

# Three-dimensional piezoelectric polymer microsystems for vibrational energy harvesting, robotic interfaces and biomedical implants

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**Piezoelectric microsystems are of use in areas such as mechanical sensing, energy conversion and robotics. The systems typically have a planar structure, but transforming them into complex three-dimensional (3D) frameworks could enhance and extend their various modes of operation. Here, we report a controlled, nonlinear buckling process to convert lithographically defined two-dimensional patterns of electrodes and thin films of piezoelectric polymers into sophisticated 3D piezoelectric microsystems. To illustrate the engineering versatility of the approach, we create more than twenty different 3D geometries. With these structures, we then demonstrate applications in energy harvesting with tailored mechanical properties and root-mean-square voltages ranging from 2 mV to 790 mV, in multifunctional sensors for robotic prosthetic interfaces with improved responsivity (for example, anisotropic responses and sensitivity of 60 mV N<sup>-1</sup> for normal force), and in bio-integrated devices with *in vivo* operational capabilities. The 3D geometries, especially those with ultralow stiffnesses or asymmetric layouts, yield unique mechanical attributes and levels of functionality that would be difficult or impossible to achieve with conventional two-dimensional designs.**

Thin-film devices in planar architectures are the basis for entire classes of technologies in flexible electronics/optoelectronics<sup>1,2</sup>, biomedical devices<sup>3,4</sup>, energy storage and conversion systems<sup>5–8</sup>, and micro/nanoelectromechanical systems<sup>9,10</sup>. Piezoelectric devices, which include mechanical sensors<sup>11–13</sup>, ultrasonic transducers<sup>14,15</sup>, energy harvesters<sup>16,17</sup>, and robots and actuators<sup>18,19</sup> adopt many of the same types of planar, multilayer layouts and device design strategies. Progress in such systems relies on advances in the materials chemistry of inorganic ceramics, organic polymers<sup>20,21</sup>, two-dimensional (2D) materials<sup>22,23</sup> and biologically derived systems<sup>24,25</sup>, with a focus on developing enhanced piezoelectric properties such as the voltage/charge coefficient and electromechanical coupling factor<sup>26–28</sup>. Progress in these systems also relies on structural designs<sup>29–31</sup> to develop optimized responses or unusual characteristics such as curvilinear shapes or stretchable mechanics. Such work is currently constrained by thin-film deposition, growth, etching and lithographic patterning techniques, which allow access only to 2D multilayer configurations and, by extension, simple, macroscale three-dimensional (3D) geometries<sup>32</sup>. Deterministic assembly

of piezoelectric material structures and device components into complex, 3D mesoscale architectures could provide valuable additional options in engineering design, as potential routes to unique properties and/or functions that would otherwise be difficult or impossible to achieve.

Among various strategies in 3D microfabrication<sup>33–35</sup>, methods that use compressive buckling associated with forces imparted by soft, elastomeric substrates are particularly attractive. These techniques operate in a high-speed, parallel fashion on devices and structures that combine the most advanced functional materials in the most sophisticated 2D configurations, simultaneously across length scales that range from nanometres to metres<sup>36–41</sup>. In this Article, we apply these methods to 2D, thin-film piezoelectric devices, creating 3D microsystems with advanced capabilities in sensing and energy harvesting through more than 20 different structures. Integration of passive components (for example, electrodes, encapsulation layers, proof mass) and the use of additional concepts in sacrificial support features allows access to 3D functional devices with ultralow stiffnesses, which are of interest in mechanical sensing and harvesting in

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the context of soft biological systems and low-frequency responses. Our experimental measurements of these devices, together with theoretical and finite element analysis (FEA) of their assembly and operation, highlight their capabilities in multidirectional energy conversion, broadband operation and multifunctional sensing.

### Piezoelectric microsystems in 3D formats

Figure 1a shows an exploded view schematic illustration and a scanning electron microscope (SEM) image of a representative 3D piezoelectric microsystem that includes a layer of piezoelectric polymer (polyvinylidene difluoride, PVDF, with thicknesses of 13  $\mu\text{m}$  and 28  $\mu\text{m}$  in thin and thick regions, respectively) and layers of metal (Cr/Au, 10 nm/100 nm in thickness) as electrodes on the top and bottom surfaces. Here, the thick region consists of an extra layer of PVDF. This design provides a spatial variation of bending stiffness in the 2D precursor, thereby yielding engineered folds at desired locations during the compressive buckling. The scheme for forming this structure begins with the use of a set of techniques in planar microfabrication (that is, photolithography, wet/dry etching) to define a 2D precursor structure. Transfer printing onto a prestrained elastomer leads to bonding at lithographically defined sites, such that release of the pre-strain geometrically transforms the system into a well-defined resultant 3D architecture (Supplementary Fig. 1). The thin regions of PVDF lead to the formation of ‘folds’ at these locations. Additional details appear in the Methods and in Supplementary Note 1. Such strategies establish routes to 3D piezoelectric microsystems with diverse geometries defined by the 2D precursor, the configuration and magnitude of the pre-strain in the elastomer substrate, and the bonding locations. Examples include 3D mesostructures that incorporate narrow filaments, plates/membranes, continuously curved features, sharp folds, single or multiple levels of hierarchy in 3D shapes and feature sizes, in multilayered configurations of active or passive materials (Fig. 1b and Supplementary Figs. 2–6).

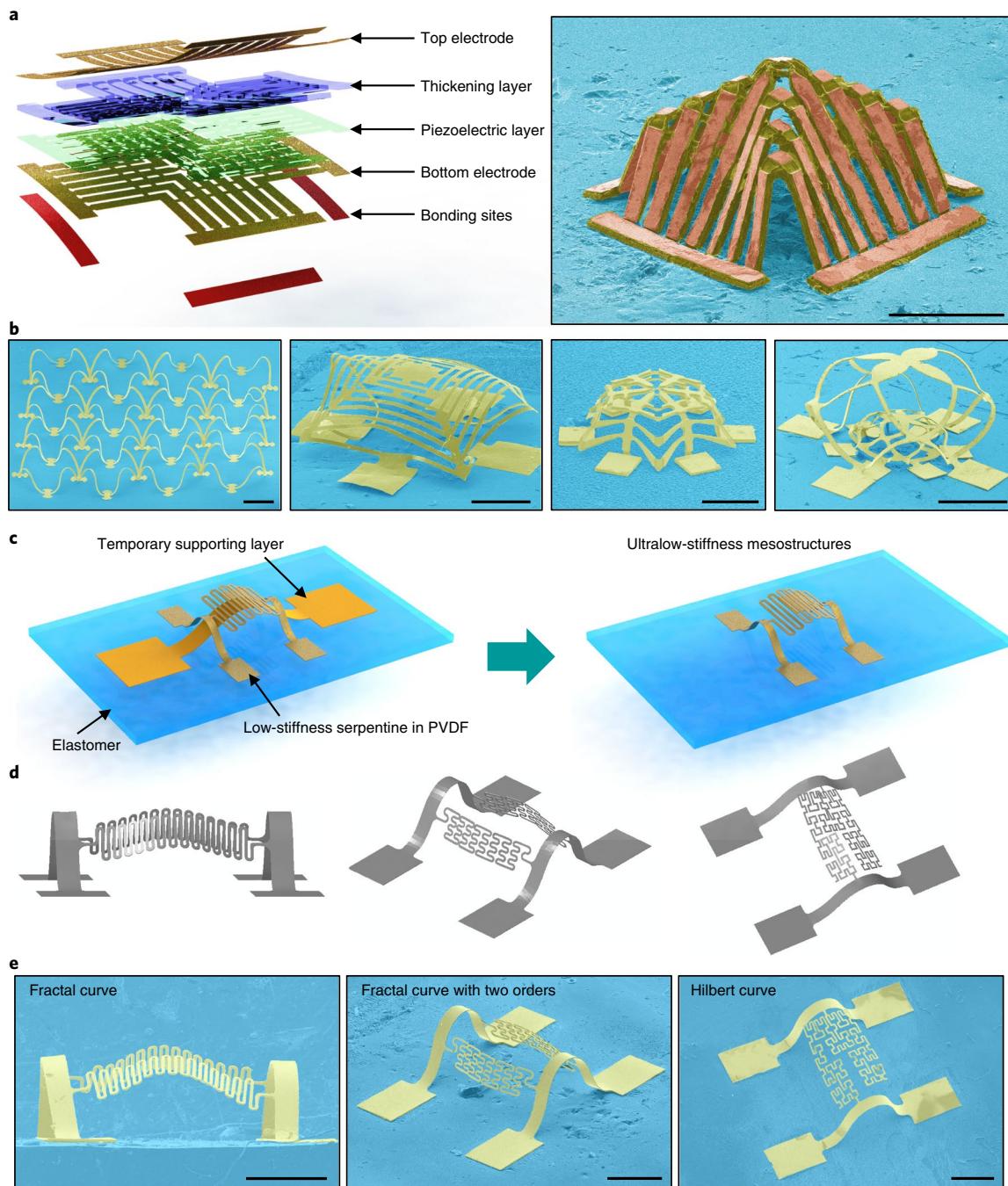
Active, piezoelectric systems represent the focus of the work presented here. In some examples in energy harvesting and related applications, structures with ultralow effective stiffnesses are of interest. Methods that achieve 3D mesostructures by controlled mechanical buckling, as described above, do not provide access to the necessary geometries, however, due to requirements for bending stiffnesses sufficient to enable the assembly process. Schemes introduced here avoid this limitation by the use of temporary structural supports designed to drive the buckling motions of associated 3D piezoelectric structures with ultralow bending stiffnesses (that is, typically more than one order of magnitude smaller than that of conventional 3D mesostructures; see Supplementary Fig. 7). As shown in Fig. 1c, the process begins with simultaneous buckling of a bilayer 2D precursor. In this example, a ribbon of polyimide (PI; 12.5  $\mu\text{m}$  in thickness) serves as an underlying support layer for a thin structure of PVDF (9  $\mu\text{m}$  in thickness) in a low-stiffness, serpentine shape. During the assembly process, the PI ribbon, bonded at its two ends to an elastomer substrate (700  $\mu\text{m}$  in thickness, pre-strain of 10%), buckles upwards, thereby imparting forces onto the back side of the PVDF serpentine to induce out-of-plane motions and a resulting 3D shape (Supplementary Fig. 8). Selectively releasing the bonding sites of the PI ribbon allows for its removal, thereby leaving an ultralow-stiffness serpentine in a 3D geometry. Without the PI, releasing the prestrain yields a serpentine with a largely 2D, wrinkled geometry because the stiffness of the structure is unable to overcome the forces of non-specific adhesion (that is, Van der Waals forces) between it and the underlying elastomer (Supplementary Fig. 9). Specifically, quantitative energetic calculations based on FEA (see Supplementary Note 2 for details) suggest that without the temporary supporting layer, the local buckling state (Supplementary Fig. 10b) in which the serpentine adheres to the substrate is more energetically favourable than the global buckling

