPU sheets with a laser cutting machine (VLS 2.30, Universal, USA). To ensure high precision laser cutting, we use a low-power multi-cutting technology to cut the TPU sheets until they are completely cut through (Fig. S1D and E). Considering the limited size of the 3D printed TPU sheets, we can only cut the series-connected bistable units into many parts. Then we solder and assemble them with an electric soldering iron (Fig. S1F and G). Finally, we assemble the prepared tandem bistable units with the elastomeric film.

2. Fabrication of bistable units by multi-material 3D printing

To achieve recoverability and the bifurcation of the deformation pathway, we introduce a temperature-responsive part (TRP) made of polyactic acid (PLA) in the TPU made bistable units. Accordingly, we manufacture two-material bistable units using multi-material additive manufacturing (Ultimaker 3), as shown in Fig. S2B. Noting that, due to the limited accuracy of the 3D printer, especially at the positions of the cut, the bistable units here are two times larger than the laser-cut specimens to ensure the accuracy of printing. We use Solidworks to design a tandem structure composed of two bistable units, as shown in Fig. S2A where the white and yellow regions represent TPU and PLA, respectively. To ensure that no debonding occurs at the connecting interface between PLA and TPU, it is necessary to allow them to overlap in a certain area when modeling. Finally, the sample is printed by the 3D printer and trimmed to a finished product, as shown in Fig. S2D.

3. Fabrication of elastomeric films by casting molding

Figure S3A-E shows the process of silicone preparation by mixing and evacuation. The elastomeric layers considered in this study are made of silicone rubber (Dragon Skin 30, Smooth-On, Inc.). We mix Part A and Part B of Dragon Skin 30 at a ratio of 1:1 and add some green paint to it. After stirring fully, there are bubbles in the silicone rubber. To ensure the stability of the mechanical properties of silicone rubber, the silicone is degassed in a vacuum chamber for a couple of minutes to remove bubbles. The molds are designed in Solidworks and 3D printed using an Objet Connex 500 printer (Stratasys). Note that, some small grooves (Fig. S3F) should be reserved in the molds to allow for some protrusions on the casted elastomeric films. The bumps can fill the holes of the tandem bistable units so that the elastomeric films can be better fixed in the bistable units. As shown in Fig. S3F-H, we overpour the prepared silicone rubber into the molds and squeeze out the excess silicone using the matched lids. Vaseline is applied to the surface of the molds and lids so that the elastomeric layers can easily separate from the molds. The elastomers are left for 10 hours at room temperature to cure. Then, we remove them from the molds and trim them for post-processing (Fig. S3J-L). Finally, the elastomeric films are obtained for assembly.

4. Assembly of bistable self-folding (BSF) elements

The assembly process of bistable units and elastomeric films requires the components shown in Fig. S4A, in which silicone adhesive (RTV silicone adhesive, MingSheng, Inc.) is used to better connect the bistable units and elastomeric layers. The following are the nine steps for fabricating the multi-step multimodal mechanical metamaterials (see, Fig. S4B).

Step 1: we apply a layer of silicone adhesive on the bumps of the elastomeric films. Similarly, the small connecting layers should also be coated with the adhesive.
Step 2: we align the protrusions of the elastomeric films with the holes of the bistable units.

Step 3: we insert the protrusions into the holes. And the silicone adhesive can connect both silicone and TPU materials very well.

Step 4: Flip the bistable units and affix the small connecting films to the position of the holes, ensuring the stability of the connection between the bistable units and elastomeric layers.

Steps 5-8: Using the same assembly method as mentioned before, another elastomeric layer can be assembled to the bistable units. And it takes 5 hours for the silicone adhesive to fully cure.

Step 9: After being stretched and released, the structure can produce two folds.

5. Material characterization

To obtain the stress-strain responses of the silicone elastomer, TPU, and PLA, uniaxial tensile tests of dog bone-shaped specimens are performed, as shown in Fig. S5. The uniaxial testing machine (Zwick Z005) is equipped with a 5 KN load cell. Specimens are fully clamped at both ends and stretched at a strain rate of 0.2 min⁻¹ at 25°C.

Material characterization of silicone elastomer. The experimentally measured stress-strain responses of three specimens of silicone elastomer are shown as the blue area (Fig. S5A). Their average is also included as the solid line. Its constitutive behavior is assumed to follow the Mooney-Rivlin hyperelastic model, with the strain energy density given by

\[ W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) \]  

(S1)

where \( I_1 \) and \( I_2 \) are the first and second invariant of \( B = (\det B)^{-1/3} B \) with \( B = F F^T \) being the left Cauchy-Green tensor of the deformation gradient \( F \). The corresponding Cauchy stress \( \sigma \) can be written as

\[ \sigma = \frac{2}{3} \left( C_{10} I_1 - C_{01} I_2 \right) I + 2C_{10}B - 2C_{01}B^{-1} \]  

(S2)

For the uniaxial tension, Eq. (S2) reduces to

\[ \sigma_{11} = 2C_{10} \left( \lambda - \lambda^{-1} \right) + 2C_{01} \left( 1 - \lambda^{-1} \right) \]  

(S3)

where \( \sigma_{11} \) is the uniaxial tensile stress and \( \lambda \) is the percentage elongation in the loading direction. By fitting Eq. (S3) against the experimental results, the material constants can be obtained as \( C_{10} = 0.157 \text{ MPa} \) and \( C_{01} = -0.037 \text{ MPa} \).

Material characterization of TPU. Similar to the previous analytical method for the silicone elastomer, the experimentally measured stress-strain response of TPU (Fig. S5B) can be assumed to follow the Mooney-Rivlin hyperelastic model. The material constants can be obtained as \( C_{10} = -10.85 \text{ MPa} \), \( C_{01} = 23.98 \text{ MPa} \).

Material characterization of PLA. The experimentally measured stress-strain response of PLA is shown in Fig. S5C, which indicates that the PLA is characterized by Young’s modulus of 2.3 GPa. Moreover, the yield strength is measured as \( \sigma_y = 55.6 \text{ MPa} \) and the corresponding yield strain is \( \varepsilon_y = 0.024 \).

Dynamic mechanical analysis (DMA). The dynamic thermomechanical properties of the materials are obtained by dynamic mechanical analysis (DMA), using a TA Instruments RSA3 dynamic mechanical analyzer. The dimensions of the PLA and TPU samples are 25 mm×8 mm×2 mm. The samples oscillate at 1 Hz to 0.02% strain. The temperature is ramped from 25°C to 90°C at a ramp rate of 1°C per minute. The temperature-dependent storage modulus and loss factor are shown in Fig. S6. It can be seen that the storage modulus of PLA is 2.3 GPa at room temperature (25°C), while it decreases to 2.7 MPa when the temperature is heated to
80°C. The phase transition of PLA from the glassy phase to the rubbery phase leads to a sharp drop in the storage modulus. The peak of the loss factor tanδ of PLA is associated with the glass transition temperature at $T_g = 75°C$.

