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## Article

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**In the field of phononics, periodic patterning controls vibrations and thereby the flow of heat and sound in matter. Bandgaps arising in such phononic crystals realize low-dissipation vibrational modes and enable applications towards mechanical qubits, efficient waveguides, and state-of-the-art sensing. Here, we combine phononics and two-dimensional materials and explore the possibility of manipulating phononic crystals via applied mechanical pressure. To this end, we fabricate the thinnest possible phononic crystal from monolayer graphene and simulate its vibrational properties. We find a bandgap in the MHz regime, within which we localize a defect mode with a small effective mass of  $0.72 \text{ ag} = 0.002 m_{\text{physical}}$ . Finally, we take advantage of graphene's flexibility and mechanically tune a finite size phononic crystal. Under electrostatic pressure up to 30 kPa, we observe an upshift in frequency of the entire phononic system by more than 350%. At the same time, the defect mode stays within the bandgap and remains localized, suggesting a high-quality, dynamically tunable mechanical system.**

## INTRODUCTION

A phononic crystal (PnC) is an artificially manufactured structure with a periodic variation of material properties e.g. stiffness, mass, or stress<sup>1</sup>. This periodic perturbation creates a meta-crystallographic order in the system leading to a vibrational band structure hosting acoustic Bloch waves, in analogy to the electronic band structure in solids<sup>1</sup>. Designing the lattice parameters of the meta-structure allows to directly manipulate phonons at various length scales<sup>2-4</sup>. This can be used to guide<sup>5-7</sup> and to focus phonons<sup>8,9</sup>, or to open a vibrational bandgap<sup>1,11-13</sup>.

Phononic bandgaps in periodic structures suppress radiation losses and allow for highly localized modes (of frequency  $f$ ) on artificial irregularities<sup>10,14,15</sup>. The quality factors ( $Q = \frac{f}{\Delta f}$ ) of these so-called defect modes are especially high<sup>16,17</sup>. In particular, resonances with  $Q > 2 \times 10^8$  have been observed at room temperature in silicon nitride (SiN) PnCs<sup>16,17</sup>. By strain engineering and thereby increasing the energy stored in the vibration (dissipation dilution)  $Q$  is increased even higher<sup>18</sup>. These measures allow the quality factor to exceed the empirical  $Q \sim m^{1/3}$  rule<sup>18,19</sup> and the vibrational periods to overcome the thermal decoherence time limit:  $\tau = \hbar Q / k_B T$ <sup>16,18</sup>. This, in turn, enables the study of quantum effects in resonators of macroscopic size – all at room temperature<sup>20,21</sup>.

There have been recent efforts to realize a PnC with dynamically tunable frequency<sup>21-32</sup>. Frequency tunability may unlock new regimes of guiding, filtering, and focusing phonons. It would furthermore allow to resonantly couple to an external optical or mechanical excitation and thus realize sensing applications with mechanical qubits and studies on quantum entanglement<sup>22</sup>. Yet, the mechanical resonances in PnCs are determined by material constants and the crystal geometry<sup>23-27</sup>, which cannot be varied easily. In principle, the mode frequencies can be controlled by changing the temperature<sup>28,29</sup> or by an external magnetic field<sup>30,31</sup>. This, however, only causes limited tunability and necessitates heating the system or inclusion of magnetic materials. While SiN, as well as other conventional low-loss materials, is very stiff and allows only limited mechanical tunability<sup>32,33</sup>, strain has been used to adjust the frequency response of elastic polydimethylsiloxane (PDMS)<sup>34</sup>. Unfortunately, low crystalline quality of the material led to limited tunability and very small  $Q$ s for mechanical modes.

Recently, PnCs made from two-dimensional (2D) materials have been considered<sup>35-37</sup>. Such materials, exemplified by graphene, feature high electrical conductivity, intrinsically low mass, high fundamental frequency, and easily accessible displacement non-linearity. Most importantly, their high tensile strength and monolayer character allows to mechanically strain them up to ten percent by application of relatively modest forces<sup>38</sup>. That invites consideration of mechanically controllable 2D-material based phononic crystals. Specifically, we expect the entire acoustic band structure of such a PnC to be highly tunable by applying mechanical pressure. Nevertheless, tunability of 2D phononic systems as well as localized defect modes in them have not been studied yet.

Here, we investigate mechanical tunability in a realistic graphene phononic crystal. First, we experimentally study the feasibility of a graphene PnC. The monolayer thickness of graphene imposes several restrictions on maximum device size, number of unit cells, and applicable patterning strategies. Nevertheless, we fabricate a suspended micron-sized monolayer graphene PnC via focused helium ion beam milling (FIB). We show phononic patterning for a range of lattice constants down to  $a = 175$  nm and probe tension redistribution in the resulting PnC by Raman spectroscopy. We then use the experimentally established parameters to compute the phononic band structure of the resulting PnC and find a bandgap from 48.8 MHz to 56.5 MHz. We use this bandgap as a phononic radiation shield that localizes a central defect mode. The defect mode positioned inside the bandgap has an effective mass of 0.72 ag, corresponding to a more than 100-fold reduction compared the fundamental resonance. This greatly increases the sensitivity for mass and force sensing applications. Finally, we computationally investigate the mechanical tunability of the PnC in an experimentally established geometry with a local electrostatic gate inducing pressure<sup>39,40</sup>. The applied pressure smears out the phononic bandgap as the out-of-plane displacement breaks the symmetry and causes perturbations of the artificial lattice, yet the mode shape of the defect mode remains highly localized. Overall, we can tune the resonance frequency of the defect mode by more than 350% and access new regimes of strain engineering.

