

# Soft 3D Microscale Vibratory Platforms for Characterization of Nanoscale Polymer Films

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## ABSTRACT

Vibrational resonances of microelectromechanical systems (MEMS) can serve as means for assessing physical properties of ultrathin coatings in sensors and analytical platforms. Most such technologies exist in largely two-dimensional (2D) configurations with limited total number of accessible vibration modes and modal displacements, thereby placing constraints on design options and operational capabilities. This study presents a set of concepts in three-dimensional (3D) microscale platforms with vibrational resonances excited by Lorentz-force actuation for purposes of measuring properties of thin film coatings. Polymer films with nanoscale thicknesses serve as test vehicles for demonstrating the advantages of these 3D MEMS for detection of multiple physical properties, such as modulus and mass, within a single polymer sample. The stability of the structure is demonstrated through multiple measurements of polymer samples using

a single platform, and via integration with thermal actuators temperature-dependent physical properties of polymer films is assessed. The findings establish unusual opportunities for interrogation of the physical properties of polymers through advanced MEMS design.

TEXT

Micro-electromechanical systems (MEMS) with tailored mechanical vibratory resonances address a range of applications in mass detection<sup>1-3</sup>, microscale rheology<sup>4-6</sup>, and energy harvesting<sup>7-9</sup>. Existing MEMS are largely based on two-dimensional (2D) geometries such as cantilevered beams<sup>10</sup>, doubly clamped bridges/wires<sup>11</sup>, and suspended plates<sup>12</sup>, which present limited numbers of accessible vibration modes and configurations of modal displacements. Consequently, the operation of most MEMS relies on a single vibration mode, thereby restricting their capability, for example, in detecting anisotropic mechanical properties in certain biological<sup>13-15</sup> and composite<sup>16-18</sup> thin-film materials, or for detecting multiple uncoupled physical properties of the same sample. Access to multimodal resonances in 2D MEMS platforms typically require non-idealities and asymmetries in nanomechanical resonators<sup>19</sup>, or advanced actuation techniques using collections of piezoelectric components<sup>20</sup> or patterned illumination with structured laser beams<sup>21</sup>, but the consequent engineering complications create challenges in scaling and adapting to complex materials systems. A potential alternative strategy relies on three-dimensional (3D) vibrational structures with multimodal and tunable resonances<sup>22-23</sup>, formed by controlled assembly. Reported methods in actuation via thin film piezoelectric elements require, however, multistep fabrication processes and they operate over a limited range of frequencies.

In this study, we introduce a set of experimental and theoretical approaches to the design and fabrication of 3D microscale frameworks as vibratory platforms with integrated circuit elements

for Lorentz-force actuation and localized heating. The assembly process exploits techniques in deterministic mechanical buckling<sup>24-27</sup> on elastomeric substrates. The key features of these systems are their 1) high resonant frequencies, accessible via a combination of Lorentz-force actuation and small characteristic dimensions, 2) diverse, multimodal motions controlled through orientation of the actuation force and design of the 3D layouts, and 3) low overall tensile stiffnesses enabled by the use of thin polymers for the structural elements. The results represent promising tools for measurement of both mass and modulus in isotropic and anisotropic thin polymer films. Demonstrations include extraction of modulus and density of coatings by use of multimodal resonances of a single, reusable measurement platform. Integrated thermal actuators allow for evaluation of the temperature dependence of the modulus, including those associated with phase transitions. Theoretical modeling and design suggest the potential for measurement of anisotropic mechanical properties in single or multilayer films.

## Results

### 3D Multimodal Vibratory Systems Actuated by Lorentz-Force

**Figure 1a** is a schematic illustration of a representative 3D system assembled on an elastomeric substrate. The overall size, as measured by the distance between the two sites at which the structure bonds to the substrate, is 800  $\mu\text{m}$ . The critical dimension, as defined by the width of the narrowest ribbon, is 100  $\mu\text{m}$ . As shown by the exploded view in **Figure 1b**, this platform consists of a layer of patterned conductive traces (Au, thickness = 300 nm) sandwiched between two layers of polyimide (PI, thickness  $\sim 2.5$   $\mu\text{m}$  for each). The PI layers not only form the structural components of the system, but they also encapsulate and protect the traces. The 3D architecture results from

mechanical buckling of a 2D precursor (see **Figure S1**) via controlled release of a prestretched elastomeric substrate, as described elsewhere<sup>24-27</sup>. The detailed fabrication process for the precursors appears in **Figure S2** and **Methods**. Briefly, layers of PI and gold are consecutively deposited on a silicon wafer and patterned using photolithography. Dissolution of a sacrificial layer of poly(methyl methacrylate) (PMMA) releases the precursor formed in this manner from the underlying wafer. The planar nature of the process then allows integration of nanoscale polymer films as test vehicles with a wide range of thicknesses and geometries. In this work, we demonstrate the use of a photodefinable epoxy (SU8), a cresol novolak resin (S1805), and a polymer brush (PNIPAm) as test films, patterned onto the 2D precursors via photopolymerization, photolithography and selective growth respectively (see **Methods** for details).

**Figure 1c** and **Figure S3** highlight the systems for Lorentz-force actuation and optical measurement. The Lorentz force follows from use of a sinusoidal voltage source (1.5V, Keithley 3390) to create an alternating current (AC) through the traces, with a permanent magnet (neodymium disc) placed underneath the 3D structure to induce a static magnetic field in the *z*-direction. The result is an AC oscillatory force at the frequency of the current, imparted onto the traces at a direction perpendicular to the current and the magnetic field. The amplitude of the Lorentz force can be estimated by  $F = B \times I_{rms} \times L_{Au} \approx 12.5 \mu\text{N}$  (magnetic field (*B*)  $\approx 0.5\text{T}$ ; rms value of electric current ( $I_{rms}$ ) = 25 mA; gold wire length ( $L_{Au}$ ) = 1 mm). The optical measurement system uses apparatus reported in previous work<sup>22</sup> based on lock-in detection of laser light scattered from the 3D structure. The amplitude of such a measurement is approximately proportional to the amplitude of the vibration. Additional details are in **Methods**. Changing the position of the magnet alters the direction of the magnetic field to allow actuation of targeted vibration modes. **Figure 1d** illustrates the lowest in-plane and out-of-plane vibration modes, obtained by 3D finite element

analysis (FEA, see **SI Section S1** for details). Experimentally, these modes are excited by placing the magnet below (denoted as mode 1, **Figure S4a**) and to the left of the 3D structure (denoted as mode 2, **Figure S4b**), respectively.