6. Finite-element simulations

Finite-element (FE) modeling is used to simulate the deformation of the mechanical metamaterials, with the commercially available software ABAQUS (version 2017). In the simulations, contact and geometrical nonlinearity are considered. A quasi-static general algorithm is adopted. Note that, to stabilize the simulations, an artificial inter-dissipation factor of $2 \times 10^{-4}$ is introduced in the FE modeling. The model is meshed using hybrid 8-node linear brick elements (i.e., C3D8H elements in ABAQUS) and mesh sensitivity is conducted to ensure numerical convergence. Mooney-Rivlin hyperelastic model is used as a material model. The material constants $C_{10}$ and $C_{01}$ of TPU and silicone elastomer are given in Fig. S5. As for the FE simulations with varying temperatures, the dependence of the modulus and loss factor upon temperature given in Fig. S6A is taken into account. A multi-step geometrically nonlinear static analysis is performed for both stretching loading and unloading. As for the loading process, a displacement controlled loading is applied to the structure, leading to snap-through of the bistable elements. The unloading is realized by removing the loading, resulting in folding due to the eccentric compression exerted by the stretched films.

7. Tensile tests at temperatures of 25°C and 80°C

Uniaxial tensile tests of three different BSF elements (i.e., Case 1, Case 2, and Case 3) are shown in Fig. S7A and B. The uniaxial testing machine (Zwick Z005) is equipped with a 50 N load cell. Specimens are fully clamped at both ends and stretched at a strain rate of 0.1 min$^{-1}$ at room temperature (25°C). The experimentally measured force-strain curves of these elements are shown as colored areas in Fig. S7A. The averages of these tests are shown as the solid lines. Then we connect these three BSF elements in series and stretch them experimentally (Fig. S7D). The measured force-strain curve is shown in Fig. S7C, in which the marked three peaks correlate to the sequential snap-throughs of Case 1, Case 2, and Case 3.

To perform tensile tests on the specimens at higher temperatures, we build a water bath tensile test platform consisting of a metal collect, a digital water bath, and a uniaxial testing machine (Zwick Z005) as shown in Fig. S8. The digital water bath can heat water to control the temperature between 25°C and 100°C. The metal collet is immersed in the digital water bath. The bottom end of the tested specimen is fixed by the collet’s gravity. The upper end is fixed by the uniaxial testing machine. During stretching, specimens are immersed in water.

Uniaxial tensile tests of two different BSF elements at temperatures of 25°C and 80°C are shown in Fig. S9. The solid and dashed lines in Fig. S9A are the average values of the experimental results shown as colored areas at 25°C and 80°C, respectively. The force-strain curves show bistability at 25°C, and monostability at 80°C. Snapshots of the experimentally observed stretching process at 80°C are shown in Fig. S9B. In Fig. S10, three BSF elements are stretched at 25°C and 80°C. The corresponding force-strain curves are shown in Fig. S10A. It can be seen that the transformation forces are $F_1 > F_2 > F_3$ at 25°C, but switching to $F_2 > F_1$ at 80°C, consistent with the FEM simulations.

Supplementary Text

1. Force-displacement response of the bistable self-folding element
In the main text, the realization of the stepwise multimodal mechanical deformation mainly depends on the varied mechanical properties of different bistable self-folding elements. The quantitative study of the force-displacement response is similar to the previous work (51) (Fig. S11). The bistable self-folding element is composed of a bistable unit and an elastomeric film as shown in Fig. S11A and B. The specific dimensions (i.e., \(a, h, l, L, b,\) and \(t\)) are marked in the figure. Note that in the text, we keep \(a, l, L,\) and \(b\) unchanged, and vary \(h\) and \(t\) to change the mechanical response.

As shown in Fig. S11C, the bistable unit is subjected to tension in the vertical direction, the four square plates undergo rotation, and the interconnect ligaments between each plate bend and can be approximated as rotational springs with torsion stiffness \(K_\theta\). Moreover, the four cantilevers bend and induce horizontal force on their adjacent square plate. The cantilevers are treated as a linear spring with stiffness \(K\), one end of which is connected to the square plate, with the other end connected to a free movable simple support. Considering the deformation of the bistable unit mainly localizes at the ligaments and the square plates can be approximated as rigid. Accordingly, a simplified mechanical model corresponding to the deformation pattern can be proposed (Fig. S11D). The model consists of one rigid square plate of dimensions \(L \times L\), one linear spring, and three rotational springs. The vertical force \(F_y\) is applied at point \(A\) so that the rigid square rotates clockwise. The inclined angle of \(AB\) is denoted by \(\theta\) representing the rotation of the plate and the vertical displacement of point \(A\) is \(V\). The compressed length of the linear spring can be obtained as \(\Delta = L(\sin \theta + \cos \theta - 1)\) and the rotation angle of the three rotational springs is \(\theta\). Therefore, the elastic deformation energy stored in the mechanical system is

\[
W = \frac{3}{2} K_\theta \theta^2 + 2KL^2 \left( \sin \theta + \cos \theta - 1 \right)^2 \tag{S4}
\]

where \(\theta = \arcsin \left( \frac{V}{L} \right)\). The vertical force \(F_y\) conjugated to \(V\) can be obtained as

\[
F_y = \frac{dW}{dV} = KL \left[ \cos \left( \arcsin \frac{V}{L} \right) + \frac{V}{L} - 1 \right] \left[ 1 - \tan \left( \arcsin \frac{V}{L} \right) \right] + \frac{3K_\theta \arcsin \left( \frac{V}{L} \right)}{L \cos (\arcsin V/L)} \tag{S5}
\]

Eq. (S5) can be written in a dimensionless form as

\[
F_y^* = \frac{F_y}{KL} = \left( \sqrt{1-V^{*2}} + V^*-1 \right) \left[ 1 - \frac{V^*}{\sqrt{1-V^{*2}}} \right] + \frac{3\beta \arcsin V^*}{\sqrt{1-V^{*2}}} \tag{S6}
\]

where \(F_y^* = F_y/KL\) and \(V^* = V/L\) are the non-dimensional force and displacement, and \(\beta = K_\theta/(KL^2)\) is the only parameter in determining the force-displacement response.

Figure S11E shows the normalized force-displacement response of the simplified model with \(\beta = 0.03\). When the normalized displacement \(V^*\) is 0 (Point A), 0.805 (Point B), and 0.968 (Point C), the normalized force \(F_y^*\) is zero, indicating two stable states (bistability). Fig. S11F shows the normalized force-displacement curves for a series of \(\beta\) ranging from 0.01 to 0.08. From Eq. (S6), one can numerically calculate the critical value of \(\beta = 0.047\) that separates bistability and monostability: when \(\beta < 0.047\), \(F_y^*\) crosses the zero horizontal line more than once, a feature of bistability, while \(F_y^*\) is always above zero for \(\beta > 0.047\). The latter is typical for monostable systems.