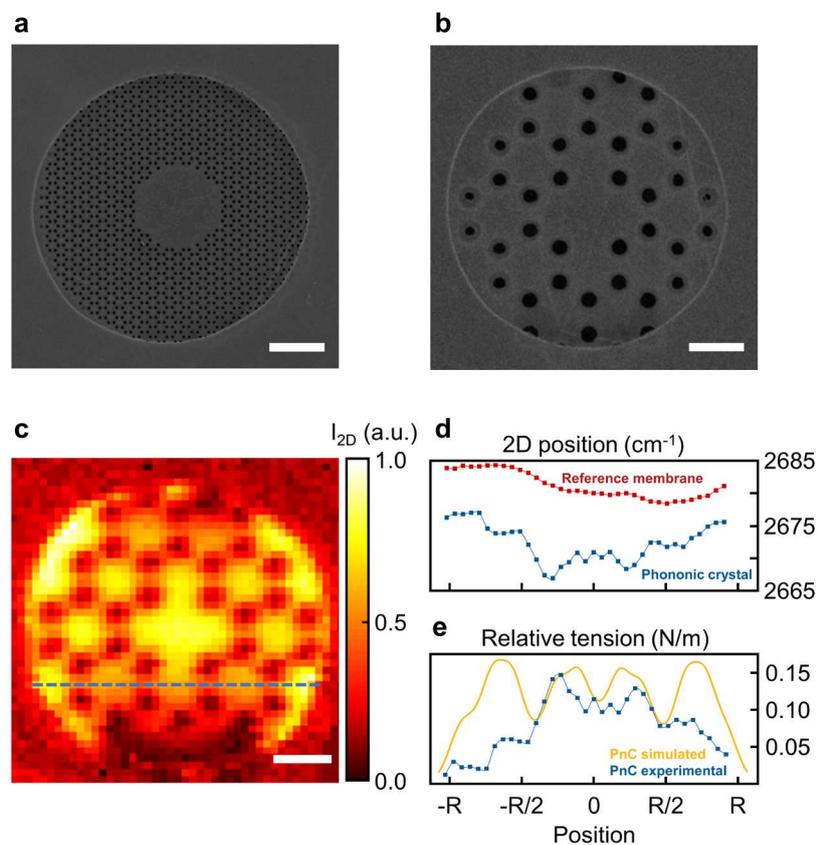
## RESULTS

**Designing a tunable phononic crystal.** Our device design of a tunable, two-dimensional phononic crystal consists of the following key elements. First, the PnC material must be freestanding to allow out-of-plane displacement. Second, it is necessary to use an electrically conductive material. In that case, an electrostatic gate electrode can be used to apply pressure and to induce tension as the membrane is pulled towards the gate. Third, the material needs to be flexible in order to allow large mechanical tunability with small pressures. Monolayer graphene with its high carrier mobility  $>200.000 \text{ cm}^2/\text{Vs}^{41}$  and large breaking strength  $>10\%^{38}$  perfectly fulfils these requirements. Finally, the device needs to host a large enough number of unit cells with sufficient periodicity to form a well-defined PnC. While this task is simple in thick SiN, it is much more challenging for fragile, freestanding monolayer graphene. To overcome this, we choose a much smaller unit cell compared to typical SiN-PnCs ( $\sim 100 \mu\text{m}$  size) and use helium FIB-milling to pattern the PnC. This direct lithography allows to pattern graphene down to 10 nm features<sup>43</sup>, whilst causing little damage to suspended graphene<sup>44,45</sup>.

A patterned prototype monolayer graphene PnC is shown in Fig. 1a. It consists of a honeycomb lattice of holes (lattice constant  $a = 350 \text{ nm}$ , hole diameter  $d = 105 \text{ nm}$ ) around a central region. Within its  $10 \mu\text{m}$  diameter the two-dimensional PnC contains more than 30 unit cells. The honeycomb lattice is inspired by Tsaturayn et al.<sup>16</sup> and results in a robust bandgap<sup>13,16,46</sup>, whilst leaving a relatively large fraction of material to ensure a stable device. Our PnC design allows us to reproducibly fabricate graphene PnCs of various sizes (Supplementary Fig. 1,2,3)<sup>42</sup>.