### Characterization of Modulus and Density Using Multimodal Resonances

The effects of the patterned polymer films under test on the vibrational frequencies of these two modes allows determination of both the modulus and density of the same sample. Dimensional analysis (see **SI Section S2** for details) and FEA suggest a linear relationship between the resonant frequency and the polymer modulus and density

$$\frac{f}{f_0} = C_E \frac{E_p h_p}{\hat{E}_{\text{Base}} h_{\text{Base}}} - C_\rho \frac{\rho_p h_p}{\hat{\rho}_{\text{Base}} h_{\text{Base}}}, \quad (1)$$

where  $f$  and  $f_0$  are the resonant frequencies with and without the polymer,  $E_p$ ,  $\rho_p$  and  $h_p$  are the polymer modulus, density and thickness,  $\hat{E}_{\text{Base}}$ ,  $\hat{\rho}_{\text{Base}}$  and  $h_{\text{Base}}$  are the effective modulus, average density and thickness of the PI/Au/PI tri-layer, respectively (see **SI Section S3** for details); and the dimensionless parameters  $C_E$  and  $C_\rho$  depend on the topology of the 3D vibratory platform, the vibration mode and the polymer patterns and the gold circuit patterns. For the 3D structure, vibration modes and polymer/gold patterns in **Figure S6**, FEA gives  $C_{E(1)} = 0.44$ ,  $C_{\rho(1)} = 0.30$  for mode 1 and  $C_{E(2)} = 0.20$ ,  $C_{\rho(2)} = 0.30$  for mode 2, as shown in **Figure 2a** and **2b**. The results indicate that the specific designs of the polymer patterns and the vibration modes enable partial decoupling of the sensitivities of resonant frequencies to the polymer modulus and density. In particular, the resonant frequency of mode 1 is 2.2 ( $C_{E(1)}/C_{E(2)}$ ) times more sensitive to the polymer modulus than that of mode 2, as measured by the slope of the curves in **Figure 2a**. Meanwhile the resonant

frequencies of both modes are sufficiently sensitive to the polymer density. The design of the polymer patterns follows from use of a FEA-based algorithm that optimizes the contribution of the polymer to the strain energy and the kinetic energy of the vibrations (see **SI Section S4** for details). The developed scaling law serves as the basis of an inverse problem to determine the polymer tensile stiffness ( $E_p h_p$ ) and mass ( $\rho_p h_p$ ) from the measured resonant frequencies (see **SI Section S5** for details).

In principle, the method is applicable to evaluation of any type of material (polymers, metals, ceramics), but the tensile stiffness ( $E_p h_p$ ) and mass ( $\rho_p h_p$ ) of the films must be at least one order of magnitude smaller than those of the base layer ( $\hat{E}_{\text{Base}} h_{\text{Base}} = 1.36 \times 10^4 \text{ N/m}$  and  $\hat{\rho}_{\text{Base}} h_{\text{Base}} = 0.013 \text{ kg/m}^2$  for the design shown in **Figure 2a**), such that the linear relationship in Eq. (1) applies. In such cases, to deduce the elastic modulus ( $E_p$ ) and density ( $\rho_p$ ) of the polymer from multimodal frequency measurements, the thickness ( $h_p$ ) of the polymer film must be known. Measurements of thickness are most easily performed prior to 3D assembly, in the precursor 2D geometry with techniques such as surface profilometry or spectroscopic ellipsometry (see **Methods** for details), with uncertainty of  $\sim 5\%$ .

As an example, **Figure 2c** and **2d** show experimental measurements of the response curves of mode 1 and 2 respectively, for the case of a patterned layer of a photodefinable epoxy (SU8; thickness = 610 nm, **Figure S8b**). The resonant frequencies are  $f_{(1)} = 30.6 \text{ kHz}$ ,  $f_{0(1)} = 28.8 \text{ kHz}$  for mode 1, and  $f_{(2)} = 28.4 \text{ kHz}$ ,  $f_{0(2)} = 27.9 \text{ kHz}$  for mode 2. The repeatability of the measurement is  $\sim 0.1\%$ , as defined by the standard deviation of four measurements performed in sequence (**Figure S7**). The effective modulus, average density and thickness of the base layer are  $\hat{E}_{\text{Base}} = 2.51 \text{ GPa}$ ,  $\hat{\rho}_{\text{Base}} = 2.41 \text{ g/cm}^3$  and  $h_{\text{Base}} = 5.4 \text{ }\mu\text{m}$ , respectively. Inserting these values into Eq. (1) yields the

modulus  $E_p = 3.8 \pm 0.4$  GPa and density  $\rho_p = 1.3 \pm 0.3$  g/cm<sup>3</sup> of the epoxy, as presented in **Figure 2e**. The combined uncertainties in thickness and frequency lead to ~10 % and ~20% uncertainties in the extracted film modulus and density, respectively (see **SI Section S5** for details). The ~5% discrepancies between the modulus and density values reported here and those in the literature (SU8 modulus 4.02 GPa; SU8 density 1.22 g/cm<sup>3</sup>) are within the uncertainties.