Then, we quantify the stiffness of the rotational spring \(K_\theta\) and the linear spring \(K\) in terms of the geometrical parameters \(l, h, L,\) and \(b,\) and the material parameters of the bistable unit (i.e., Young’s modulus \(E\) and Poisson’s ratio \(\nu\)). Through dimensional analysis, \(K_\theta\) and \(K\) can be assumed to have the following form of
\[ K_\theta = \alpha_1 Eb l^2 \]  
\[ K = \frac{\alpha_2 Eb h^3}{L^2} \]  
(S7)  
(S8)

where \( \alpha_1 \) and \( \alpha_2 \) are the dimensionless coefficients to be determined. The determination of \( \alpha_1 \) and \( \alpha_2 \) is discussed in detail in the previous work (51). They can be obtained as \( \alpha_1 = 0.08 \) and \( \alpha_2 = 0.056/(h/L + 0.054) - 0.037 \). Once \( K_\theta \) and \( K \) are determined, \( \beta \) can be expressed as

\[ \beta = \frac{K_\theta}{KL^2} \frac{l^2 L}{\alpha_2 h^3} = \frac{0.08(l/L)^2}{[0.056/(h/L + 0.054) - 0.037](h/L)^3}. \]  
(S9)

A series of FE simulations for different sets of geometrical parameters \( l/L \) and \( h/L \) are conducted and are shown in the phase diagram as symbols (Fig. S11G), with \( \beta = 0.047 \) denoted by the dashed line, i.e.,

\[ l/L = \sqrt{\frac{0.033}{h/L + 0.054} - 0.022}(h/L)^3. \]  
(S10)

Next, a quantitative study of the elastomeric layer is carried out as follows. Because the elastomeric layer is made of silicone elastomer, an incompressible Mooney-Rivlin hyperelastic constitutive relation can be used, with the uniaxial tensile stress \( \sigma_{11} \) and elongation in the loading direction \( \lambda \) given by Eq. (S3). And \( \lambda \) can be expressed as

\[ \lambda = 1 + D/2a \]  
(S11)

where \( D \) is the elongation of the elastomeric layer. By submitting Eq. (S11) into Eq. (S3), the tensile stress \( \sigma_{11} \) can be expressed as a function of the elongation \( D \). As shown in Fig. S11H, the tensile force of the elastomeric layer can be expressed as

\[ F_D = \sigma_{11} \cdot A = \sigma \frac{8aLt}{2a + D}. \]  
(S12)

Combining Eqs. (S3), (S11), and (S12), the tensile force \( F_D \) can be explicitly expressed as a function of elongation \( D \).

2. Analysis of the folding process

The folding process of the BSF element can be divided into two stages (Fig. S12A). First, the element is stretched until the BSF element reaches its second stable state, and the elastomeric layer is highly stretched (State 2). When the stretching load is released, the element undergoes out-of-plane bending, since the eccentric compression imposed by the stretched elastomeric layer gives a folding angle \( \phi_0 \) (State 3). For a better illustration, side views of the deformed states (State 2) are given in Fig. S12B. As for state 2, after releasing the stretching force \( F \), the resistant force \( F_D \) sustained by the elastomeric layer can result in a bending moment of \( M = (b+t)F_D/2 \). Therefore, the bending moment induces out-of-plane deformation of the BSF element.

To determine the folding angle \( \phi_0 \), the folded element (Fig. S12C) can be simplified as a rod-spring model (Fig. S12D). Considering that the modulus of the constituent material of the bistable unit is much greater than that of the elastomeric film (silicone elastomer), the folded bistable unit can be represented by the rigid rods connected by a rotational spring with torsional stiffness \( K_\phi \). The elastomeric film can be approximated by a spring with stiffness \( K_D \), two
ends of which are connected to Points D and E. To ensure that the system is in a balanced state, the line spring needs to produce an initial elongation of \( D_0 \) for a given folding angle \( \phi_0 \) (the rotational angle of the torsional spring). It is noted that the thicknesses of the bistable unit and the elastomeric layer are \( b \) and \( t \) and can be represented by the rigid rods BD and EF in the model of equivalent length \( t_e = (b + t)/2 \).

Through a geometric and equilibrium analysis, the folding angle \( \phi_0 \) can be obtained quantitatively. Following a geometric analysis, its geometric relationship is shown in Fig. S12E, in which the length of AC is \( S = d + 2a + 0.85L/\sin(\phi_0/2) \) and that of the elastomeric layer is \( 2a + D_0 \). The elongation of the elastomeric layer \( D_0 \) is then be given by

\[
D_0 = 2(S - d) \sin\left(\frac{\phi_0}{2}\right) - 2t_e \cos\left(\frac{\phi_0}{2}\right) - 2a. \tag{S13}
\]

As to the equilibrium analysis, we consider the right half rigid rod of the model (Fig. S12F), where \( M \) refers to the bending moment induced by the rotational spring, \( F_x \) and \( F_y \) are the forces in the horizontal and vertical directions, respectively, and \( F_D \) is the force sustained by the elastomeric layer. And \( M \) can be expressed in terms of \( F_D \) as

\[
M = F_D \cos\left(\frac{\phi_0}{2}\right)(S - d) + F_D \sin\left(\frac{\phi_0}{2}\right)t_e. \tag{S14}
\]

Considering the identity \( M = K_\phi(\pi - \phi_0) \), Eq. (S14) can be expressed as

\[
F_D = \frac{K_\phi(\pi - \phi_0)}{\cos\left(\frac{\phi_0}{2}\right)(S - d) + t_e \sin\left(\frac{\phi_0}{2}\right)}. \tag{S15}
\]

in which \( F_D \) is a function of \( D_0 \) given by Eq. (S12), and \( K_\phi \) is the equivalent torsional stiffness. To determine \( K_\phi \), we need to simulate the response \( \pi - \phi \) of the bistable unit to the quasi-static bending moment \( M \) (i.e., \( K_\phi = M/(\pi - \phi) \) shown in Fig. S12G). Dimensional analysis shows that \( K_\phi \) can be assumed to have the following form of

\[
K_\phi = \alpha Eb^2L \tag{S16}
\]

where \( \alpha \) is a dimensionless coefficient determined by fitting the obtained FE results shown in Fig. S12H, giving \( \alpha = 0.00291b/L + 0.00074 \). Combining Eqs. (S12), (S13), (S15), and (S16), the folding angle \( \phi_0 \) can be obtained numerically.

To validate the above theory, the experiment and FE simulation are conducted for a folded bistable self-folding element with the dimensions \( b = 1.5 \) mm, \( t = 0.75 \) mm, \( a = 10 \) mm, and \( L = 2.5 \) mm. The corresponding results are given in Fig. S12I, in conjunction with the corresponding theoretical prediction. Excellent agreement among them indicates the reliability of the theory.