Next, we map the tension within the produced structures, because the total tension and its redistribution upon cutting affect the properties of the PnC. For the honeycomb lattice, we expect tension hot spots in the thin ribbons and relaxation in the centres of the hexagons. This redistribution effect has been demonstrated in SiN at length scales of tens of micrometers<sup>47</sup>. We now use Raman spectroscopy to probe this effect on a much smaller length scale. To this end, we fabricate another prototype device (Fig. 1b) with lattice constant  $a = 2 \mu\text{m}$  and spatial features comparable to the size of a focused laser spot. The intensity map of the 2D-Raman mode of graphene for this device is shown in Fig. 1c. The intensity of the 2D-mode corresponds to the amount of material while its spectral position depends on

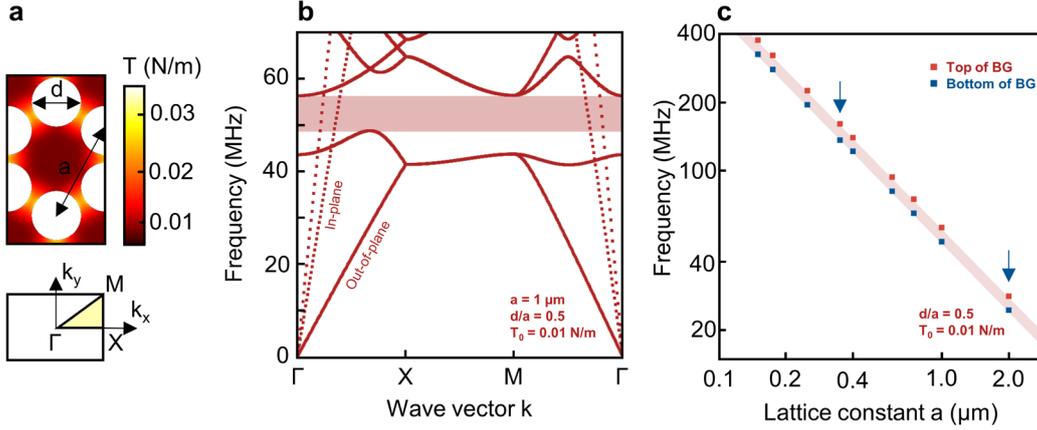
the tension in the material<sup>48,49</sup>. In the pizza-like image one can clearly see the removed material from the drop in intensity and identify the honeycomb lattice. In Fig. 1d, we compare the spectral position of the Raman 2D-mode for a graphene PnC (blue) along the dashed line shown in Fig. 1c to an unpatterned graphene membrane (red). The quasi-periodic variations in the PnC device, that are absent in the unpatterned reference, correspond to the redistributed tension. We compare the extracted relative tension (Fig. 1e, blue) to a simulation (Fig. 1e, yellow) and find the expected signatures of tension redistribution – higher tension between the holes and lower tension in the middle of the hexagons (see Supplementary section IV).



**Figure 1 | Graphene phononic crystals and tension redistribution.** **a,b**, Helium ion micrographs of prototype monolayer graphene phononic crystal devices with lattice constants 350 nm and 2  $\mu\text{m}$ , respectively. The phononic pattern, a honeycomb lattice of holes with a defect in its centre allows us to localize a vibrational defect mode. Scale bar length is 2  $\mu\text{m}$ . **c**, Intensity map of the Raman-active 2D mode of graphene for the device shown in **(b)**. The periodic pattern is clearly visible. **d**, Raman 2D-mode position along a line cut (dashed line in **(c)**) for a PnC (blue) and reference membrane (red). The PnC shows a periodic variations of much larger amplitude compared to the fluctuation in the reference sample. **e**, Comparison of the relative tension extracted from Raman measurements (blue) to the simulated tension distribution (yellow) confirming the redistribution of tension upon patterning. The simulation includes spatial broadening due to the finite size of the laser spot.

**Simulations.** Having experimentally established the feasibility of a suspended graphene PnC, we use our findings to simulate its phononic properties. We employ two independent simulation approaches. First, we calculate the phononic band structure for an infinitely repeated unit cell (“infinite model”). This model is well-accepted and fast<sup>16–18</sup>. However, due to the size limits of suspended graphene, our devices are smaller than typical SiN-PnCs (mm size)<sup>16–18</sup> and contain fewer unit cells. Furthermore, we want to apply pressure to the entire system and investigate localized modes in the bandgap. Therefore, we also simulate a more realistic system of finite size (“finite model”). For both models, we use the honeycomb lattice with feasible parameters and account for tension redistribution upon fabrication (Fig. 1d,e). We choose a lattice constant  $a = 1 \mu\text{m}$ , a filling factor of  $d/a = 0.5$  (slightly larger than in Fig. 1) and an initial tension of  $T_0 = 0.01 \text{ N/m}$ , a realistic value for clean monolayer graphene<sup>39,50</sup>. We start with discussing our simulation results, and then address the question of experimental signatures and their detectability.

**Infinite model.** To calculate a complete band structure for the infinite honeycomb lattice, we apply periodic Floquet boundary conditions to the unit cell (Fig. 2a) and calculate the eigenfrequencies for the wavevector  $k$  along the high symmetry lines of the first Brillouin zone (1.BZ). The resulting band structure is shown in Fig. 2b. We find a mixture of in-plane (dashed lines) and out-of-plane modes (solid lines). From the slope of the out-of-plane modes in Fig 2b, we determine the speed of sound  $v_g = \frac{\partial\omega}{\partial k} = 13\text{m/s}$ . In the range from 48.8 MHz to 56.5 MHz (red shaded area) we find a bandgap for out-of-plane modes. This quasi-bandgap (in-plane modes are still present) has a gap-to-midgap ratio of 14.6%. The in-plane modes do not couple to out-of-plane modes<sup>51</sup> and therefore do not hinder radiation shielding. The bandgap originates from Bragg scattering, with each hole acting as a scatterer for out-of-plane oscillations. Upon negative interference conditions, directional Bragg bandgaps open at the high symmetry points. Where these gaps overlap radiation shielding becomes possible, because wave propagation is isotropically forbidden<sup>10</sup>. The bandgap position depends on the lattice constant  $a$ . With our fabrication schema we can tailor the bandgap centre from 350 MHz to 26 MHz by varying  $a$  from 175 nm to 2  $\mu\text{m}$  (Fig 2c, devices in Supplementary Fig. 2,3). Overall, the simulations in the infinite model suggest the possibility of a large quasi-bandgap, which we will next use to control phonons.