### **Robustness and Reusability of 3D Vibrators**

The elastic, reversible nature of the 3D assembly process allows repetitive use of these measurement platforms. Specifically, after measuring one polymer sample, applying tensile strain on the underlying elastomeric substrate fully returns the 3D structure to its unbuckled, 2D form, thus allowing the removal of the film under test and application of another material (see **Figure S9** for the schematic illustration). Release of the tensile strain re-assembles the 3D structure for frequency measurements. Cycling tests that involve 500 cycles of stretching/release reveal a resonance frequency shift of less than 0.3% (from  $34.90 \pm 0.03$  kHz to  $35.00 \pm 0.03$  kHz), despite an increase in circuit resistance of ~20% (**Figure S10**) perhaps due to some level of plastic yielding in the gold layer. Because the gold (300 nm) is much thinner than the PI (~5-6  $\mu$ m) and because it is positioned at the neutral mechanical plane, its plastic yielding has a minor effect on the overall resonant frequency. As a demonstrating example, **Figure 3b** shows a set of experimental results on two cresol novolak resin (S1805) films with different thicknesses ( $450 \pm 30$  nm and  $650 \pm 40$  nm), actuated with mode 1. The scanning electron microscope images of the device before and after the novolak resin is spin coated (**Figure S11**) indicate full coverage and good uniformity of the resulted film. Note that after washing away S1805 with acetone, the response curve of the 3D

vibrator matches the initial polymer-free state exceptionally well, indicating good robustness and repeatability of the system, as well as effectively complete removal of S1805.

### **Integration with Thermal Actuators for Temperature-Dependent Measurements**

The compatibility with planar fabrication technologies allows integration of additional actuating and sensing elements, via a few additional fabrication steps (see **Methods** for fabrication details). As a specific example, thermal actuators allow studies of modulus as a function of temperature. **Figure 4a** shows schematic illustrations of a modified 3D platform that includes this functionality. A bilayer of photodefinable epoxy (SU8, thickness =  $40\pm 2$  nm) and a poly(N-isopropylacrylamide) (PNIPAm) brush (thickness =  $270\pm 20$  nm) patterned directly on top of the thermal actuators serve as the test sample (see **Methods** for experimental details). **Figure 4b** shows the FEA calibration of the temperature change versus direct current input for the thermal actuator (see **SI Section S1** for details). The temperature change at the polymer region is uniform, as illustrated by the inset of **Figure 4b**. The experimental results in **Figure 4c** show as expected that the resonant frequency decreases as the temperature increases. Measurements for three cycles of heating are in **Figure 4c**, where the peak temperature is  $80^\circ\text{C}$  in the polymer in cycle 1 (denoted by the blue solid triangles) and  $300^\circ\text{C}$  in cycles 2 (red solid circles) and 3 (black solid squares). The sharp,  $\sim 0.5\pm 0.05$  kHz decrease in resonant frequency at  $40^\circ\text{C}\sim 50^\circ\text{C}$  corresponds to the glass transition of PNIPAm, while the  $\sim 0.2\pm 0.05$  kHz decrease at  $100^\circ\text{C}\sim 150^\circ\text{C}$  corresponds to that of SU8. The measured glass transition temperature matches well with the literature values for SU8<sup>29</sup>, whereas it is lower than the reported values for bulk PNIPAm<sup>30</sup>, as expected for this material which contains short polymer chain lengths and has a small thicknesses<sup>31-36</sup>. During the glass transition, the change in polymer

mass ( $\rho_p h_p$ ) is negligible because of mass conservation; the change in polymer thickness is also small (<1%) due to the small coefficient of thermal expansion<sup>37-38</sup> and the small change in temperature across the glass transition. The changes in the moduli of PNIPAm and SU8 films are thus determined from the corresponding resonant frequency shifts to be  $\Delta E_{\text{PNIPAm}} = 1.1 \pm 0.2$  GPa and  $\Delta E_{\text{SU8}} = 4 \pm 1$  GPa, respectively (**Figure 4d**, see **SI Section S6** for detailed calculations). The difference between the measured change in modulus of SU8 across the glass transition is consistent with the literature value<sup>29</sup> (3.7 GPa), to within experimental uncertainties. Finally, the further reduction in resonant frequency above 200 °C is likely attributable to the onset of softening of PI itself.<sup>39</sup>

### **Potential for Characterization of Anisotropic Elastic Moduli Using Multimodal Resonances**

The multimodal resonances of the 3D structures, and the ability to pattern polymers in a precise manner provide routes to determine the elastic moduli of anisotropic materials. **Figure 5** presents schematic illustrations of the 3D vibratory structure with patterned test films that have transversely isotropic elastic moduli (for instance, oriented polystyrene-block-polydimethylsiloxane films<sup>40</sup>). The optimized patterns are shown in **Figure S12**, with the optimization algorithm presented in the **SI Section S4**. Briefly, the optimization algorithm uses FEA results for the distributions of strain components. To enhance the sensitivity of the resonant frequencies to the longitudinal component of the modulus, the test polymer should be patterned in locations with the largest strain along the longitudinal direction. The same consideration holds true for the transverse modulus component. Optimized patterns of typical polymers balance these two considerations (see **SI Section 4** for details). For the current design, the resonant frequency of mode 2 is roughly 2.5 times more

sensitive to the transverse modulus ( $E_{P(t)}$ ) than the mode 1 (**Figure 5b**), while the latter is roughly 1.5 times more sensitive to the longitudinal modulus ( $E_{P(l)}$ , see **Figure 5c**) than the former, as measured by the slopes of the curves in **Figure 5b** and **5c**. This decoupling allows determination of both the longitudinal and transverse moduli by measuring the frequency of mode 1 and 2 with and without the polymer patterns, assuming that the polymer density and thickness are known.