3. Temperature-dependent response of the bistable self-folding element

To overcome the limitations of irreversible deformation and single deformation pathway, we introduce a temperature-responsive part (TRP) into the bistable self-folding element by partially replacing the cantilever (Fig. S13A). To quantitatively analyze its behavior, we analyze a bi-material cantilever with one end fixed and the other end subject to force \( F \) (Fig. S13B). The Euler-Bernoulli beam theory is adopted. The bi-material cantilever is equivalent to a T-shaped cantilever made of one material as shown in Fig. S13C. According to the cross-
section of the T-shaped cantilever (Fig. S13D), the position of the neutral axis \( c \) relative to the interface can be obtained as
\[
c = \frac{(E_1h_1^3 - E_2h_2^3)}{2(E_1h_1 + E_2h_2)} \quad (S17)
\]
where Young’s moduli \( E_1 \) and \( E_2 \), and thickness \( h_1 \) and \( h_2 \) refer to PLA and TPU, respectively. The moment of inertia can be given as
\[
I = \frac{1}{12}bE_2h_2^3 + bE_1\left(\frac{h_1}{2} - c\right)^2 + \frac{1}{12}bh_2^3 + bh_2\left(\frac{h_2}{2} + c\right)^2. \quad (S18)
\]
The equivalent stiffness of the cantilever is thus
\[
K = \alpha \frac{E_2I}{L^2} = \alpha\left[\frac{bE_2h_2(h_1 + h_2)^2}{4(E_1h_1 + E_2h_2)L^2} + \frac{b}{12L^2}(E_1h_1^3 + E_2h_2^3)\right] \quad (S19)
\]
in which dimensionless coefficient \( \alpha \) can be obtained empirically as
\[
\alpha = -1.087 + 0.692 \arctan \left[ \left(\frac{E_1}{E_2}\right)^{1/3} \frac{h_1}{L} + \frac{h_2}{L} \right] + \frac{0.852}{0.0651 + \left(\frac{E_1}{E_2}\right)^{1/3} \frac{h_1}{L} + \frac{h_2}{L}} \quad (S20)
\]
Therefore, \( \beta \) mentioned can be given as
\[
\beta = \frac{K}{KL^2} = \frac{\alpha E_1h_1^2}{\alpha E_2I L^2} = \frac{\alpha_1}{\alpha} \frac{bL^2}{T} \quad (S21)
\]
Combining Eqs. (S18), (S20), and (S21), the parameter \( \beta \) can be obtained numerically.

At room temperature (25°C), the moduli of TPU and PLA are \( E_2 = 78.2 \text{ MPa} \) and \( E_1 = 2.3 \text{ GPa} \), respectively. When the temperature rises to 80°C, \( E_2 = 40 \text{ MPa} \) and \( E_1 = 2.7 \text{ MPa} \). As for a temperature-responsive bistable unit with dimensions \( h_1 = 3 \text{ mm} \), \( h_2 = 1 \text{ mm} \), \( l = 0.4 \text{ mm} \), \( L = 5 \text{ mm} \), and \( b = 3 \text{ mm} \) (Fig. S13E), the parameter \( \beta \) can be obtained as \( \beta = 0.0103 \) for \( T = 25^\circ \text{C} \) and \( \beta = 0.0805 \) for \( T = 80^\circ \text{C} \). Submitting these \( \beta \) into Eq. (S6), it can be seen that the unit changes from bistable to monostable by increasing the temperature from 25°C to 80°C (Fig. S13F). Similarly, for another unit with \( h_1 = 4 \text{ mm} \) and \( h_2 = 1 \text{ mm} \), the switch between bistability and monostability can also be achieved by changing temperature (Fig. S13G and H).

4. Force-displacement response of a series of BSF elements

As for a structure consisting of a series of BSF elements, its multistability inherits from the BSF elements. When the BSF element is subject to a force controlled loading, snap-through occurs at the peak as shown in Fig. S14A. When entering the negative stiffness regime, a BSF element is kinetically deterministic while the multistable structure is not, because the negative stiffness units are connected in series and there is more than one equilibrium state. As shown in Fig. S14C for a structure with three identical BSF elements (defined as unit a, unit b, and unit c), the structure deforms uniformly before reaching the peak force \( F_{\text{max}} \) and the three units have the same deformation and force-displacement curve. Beyond \( F_{\text{max}} \), one of the units (unit a) is stretched to its second stable state, and the other two units shrink. Upon further stretching, unit b and unit c reach their second stable state in turn. Namely, upon deformation of one unit, all the others have to rearrange their deformation, i.e., the behavior is nonlocal, and the order of deformation is to some extent random. With the increasing number of BSF elements, the path becomes increasingly smooth and gradually converges into the flat snap-through path.
Due to the multistability, the structure has multiple stable states and stable lengths, and the loading process may start from and stop at any stable length. It can be seen from Fig. S14A that the curve can be divided into three parts, i.e., Stage A, Stage B, and Stage C, as shown in Fig. S14B. To obtain the loading curve of arbitrary stable length, we formulate the theoretical stiffness using a model composed of a series of nonlinear springs as shown in Fig. S14D and E. The number of the units in Stage A, Stage B, and Stage C are denoted as \( n_A \), \( n_B \), and \( n_C \), respectively, where \( n = n_A + n_B + n_C \) is the total number of units and \( n_B \) satisfies \( n_B = 0 \) or \( 1 \).

The deformation process can be discussed in two cases: the first case (Fig. S14D) is that all units are in Stage A or Stage C; the second case (Fig. S14E) is that only one unit is in Stage B, and the rest of the units are in Stage A or Stage C. The BSF elements can be modeled as nonlinear springs, and the constitutive relation is summarized in Fig. S14B, expressed as

\[
F_i = K_i(V_i)
\]

where \( V_i \) is the displacement of the \( i \)th BSF element. Here, we only consider the regime when the force is less than \( F_{\text{max}} \). Considering the series connection, the force \( F \) applied to the structure satisfies

\[
F = F_i
\]

and the total displacement is the summation of those of all units,

\[
V = \sum_{i=1}^{n_A} V_{iA} + n_B V_{iB} + \sum_{i=1}^{n_C} V_{iC} = \sum_{i=1}^{n_A} K_{iA}^{-1}(F) + n_B K_{iB}^{-1}(F) + \sum_{i=1}^{n_C} K_{iC}^{-1}(F)
\]

where the \( V_{iA} \), \( V_{iB} \), and \( V_{iC} \) refer to the displacement of the \( i \)th unit in Stage A, Stage B, and Stage C, respectively, and \( K_{iA}^{-1} \), \( K_{iB}^{-1} \), and \( K_{iC}^{-1} \) are the inverse functions of Eq. (S22).

Figure S14F shows the theoretically predicted force-strain curves of a multistable structure consisting of three identical units with parameters given in Fig. S14A and B. The dotted line is obtained theoretically. Considering the series-connected structure is loaded by a displacement controlled loading, the black solid line in the figure is the actual force-strain curve. Similarly, the force-strain curve of a structure with nine identical units connected in series is shown in Fig. S14G. A zigzag-shaped force plateau with the value of \( F_{\text{max}} \) is present.