**Figure 2 | Band structure calculations of an infinite graphene phononic crystal.** **a**, Unit cell of the honeycomb lattice with redistributed tension (top) and the corresponding first Brillouin zone (bottom). **b**, Phononic band structure for the unit cell shown in (a). In-plane modes are shown as dashed lines, out-of-plane modes as solid lines, and the corresponding quasi-bandgap region as red shaded area. **c**, Top (red) and bottom (blue) of the bandgap vs. lattice constant. The blue arrows indicate the lattice constant of the devices from Fig. 1a,b.

**Finite model.** To study a realistic device of finite size under electrostatic pressure and to implement a defect into the phononic pattern, we conduct a second, independent simulation (“finite model”). In this model, we consider a finite number of unit cells of the honeycomb lattice (same  $a$ ,  $d/a$ , and  $T_0$  as before) and employ fixed boundary conditions along the PnC’s perimeter. We choose a circular device as such a geometry allows uniform suspension and shows little edge effects. To create a defect, we translate six holes within the lattice, leaving a hexagonal defect<sup>16</sup>, as sketched in Fig. 3a for a device with a device diameter of  $30.6 \mu\text{m}$  and a hexagonal defect diagonal of  $1.9 \mu\text{m}$ . Freestanding graphene devices of that size have been fabricated<sup>52</sup> and the central defect area is large enough to measure resonances interferometrically<sup>53,54</sup>. Next, we simulate the first 1500 eigenfrequencies and the corresponding spatial mode shape. In Fig 3b, we plot the frequencies  $f$  vs. mode number  $N$  for the PnC (blue) and compare it to an unpatterned graphene membrane as reference (green). The graph for the PnC shows signs of a bandgap, as we observe an initial flattening of the curve, followed by a sudden increase. This region of reduced mode density coincides exactly with the bandgap from our infinite model (blue shaded area) and stands in contrast to the unpatterned membrane for which the frequencies gradually increase with mode number. The second indication of the bandgap is evident when we examine the effective mass of the modes:

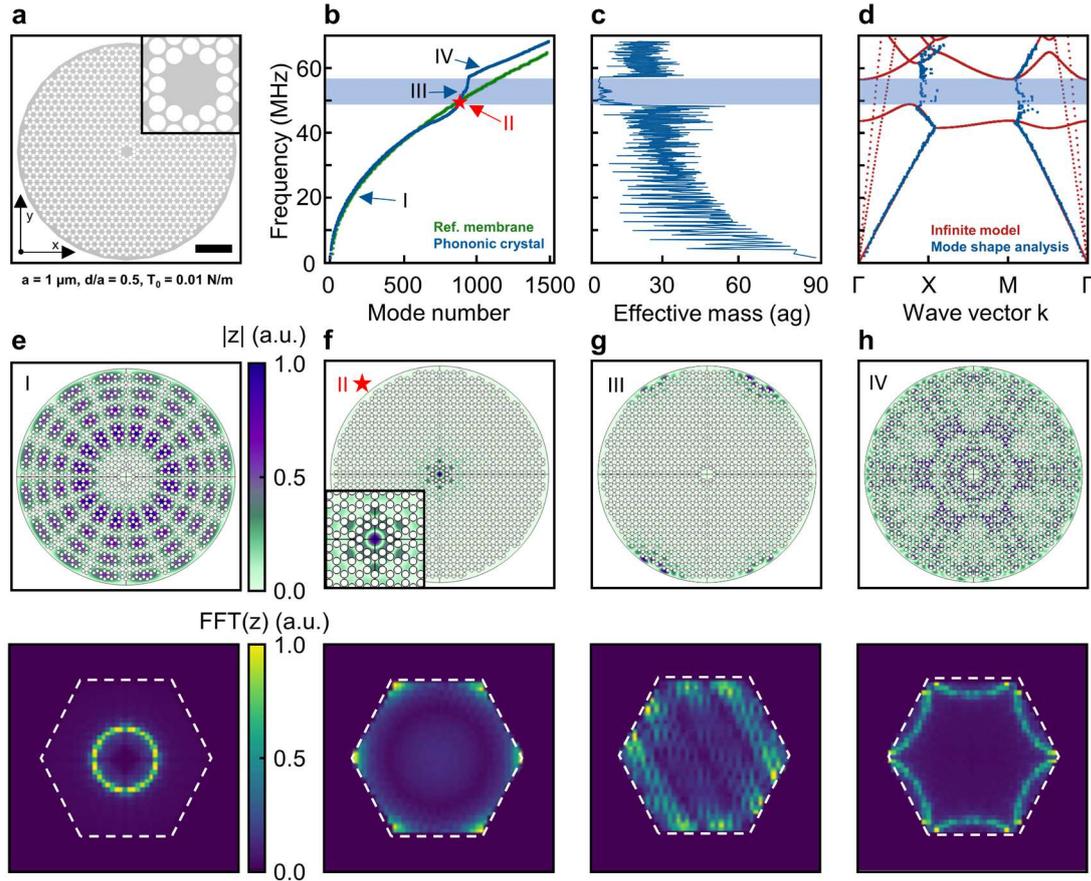
$$m_{\text{eff}} = \rho_{2D} \iint \frac{z^2}{z_{\text{max}}^2} dx dy,$$