state (Supplementary Fig. 10c), due to the adhesion energy and the low stiffness of the serpentine mesostructure. (In contrast, the straight ribbon geometry of the supporting layer yields a comparatively much larger stiffness, even for the case of similar thicknesses, thereby enabling delamination from the elastomer to yield an arch shape<sup>42</sup>.) This process induces transformation of the serpentine into a 3D configuration. Subsequently, the supporting layer collapses to the substrate in a very short period of time (~0.3 ms, Supplementary Fig. 11a) upon detaching its bonding sites. After removing the supporting layer, this global buckling state remains, as it corresponds to a state of local energy minimum. Numerical examples of this energetic analysis appear in Supplementary Note 3 and Supplementary Figs. 10 and 11.

Subsequent stretching of the elastomer allows the ultralow-stiffness 3D mesostructures to shift back to their original 2D planar shapes. Different from the buckling process, which requires an intimate contact between the fractal curves, the supporting layer and the underlying elastomer, the stretching process can yield only comparatively poor contact and lower adhesion between the fractal curves and the elastomer due to surface roughness<sup>43</sup>. The ultralow-stiffness 3D mesostructures can, therefore, switch between 2D and 3D geometries through stretching and releasing of the elastomer (Supplementary Fig. 12). Examples of ultralow-stiffness 3D mesostructures formed in this manner in PVDF (9  $\mu\text{m}$  in thickness, 20  $\mu\text{m}$  in ribbon width) include simple fractal curves (the left frame in Fig. 1d,e, stiffness: 0.26 N m<sup>-1</sup>), and those with two orders (the middle frame in Fig. 1d,e and Supplementary Fig. 13, stiffness: 0.18 N m<sup>-1</sup>) and Hilbert geometries (the right frame in Fig. 1d,e, stiffness: 0.084 N m<sup>-1</sup>). The good agreement between FEA (Fig. 1d) and experimental results (Fig. 1e) highlights the level of control and reproducibility that is possible. These concepts expand the range of design opportunities in low-stiffness 3D structures beyond those possible with previously published schemes.

### Energy harvesting from 3D piezoelectric microsystem

Such classes of ultralow-stiffness 3D mesostructures in functional piezoelectric materials offer complex modes of motion induced by vibrations, thereby creating interesting opportunities in energy harvesting, especially for small-scale systems. Figure 2a–d and Supplementary Figs. 14a and 15–16 feature an array of 3D serpentine as an example that is engineered for multidirectional energy harvesting. This device consists of PVDF (9  $\mu\text{m}$  in thickness, 50  $\mu\text{m}$  in ribbon width, 3 mm in lateral dimension, 1.2 mm in vertical displacement) with top (Cr/Au/Ti, 10 nm/100 nm/20 nm in thickness) and bottom (Cr/Au, 10 nm/100 nm in thickness) electrodes, as shown in the inset in Fig. 2a and Supplementary Fig. 14a. Harmonic vibrations can excite selected vibrational modes of the 3D structure, thereby producing electrical power that can be harvested through contacts to the metal electrodes. For out-of-plane vibrations (that is, vertical direction; peak accelerations of 4g), the device generates root-mean-square (RMS) voltages over 1 mV between 600 Hz to 700 Hz (Fig. 2a). Unlike traditional harvesters that use simple cantilever geometries, the 3D serpentine configuration enables operation for motions in multiple directions. For example, Fig. 2b shows that in-plane vibrations (that is, lateral direction) induce comparable electrical outputs. In both cases, the peak output RMS voltage (2.02 mV under out-of-plane vibration, 1.73 mV under in-plane vibration) occurs at ~650 Hz. Figure 2c presents the time dependence of the output voltage and corresponding fast Fourier transform (FFT). The single, well-defined peak at ~657 Hz is consistent with a linear mechanical response. The resonance lies in the midrange of the audible spectrum, thereby creating opportunities in harvesting energy from the sounds of the human voice, musical instruments and ambient noise. Figure 2d shows an example of converting sound (657 Hz) from a commercial speaker (Logitech, S-150) into electrical power. The feature of multidirectional energy