Considering three BSF elements with different geometric dimensions (i.e., Case 1, Case 2, and Case 3), the corresponding force-displacement curves are shown in Fig. S15A, and each curve can be divided into three stages, as shown in Fig. S15B. In the series-connected structure, the number of Case 1, Case 2, and Case 3 can be represented by \( n_1 \), \( n_2 \), and \( n_3 \). Firstly, as for the multistable structure with \( n_1 = n_2 = n_3 = 1 \), there is a force-strain curve with three zigzag-shaped force plateaus of different heights (Fig. S15C). When we change the composition ratio of the three units (i.e., \( n_1 = 3 \), \( n_2 = 2 \), and \( n_3 = 1 \)), the strain range corresponding to the three force plateaus in the force-strain curve can be tuned, see Fig. S15D.

5. Design strategy

For the transformation of 1D structures into stable 2D structures, the flowchart of the design strategy is summarized in Fig. S16, with the construction process of a three-step deployable mechanical metamaterial shown in Fig. S16B. First, we need choose the number of steps for the deformation of a metamaterial (e.g., \( n=3 \) for the shown example). Second, three different rod-dot models are chosen for assembly as shown in Fig. S16B, in which blue, pink, and green dots indicate hinges with different deformation orders, and hollow and solid dots represent concave and convex rotations respectively. Third, we arrange the three rod-dot models in a different order and switch the rotation of each dot (i.e., concave or convex) to form
a 1/4 unit cell. There are $3 \times 2^3 = 48$ different combinations for the 3-step deformation. Similarly, as for an $n$-step deformation metamaterial, there are $n!2^n$ different combinations. **Fourth**, change the rotation direction of the dots in 1/4 unit cell obtained before. **Fifth**, connect them in series to form a 1/2 unit cell. **Sixth**, rotate the half unit 180° to obtain another 1/2 unit cell. **Seventh**, glue the two 1/2 unit cells obtained before to form a unit cell. **Eighth**, we need determine whether there are intersections of rods in the deployment process. If yes, the arrangement cannot form a deployed metamaterial and is abandoned. Otherwise, we proceed to the next step. **Ninth**, we need determine whether there is overlap, i.e., whether the deformation of two 1/2 unit cells is identical. If yes, the cellular structure cannot be obtained through deployment. If not, we go to the next step. **Tenth**, the unit cell can then be tessellated to form a 3-step deformation mechanical metamaterial. Similarly, a 1D-to-2D deployable mechanical metamaterial that has $n$-step deformation can be realized.

Based on the above design strategy, we used MATLAB code to generate all the 2D deformation modes of $n$-step deployable mechanical metamaterials. The obtained deformation modes of 3- and 4-step mechanical metamaterials with different combinations are summarized in Fig. S17 and Fig. S18, respectively. For the 4-step metamaterials, there are 128 possible structures existing through calculations, and for the sake of brevity, only 22 of them are given in Fig. S18.

As to the design strategy of transforming 2D structures into stable 3D structures, its flowchart is given in Fig. S19A. The construction procedure of a two-step deployable mechanical metamaterial is shown in Fig. S19B. Similar to the cases of 1D structures into stable 2D structures, **first**, we need choose the number (e.g., $n=2$) of steps for the deformation of a metamaterial. **Second**, two different rod-dot models are chosen for assembly as shown in Fig. S19B. **Third**, we arrange the two rod-dot models in a different order and switch the rotations of each dot (i.e., concave or convex) to form a 1/8 unit cell. **Fourth**, we change the rotation direction of the dots in 1/8 unit cell obtained before. **Fifth**, we connect them in series to form a 1/4 unit cell. **Sixth**, it is rotated by 90°, 180°, and 270°, respectively, to obtain three more 1/4 unit cells. They are then combined to form a unit cell. Considering that the 2D unit cells should be able to densely tessellate the whole plane, we choose to construct the unit cell in an orthogonal pattern. There are, of course, other different ways of construction. **Seventh**, we need determine whether there are intersections of rods in the deployment process. If yes, the arrangement cannot form a deployed metamaterial. If not, continue to the next step. **Eighth**, we arrange four unit cells to get the desired close-packed or non-close-packed structure. Again, a 2D-to-3D deployable mechanical metamaterial with $n$-step deformation can be similarly realized.

The deformation modes of the 2-, 3-, 4-step mechanical metamaterials with different combinations, obtained using a MATLAB code, are shown in Fig. S20, S21, and S22, respectively. For each case, the first row shows the constituent unit cells. The second and the third rows show the close-packed and non-close-packed 2×2 metamaterials, respectively.

### 6. Multi-step deformation

In the main text, we have realized deployable mechanical metamaterials with 1-step, 2-step, and 3-step deformation. The specific geometric dimensions of the bistable self-folding elements in the metamaterials are given in Fig. S23. As for the metamaterial with 1-step deformation, all the constituent elements are the same and the geometric parameters are $h = 2.5$ mm and $t = 0.75$ mm (Fig. S23A). As for 2-step deformation, there are two different constituent elements (i.e., Case 1 with $h = 1.75$ mm , $t = 0.75$ mm and Case 2 with $h = 2.5$ mm , $t = 0.75$ mm), as shown in Fig. S23B. As for 3-step deformation, there are three different constituent elements (i.e., Case 1 with $h = 1.75$ mm, $t = 0.4$ mm, Case 2 with
$h = 1.75 \text{ mm}, \ t = 0.75 \text{ mm}$, and Case 3 with $h = 2.5 \text{ mm}, \ t = 0.75 \text{ mm}$), as shown in Fig. S23C.

In fact, $n$-step multimodal mechanical metamaterials can be designed according to the design method mentioned in the main text. Here, a 4-step multimodal mechanical metamaterial is discussed as a typical example.

As shown in Fig. S24A, blue, pink, green, and purple dots indicate the rotation of the first, second, third, and fourth steps, driving two rods to produce relative rotation. Here, four bistable self-folding elements with different geometric dimensions (Case 1, Case 2, Case 3, and Case 4 in Fig. S24B) are used to represent the four steps of deformation. They share the same structural design ($a = 11 \text{ mm}, \ b = 1.5 \text{ mm}, \ L = 2.5 \text{ mm}$) but differ in $h$ and $t$ (Fig. S24C). Figure S24B shows the FE simulated stretching force as a function of strain for the four samples, in which the maximum of the force gradually increases from Case 1 to Case 4. Then we design a 4-step deformation mechanical metamaterial composed of four kinds of bistable self-folding elements (Fig. S24D). The one-dimensional structure can be sequentially transformed into Mode 1, Mode 2, Mode 3, and Mode 4 in four steps. Meanwhile, FE simulation of the deformation process (Fig. S24E) is present and the results are consistent with our designed configurations. Then we tessellate the four samples (Case 1, Case 2, Case 3, and Case 4) into a $3 \times 2$ structure (Fig. S24F) in the designed order, showing that multiple deformation modes can be realized by stepwise loading.