where  $\rho_{2D}$  is the areal density of graphene and  $z$  ( $z_{\text{max}}$ ) is the (maximum) vibration amplitude in  $z$ -direction. For the fundamental mode we obtain  $m_{\text{eff}} = 80.9 \text{ ag} = 0.252 m_{\text{physical}}$ , which roughly matches the literature value for a uniform, circular membrane of  $m_{\text{eff}} = 0.269 m_{\text{physical}}$  (zerth order Bessel function)<sup>55</sup>. We observe a pronounced drop of  $m_{\text{eff}}$  in the bandgap region (Fig. 3c). This observation is consistent with localized modes inside the bandgap, which typically show a small average displacement resulting in a reduced effective mass<sup>18</sup>.

Finally, we directly extract the band structure from the results of the finite model and compare it to that of the infinite model. To accomplish this, we analyse the mode shape of each resonance following ref. <sup>56</sup>. Specifically, we take the spatial FFT of each mode shape to find its representation in reciprocal space and to assign a wave vector  $k$  to each mode. In Figs. 3e-h, we show real space (top) and reciprocal space (bottom) plots of exemplary modes. Mode I (20.2 MHz – Fig. 3e) is below the bandgap and resembles a higher order Bessel mode in real space, which transforms to a near-uniform circle in momentum space. A higher frequency mode IV (60.7 MHz – Fig. 3h) is situated above the bandgap. For this mode, we observe zone folding as the mode reaches out beyond the 1.BZ (dashed white line). Analysing all 1500 modes lets us restore the dispersion relation beyond the 1.BZ (Fig.3d, blue markers), which almost perfectly matches the band structure from the infinite model (red solid lines). From our observations of reduced mode density (Fig. 3b), drop in effective mass (Fig. 3c), and mode shape-analysis (Fig. 3d), we confirm the presence of a bandgap for out-of-plane modes in a realistic system of finite size.

Next, we examine the modes located within the bandgap and identify the defect mode. In Fig. 3g, we show a typical bandgap mode in real (top) and  $k$ -space (bottom). As most modes in the bandgap, this mode is localized at the edges of the PnC in the real space. However, one mode at frequency 49.9 MHz is localized at the central defect (Fig. 3f). We therefore identify it as our defect mode. The  $m_{\text{eff}}$  of the mode is 0.724 ag, which is more than a factor 100 smaller than the fundamental mode of the system and orders of magnitude lower than for any reported SiN defect mode<sup>16-18</sup>. This small  $m_{\text{eff}}$  corresponds to the high degree of spatial localization expected for localized modes. Indeed, within the bandgap any

transmission is heavily damped<sup>57,58</sup> and no extended waves are present<sup>1,10</sup>. We note that the relative position of the defect mode within the bandgap is purely determined by the device geometry. Consequently, our design works for any initial tension  $T_0$  in the graphene membrane. Overall, our model confirms the vibrational bandgap for system of finite size and a localized defect mode with extremely small effective mass within that bandgap.



**Figure 3 | Finite size model of a graphene phononic crystal.** **a**, Device geometry for the finite system simulations (scale bar is  $5 \mu\text{m}$ ). A central “defect” region is designed to localize one vibrational mode and decouple it from its environment **b**, The first 1500 simulated eigenfrequencies vs. mode number for a phononic crystal device (blue) and a circular membrane without patterning (green). The bandgap region from the infinite model is shown in blue. **c**, Effective mass for each mode. The modes within the bandgap (blue) show a more than 100-fold decrease in effective mass compared to the fundamental mode. **d**, Band structure calculated from the finite model via mode-shape analysis (blue) along with the band structure from the infinite model (red). The low energy acoustic branches fit well, and the bandgap regions coincides with the simulated results from the infinite model (red). **e-h** Exemplary mode shapes in real (top) and reciprocal space (bottom) for: **(e)** a mode below the bandgap **(I)**, **(f)** the defect mode **(II)**, **(g)** another highly localized mode in the bandgap **(III)** and **(h)** a mode above the gap **(IV)**.