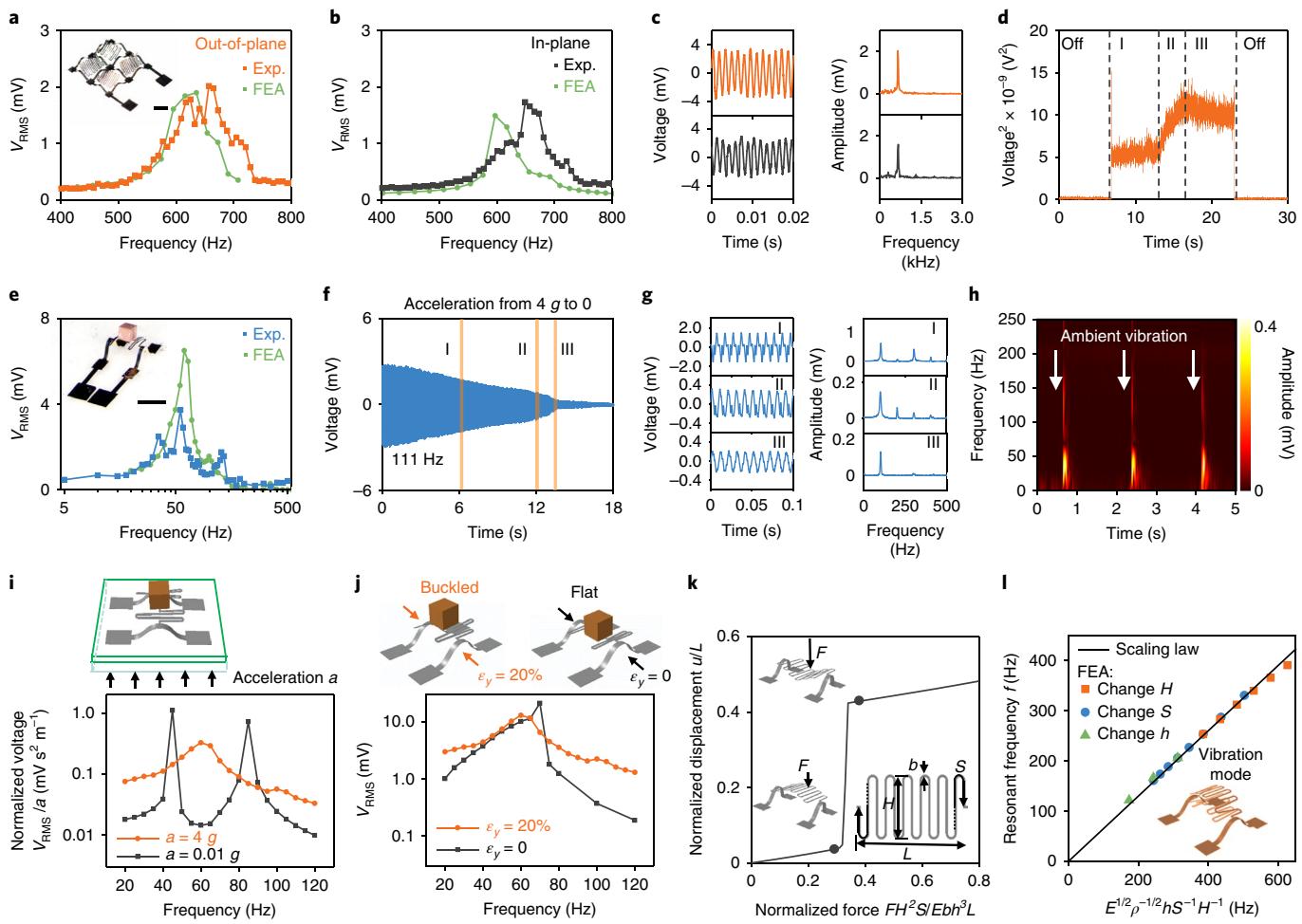


**Fig. 1 | 3D mesoscale piezoelectric frameworks and ultralow-stiffness mesostructures.** **a**, Exploded-view schematic illustrations (left) and SEM image (right) of a 3D PVDF mesostructure with top and bottom metal electrodes. **b**, SEM images of representative 3D mesoscale networks in PVDF, including an array of filamentary serpentines (left), mixed collection of membranes and filaments (middle left), folded sheets (middle right), and overlapping networks (right). **c**, Process for assembly of mesostructures with ultralow stiffnesses. **d,e**, FEA predictions (**d**) and corresponding SEM images (**e**) of ultralow-stiffness PVDF mesostructures, including examples that consist of first- (left) and second-order (middle) fractal curves and a Hilbert geometry (right). Scale bars, 500  $\mu$ m.

harvesting allows the device to operate independently of the relative position between the device and the speaker. Regions I, II and III in Fig. 2d correspond to locating the speaker next to the device, gradually changing the position of the speaker, and placing the speaker above the device, respectively.

Broadband operation is advantageous for many applications in energy harvesting. Figure 2e-h shows a design configured towards this goal. The structure involves a buckled bi-stable serpentine (PVDF, 9  $\mu$ m in thickness, 50  $\mu$ m in ribbon width) and a proof mass

(Cu, 500  $\mu$ m  $\times$  500  $\mu$ m  $\times$  500  $\mu$ m). The overall lateral dimensions are 3 mm  $\times$  3 mm and the height is 1.5 mm (the inset in Fig. 2e and Supplementary Fig. 14b). Here, the buckled shape provides two stable states, with a serpentine layout that, together with the proof mass, reduce the energy barrier between these two states, thereby facilitating the excitation of nonlinear vibrational responses. As such, under an out-of-plane vibration with an acceleration of 4g, this 3D system can generate electrical power across a range of frequencies spanning two orders of magnitude (that is, from 5 Hz to



**Fig. 2 | 3D vibratory piezoelectric energy harvesters.** **a–c**, Electrical output characteristics of a 3D serpentine array for multidirectional energy harvesting, including RMS voltages at different frequencies for out-of-plane (**a**) and in-plane (**b**) vibrations, with time-domain output voltage and fast Fourier transform results (**c**) that highlight a resonance at 657 Hz. **d**, Power spectrum of the 3D serpentine array when excited at 657 Hz with a commercial speaker. The inset in **a** features an optical image of the device. **e**, RMS voltages from a bi-stable 3D serpentine structure with an integrated test mass for broadband energy harvesting. **f,g**, Time-domain output voltage from a bi-stable 3D serpentine structure at 111 Hz under different accelerations (**f**), with a magnified view and fast Fourier transform (**g**). **h**, Spectrogram of the output voltage associated with periodic ambient vibration. The inset in **e** shows an optical image of the device. **i**, FEA prediction of the dependence of the normalized open-circuit voltage (RMS) on frequency in a regime of nonlinear (acceleration 4 g) and linear (acceleration 0.01 g) vibratory response. **j**, FEA prediction of the dependence of the open-circuit voltage (RMS) on frequency for a buckled and flat serpentine pattern. **k**, Scaling law for the relationship between the nonlinear normalized displacement and the normalized force for the buckled serpentine pattern. **l**, Scaling law for the resonant frequency of the buckled serpentine pattern ( $\epsilon_y = 10\%$ ). The solid line in **l** is a linear fit to the data. Scale bars, 1 mm.

500 Hz, Fig. 2e). Supplementary Fig. 17 summarizes representative time-domain output voltages with corresponding FFT analyses. Figure 2f presents the voltage under different accelerations. The magnified view and corresponding fast Fourier transform (FFT) in Fig. 2g illustrate the transition from nonlinear to linear response as the vibratory acceleration decreases in magnitude. A device with this construction has, by extension, the ability to harvest energy from broadband ambient vibrations. As an example, an impulsive force delivered to the supporting surface of this type of device induces damped vibrations of the serpentine, thereby generating electrical power with gradually decreased amplitude. Figure 2h shows the spectrogram of the output voltage produced by periodic delivery of the force. The corresponding time-domain output voltage appears in Supplementary Fig. 18. A lower level of vibrations with an acceleration of 0.1 g is also detectable by introducing filters to reduce the noise (Supplementary Fig. 19).

FEA further demonstrates that these nonlinear responses lead to capabilities in energy harvesting that extend over a much broader

range of frequencies than that possible with a linear response. As shown in Fig. 2i, the frequency spectrum of the normalized output voltage shows peaks at discrete values during linear vibrations. By contrast, the large output voltages extend over a comparatively wide frequency range during nonlinear vibrations. Furthermore, the 3D serpentine formed via controlled buckling has a notably larger output voltage than that of the 2D serpentine over a wide range of frequencies, as shown by the FEA result in Fig. 2j and experimental results in Supplementary Fig. 20. The full-width at half-maximum values are about 30 Hz and 18 Hz for the bi-stable 3D serpentine and flat 2D serpentine, respectively.

These classes of 3D piezoelectric energy harvesters clearly offer mechanical properties that are beneficial in vibrational energy harvesting. Many other designs with tailored mechanical properties are also possible. Supplementary Figs. 14c and 21,22 highlight a 3D spiral piezoelectric device in a multilayer format. Key considerations for this device include a spiral shape that reduces its stiffness and a 3D geometry that provides space for large-amplitude vibrations.

The result is an energy harvester with a low resonant frequency and a wide operating frequency range (Supplementary Fig. 21), with the ability to harvest energy from air flow (Supplementary Fig. 22). This geometric diversity creates possibilities for designs specifically tailored to application requirements.

Further refinements in layout can lead to an improved output of 3D vibratory energy harvesters while retaining their unique mechanical properties (for example, low resonant frequency, wide bandwidth), thereby creating opportunities for their use in practical applications. For example, adding a PI supporting layer (Supplementary Fig. 23) increases the open-circuit output voltage by two orders of magnitude, due mainly to the offset of the PVDF relative to the neutral mechanical plane, leading to an increased membrane strain during bending deformations. The open-circuit output voltage can also be improved by optimizing the pattern of the electrode (Supplementary Fig. 24b). Based on guidance from FEA results for the spatial distribution of voltage (without electrodes; Supplementary Fig. 24c), the electrode can be designed to cover regions of positive voltage and to avoid those with negative voltage. The result increases the net output voltage by a factor of 2.2 (Supplementary Fig. 24d). Experimental examples of using a supporting layer and patterned electrode to enhance the output voltages appear in Supplementary Figs. 25–27. A 3D bi-stable serpentine structure with larger dimensions and patterned electrode can produce an RMS voltage around 0.79 V (Supplementary Fig. 26).

### Fundamental studies of energy harvesting

As highlighted in the previous sections, broadband vibrational energy harvesting results from nonlinear deformations due to the combined effect of ultralow-stiffness serpentine microstructures and compressive buckling. A platform that allows further study of the underlying physics involves a buckled serpentine pattern as shown in Supplementary Fig. 28, where an applied force ( $F$ ) serves as an external mechanical stimulus. When the magnitude of this force increases beyond a critical point ( $F_c$ ), the serpentine switches from a geometry with negative vertical curvature to one with positive curvature, leading to a highly nonlinear displacement–force relation (Fig. 2k). The following scaling law captures the relationship among the displacement ( $u$ ), force ( $F$ ) and the geometry/material parameters for this case

$$\frac{u}{L} = U \left( \frac{FH^2S}{Ebh^3L} \right) \quad (1)$$

where  $L$ ,  $H$ ,  $b$  and  $S$  are the serpentine length, height, width and total arc length, respectively;  $h$  is the thickness,  $E$  is the material Young's modulus and  $U$  is a nonlinear function that depends on the compressive strain ( $\epsilon_y$ ) applied to the serpentine pattern during compressive buckling. When the compressive strain  $\epsilon_y = 0$ , the  $\frac{u}{L}$  versus  $\frac{FH^2S}{Ebh^3L}$  relationship is almost linear (Supplementary Fig. 29), due to the flat, 2D geometry of the serpentine in this situation. The 3D shape of the serpentine pattern formed via compressive buckling is, in this sense, crucial in realizing nonlinear deformations. The scaling law is consistent with FEA for typical serpentine geometries (with  $L \ll S$  and  $h \ll b$ ), as shown in Supplementary Fig. 30 and Supplementary Note 4.