## Conclusions

In summary, the results presented here demonstrate the applicability of 3D vibratory platforms to characterize the properties of thin film materials, with demonstrations on three types of polymers. With the current device parameters and optical measurement setups, modulus and density of any thin film with sufficiently small tensile stiffness and mass can be reliably, and independently, measured on the same polymer sample. The robustness and reusability of these 3D systems, together with their utility in evaluating temperature-dependent film properties, creates a broad range of possible uses. Theoretical modeling and design suggest opportunities for measuring anisotropic materials properties, specifically the transverse isotropic moduli of thin films. The assembly approaches can be applied to 3D structures with reduced dimensions, increased operating frequencies and enhanced sensitivity.

## Methods

**Fabrication of 3D Vibratory Platforms.** Fabrication began with spin coating a bilayer of poly(methyl methacrylate) (950 PMMA A4, MicroChem, thickness  $\sim 200$  nm) and polyimide (PI 2545, HD MicroSystems, thickness  $\sim 2.5$   $\mu\text{m}$ ) on a silicon wafer. Depositing Cr (thickness  $\sim 5$  nm)

and Au (thickness  $\sim 300$  nm) by electron beam evaporation followed by photolithography and wet etching defined the circuits. Spin coating another layer of PI (thickness  $\sim 2.5$   $\mu\text{m}$ ) followed by deposition and patterning of Cr (thickness  $\sim 5$  nm) and Au (thickness  $\sim 50$  nm) defined a hard mask in the shape of 2D devices, where the external electrodes were exposed for connections with the function generator. Oxygen plasma etching (March Jupiter III RIE) removes the unmasked PI. After removing the etching mask by wet etching, a layer of polymer was patterned on top to serve as the measurand (see below for details). The underlying PMMA layer was dissolved by immersion in acetone overnight to allow retrieval of the device onto a piece of water-soluble tape. A thin layer of silicon oxide (thickness  $\sim 50$  nm) deposited on the back side of the sample by electron beam evaporation created the necessary surface chemistry for strong adhesion to the elastomer platform at the bonding sites. The non-bonding sites were protected from  $\text{SiO}_2$  deposition by a Kapton shadow mask created by laser cutting. A silicone elastomeric substrate (Dragon Skin 10) was stretched to a desired prestrain (30-40%) using a customized, biaxial stage. After exposing the elastomer and the 2D precursors (still on water-soluble tape) to ultraviolet (UV) induced ozone (Jelight UVO-Cleaner, Model 144AX), the two were laminated together and then baked in a convection oven at  $70$   $^\circ\text{C}$  to yield strong adhesion at the bonding sites where  $\text{SiO}_2$  was deposited. Finally, dissolving the tape in warm water and slowly releasing the prestrain completed the assembly process. A schematic illustration of the procedures can be found in Figure S2.

**Patterning and Characterizing the Thicknesses of the Polymer Films.** Patterning the SU8 film used in Figure 2 began with spin coating the SU8 precursor (SU8 2000.5, MicroChem) onto the 2D devices at 2000 rpm for 40 s. After prebaking at  $95$   $^\circ\text{C}$  for 1 min, the film was exposed under an iron oxide photomask with 365 nm UV light at an intensity of  $60$   $\text{mJ}/\text{cm}^2$ . Post exposure baking

at 95 °C for 1 min and developing in SU8 developer (MicroChem) yielded the desired patterns on the 2D precursors. The sample was then hard bake at 200 °C for 10 min. The thickness was characterized using a profilometer (Dektak 3030) while the 2D precursors were still on silicon wafer.

Patterning of the S1805 films used in Figure 3 began by forming the 3D vibratory platforms as described in the previous section and stretching the elastomeric substrate to fully recover its 2D form. After rinsing the sample with acetone, isopropanol alcohol, and DI water, the S1805 precursor (MicroChem) was spin coated at 4000 rpm (cycle 2, Figure 3b) or 2000 rpm (cycle 3, Figure 3c), followed by baking at 110 °C for 3 min. The elastomeric substrate was then relaxed to return the sample to its 3D form. After each cycle, the sample was thoroughly rinsed with acetone to completely remove the S1805 film.

Patterning of the bilayer of SU8 and poly(N-isopropylacrylamide) (PNIPAm) brush used in Figure 4 began by spin coating SU8 precursor (SU8 2002, MicroChem), diluted using cyclopentanone with a 1:5 volume ratio, onto the 2D devices at 4000 rpm for 40 s. After prebaking at 95 °C for 1 min, the film was exposed under an iron oxide photomask with 365 nm UV light at an intensity of 50 mJ/cm<sup>2</sup>. Post exposure baking at 95 °C for 1 min and developing in SU8 developer (MicroChem) defined the desired patterns on the 2D precursors. The sample was then hard baked at 200 °C for 10 min. The PNIPAm brush was selectively grown on the SU8 patterns using surface-initiated atom transfer radical polymerization (SI-ATRP) using previously reported procedures<sup>[S1]</sup>. Briefly, SU8 surfaces were activated using oxygen plasma for 3 minutes (150 W, March Plasmod GCM-200) and then functionalized with the ATRP initiator (11-(2-Bromo-2-methyl)propionyloxy) undecyltrichlorosilane (BMPOUTS). Devices were placed in a 1 mM

solution of BMPOUTS in anhydrous hexanes at room temperature. After 24 hours, substrates were removed, sonicated in hexanes, dried using a nitrogen stream, and placed in a reaction vessel under argon. NIPAM (4.85 g, 42.9 mmol) was diluted with MeOH (15 mL) and H<sub>2</sub>O (7 mL) and degassed in a separate Schlenk flask. Then, 1,1,4,7,10,10-hexamethyltriethylenetetramine (HMTETA, 242  $\mu$ l, 0.89 mmol) and CuBr (53.8 mg, 0.38 mmol) were added to the monomer solution under positive argon flow. Once dissolved, the flask was then sealed and the mixture was transferred to the reaction vessel containing the substrates. Substrates were removed after 10 minutes, sonicated in methanol, ethanol, and water to remove the film residuals in the non-SU8 regions, and then dried in a nitrogen stream.