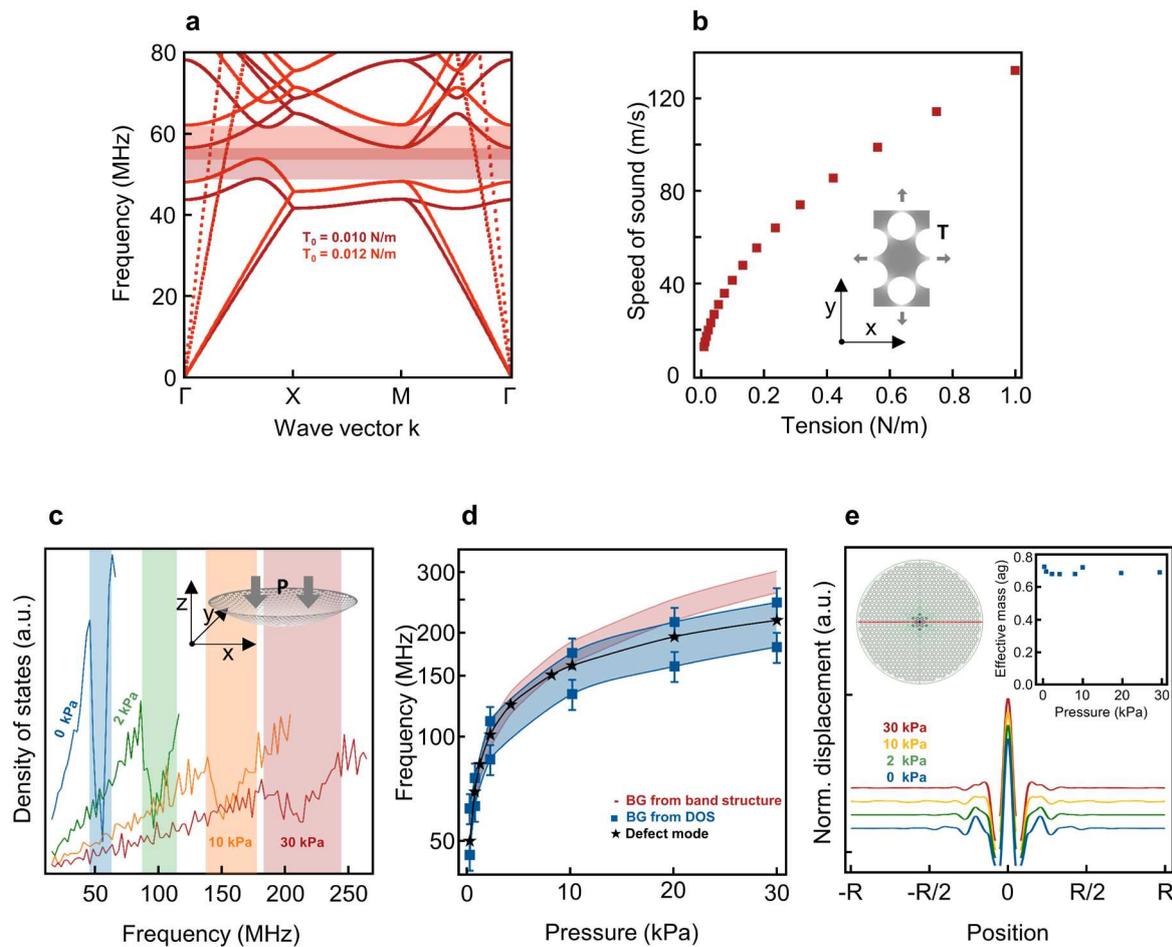
**Phononic crystal tuning.** We now show the key advantage of our graphene PnC – dynamic and rapid frequency tuning of the bandgap and of the defect mode. To demonstrate this, we model our graphene

PnC under pressure, which is applied by an electrostatic gate. The pressure causes displacement of the suspended membrane and increases the in-plane tension. We initially approximate this effect in first order in our infinite model by neglecting out-of-plane displacement and simply increasing the in-plane tension. In Fig. 4a, we plot the band structure for  $T_0 = 0.010$  N/m (red) and  $T_0 = 0.012$  N/m (orange). We observe a frequency increase of the out-of-plane modes and thus an upshift of the quasi-bandgap by 10%. The speed of sound  $v_g$  rises from 13 m/s to 130 m/s in the range of tension from 0.01 N/m to 1 N/m (Fig. 4b). The system behaves as a thin membrane under tension and the resonance frequencies scale directly with tension:  $f \propto \sqrt{T_0}$ <sup>55</sup>. This scaling makes our system highly sensitive to tension and in combination with the mechanical flexibility of monolayer graphene allows for broad frequency tuning.

Having demonstrated the overall tunability of our system, we now simulate the effect of electrostatic pressure on the phononic system and the defect mode in a realistic device. To do so, we switch to the finite model and apply pressure in negative z-direction causing largest displacement in the centre of the device and perturbing the lattice. In our simulations we stick to experimentally reported pressure values and apply a maximum of 30 kPa<sup>39</sup>. To investigate the influence of pressure on the bandgap, we compute the density of states,  $DOS = dN/df$ , and plot it vs. pressure in Fig. 4c. In this plot, the bandgap is distinguished by a reduced DOS. While at zero pressure the bandgap region is obvious, for higher pressures the drop becomes less pronounced (Fig. 4c). We attribute this smearing out to a symmetry breaking and perturbation of the phononic crystal. Nevertheless, we can estimate the top and bottom of the bandgap, which we show in Fig 4d as blue squares. A clear bandgap pressure tuning by more than 300% is evident. To verify the bandgap tuning, we follow another approach and estimate the bandgap (Fig. 4d, red) by averaging the tension in our infinite model (details in Supplementary info). For small pressures, both approaches agree within uncertainty. Yet at higher pressures slightly different scaling becomes evident, which we attribute the difference to the z-component of the tension, which is non-zero for the finite model.