The ultralow-stiffness serpentine pattern allows nonlinear deformations to follow from forces with small magnitudes. For a representative set of geometric and material parameters,  $L = H = 1$  mm,  $S = 10L$ ,  $h = 9\text{ }\mu\text{m}$ ,  $b = 50\text{ }\mu\text{m}$ ,  $E = 2\text{ GPa}$  (PVDF) and  $\epsilon_y = 10\%$ , the scaling law equation (1) indicates that the critical force required to actuate nonlinear deformations can be as low as  $2\text{ }\mu\text{N}$ , in a range accessible to biological stimuli such as the beating of muscle cells<sup>44,45</sup>. By comparison, a straight ribbon with otherwise similar geometries and materials requires a force with a magnitude  $\sim 250$  times larger

than this value to actuate nonlinear deformations, according to FEA (Supplementary Fig. 31).

The scaling law in equation (1) also suggests that the serpentine pattern has a low resonant frequency, because of its low stiffness. The stiffness, defined as  $K = \frac{\partial F}{\partial u} \Big|_{u=0}$ , is linearly proportional to  $\frac{Ebh^3}{HS}$  (see FEA validation in Supplementary Fig. 32 and Supplementary Note 4). Dimensional analysis suggests that the equivalent mass of vibration ( $M$ ) is linearly proportional to  $\rho bhS$ , where  $\rho$  is the material density. Therefore, the linear resonant frequency ( $f$ ) obeys the scaling law,

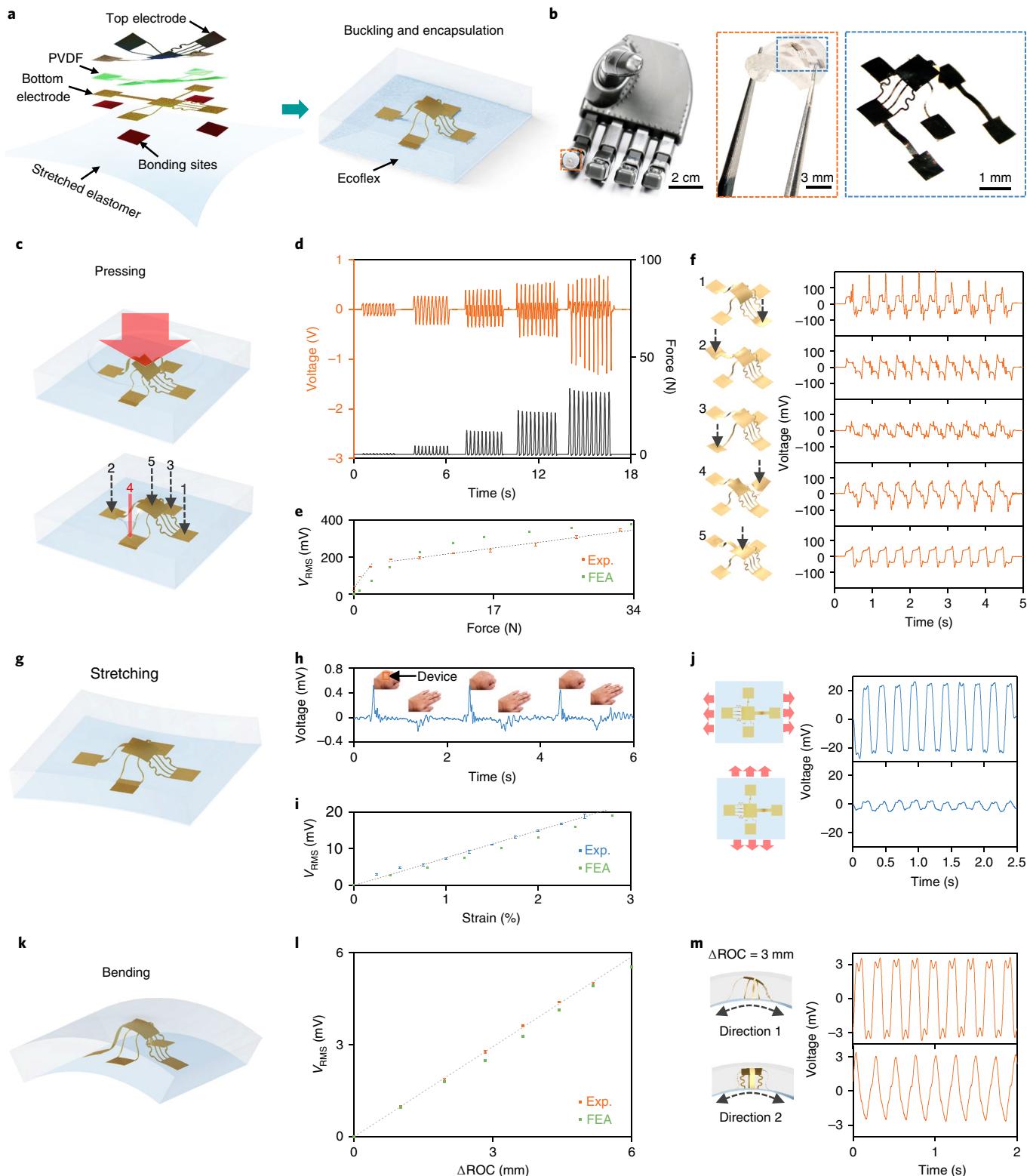
$$f = \alpha \sqrt{\frac{K}{M}} = \alpha \frac{h}{HS} \sqrt{\frac{E}{\rho}} \quad (2)$$

where  $\alpha$  is a dimensionless constant that depends on the vibration mode and the compressive strain  $\epsilon_y$ . With  $\alpha = 0.65$ , this relationship agrees reasonably well with FEA results for the first-order vibrational mode, as shown in Fig. 2l ( $\epsilon_y = 10\%$ ). Additional details concerning the validation of equation (2) are in Supplementary Fig. 33 and Supplementary Note 4. For representative parameters,  $H = 2$  mm,  $S = 15H$ ,  $h = 9\text{ }\mu\text{m}$ ,  $E = 2\text{ GPa}$  and  $\rho = 1800\text{ kg m}^{-3}$ , the resonant frequency is below 100 Hz. By comparison, at similar length scales and with similar materials, 3D structures without this type of serpentine geometry typically have resonant frequencies larger than 10 kHz (ref. <sup>46</sup>). The serpentine design is an important, enabling aspect of efficient energy harvesting from low-frequency mechanical stimuli, such as machine-induced vibrations (10–100 Hz)<sup>47</sup> and human motions (<10 Hz)<sup>48</sup>.