7. Temperature-induced recoverable deformation

By introducing the temperature-responsive part (TRP) into the bistable self-folding element to partially replace the cantilever, the folded elements can return to the initial state when they are heated. The principle of this phenomenon has been discussed in Fig. 4 and Fig. S13. As shown in Fig. S25, we use the FE method to explore the recoverable deformation of a structure. The temperature dependence of the material properties is considered in the simulations. At room temperature (25°C), the unit cell is stretched and released, resulting in a folded hexagonal structure (Fig. S25). Then the temperature is raised to 80°C. Each element in the unit cell becomes monostable, causing the disassembly process of the folded structure by snapshots. Compared with the process shown in Fig. 4E and F of the main text, the FE simulated results are similar to the deformation observed in experiments.

Furthermore, we use the temperature-responsive bistable self-folding elements to form a 3D unit cell experimentally (Fig. S26A). The 2D structure can be stabilized in a 3D structure by load stretching and releasing. Then the 3D structure is immersed in water at a temperature of 80°C, resulting in a rapid decrease in the modulus of the PLA part in the cantilever. As a result, the folded elements in the structure become monostable and return to their original states. We also use the FE method to explore the recoverable deformation (Fig. S26B). The FE simulated results are in good agreement with the experiments.

8. Temperature-induced bifurcation of deformation pathways

In the main text, Figure 5 presents two deformation pathways for the 3-step deformation metamaterial by adjusting the temperature. Here, to demonstrate the diversity of deformation pathways, we introduce a 4-step deformation metamaterial to realize the multiple bifurcations. Figure S27 shows the schematic illustration of the temperature-induced bifurcation of 4-step deformation pathways. At room temperature (25°C), four configurations (Mode 1, 2-1, 3-1, and 4) of the unit cell can be produced by 4-step loading, forming Pathway 1. To realize Pathway 2, we put the structure of Mode 1 in Pathway 1 into an environmental bath with the temperature being 80°C, and the positions of pink and purple dots are swapped, producing the configurations of Mode 1, 2-2, 3-2, and 4. As for Pathway 3, we put the structure of Mode 2-2 in Pathway 2 into a room temperature environment again, the positions of green and purple
dots are swapped, producing the configurations of Mode 1, 2-2, 3-3, and 4. As for Pathway 4, the structure of Mode 2-1 in Pathway 1 is put into an environment of 80°C, and the positions of green and purple dots are swapped, producing the configurations of Mode 1, 2-1, 3-3, and 4. In summary, we achieve four deformation pathways of one structure by changing the temperature.

Then, we verify the above design strategy through FE simulations. Figure S28A shows four bistable self-folding elements with the same cantilever of \( h = 6.2 \text{ mm} \), in which the width of the TRP (\( h_i \)) gradually increases from 1 to 4. The results of the FE simulations (W-V in Fig. S28B) indicate that the maximum strain energy satisfies \( W_4 > W_3 > W_2 > W_1 \) at room temperature (25°C). When the temperature is 80°C, the strain energy responses of the four elements are reversed compared to the previous one, causing the maximum strain energy to become \( W_1 > W_2 > W_3 > W_4 \). As shown in Fig. S28C, we then connect the four elements in series in the order of 1-2-3-4 to form the desired 4-step deformation metamaterial. Figure S28D shows the results of the FE simulation, realizing four deformation pathways by changing temperature.

9. Tunable force transmissibility

Considering that the multiple modes of the structure can produce different mechanical responses, the multi-step multimodal metamaterials can be used to tune force transmissibility. Here, we use a two-step deformation mechanical metamaterial that has two deformation modes to realize the switch between an isolating mode and a responsive mode in the low frequency range (e.g., lower than 30Hz). As shown in Fig. S29A, we construct a 2D 2-step deformation metamaterial that can be transformed into two deformed configurations (Mode 1 and Mode 2). The thicknesses of the elastomeric layers and the bistable units are 0.75 mm and 1.5 mm, respectively. The difference between the two constituent elements lies in the width of the cantilever. One is 2.5 mm and the other is 1.75 mm. Note that there are three layers of the 2D structure in the z-direction to prevent the out-of-plane buckling during the compression. Then displacement controlled quasi-static compression tests of Mode 1 and Mode 2 are conducted using the material test machine – Zwick (model Z005) with a cross-head loading rate of 2 mm/min. The force is measured by the load cell with a resolution of 0.01 N. Note that Vaseline needs to be applied to the surface of the indenter to reduce the friction between the indenter and the samples. During the test, an HD video camera is used to record the global as well as local deformation. The measured uniaxial compressive force-strain curves of Mode 1 and Mode 2 are given in Fig. S29B, clearly showing that the two modes correspond to two different mechanical responses. Specifically, at the load of 0.92 N, Mode 1 and Mode 2 exhibit the positive stiffness of \( k_1 = 0.1097 \text{ N/mm} \) and \( k_2 = 0.1514 \text{ N/mm} \). As for Mode 2, the stiffness decreases to \( k_2' = 0.1175 \text{ N/mm} \) when the load is 3.9 N. A series of measured shots of the deformed patterns of Mode 1 and Mode 2 are presented in Fig. S29C and D.

Next, vibration tests are conducted to determine the tunability of force transmissibility in the low frequency range. The experimental setup is shown in Fig. S29E and F. The signal generator (AFG2021 from Tektronix Inc.) generates signals of different frequencies, which are amplified by the power amplifier (HEAS-50 from Nanjing Foneng Inc.). Finally, the amplified signals are transmitted to the vibration table to generate vibration. The vibration table includes an electromechanical shaker (HEV-50 from Nanjing Foneng Inc.) to generate vertical vibrations at various frequencies, two identical acceleration sensors (LC0101 from Lance Technologies Inc.) attached on the bottom and top surfaces to record the input and output accelerations \( a_{in} \) and \( a_{out} \), respectively, and the deformed structure (Mode 1 or Mode 2) as an isolator. Here, tapes are used between the isolator and the acrylic boards to ensure that the
acrylic boards cannot detach during vibration. Finally, the data are collected through a dynamic data collection device (National Instruments NI 6341 DAQ card and LABVIEW program).

Figure S29G shows the transmissibility at frequencies from 1 to 25 Hz for Mode 1 and Mode 2 under the weight of 92 g. The results show that the resonance peak of Mode 1 (9 Hz) is significantly smaller than that of Mode 2 (13 Hz) due to the difference in compression stiffness. The isolators at Mode 1 can isolate vibrations when the frequency is higher than 13 Hz. As for Mode 2, vibration isolation requires that the frequency is higher than 19 Hz. Moreover, considering that the stiffness of Mode 2 decreases significantly as the strain increases, the transmissibility of Mode 2 under different loads is shown in Fig. S29H. It can be seen that the resonance peak under the mass of 390 g is smaller than that under 90 g.