Next, we investigate tunability of the defect mode. Upon applying 30 kPa pressure to a device with an initial tension of 0.01 N/m, the resonance frequency of the defect mode upshifts from 49.9 to 217.5 MHz (black stars Fig. 4d). Since the bandgap is smeared under pressure (Fig. 4c), it is important to

check the localization of the defect mode. Hence, we inspect a line cut through the centre of the device and plot the normalized mode shape vs. pressure in Fig. 4e. The shape remains virtually unchanged and the mode retains its localization. The effective mass also remains almost unchanged (inset Fig. 4e). Summarizing, we have shown a tunable speed of sound and realized an upshift of the defect mode resonance under pressure, whilst maintaining its localization. Such a more-than-four-fold frequency increase is unprecedented and remains elusive in any other phononic systems<sup>21-32</sup>.



**Figure 4 | Mechanically tunable graphene phononic crystal.** **a**, Band structure for initial tension values  $T_0 = 0.010$  N/m (red) and  $T_0 = 0.012$  N/m (orange). The entire out-of-plane branch scales strongly with tension. The position and width of the bandgap are equally tension-dependent. **b**, Speed of sound for the out-of-plane modes extracted from (a) vs. tension. **c**, Density of states calculated from the finite model as a function of pressure applied to the suspended PnC ( $T_0 = 0.010$  N/m). **d**, Pressure dependence of resonance frequency of the central defect mode (stars), of the bandgap from infinite model (red shaded), and of the bandgap extracted from the density of states (blue squares). The defect mode remains within the bandgap even at high pressures. **e**, Line cut for the spatial profile of the defect mode at different pressures (vertically offset for clarity). Even at large applied loads, the mode shape remains localized and the effective mass (inset) stays constant.

## Discussion and Outlook

So far, we have presented fabrication and modelling of a tunable graphene phononic crystal with a highly localized defect mode. It is now instructive to discuss experimental signatures of this mode. The extended non-bandgap-modes in our devices (Fig. 3e,h) are tightly spaced in frequency and mostly have spatial features too fine to be resolved via conventional diffraction-limited optics. At the same time, the extent of the defect mode is in the size of microns (Fig. 3f) and we therefore expect to detect the defect mode experimentally via interferometric read-out<sup>53,54</sup> (simulation in Supplementary Fig. 8). We also note that this mode has a non-zero net displacement and can be directly actuated via electrostatic drive. It will be straightforward to distinguish the defect mode from other modes by its localization in the centre of the device and its high quality factor. Indeed, the quality factor is defined by:  $Q = E_{\text{stored}}/E_{\text{diss}}$ , where  $E_{\text{diss}}$  is the dissipated energy per oscillation including all dissipation mechanisms, and the numerator depicts a mode's total energy. As the mode shape shows zero displacement near the clamping points, we expect strongly suppressed bending losses. Additionally, the phononic shield hinders radiation losses into the substrate, which become especially important at higher frequencies<sup>17</sup>. By applying pressure, we increase the stiffness of the resonator. This increases the energy stored in the system<sup>18</sup> and further enhances the quality factor. The demonstrated level of strain control in our system invites future studies on dissipation dilution via strain engineering following Ghadimi's work<sup>18</sup>. We also note that our results can be easily extended to the entire family of two-dimensional materials. Moreover, we are able to pattern these materials at the scale that is necessary for the operation of photonic crystals<sup>59</sup>, one could therefore confine light and mechanical motion in small spaces and strongly enhance phonon-photon interaction<sup>3,60</sup>. It will be challenging to achieve sufficient uniformity in the graphene membrane in order to generate a spatially uniform bandgap. Monolayer graphene is rather sensitive to surface corrugations<sup>39</sup>, so using thin multilayer would be a solution. The increased uniformity in multilayer graphene comes along with a decreased tunability, yet we expect more than 100% relative tuning for up to ~35 layers (Supplementary Fig. 9). For our graphene PnC, we do not expect to reach  $Q$ s comparable to SiN. Nevertheless we estimate  $m_{\text{eff}}$  of our defect mode to be at least eight orders of magnitude lower than in other 2D-SiN-PnCs<sup>16</sup>. This immensely increases the

measurement rate of quantum states  $\Gamma_{\text{meas}} \propto 1/m_{\text{eff}}$  and decreases thermomechanical noise<sup>16</sup>. The frequencies in our system are controlled by simply adjusting a gate voltage, and we expect the tuning to take place on time scales comparable to regular graphene resonators and therefore achieve tuning bandwidths in the high kHz regime<sup>61</sup>.

## Conclusion

In summary, we have fabricated and simulated a tunable PnC made from monolayer graphene. For an experimentally-informed honeycomb lattice structure, we find a robust vibrational bandgap in the MHz range. The bandgap persists for a finite-size system and we use it to localize a central defect mode and shield it from its surroundings. This defect mode shows a very small effective mass of 0.724 ag, orders of magnitude smaller compared to traditional PnCs. As our central result, we demonstrate a frequency upshift of the defect mode as well as the entire phononic system by more than 350% by applying an experimentally feasible pressure of 30 kPa to the system. While the bandgap smears out due to out-of-plane displacement perturbing the lattice, the defect mode stays within the bandgap and remains highly localized. We propose realistic experimental signatures of the defect mode and differentiation from other modes in the system. Overall, our design of a 2D-material based phononic crystal adds a new knob to dynamically and rapidly tune frequencies in a broad range of phononic applications. Lastly, our results invite future experiments as our approach allows adjustable coupling of a PnC to external systems and may lead to better understanding of the dissipation mechanisms in graphene.