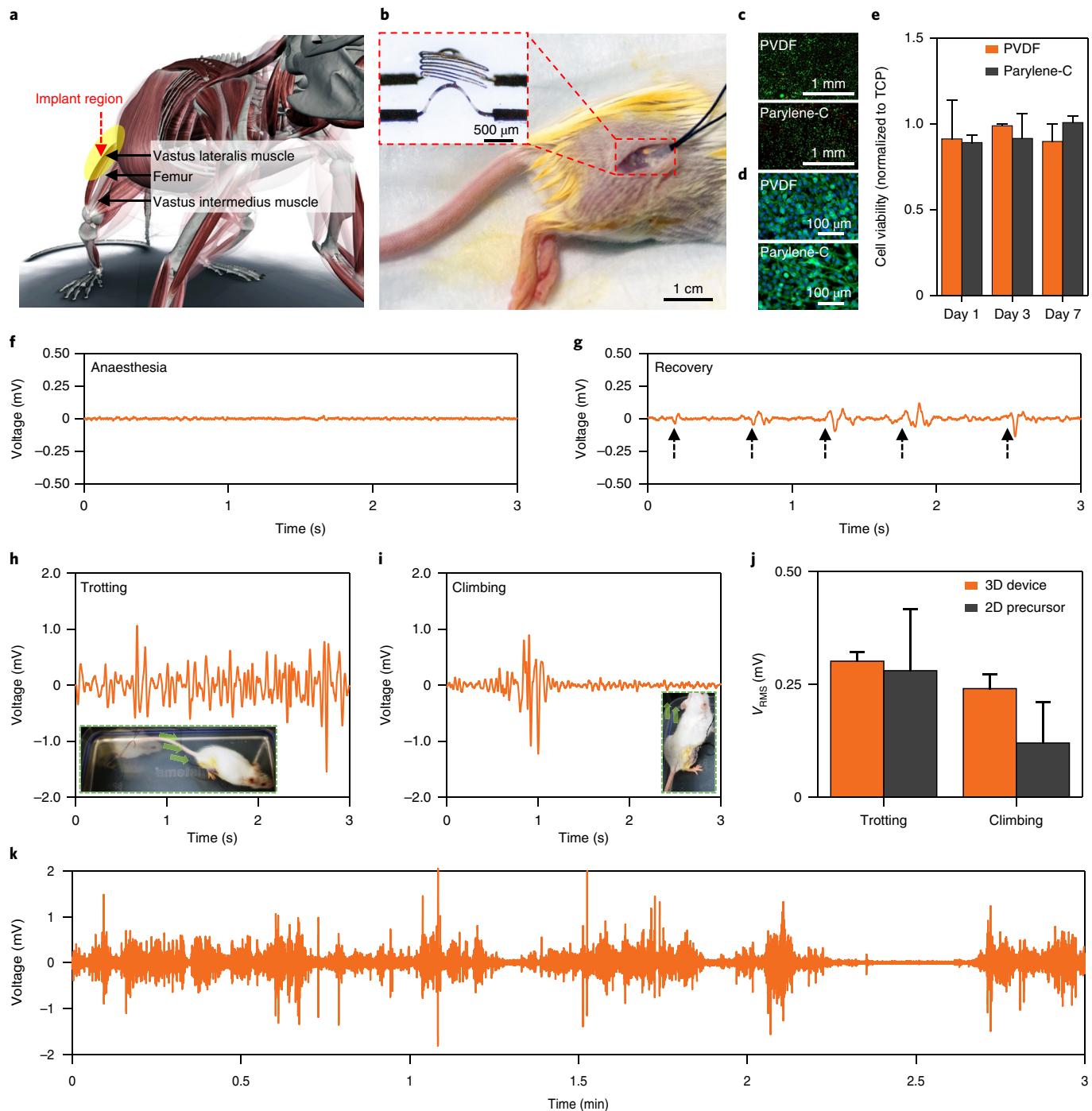
### 3D piezoelectric microsystems as robotic interfaces

In addition to vibrations, other types of mechanical stimuli such as pressure, normal force, stretching and bending from biological or environmental sources can induce responses. Designs that operate effectively in such cases can broaden uses in energy harvesting and expand applications to mechanical sensing (Fig. 3 and Supplementary Figs. 34–40). Figure 3a,b presents exploded schematic illustrations and optical images of a representative 3D device. Here, the 3D geometry offers an increased number of degrees of freedom in deformation compared to 2D alternatives, thereby creating interesting design possibilities. Encapsulating these 3D structures with a soft silicone elastomer (1 mm in thickness, Ecoflex, Smooth-On) physically protects the device, while maintaining its deformability (the middle frame in Fig. 3b).

Figure 3c–f and Supplementary Fig. 37 present some results for application of normal forces (pressing) over large and small areas. The large area (1 cm by 1 cm) case can lead to global deformation of the 3D piezoelectric structure (the top frame in Fig. 3c)<sup>49</sup>, thereby generating peak voltages > 1 V (RMS voltage  $\sim 350\text{ mV}$ ) when the magnitude of the force is 32 N. Measurable output occurs for forces as small as 30 mN. The output RMS voltage changes with the magnitude of the force (Fig. 3e), with high sensitivity ( $60\text{ mV N}^{-1}$ ) at small forces (that is, forces below 3 N) and comparatively low sensitivity ( $5.7\text{ mV N}^{-1}$ ) at large forces (that is, forces above 3 N) due to the increased slope of the stress-strain response of the elastomer<sup>50</sup>. The encapsulation materials and the 3D geometries can be selected for desired sensitivity. Forces applied over small areas induce only local deformations. Controlling the displacement of a cylinder with a diameter of 0.65 mm yields forces over a small circular region with magnitudes from 0 to 91 mN (Supplementary Fig. 38). Here, asymmetric designs (the right frame in Fig. 3b and Supplementary Fig. 34) allow the position of the applied force to be determined from the shape of the output signal. Figure. 3f shows the time-domain output voltage in the case of application of force over a small circular region (diameter: 0.65 mm, displacement of the cylinder: 1 mm) at five different positions. Such distinct response shapes in these five cases



**Fig. 3 | Impact-based 3D piezoelectric energy harvester and sensors.** **a**, Exploded view of the 2D geometry (left) and schematic illustration of the impact-based 3D piezoelectric energy harvester (right). **b**, Optical images of the device attached to a robotic arm (left), the encapsulated device (middle), and the 3D geometry before encapsulation (right). **c**, Schematic illustration of the device under pressure applied over a large area (top) and a small area (bottom). **d**, Time-domain output voltage (orange curve) with different external forces (dark grey curve) applied over a square area ( $1\text{cm}^2$ ). **e**, The dependence of RMS voltage on the applied force. **f**, Time-domain output voltage with external forces applied over different small circular regions (0.65 mm in diameter). **g**, Schematic illustration of the device under stretching. **h**, Demonstration of harvesting mechanical energy from human skin by attaching the device onto the dorsal surface of a hand. **i**, Dependence of RMS voltage on the applied strain. **j**, Time-domain output voltage with strains applied in different directions. **k**, Schematic illustration of the device under bending. **l**, Dependence of RMS voltage on the change of radius of curvature at different bending directions. **m**, Time-domain output voltage by bending along different directions. Error bars in **e**, **i** and **l** correspond to the calculated standard error deviation from three measurements. Grey dashed lines are linear fits to the data.



**Fig. 4 | 3D piezoelectric devices as biomedical implants.** **a**, Schematic illustration of the location of a subdermal implant. **b**, Optical image of a 3D piezoelectric energy harvester implanted into the hind leg of a mouse. The inset shows a magnified optical image of the device. **c**, Representative fluorescence images from a cytotoxicity assay (live/dead test) for fibroblasts after three days of culturing on PVDF and parylene-C. Green indicates live cells and red indicates dead cells. **d**, Representative fluorescence images from F-actin staining for fibroblasts after seven days of culturing on PVDF and parylene-C. Green indicates actin filaments and blue indicates cell nuclei. **e**, Normalized cell viability data based on alamar blue assay for days 1, 3 and 7, proving no signs of cytotoxicity for all the groups. Error bars correspond to the calculated standard error deviation from three measurements. **f**, Output voltage of the 3D piezoelectric device when the mouse is anaesthetized. **g**, Output voltage when the mouse begins to recover but shows no obvious movement. **h**, **i**, Output voltage under different mouse behaviours: trotting (**h**), climbing (**i**). Insets show corresponding images of the mouse behaviour. **j**, Comparison of the output voltage from the 3D device and its 2D precursor under different behaviours of the animal. Data correspond to averages of five different trotting and climbing behaviours. Error bars correspond to calculated standard error deviation. **k**, Output voltage of the 3D piezoelectric device as the mouse freely moves inside an enclosure for three minutes.

can be attributed to the different types of associated deformations, as illustrated in the left frame of Fig. 3f. As the displacement of the cylinder decreases to 0.75 mm, the differences in response shapes

become less obvious. Smaller displacements (for example, 0.5 mm and 0.25 mm) yield similar responses for different pressing positions (Supplementary Fig. 39). Other types of 3D mesostructures,

including but not limited to those with filaments and membranes, are also feasible for energy harvesting and sensing of normal pressure (Supplementary Fig. 40).

Figure 3g–j presents a schematic diagram and the results of energy harvesting from uniaxial stretching, a mode of deformation that can be relevant for interfaces to biomechanical sources of motion. An example is in adhesive integration (Silbione RT Gel 4717 A/B, Bluestar Silicones) of a device on the skin. Figure 3h shows that a device attached on the dorsal surface of the hand produces a peak voltage  $> 0.5$  mV as the gesture switches between closed fist and open hand. The linear relationship between the RMS voltage and applied strain provides the basis for sensing of strain (Fig. 3i). The asymmetric design allows determination of the direction of stretching from the amplitudes of the output signals if the value of the applied uniaxial strain is known. For example, under the same strain of 2.5%, the RMS voltage along two directions differs by more than a factor of seven (that is, 18.5 mV and 2.5 mV), as shown in Fig. 3j.