10. Autonomous untethered robotic material

The self-folding process of the metamaterial can be divided into two stages: the first is stretching, and the second is assembly after releasing the load. As shown in Fig. S30, the initial state of the material is completely stretched. Then, the applied tensile load is released, causing complete self-folding of the metamaterial at an extremely fast speed. The characteristic of the rapid self-folding is attractive compared with the traditional deployable structures.

Here, considering the characteristic of rapid self-folding, we construct an autonomous untethered robotic material by combining a microcontroller, drivers, and a deployable metamaterial (Fig. S31). It can sense the moving velocity and determine whether to expand. The microcontroller (Fig. S31A) contains a lithium battery and a microcontroller integrated with a gyro sensor. The drivers can sense the moving velocity and can transmit a signal to the magnetic valve. The drivers (Fig. S31B) contain pressure canisters, magnetic valves, and air cylinders. The pressure canisters made of silicone are filled with compressed air and are connected to the normally closed magnetic valves. The air cylinders are connected to the other end of the magnetic valves. When the signal exported by the control part is transmitted to the magnetic valves, the magnetic valves can quickly switch from the close state to the open state, causing the compressed air in the pressure canisters to push the piston of the air cylinders to move (see movie S6). As shown in Fig. S31D and E, we integrate the three parts to a thin board to construct the robotic material, in which both ends of the deployable metamaterials are prestretched with ropes.

11. Impact tests for deployed and undeployed structures

We perform drop tests to explore the protective performance of the metamaterials in both deployed and undeployed states. As shown in Fig. S32A and B, a custom impact testing apparatus is developed, including a supporting frame, an acceleration sensor, a dynamic data collection device, and an untethered robotic material. The robotic material can be dropped from different heights to generate various levels of impulse loading. The accelerometer is rigidly attached to the top surface of the robotic material to measure acceleration data. A comparative investigation of the acceleration responses of the deployed and undeployed robotic material is carried out to analyze its cushion property for different states. When the deployed metamaterial is released from a height of 30 cm (Fig. S32D), the acceleration response (Fig. S32C) shows that its peak acceleration is 13G. For comparison, the undeployed metamaterial (i.e., prestretched state) is released from the same height (Fig. S32F). The measured acceleration response in Fig. S32E shows that the peak of acceleration is higher than the previous and increases from 13G to 22G. Through this comparative study, we show their potential use in protective equipment. There are still some areas that can be improved in the future, such as choosing the elastic film with a larger Young’s modulus to have a higher bearing capacity.

12. Bistable units at different scales
Figure 33A shows fabricated bistable units at both millimeter- and centimeter-level. They are prepared by three processes: 3D printing (PµSL), laser cutting, and 3D printing (FDM). The millimeter-level bistable units, shown as specimens (1) to (5) in Fig. 33A, are made of an ultra-tough-low-viscosity (UTL) resin and printed by projection micro stereolithography with the 3D printer micro Arch P150 (Boston Micro Fabrication). The enlarged view of the bistable unit in Fig.33A shows that this printing technique can print the specimens with good accuracy. Then we choose three millimeter-level units (specimen 1, 2, and 4) for tensile testing as shown in Fig. S33B, to demonstrate their experimental feasibility.
Fig. S1.
**Fabrication of tandem bistable units by laser cutting.** (A and B) 3D printing process for manufacturing 1.5 mm thick TPU sheets using an Ultimaker 3 printer. (C to E) Laser cutting process of TPU to obtain tandem bistable units. (F and G) Soldering and assembling with an electric soldering iron. (H) Assembly with elastic films.
Fig. S2.
**Fabrication of bistable units by multi-material 3D printing.** (A) Multi-material design of bistable units. (B) 3D printer that can print two-material structures. (C) The 3D printed bistable units. (D) The final specimen after trimming.
Fig. S3.
Fabrication of elastomeric films. (A to E) Preparation of silicone by mixing and evacuation. (F to I) Casting and molding of silicone. (J to L) Demolding and trimming of elastomeric films.
Fig. S4.
Assembly of bistable self-folding (BSF) elements. (A) Components of the manufacturing process. (B) Snapshots of the 9 steps to fabricate multi-step multimodal mechanical metamaterials.
Fig. S5.
Measured nominal stress-strain responses of three materials at room temperature (25°C). (A) Silicone elastomer, (B) TPU, and (C) PLA.
Fig. S6.
Dynamic mechanical analysis (DMA) of PLA and TPU. (A) Storage modulus. (B) Loss factor tanδ.
Fig. S7.

**Tensile tests at room temperature.** (A) Stretching force as a function of strain for three BSF elements. (B) Snapshots of Case 1, Case 2, and Case 3. (C) Relationship between the tensile force and strain for a model consisting of three elements in series. (D) Transformation snapshots of the serial-connected model.
Fig. S8.
Water bath and tensile test platform.
**Fig. S9.**

**Tensile tests at 25°C and 80°C for two specimens.** (A) Experimentally measured force-strain behaviors of BSF elements at temperatures of 25°C and 80°C. (B) Images of the experimentally observed stretching process at 80°C.
Fig. S10. Tensile tests at 25°C and 80°C for three specimens. (A) Experimentally measured force-strain curves for three BSF elements at 25°C and 80°C. (B) Images of the experimentally observed stretching process at 80°C.
Fig. S11.

Theoretical model of the bistable self-folding (BSF) element. (A) BSF element. (B) Exploded illustration of the BSF element, including a bistable unit and an elastomeric layer. (C) The deformation pattern of the bistable unit during stretching and the equivalent mechanical model. (D) The rotatable square spring model corresponding to a quarter of the bistable unit. (E) Normalized force-displacement response for $\beta = 0.3$. (F) Normalized force-displacement responses for various $\beta$ in the range between 0.01 and 0.08. (G) Phase diagram of bistable and monostable properties for two dimensionless parameters $h/L$ and $l/L$. (H) Mechanical model of the elastomeric layer.
Theoretical model of the self-folding process. (A) The folding process of the BSF element. (B) The side view of the unfolded BSF element and the equilibrium analysis. (C) The folded BSF element. (D) The rigid rod spring model corresponding to the folded BSF element. (E) Geometric analysis. (F) Equilibrium analysis. (G and H) Determination of $K$. (I) The folding angle of the BSF element obtained by experiment, FEM simulation, and theory.
**Fig. S13.**

**Theoretical model of the temperature-responsive BSF element.** (A) A BSF element composed of two materials (PLA and TPU), with an inset denoting a large view of temperature-responsive part (TRP). (B) Bi-material cantilever with one end fixed extracted from the element. (C) T-shaped equivalent cantilever. (D) Cross profile of the T-shaped cantilever. (E and G) Two bistable units with different values of $h_1$. (F and H) Normalized force-displacement responses for the sample with $h_1 = 3 \text{ mm}$ and $h_2 = 1 \text{ mm}$ and the sample with $h_1 = 4 \text{ mm}$ and $h_2 = 1 \text{ mm}$. 