The thickness of the PNIPAm brush was determined using spectroscopic ellipsometry (VASE, J.A. Wollam Co) on a control sample. Ellipsometric parameters ( $\Psi$ ,  $\Delta$ ) were measured at three angles of incidence (65°, 70°, 75°) and from 400 nm to 800 nm. Data was analyzed by WVASE software using a three-layer model. Software-supplied refractive indices were used for silicon (substrate) and silicon dioxide (2 nm). Data was fit to a Cauchy layer model, with fixed ( $A_n$ ,  $B_n$ ) values of (1.45, 00.1) and no optical absorption. Control samples were prepared by growing the PNIPAm brush off a silicon wafer that had been patterned with SU8 using the identical conditions as the actual samples. The SU8 layer was determined to be 39.6 nm. After polymerization, the total thickness of the polymer layers (SU-8 and PNIPAM) was determined to be 312.9 nm. Thus, the NIPAM brush thickness is approximately 273.3 nm.

**Lorentz-force Actuation System and Optical Measurement System.** A function generator (Keithley 3390) applied 1.5 V sinusoidal voltage to the 3D vibratory structure while placed in a

static magnetic field induced by a permanent neodymium disc magnet. The Lorentz-force, perpendicular to both the electric current and the magnetic field, oscillated at the frequency of the applied voltage to drive vibrations of the structure.

An optical measurement system was custom-built to measure the dynamics of the 3D vibratory platforms (Figure S3). A focusing lens and mirror delivered a focused laser beam onto the center of the 3D structure. A microscope facilitated alignment. The 3D structure was firmly mounted onto a mechanical stage capable of translation in X-, Y-, and Z- directions and tilt with respect to X- and Y- axes. Light scattered from the 3D structure was reflected by a second mirror, collected by a second lens and directed to a photodetector (Thorlabs, DET110) for intensity measurements. The fluctuations in the scattering intensity due to the 3D vibrations generated photocurrent in the photodetector with the same overall time dependence. The system was tuned such that the amplitude of the photocurrent responded linearly to the amplitude of the input voltage. Within the operational range of frequency, the amplitude of the fluctuating photocurrent is directly proportional to the vibration amplitude, which peaked at the resonant frequency. A lock-in amplifier (SRS 830, Stanford Research Systems) was used to record the amplitude of the photocurrent.

A Labview program was used to sweep the desired range of frequency and convert data from the lock-in amplifier. At each frequency, the measurement was repeated 64 times to ensure minimum fluctuations, and the increment of frequency was 50 Hz. The error of the system was determined to be  $\sim 50$  Hz.

## FIGURES

Figure 1. 3D microstructure actuated by Lorentz force. a) Schematic illustrations of the 3D structure, assembled on an elastomer substrate. b) Exploded view of the layered composition with integrated conductive traces. c) Schematic illustration of the measurement system, consisting of a magnet and current source for Lorentz-force actuation and a laser apparatus for detection of vibrational motions. d) FEA views of the two distinct vibration modes of the vibrator, achieved by placing the permanent magnet at the bottom (top frame) and at the side (bottom frame). The amplitudes of the vibrations are exaggerated for viewing purposes.

Figure 2. Measurement of modulus and density using a single 3D microstructure. a)-b) FEA result for variation of the normalized resonant frequency with the normalized modulus (a) and the normalized density (b), for both vibration mode 1 and 2. c)-d) Measurement results of the normalized amplitude vs. frequency for mode 1 (c) and mode 2 (d). e) Modulus (left frame) and density (right frame) determined by the 3D vibratory structure, compared with the literature values.

Figure 3. Reusability of the 3D vibratory structure. a) Maximum strain in the gold layer obtained by FEA, which is below its yield strain (left frame); Exploded view of the layered composition of the vibratory platform (right frame). b) FEA views of the vibration mode in this measurement (inset), and a series of measurement results on the same platform using different testing subjects. The amplitudes of the vibrations are exaggerated for viewing purposes.

Figure 4. 3D vibratory platform integrated with thermal actuators. a) Schematic illustration of the 3D architecture integrated with thermal actuators, circuits for Lorentz force actuation and patterned polymer film (left frame), and magnified, exploded view of the section containing the thermal actuators (right frame). b) Calibration of temperature vs. supplied electric current. c) Experimental

results for the variation of the resonant frequency with temperature. d) Change in modulus of PNIPAm (left frame) and SU8 (right frame) determined from the results in c).

Figure 5. Theoretical investigation of measurement of anisotropic elastic moduli using a 3D vibratory platform. a) Schematic illustration of the 3D vibratory platform with a thin films that has anisotropic elastic moduli. In this case, the thin film has have different elastic moduli along the transverse ( $t$ ) and longitudinal ( $l$ ) directions. b)- c) FEA results of normalized resonant frequency for mode 1 and mode 2 with normalized modulus in transverse ( $t$ ) (b) and longitudinal ( $l$ ) (c) directions, respectively.

## ASSOCIATED CONTENT

### **Supporting Information**

Section S1. Finite element analysis (FEA)

Section S2. The scaling law in Eq. (1)

Section S3. Effective modulus and average density of an  $n$ -layer composite

Section S4. Algorithm that optimizes the polymer patterns

Section S5. Determination of the polymer modulus and density from the resonant frequency of two vibration modes

Section S6. Determination of the polymer tensile stiffness change during glass transition

Figure S1-S12.

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## ABBREVIATIONS

MEMS, micro-electromechanical systems; FEA, finite element analysis; PI, polyimide; 2D, two-dimensional; 3D, three-dimensional; PNIPAm, Poly(N-isopropylacrylamide); PMMA, poly(methyl methacrylate); AC, alternating current; DC, direct current.