A bending mode of operation is also possible (Fig. 3k–m). Changes in the radius of curvature ( $\Delta$ ROC) define the mechanical stimulus in this case. Increasing the magnitude of this change increases the output voltage in a linear fashion (Fig. 3l). Different directions of bending also result in distinguishable amplitudes in the electrical response (Fig. 3m). For example, bending at a  $\Delta$ ROC of  $\sim 3$  mm along direction 1 and direction 2 (illustrated in the left frame of Fig. 3m) leads to RMS voltages of 2.8 mV and 2.1 mV, respectively. A key appealing aspect of this 3D piezoelectric device is that the amplitudes of its output voltage change with the magnitude as well as the direction of the mechanical stimuli, and the shapes of its output signals vary with the location and type of mechanical stimuli, with corresponding capabilities in detection. Examples include, but are not limited to, distinguishing the positions of an applied force by comparing the shapes of the output signal (Fig. 3f) and measuring the intensity or direction of uniaxial strain based on the amplitude of the output voltage (Fig. 3i,j). These additional options in mechanical sensing suggest potential routes for developments of robotic prosthetic interfaces. Further integrating multiple devices over a large area in a distributed manner allows for gesture recognition, spatio-temporal mapping of motions, and other applications.

### Implantation of 3D piezoelectric microsystems

These 3D piezoelectric systems are well configured for interactions with biological systems, in three dimensions. One example involves implantation in a region in the hind leg of a female mouse, as illustrated in Fig. 4a,b. Here, a conformal coating of parylene-C (2  $\mu$ m in thickness) encapsulates the 3D device (the inset in Fig. 4b) to prevent the penetration of biofluids. Figure 4c–e and Supplementary Fig. 41 support biocompatibility of the encapsulation layer and the functional piezoelectric material (PVDF, 9  $\mu$ m in thickness). Specifically, fibroblast cells show no signs of cytotoxicity when cultured on films of PVDF or parylene-C for up to three days (Fig. 4c); they remain adhered to the surface with appropriate morphology after seven days (Fig. 4d). Observations indicate no significant differences in cell viability among tissue culture plastic (TCP), PVDF and parylene-C groups (Fig. 4e).

After implantation, the device shows no output signal (Fig. 4f) until the mouse gradually recovers from anaesthesia and begins with small movements, yielding small voltages of  $\sim 100$   $\mu$ V (Fig. 4g). Following complete recovery, the mouse exhibits normal locomotor activity inside an enclosure (23 cm by 16 cm in area, 16 cm in height), including but not limited to trotting and climbing. Such motion induces output voltages with peak amplitudes  $> 1$  mV (Fig. 4h,j). Here, the 3D geometry allows the device to deform easily under external forces, thereby enabling efficient coupling to contractile motions of surrounding muscles without irritation or significant mechanical constraints on natural movements. By comparison, the

corresponding 2D counterpart (Supplementary Fig. 42) implanted at the same region exhibits reduced stability and output performance. Specifically, the output voltage of the 2D precursor under trotting behaviour has a comparable amplitude to the corresponding 3D device, although its output under climbing behaviour is lower (Fig. 4j). In both cases, the 2D precursor fails to produce output signals with repeatable amplitudes, due to insufficient deformation in certain cases (Supplementary Fig. 43 and Supplementary Video 1). For example, similar trotting behaviours can result in RMS voltages that span one order of magnitude (that is, from 0.04 mV to 0.4 mV). Figure 4k and Supplementary Video 2 illustrate the stability of output signals from the 3D piezoelectric device with a mouse moving freely inside the enclosure. The device can still produce electrical signals under trotting behaviours after implantation for  $\sim 48$  hours (Day 3, Supplementary Fig. 44). FEA study of typical types of deformation further supports the use of the device in this application. Under pressure, the output voltage is larger than that of the 2D precursor by one order of magnitude (Supplementary Fig. 45a), because the underlying dermis constrains the deformation of the 2D structure. Under bending deformation, the output voltages of the 2D and 3D devices are of the same order of magnitude (Supplementary Fig. 45b). Consistency between the output amplitude and motion intensity suggests the feasibility of using such 3D piezoelectric microsystems as biomedical implants for energy harvesting and sensing.

### Conclusions

Our results provide pathways for extending piezoelectric microsystems to three dimensions in a collection of architectures that are configured for applications in energy harvesting, robotic prosthetic interfaces and biomedical implants. The experimental results, computational models and theoretical studies reveal the possibilities of using 3D piezoelectric microsystems for multidirectional, broadband and/or low-frequency energy harvesting from vibrations of various types; for mechanical to electrical energy conversion from a wide range of applied forces; for sensing the intensity and distinguishing the direction/position of mechanical stimuli; and for in vivo biomechanical energy harvesting and sensing. Our mechanically guided 3D assembly approach allows 3D piezoelectric microsystems to leverage the most advanced micro/nanofabrication techniques and sophisticated active materials, in a diverse set of 3D geometries. The structures can offer ultralow-stiffness and asymmetric designs, as well as controlled thickness profiles, coherently coupled multilayers, selective bonding configurations and gradient arrays that result from non-uniform distributions of prestrain.

### Methods

**Fabrication of 3D piezoelectric mesostructures.** Preparation of 2D precursors in PVDF began with spin coating (1,000 r.p.m. for 60 s) and curing (70 °C for 1 h) polydimethylsiloxane (PDMS) (1:10, Sylgard 184, Dow Corning) on silicon wafers. Laminating a film of PVDF (9  $\mu$ m in thickness, Piezotech) onto these substrates and depositing Cr/Au (5 nm/50 nm in thickness) via electron beam evaporation formed a hard mask for patterning the PVDF by dry etching with O<sub>2</sub>. Next, photolithography defined a pattern of resist (AZ 5214E, MicroChem) for wet etching of the Cr/Au and dry etching of PVDF to complete the fabrication of the 2D precursor structures. After removing the hard mask (Cr/Au), a piece of poly(vinyl alcohol) (PVA) tape allowed retrieval of the PVDF structures from the PDMS-coated wafer to expose their back sides for deposition of Ti/SiO<sub>2</sub> (10 nm/100 nm in thickness) via electron beam evaporation through a shadow mask to serve as the bonding sites. The process of compressive buckling began with stretching of a flat silicone elastomer substrate using a custom mechanical stage. Exposing the 2D structures and the surface of this elastomer (Dragon Skin, Smooth-On) to ultraviolet-induced ozone created chemical functionalization to facilitate bonding. Laminating the 2D precursors/PVA tape onto the stretched elastomer and then heating at 70 °C for 10 min led to the formation of strong siloxane bonds at the bonding sites. Dissolving the PVA with water and releasing the prestrain transformed the 2D precursors into 3D mesostructures.

**Fabrication of 3D folded piezoelectric mesostructures.** The process began by laminating a film of PVDF (28  $\mu$ m in thickness, TE Connectivity) onto PDMS-coated wafers according to the steps outlined in the previous section. Depositing

Cr/Au (5 nm/50 nm in thickness) via electron beam evaporation defined a hard mask for dry etching the PVDF. Two steps of photolithography, wet etching of Cr/Au and dry etching of PVDF defined 2D patterns (28  $\mu\text{m}$  in thickness) with localized regions of reduced thickness (5–13  $\mu\text{m}$  in thickness). The remaining steps followed the procedures mentioned above.

**Fabrication of 3D ultralow-stiffness piezoelectric mesostructures.** Fabrication and transfer of 2D precursors followed the procedures for the 3D piezoelectric mesostructures described above. An additional layer of polyimide (12.5  $\mu\text{m}$  in thickness) patterned by photolithography and dry etching with bonding sites defined by patterns of  $\text{Al}_2\text{O}_3/\text{Ti}/\text{SiO}_2$  (50 nm/10 nm/100 nm in thickness) served as a supporting layer. After aligning this layer with the 2D precursor of PVDF on a PVA tape, the same compressive buckling steps as described previously transformed the precursor and supporting layer into a 3D geometry. Applying HCl to the bonding sites of the supporting layer dissolved the  $\text{Al}_2\text{O}_3$ , thereby releasing this layer from the silicone elastomer to yield 3D PVDF mesostructures with ultralow stiffnesses.

**Fabrication of vibration-based energy harvesters.** Fabrication began with deposition of Cr/Au/Ti (10 nm/100 nm/20 nm in thickness) on one side of a film of PVDF (9  $\mu\text{m}$  in thickness), and Cr/Au (10 nm/100 nm in thickness) on the other side. Laminating the PVDF onto a PDMS-coated wafer with the Cr/Au/Ti side facing up allowed for further steps in photolithography, wet etching of Cr/Au/Ti, and dry etching of PVDF to define the 2D precursors. Sonication separated the exposed Cr/Au at the bottom side from the metal patterns at the top side, thereby preventing short circuits between the top and bottom metal layers. After retrieving the 2D precursors with PVA tape, electron beam evaporation of Ti/SiO<sub>2</sub> (10 nm/100 nm in thickness) through a shadow mask defined the bonding sites. Aligning a supporting layer, with  $\text{Al}_2\text{O}_3/\text{Ti}/\text{SiO}_2$  (50 nm/10 nm/100 nm in thickness) at the bonding sites, onto the same PVA tape completed the fabrication and transfer of the 2D precursors. The remaining compressive buckling steps followed the same procedures for fabrication of 3D ultralow-stiffness mesostructures in PVDF.