Fig. S14.
Illustration of the loading curve of the multistable structure consisting of a series of BSF elements with the same geometric and material properties. (A) The force-displacement curve for a BSF element with \( h = 2.5 \text{ mm} \) and \( t = 0.75 \text{ mm} \). (B) The nonlinear constitutive relations of the three stages. (C) Snapshots of the deformation of the series-connected structure with three identical BSF elements. (D and E) Schematic of the series-connected nonlinear spring model. (F) Loading force-strain curve for a multistable structure with three identical BSF elements. (G) The curve for a multistable structure with nine identical BSF elements.
**Fig. S15.**  
Illustration of the loading curve of a multistable structure with three different BSF elements. (A) The force-displacement curve for three different BSF elements with $h = 1.75$ mm and $t = 0.4$ mm for Case 1, $h = 1.75$ mm and $t = 0.75$ mm for Case 2, $h = 2.5$ mm and $t = 0.75$ mm for Case 3. (B) The nonlinear constitutive relations of three stages for three different units. (C and D) Loading force-strain curves for a multistable structure with $n_1 = n_2 = n_3 = 1$ and $n_1 = 3$, $n_2 = 2$, $n_3 = 1$. 
Fig. S16.
Design strategy for 1D-to-2D structures. (A) Flowchart of the 1D to 2D deformation design strategy. (B) A design flow for a three-step deformation mechanical metamaterial.
Fig. S17.
Transformations of 1D structures into 2D structures via 3-step loading pathways.
Fig. S18.
Transformations of 1D structures into 2D structures via 4-step loading pathways.
Fig. S19.

**Design strategy for 2D-to-3D structures.** (A) Flowchart of the 1D to 2D deformation design strategy. (B) Design flow for a three-step deformation mechanical metamaterial.
Fig. S20.
Transformations of 2D structures into 3D structures via 2-step loading pathways.
Fig. S21.
Transformations of 2D structures into 3D structures via 3-step loading pathways.
Fig. S22.
Transformations of 2D structures into 3D structures via 4-step loading pathways.
Fig. S23.
Geometrical dimensions of multi-step multimodal mechanical metamaterials. (A) 1-step deformation. (B) 2-step deformation. (C) 3-step deformation. (D and E) Two-dimensional structures.
Fig. S24.

4-step deformation mechanical metamaterials. (A) Schematic of the rotating parts. (B) Force-strain responses of four BSF elements. (C) The BSF element. (D) Schematic illustration of the 4-step deformation using the rod-dot models. (E and F) FEM simulated and experimentally measured 4-step deformation.
Fig. S25.
Temperature-induced recoverable deformation of 2D structures, predicted by FEM simulations.
Fig. S26.
Temperature-induced recoverable deformation of 3D structures. (A) Experimental snapshots of the deformation process. (B) FEM simulated snapshots of the deformation process.
Fig. S27.

Temperature-induced bifurcation of 4-step deformation pathways. By changing the temperature, the metamaterial can be reprogrammed to undergo different deformation pathways.
Fig. S28.
Reprogramming of a 4-step deformation pathway in a metamaterial by temperature-guided bifurcation. (A) Four BSF elements with \( h = 6.2 \) mm (① \( h_1 = 0 \) mm, ② \( h_1 = 1 \) mm, ③ \( h_1 = 1.7 \) mm, ④ \( h_1 = 2.5 \) mm). (B) Dependence of stretching force \( F \) on displacement \( V \) applied to the elements for \( T = 25^\circ C \) and \( T = 80^\circ C \). The deformation pathway is determined by the kinematically obtainable configurations with the minimum strain energy \( (W) \). (C) Schematic of a series-connected structure composed of eight bistable self-folding elements. (D) Temperature-induced bifurcation of the deformation pathway through FEM simulation.
Controllable force transmissibility for different modes. (A) Photographs of the deployable metamaterial in Mode 1 and Mode 2. (B) Force-strain relationships of Mode 1 and Mode 2. (C and D) Snapshots of sequential deformations of Mode 1 and Mode 2. (E and F) Experimental setup used to measure acceleration transmissibility. (G) Acceleration transmissibility of the deployable metamaterial in Mode 1 and Mode 2 under various vibration frequencies from 1 to 25 Hz. (H) Acceleration transmissibility of the deployable metamaterial in Mode 2 under different loads.
Fig. S30.
Snapshots of the deployable metamaterials after releasing the load.
Fig. S31.

**Fabrication of an autonomous, untethered robotic metamaterial.** (A) A microcontroller unit including a microcontroller, a gyro sensor, and a lithium battery is used to sense the velocity and control the deployment of the metamaterial. (B) Drivers consisting of air cylinders, magnetic valves, and pressure canisters to generate driving force. (C) A deployable metamaterial. (D and E) We integrate the above three parts on a thin board to form an autonomous, untethered robotic metamaterial.
Fig. S32.
Impact tests for deployed and undeployed structures.
Fig. S33.
Bistable units of different scales. (A) Seven bistable units obtained from three different fabrication processes. (B) Tensile experiments on millimeter-level bistable units.
Movie S1.

**Design strategy.** We demonstrated our design strategy using a model of rods and dots. The colored dots represent the hinges that can be rotated in a designed order induced by excitations. The elements of different geometrical dimensions correspond to different deformation sequences.

Movie S2.

**Experimental and simulated verification: 1D structures transform to 2D configurations.** 1D mechanical metamaterials with multi-step multimodal deformation are constructed using TPU plates and silicone layers. Following our design framework, multiple 2D deformation modes can be obtained through multi-step loading. Simulated and experimental results verify our design strategy.

Movie S3.

**Experimental and simulated verification: 2D structures transform to 3D configurations.** 2D mechanical metamaterials with multimodal multi-step deformation are constructed using TPU plates and silicone layers. Following our design framework, multiple 3D deformation modes can be obtained through multi-step loading. Simulated and experimental results verify our design strategy.

Movie S4.

**Temperature-induced reversibility of the deformed metamaterials.** Temperature-responsive parts (TRPs) are introduced into the bistable self-folding element. By changing the temperature (i.e., switching between 25°C and 80°C), the element can switch between monostability and bistability, so as to realize recoverable deformation of the metamaterial.

Movie S5.

**Reprogramming of a metamaterial through temperature-induced bifurcation.** Temperature-responsive parts (TRPs) are introduced into the bistable self-folding element. By changing the temperature (i.e., switching between 25°C and 80°C), the deformation pathways can be reprogrammed through modal bifurcation that is guided by adjustment of the width of the TRPs.

Movie S6.

**Applications.** We demonstrate tunable force transmission and an autonomous, untethered metamaterial robot, both constructed using our design framework.