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# Supporting Information for “Soft 3D Microscale Vibratory Platforms for Characterization of Nanoscale Polymer Films”

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## Section S1. Finite element analysis (FEA)

**2D to 3D transformation induced by the compressive strain.** FEA was conducted by the commercial software ABAQUS (version 6.14, standard). The post-buckling analysis predicts the buckled 3D shape induced by the compressive strain. The structure was discretized by Four-node finite-strain shell element, with at least ten elements along the narrowest ribbon to guarantee convergence.

**Vibration actuated by the Lorentz force.** The electrical analysis imported the buckled 3D shape obtained from the post-buckling analysis and predicted the electric current ( $\mathbf{J}$ ). The Lorentz force per unit volume was then calculated as  $\mathbf{J} \times \mathbf{B}$  by a home-made python script and exported to the following steady-state analysis of vibration. The steady-state analysis obtained the frequency spectrum of the vibration displacement and then the resonant frequency.

**Temperature change vs. direct electrical current for the micro heater.** The coupled thermal-electrical analysis imported the buckled 3D shape and obtained the temperature change due to Joule heating. The power per unit volume ( $P$ ) of the Joule heating is related to the electrical current by  $P = \frac{\mathbf{J} \cdot \mathbf{J}}{\sigma}$ , where  $\sigma$  is the electrical conductivity. Eight node thermal-electric element was adopted.

**Material properties.** The Young's modulus ( $E$ ), Poisson's ratio ( $\nu$ ) and density ( $\rho$ ) of the polyimide (PI), gold (Au) and substrate are  $E_{PI}=2.5$  GPa,  $\nu_{PI}=0.34$  and  $\rho_{PI}=1420$  kg/m<sup>3</sup>;  $E_{Au}=78$  GPa,  $\nu_{Au}=0.44$ ,  $\rho_{Au}=19320$  kg/m<sup>3</sup>;  $E_{Substrate}=166$  kPa,  $\nu_{Substrate}=0.49$ ,  $\rho_{Substrate}=1070$  kg/m<sup>3</sup>. The thermal conductivity of PI and Au is 0.12 W/(m·K) and 320 W/(m·K), respectively. The electrical

conductivity of Au as a function of temperature is given in Table S1. The convection coefficient between the structure and air is  $10 \text{ W}/(\text{m}^2 \cdot \text{K})$  [s2].

Table S1. Electrical conductivity of Au [s3]

Temperature (°C)	Electrical conductivity (S/m) $\times 10^7$
0	4.88
20	4.52
25	4.43
27	4.40
127	3.22
227	2.52
327	2.05
427	1.72

## Section S2. The scaling law in Eq. (1)

As a first step, a scaling law was derived via dimensional analysis for the resonant frequency when a thin layer of polymer is deposited homogeneously on a PI base layer (modulus--  $E_{PI}$ , density--  $\rho_{PI}$  and thickness  $h_{PI}$ ). For a vibration mode dominated by bending deformation, the resonant frequency is<sup>[s4]</sup>

$$f = \alpha \sqrt{\frac{K}{M}}, \quad (S1)$$

where the bending stiffness

$$K = \frac{E_{PI}^2 h_{PI}^4 + 4E_{PI}E_p h_{PI}^3 h_p + 6E_{PI}E_p h_{PI}^2 h_p^2 + 4E_{PI}E_p h_{PI} h_p^3 + E_p^2 h_p^4}{12(E_{PI}h_{PI} + E_p h_p) L^2}, \quad (S2)$$

the mass

$$M = (\rho_p h_p + \rho_{PI} h_{PI}) L^2, \quad (S3)$$

and  $L$  is the in-plane size of the structure. Considering that the polymer thickness ( $h_p$ ) is much smaller than the PI thickness, the ratio of the resonant frequency with/without the polymer is

$$\frac{f}{f_0} = \frac{3}{2} \frac{E_p h_p}{E_{PI} h_{PI}} - \frac{1}{2} \frac{\rho_p h_p}{\rho_{PI} h_{PI}}. \quad (S4)$$

For the 3D vibrators presented in the main text, an Au layer is sandwiched between two PI layers. Therefore, the PI modulus  $E_{PI}$  and density  $\rho_{PI}$  should be revised to the effective modulus  $\hat{E}_{Base}$  and average density  $\hat{\rho}_{Base}$  of the PI/Au/PI tri-layer (see **Section S3**). Equation (S4) is for the structure with the polymer depositing on the entire surface of the base layer. When the polymer is patterned

on a selected region such as the one shown in Figure S6a, the parameters  $C_E$  and  $C_\rho$  replace  $\frac{3}{2}$  and  $\frac{1}{2}$ , respectively. These considerations lead to the scaling law Eq. (1).

### Section S3. Effective modulus and average density of an $n$ -layer composite

The effective modulus of an  $n$ -layer composite is

$$\hat{E} = \frac{12}{h^3} \left( \frac{D_3}{3} - D_2 y_m + D_1 y_m^2 \right), \quad (\text{S5})$$

with

$$D_1 = \sum_{i=1}^n E_i h_i, \quad D_2 = \sum_{i=1}^n E_i (y_i^2 - y_{i-1}^2), \quad D_3 = \sum_{i=1}^n E_i (y_i^3 - y_{i-1}^3), \quad y_m = \frac{D_2}{2D_1}, \quad h = \sum_{i=1}^n h_i, \quad (\text{S6})$$

where  $E_i$  and  $h_i$  is the modulus and thickness of the  $i$ -th layer respectively;  $y_0 = 0$  and

$y_i = y_{i-1} + h_i$  for  $i=1, 2, \dots, n$ . The average density is

$$\hat{\rho} = \frac{1}{h} \sum_{i=1}^n \rho_i h_i, \quad (\text{S7})$$

where  $\rho_i$  is the density of the  $i$ -th layer.