**Fabrication of impact-based energy harvester and sensor.** Fabrication of impact-based energy harvesters followed the procedures outlined for fabrication of vibration-based energy harvesters. Subsequent steps included drop casting and curing of silicone elastomer (Ecoflex 0050, Smooth-On) around the 3D mesostructures, and patterning silicone elastomer by use of a CO<sub>2</sub> laser cutting system (Universal Laser Systems).

**Fabrication of 3D energy harvesters with supporting layers.** Fabrication of 3D energy harvesters with supporting layers followed the steps outlined for fabrication of vibration-based energy harvesters. Subsequent conformal deposition of parylene-C (10  $\mu\text{m}$  in thickness) on the harvester and dry etching of parylene-C at the top side of the structure completed the process.

**Fabrication of 3D energy harvesters with patterned electrodes.** Fabrication of 3D energy harvesters with patterned electrodes began with preparation of 2D precursors in PVDF as described in the fabrication of 3D piezoelectric mesostructures. Additional steps involved deposition of Cr/Au (10 nm/100 nm in thickness) on both sides of the 2D precursors through shadow masks. The transfer and buckling steps followed the procedures mentioned above.

**Fabrication of 3D piezoelectric microsystems for biomedical implants.** Fabrication of 3D piezoelectric microsystems for biomedical implants followed the procedures for fabrication of vibration-based energy harvesters. Subsequent steps included conformal deposition of parylene-C (2  $\mu\text{m}$  in thickness) onto the 3D mesostructures.

**Measurement of vibration-based energy harvesters.** The data acquisition system included a vibration generator (U56001, 3B Scientific), a function generator (FG100, 3B Scientific), a vibration meter (AR63B, Smart Sensor) and a PowerLab computer interface (Model 8/35, ADInstruments). The vibration generator powered by the function generator produced periodic harmonic vibrations at programmable frequencies between 5 Hz and 800 Hz, at an acceleration of 4g. The vibration meter provided a means to calibrate the accelerations at different frequencies before testing the energy harvesters. The PowerLab system allowed recordings of output voltage through a 1 M $\Omega$  probe at a sampling rate of 200,000 samples s<sup>-1</sup>. A notch filter at 60 Hz attenuated noise from the electrical lines.

**Measurement of impact-based energy harvesters and sensors.** The data acquisition hardware included a PowerLab system, a high-precision linear stage (ATS100-150; Aerotech, Inc.) with a LabVIEW control interface, a load cell (FC2231, TE Connectivity), an impedance analyser (Keysight E4990A) and a semiconductor parameter analyser (Agilent, 4155 C). The linear stage produced periodic linear motions to press, stretch or bend the impact-based energy harvesters. The pressing tests used the load cell to measure the magnitude of the applied force. In the stretching tests, motions of the linear stage yielded different

strains applied to the impact-based energy harvester. In the bending tests, the impact-based energy harvester was attached to a PI film (75  $\mu\text{m}$  in thickness), such that displacements changed its radius of curvature. The PowerLab system recorded the output voltage during pressing, stretching and bending tests via measurements through a 100 M $\Omega$  probe with a low-pass filter (30 Hz, to attenuate noise signal at high frequencies) at a sampling rate of 200,000 samples s<sup>-1</sup>. The impedance analyser determined the magnitude of the impedance at frequencies between 20 Hz and 100 kHz. The semiconductor parameter analyser measured the current–voltage responses over a range of voltages from 0 to 50 V.

**Cell seeding and assessment of cell viability.** PVDF (9  $\mu\text{m}$  in thickness) and parylene-C (4  $\mu\text{m}$  in thickness) films were cut into small pieces (5.4 mm in diameter) and placed into a 96-well TCP plate. The plate was then sterilized using ethylene oxide (EtO) gas. L929 fibroblasts (passage number: 5) were seeded onto TCP, PVDF and parylene-C surfaces at a density of 10,000 cells cm<sup>-2</sup>. Cell viability at days 1, 3 and 7 was tested with alamarBlue assay. Briefly, cells were incubated with 100  $\mu\text{l}$  Dulbecco's Modified Eagle Medium (DMEM) culture medium supplemented with 10% alamarBlue solution in a cell culture incubator for 3 h. Then, the type medium was transferred into a new 96-well plate. The fluorescence intensity at 590 nm was read to evaluate cell viability. The viability data of the cells cultured on PVDF and parylene-C films were normalized based on the control cells, which were cultured on TCP. Cell viability was assessed by a live/dead kit (Invitrogen). Live/dead staining were performed at day 3 to confirm the influence of PVDF and parylene-C on cell viability. Cells were incubated with serum-free medium supplemented with 4  $\mu\text{M}$  calcein AM and 2  $\mu\text{M}$  ethidium homodimer-1 in a cell culture incubator for 30 min. To assess the cell attachment and spreading, cells were fixed using 4% paraformaldehyde followed by permeabilization using 0.2% triton X-100 at day 7. Then, the cells were blocked with 1% Bovine Serum Albumin (BSA) solution and stained with Alexa Fluor 488 Phalloidin and DAPI to check the cell morphology on PVDF and parylene-C films.

**Evaluation of 3D piezoelectric device in animal models.** All procedures associated with animal studies followed recommendations in the Guide for the Care and Use of Laboratory Animals of the National Institutes of Health. The Institutional Animal Care and Use Committee (IACUC) at Northwestern University (Protocol No. IS00005877) approved the protocol. Female mice (CD1, age at initiation of the treatment: at least 6 weeks, but not more than 15 weeks, purchased from Charles River Laboratories) were acclimated up to 5 days before surgery. Animals were anaesthetized using isoflurane gas during the implantation surgery and measurement. Fur removal and surgical exposure enabled the insertion of a 3D energy harvester into the subcutaneous region near the right flank. Bioabsorbable suturing and gluing closed the surgically exposed region with two biocompatible wires probing externally for signal readout (Supplementary Fig. 46). A PowerLab system enabled collection of the data through a 1 M $\Omega$  probe with a bandpass filter (0.1–30 Hz, to attenuate noise signals at high frequencies and avoid a pyroelectric signal from temperature fluctuations) at a sampling rate of 200,000 samples s<sup>-1</sup>.

**Reporting Summary.** Further information on experimental design is available in the Nature Research Reporting Summary linked to this article.

## Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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## References

1. Kim, D. H. et al. Epidermal electronics. *Science* **333**, 838–843 (2011).
2. Larson, C. et al. Highly stretchable electroluminescent skin for optical signaling and tactile sensing. *Science* **351**, 1071–1074 (2016).
3. Tian, B. et al. Macroporous nanowire nanoelectronic scaffolds for synthetic tissues. *Nat. Mater.* **11**, 986–994 (2012).
4. Hwang, G. T. et al. Self-powered cardiac pacemaker enabled by flexible single crystalline PMN-PT piezoelectric energy harvester. *Adv. Mater.* **26**, 4880–4887 (2014).
5. Fan, F. R., Tian, Z. Q. & Lin Wang, Z. Flexible triboelectric generator. *Nano Energy* **1**, 328–334 (2012).
6. Yin, J. et al. Generating electricity by moving a droplet of ionic liquid along graphene. *Nat. Nanotech.* **9**, 378–383 (2014).
7. Jinno, H. et al. Stretchable and waterproof elastomer-coated organic photovoltaics for washable electronic textile applications. *Nat. Energy* **2**, 780–785 (2017).
8. Zhao, J. et al. Air-stable and freestanding lithium alloy/graphene foil as an alternative to lithium metal anodes. *Nat. Nanotech.* **12**, 993–999 (2017).

9. Li, M., Tang, H. X. & Roukes, M. L. Ultra-sensitive NEMS-based cantilevers for sensing, scanned probe and very high-frequency applications. *Nat. Nanotech.* **2**, 114–120 (2007).
10. Kan, T. et al. Enantiomeric switching of chiral metamaterial for terahertz polarization modulation employing vertically deformable MEMS spirals. *Nat. Commun.* **6**, 8422 (2015).
11. Nguyen, T. D. et al. Piezoelectric nanoribbons for monitoring cellular deformations. *Nat. Nanotech.* **7**, 587–593 (2012).
12. Wu, W., Wen, X. & Wang, Z. L. Taxel-addressable matrix of vertical-nanowire piezotronic transistors for active and adaptive tactile imaging. *Science* **340**, 952–957 (2013).
13. Yu, X. et al. Needle-shaped ultrathin piezoelectric microsystem for guided tissue targeting via mechanical sensing. *Nat. Biomed. Eng.* **2**, 162–172 (2018).
14. Yang, J. M. et al. Simultaneous functional photoacoustic and ultrasonic endoscopy of internal organs *in vivo*. *Nat. Med.* **18**, 1297–1302 (2012).
15. Hu, H. et al. Stretchable ultrasonic transducer arrays for three-dimensional imaging on complex surfaces. *Sci. Adv.* **4**, eaar3979 (2018).
16. Wang, X., Song, J., Liu, J. & Zhong, L. W. Direct-current nanogenerator driven by ultrasonic waves. *Science* **316**, 102–105 (2007).
17. Dagdeviren, C. et al. Conformal piezoelectric energy harvesting and storage from motions of the heart, lung, and diaphragm. *Proc. Natl Acad. Sci. USA* **111**, 1927–1932 (2014).
18. Baek, S. H. et al. Giant piezoelectricity on Si for hyperactive MEMS. *Science* **334**, 958–961 (2011).
19. Ma, K. Y., Chirarattananon, P., Fuller, S. B. & Wood, R. J. Controlled flight of a biologically inspired, insect-scale robot. *Science* **340**, 603–607 (2013).
20. You, Y. M. et al. An organic-inorganic perovskite ferroelectric with large piezoelectric response. *Science* **357**, 306–309 (2017).
21. Curry, E. J. et al. Biodegradable piezoelectric force sensor. *Proc. Natl Acad. Sci. USA* **115**, 909–914 (2018).
22. Wu, W. et al. Piezoelectricity of single-atomic-layer MoS<sub>2</sub> for energy conversion and piezotronics. *Nature* **514**, 470–474 (2014).
23. Zhu, H. et al. Observation of piezoelectricity in free-standing monolayer MoS<sub>2</sub>. *Nat. Nanotech.* **10**, 151–155 (2015).
24. Lee, B. Y. et al. Virus-based piezoelectric energy generation. *Nat. Nanotech.* **7**, 351–356 (2012).
25. Chen, X., Mahadevan, L., Driks, A. & Sahin, O. Bacillus spores as building blocks for stimuli-responsive materials and nanogenerators. *Nat. Nanotech.* **9**, 137–141 (2014).
26. Li, M. et al. Revisiting the  $\delta$ -phase of poly(vinylidene fluoride) for solution-processed ferroelectric thin films. *Nat. Mater.* **12**, 433–438 (2013).
27. Li, F. et al. Ultrahigh piezoelectricity in ferroelectric ceramics by design. *Nat. Mater.* **17**, 349–354 (2018).
28. Yan, Y., Zhou, J. E., Maurya, D., Wang, Y. U. & Priya, S. Giant piezoelectric voltage coefficient in grain-oriented modified PbTiO<sub>3</sub> material. *Nat. Commun.* **7**, 13089 (2016).
29. Qin, Y., Wang, X. & Wang, Z. L. Microfibre-nanowire hybrid structure for energy scavenging. *Nature* **451**, 809–813 (2008).
30. Qi, Y. et al. Enhanced piezoelectricity and stretchability in energy harvesting devices fabricated from buckled PZT ribbons. *Nano Lett.* **11**, 1331–1336 (2011).
31. Han, M. et al. r-shaped hybrid nanogenerator with enhanced piezoelectricity. *ACS Nano* **7**, 8554–8560 (2013).
32. Saadon, S. & Sidek, O. A review of vibration-based MEMS piezoelectric energy harvesters. *Energy Convers. Manag.* **52**, 500–504 (2011).
33. Meza, L. R., Das, S. & Greer, J. R. Strong, lightweight, and recoverable three-dimensional ceramic nanolattices. *Science* **345**, 1322–1326 (2014).
34. Cavallo, F. & Lagally, M. G. Nano-origami: Art and function. *Nano Today* **10**, 538–541 (2015).
35. Sydney Gladman, A., Matsumoto, E. A., Nuzzo, R. G., Mahadevan, L. & Lewis, J. A. Biomimetic 4D printing. *Nat. Mater.* **15**, 413–418 (2016).
36. Xu, S. et al. Assembly of micro/nanomaterials into complex, three-dimensional architectures by compressive buckling. *Science* **347**, 154–159 (2015).
37. Yan, Z. et al. Mechanical assembly of complex, 3D mesostructures from releasable multilayers of advanced materials. *Sci. Adv.* **2**, e1601014 (2016).
38. Yan, Z. et al. Deterministic assembly of 3D mesostructures in advanced materials via compressive buckling: A short review of recent progress. *Extrem. Mech. Lett.* **11**, 96–104 (2017).
39. Fu, H. et al. Morphable 3D mesostructures and microelectronic devices by multistable buckling mechanics. *Nat. Mater.* **17**, 268–276 (2018).
40. Lee, W. et al. Two-dimensional materials in functional three-dimensional architectures with applications in photodetection and imaging. *Nat. Commun.* **9**, 1417 (2018).
41. Zhang, Y. et al. Printing, folding and assembly methods for forming 3D mesostructures in advanced materials. *Nat. Rev. Mater.* **2**, 17019 (2017).
42. Wang, S. et al. Mechanics of curvilinear electronics. *Soft Matter* **6**, 5757–5763 (2010).
43. Persson, B. N. J., Albohr, O., Tartaglino, U., Volokitin, A. I. & Tosatti, E. On the nature of surface roughness with application to contact mechanics, sealing, rubber friction and adhesion. *J. Phys. Condens. Matter* **17**, 1–62 (2005).
44. Yin, S. et al. Measuring single cardiac myocyte contractile force via moving a magnetic bead. *Biophys. J.* **88**, 1489–1495 (2005).
45. Raman, R. et al. Optogenetic skeletal muscle-powered adaptive biological machines. *Proc. Natl Acad. Sci. USA* **113**, 3497–3502 (2016).
46. Ning, X. et al. 3D tunable, multiscale, and multistable vibrational micro-platforms assembled by compressive buckling. *Adv. Funct. Mater.* **27**, 1605914 (2017).
47. Kulah, H. & Najafi, K. Energy scavenging from low-frequency vibrations by using frequency up-conversion for wireless sensor applications. *IEEE Sens. J.* **8**, 261–268 (2008).
48. Zi, Y. et al. Harvesting low-frequency (<5 Hz) irregular mechanical energy: A possible killer application of triboelectric nanogenerator. *ACS Nano* **10**, 4797–4805 (2016).
49. Humood, M. et al. Fabrication and deformation of 3D multilayered kirigami microstructures. *Small* **14**, 1703852 (2018).
50. Ranzani, T., Gerboni, G., Cianchetti, M. & Menciassi, A. A bioinspired soft manipulator for minimally invasive surgery. *Bioinspir. Biomim.* **10**, 035008 (2015).

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## Author contributions

M.H., H.W., Z.Y., Y.Z., Y.H. and J.A.R. conceived the idea and designed the research. M.H., Y.Y., C.L., Z.Y. and J.L. performed micro-fabrication of all structures and devices. M.H., C.L., W.B. and H.Z. performed measurements and analysed the experimental data. M.H., W.B. and I.K. evaluated the devices in animal models. X.W. and B.A. performed cell seeding and cell viability assay under the supervision of G.A.A. H.W., Y.Z. and Y.H. led the mechanical modelling and theoretical studies, with assistance from H. Li, Y.X. and H. Luan. M.H., H.W., Y.Z., Y.H. and J.A.R. wrote the manuscript.

## Competing interests

The authors declare no competing interests.

## Additional information

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Sample size	Sample size was n=3 mice for the measurements of 3D devices. Output signals of the 3D and 2D devices were measured three times after implantation. Duration of each measurement ranges from 1 min to 5 min. Over twenty similar mouse behaviors appeared in the measurements. Three 3D devices were implanted.
Data exclusions	No data were excluded from the analyses.
Replication	Data measured from the implanted 3D and 2D devices were reproducible. Similar output of the device appeared in three different measurements which include over twenty mouse behaviors.
Randomization	All devices and animals tested were selected randomly.
Blinding	The investigators were blinded to group allocation during data collection and analysis (i.e., the assessment of cell viability and comparison of the output voltage of implanted 3D and 2D devices).

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<input checked="" type="checkbox"/>	<input type="checkbox"/> ChIP-seq
<input checked="" type="checkbox"/>	<input type="checkbox"/> Flow cytometry
<input checked="" type="checkbox"/>	<input type="checkbox"/> MRI-based neuroimaging

## Animals and other organisms

Policy information about [studies involving animals; ARRIVE guidelines](#) recommended for reporting animal research

Laboratory animals	Species: Mice. Strain: CD1, Charles River, Crl:CD1(ICR). Sex: female. Age at initiation of the treatment: At least 6 weeks, but not more than 15 weeks.
Wild animals	N/A
Field-collected samples	N/A