## Section S4. Algorithm that optimizes the polymer patterns

### Polymer patterns for determining polymer modulus and density of isotropic material. An

algorithm was developed to select the polymer patterns such that the resonant frequencies of the two vibration modes have decoupled sensitivities to the polymer modulus and density. FEA first

predicts the distribution of the ratio  $\frac{W_{\text{Strain}}}{W_{\text{Kinetic}}}$  without polymer for vibration mode 1 and mode 2,

respectively;  $W_{\text{Strain}}$  and  $W_{\text{Kinetic}}$  are the strain energy density and the kinetic energy density of

vibration, respectively. For vibration mode 1, the entire region ( $\Omega$ , area  $A$ ) of the base layer is

divided into  $\Omega_{\text{Polymer}(1)}$  (area  $A_{\text{Polymer}(1)}$ ) and  $\Omega_{\text{NoPolymer}(1)}$ , with  $\Omega_{\text{Polymer}(1)} \cup \Omega_{\text{NoPolymer}(1)} = \Omega$ ,

$\Omega_{\text{Polymer}(1)} \cap \Omega_{\text{NoPolymer}(1)} = \emptyset$  and  $A_{\text{Polymer}(1)} = \beta_{(1)}A$ .  $\beta_{(1)}$  is an optimization variable. The region

$\Omega_{\text{Polymer}(1)}$  is selected such that for an arbitrary point in  $\Omega_{\text{Polymer}(1)}$ , the ratio  $\frac{W_{\text{Strain}}}{W_{\text{Kinetic}}}$  is larger than

that of an arbitrary point in  $\Omega_{\text{NoPolymer}(1)}$ . Similarly for vibration mode 2, a region  $\Omega_{\text{Polymer}(2)}$  with

area  $A_{\text{Polymer}(2)} = \beta_{(2)}A$  is selected such that for an arbitrary point in  $\Omega_{\text{Polymer}(2)}$ , the ratio  $\frac{W_{\text{Strain}}}{W_{\text{Kinetic}}}$  is

smaller than that of an arbitrary point in  $\Omega_{\text{NoPolymer}(2)}$  ( $\Omega_{\text{Polymer}(2)} \cup \Omega_{\text{NoPolymer}(2)} = \Omega$ ,

$\Omega_{\text{Polymer}(2)} \cap \Omega_{\text{NoPolymer}(2)} = \emptyset$ ).  $\beta_{(2)}$  is also an optimization variable. The polymer patterns are the

intersection of  $\Omega_{\text{Polymer}(1)}$  and  $\Omega_{\text{Polymer}(2)}$ . With the polymer patterns obtained in this manner, FEA

predicts the relationship of the resonant frequency versus the polymer modulus and density for the

two vibration modes. A parameter study on the variables  $\beta_{(1)}$  and  $\beta_{(2)}$  suggests that with

$\beta_{(1)} = \beta_{(2)} = 0.68$ , the sensitivities of the two vibration modes to the polymer modulus and density

are reasonably well decoupled (**Figure 2a and 2b**). The resultant polymer patterns are presented in **Figure S5a**.

**Polymer patterns for determining longitudinal modulus and transverse modulus of transversely isotropic material.** A similar algorithm selects the polymer patterns presented in **Figure S11** such that the resonant frequencies of the two vibration modes have decoupled sensitivities to the longitudinal modulus and the transverse modulus. FEA first predicts the distribution of the ratio  $\left| \frac{\varepsilon_l}{\varepsilon_t} \right|$  without polymer for vibration mode 1 and mode 2, respectively;  $\varepsilon_l$  and  $\varepsilon_t$  are the normal strain of vibration along the longitudinal direction and the transverse direction, respectively. Region  $\Omega_{\text{Polymer}(1)}$  is selected such that for an arbitrary point in  $\Omega_{\text{Polymer}(1)}$ , the ratio  $\left| \frac{\varepsilon_l}{\varepsilon_t} \right|$  is larger than that of an arbitrary point in region  $\Omega_{\text{NoPolymer}(1)}$ . Region  $\Omega_{\text{Polymer}(2)}$  is selected such that for an arbitrary point in  $\Omega_{\text{Polymer}(2)}$ , the ratio  $\left| \frac{\varepsilon_l}{\varepsilon_t} \right|$  is smaller than that of an arbitrary point in region  $\Omega_{\text{NoPolymer}(2)}$ . The polymer patterns are the intersection of  $\Omega_{\text{Polymer}(1)}$  and  $\Omega_{\text{Polymer}(2)}$ .  $\beta_{(1)}$  and  $\beta_{(2)}$  are optimization variables. A parameter study on  $\beta_{(1)}$  and  $\beta_{(2)}$  suggests that with  $\beta_{(1)} = \beta_{(2)} = 0.62$ , the sensitivities of the two vibration modes to the longitudinal modulus and the transverse modulus are reasonably well decoupled (**Figure 5b and 5c**). The resultant polymer patterns are presented in **Figure S11**.

**Section S5. Determination of the polymer modulus and density from the resonant frequency of two vibration modes**

Equation (1) leads to the following formula to determine the polymer modulus and density

$$E_P = \frac{\hat{E}_{\text{Base}} h_{\text{Base}}}{h_P} \frac{C_{\rho(1)} (\hat{f}_{(2)} - 1) - C_{\rho(2)} (\hat{f}_{(1)} - 1)}{C_{E(2)} C_{\rho(1)} - C_{E(1)} C_{\rho(2)}}, \quad (\text{S8})$$

$$\rho_P = \frac{\hat{\rho}_{\text{Base}} h_{\text{Base}}}{h_P} \frac{C_{E(1)} (\hat{f}_{(2)} - 1) - C_{E(2)} (\hat{f}_{(1)} - 1)}{C_{E(2)} C_{\rho(1)} - C_{E(1)} C_{\rho(2)}}, \quad (\text{S9})$$

where  $\hat{f}_{(1)} = \frac{f_{(1)}}{f_{0(1)}}$  and  $\hat{f}_{(2)} = \frac{f_{(2)}}{f_{0(2)}}$  are the ratio of the resonant frequency with/without the polymer

for vibration mode 1 and 2, respectively. The uncertainties in the determined polymer modulus and density caused by the uncertainties in the experiment results of the resonant frequencies are

$$\delta E_P = \frac{\hat{E}_{\text{Base}} h_{\text{Base}}}{h_P} \frac{C_{\rho(1)} \delta \hat{f}_{(2)} - C_{\rho(2)} \delta \hat{f}_{(1)}}{C_{E(2)} C_{\rho(1)} - C_{E(1)} C_{\rho(2)}} - \frac{\delta h_P}{h_P} E_P, \quad (\text{S10})$$

$$\delta \rho_P = \frac{\hat{\rho}_{\text{Base}} h_{\text{Base}}}{h_P} \frac{C_{E(1)} \delta \hat{f}_{(2)} - C_{E(2)} \delta \hat{f}_{(1)}}{C_{E(2)} C_{\rho(1)} - C_{E(1)} C_{\rho(2)}} - \frac{\delta h_P}{h_P} \rho_P, \quad (\text{S11})$$

respectively, where

$$\delta \hat{f}_{(1)} = \frac{\delta f_{(1)}}{f_{(1)}} - \frac{f_{(1)}}{f_{0(1)}} \frac{\delta f_{0(1)}}{f_{0(1)}}, \quad \delta \hat{f}_{(2)} = \frac{\delta f_{(2)}}{f_{(2)}} - \frac{f_{(2)}}{f_{0(2)}} \frac{\delta f_{0(2)}}{f_{0(2)}}, \quad (\text{S12})$$

$\delta f_{(1)}$ ,  $\delta f_{0(1)}$ ,  $\delta f_{(2)}$  and  $\delta f_{0(2)}$  are the uncertainties in the resonant frequency with/without polymer of vibration mode 1 and 2, respectively;  $\delta h_P$  is the uncertainty in the polymer thickness.

## Section S6. Determination of the polymer modulus change during glass transition

The SU8 and PNIPAm are much thinner than the base layer, which simplifies the resonant frequency of the structure in Figure 4a as

$$\frac{f}{f_0} = C_E \frac{E_{\text{SU8}} h_{\text{SU8}}}{\hat{E}_{\text{Base}} h_{\text{Base}}} + C_E \frac{E_{\text{PNIPAm}} h_{\text{PNIPAm}}}{\hat{E}_{\text{Base}} h_{\text{Base}}} - C_\rho \frac{\rho_{\text{SU8}} h_{\text{SU8}}}{\hat{\rho}_{\text{Base}} h_{\text{Base}}} - C_\rho \frac{\rho_{\text{PNIPAm}} h_{\text{PNIPAm}}}{\hat{\rho}_{\text{Base}} h_{\text{Base}}}, \quad (\text{S13})$$

where  $E_{\text{SU8}}$ ,  $\rho_{\text{SU8}}$ ,  $h_{\text{SU8}}$ ,  $E_{\text{PNIPAm}}$ ,  $\rho_{\text{PNIPAm}}$  and  $h_{\text{PNIPAm}}$  are the modulus, density and thickness of SU8 and PNIPAm, respectively. During the glass transition of PNIPAm, the SU8 tensile stiffness ( $E_{\text{SU8}} h_{\text{SU8}}$ ) does not change because the glass transition temperature of SU8 is much higher than that of PNIPAm. Meanwhile, the change in the polymer mass and thickness are also negligible as discussed in the main text. Therefore, the PNIPAm modulus change is

$$\Delta E_{\text{PNIPAm}} = \frac{\hat{E}_{\text{Base}} h_{\text{Base}} \Delta f_{\text{PNIPAm}}}{C_E h_{\text{PNIPAm}} f_0}, \quad (\text{S14})$$

where  $C_E = 0.53$  is determined by FEA and  $\Delta f_{\text{PNIPAm}}$  is the resonant frequency change during glass transition. Similarly, during the glass transition of SU8, the SU8 tensile stiffness change is

$$\Delta E_{\text{SU8}} = \frac{\hat{E}_{\text{Base}} h_{\text{Base}} \Delta f_{\text{SU8}}}{C_E h_{\text{SU8}} f_0}, \quad (\text{S15})$$

where  $\Delta f_{\text{SU8}}$  is the resonant frequency change. The uncertainties in the determined modulus change are

$$\delta(\Delta E_{\text{PNIPAm}}) = \frac{\delta(\Delta f_{\text{PNIPAm}})}{\Delta f_{\text{PNIPAm}}} \Delta E_{\text{PNIPAm}} - \frac{\delta h_{\text{PNIPAm}}}{h_{\text{PNIPAm}}} \Delta E_{\text{PNIPAm}}, \quad (\text{S16})$$

$$\delta(\Delta E_{\text{SU8}}) = \frac{\delta(\Delta f_{\text{SU8}})}{\Delta f_{\text{SU8}}} \Delta E_{\text{SU8}} - \frac{\delta h_{\text{SU8}}}{h_{\text{SU8}}} \Delta E_{\text{SU8}}, \quad (\text{S17})$$

for PNIPAm and SU8 respectively, where  $\delta(\Delta f_{\text{PNIPAm}})$ ,  $\delta h_{\text{PNIPAm}}$ ,  $\delta(\Delta f_{\text{SU8}})$  and  $\delta h_{\text{SU8}}$  are the uncertainties in the resonant frequency changes and thicknesses of PNIPAm and SU8 respectively.

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