## METHODS

**Sample synthesis.** Single layer graphene was synthesized on the copper substrate by chemical vapor deposition (CVD). The mixture of methane (5 sccm), hydrogen (10 sccm), and argon (5 sccm) was introduced into the CVD chamber, which was kept at 1035 °C. The growth time was 7 min. After the growth, graphene was transferred by the well-known fishing method onto a perforated SiN membrane, covered by a thin layer of Cr/Au (5 nm/35 nm) to electrically contact the graphene.

**Device patterning.** The patterning was carried out in a He-Ion microscope (Orion Nanofab). The holes for the PnC design were milled starting from the perimeter spiralling in towards the centre of the device.

Patterning parameters: Dwell time of 1.5 ms, pixel spacing of 1 nm at a beam current of 4-5 pA. Machine settings:  $2 \times 10^{-6}$  Torr He,  $U_{\text{acc}} = 30$  kV, UBIV = 34 kV, aperture  $2 \mu\text{m}$ . The Supplementary section I. provides more detail.

**Raman Spectroscopy.** Raman mapping was performed on a Horiba Xplora Raman spectrometer equipped with a xy-Piezo stage in backscattering configuration using a 100x (NA 0.9) objective and 532 nm excitation. Spectra were acquired with a laser power of 0.5 mW to avoid heating and an integration time of 3s per spectrum, and then fitted using a single Lorentzian to obtain the intensity (integrated area) and the position of the graphene 2D-Raman mode shown throughout Fig.1. Tension values were derived from the 2D-mode position following standard procedures from the literature, see Supplementary Information section IV.

**Simulations.** For the finite element modelling we use COMSOL Multiphysics (Version 5.5) and assume the following material parameters for monolayer graphene: Young's modulus  $E_{2D} = 1.0 \text{ TPa}^{38}$ , Poisson's ratio of  $\nu = 0.15$ , thickness of  $h = 0.335 \text{ nm}$  and a density of  $\rho = \frac{\rho_{2D}}{h} = 2260 \text{ kg/m}^3$ . The initial tension  $T_0 = 0.01 \text{ N/m}$  thus corresponds to an initial strain:  $\epsilon_0 = \frac{T_0}{E_{2D}} \approx 0.003\%$ . Both simulation models (finite and infinite) consist of two steps/studies. A first stationary study to calculate the redistributed tension is followed by eigenfrequencies studies. For the infinite model, we parametrize the wave vector  $k$ , implement it via periodic boundary conditions and directly obtain the band structure. For the infinite model, we calculate the eigenfrequencies and mode shapes, which then analysed in an external script carrying out mode-shape-analysis to assign a  $k$  value to each frequency  $f$ . More details in Supplementary section II.

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## Author contributions

J.N.K. conceived the idea. Suspended graphene devices were fabricated by K.W., S.K. and J.N.K. He-FIB patterning procedures were developed and carried out by K.H. and V.D. at HZB Berlin. S.H. acquired and analysed Raman spectroscopy data. Sample design and FEM-modelling was performed by J.N.K. with participation by K.W. J.N.K. and K.I.B. co-wrote the paper with input from all authors. K.I.B. supervised the project. All authors discussed the results.

## References

1. Kushwaha, M. S., Halevi, P., Dobrzynski, L. & Djafari-Rouhani, B. *Acoustic band structure of periodic elastic composites. Physical Review Letters* **71**, (1993).
2. Hopkins, P. E. *et al.* Reduction in the thermal conductivity of single crystalline silicon by phononic crystal patterning. *Nano Lett.* **11**, 107–112 (2011).
3. Maldovan, M. Sound and heat revolutions in phononics. *Nature* **503**, 209–217 (2013).
4. Mousavi, S. H., Khanikaev, A. B. & Wang, Z. Topologically protected elastic waves in phononic metamaterials. *Nat. Commun.* **6**, 1–7 (2015).
5. Balram, K. C., Davanço, M. I., Song, J. D. & Srinivasan, K. Coherent coupling between radiofrequency, optical and acoustic waves in piezo-optomechanical circuits. *Nat. Photonics* **10**, 346–352 (2016).
6. Olsson, R. H. & El-Kady, I. Microfabricated phononic crystal devices and applications. *Meas. Sci. Technol.* **20**, 13 (2009).
7. Khelif, A., Choujaa, A., Benchabane, S., Djafari-Rouhani, B. & Laude, V. Guiding and bending of acoustic waves in highly confined phononic crystal waveguides. *Appl. Phys. Lett.* **84**, 4400–4402 (2004).
8. Lin, S. C. S., Huang, T. J., Sun, J. H. & Wu, T. T. Gradient-index phononic crystals. *Phys. Rev. B - Condens. Matter Mater. Phys.* **79**, (2009).
9. Zhao, J., Bonello, B., Marchal, R. & Boyko, O. Beam path and focusing of flexural Lamb waves within phononic crystal-based acoustic lenses. *New J. Phys.* **16**, (2014).
10. Sigalas, M. M. & Economou, E. N. *Comment on acoustic band structure of periodic elastic composites. Physical Review Letters* **75**, (1995).
11. Benchabane, S., Khelif, A., Rauch, J. Y., Robert, L. & Laude, V. Evidence for

- complete surface wave band gap in a piezoelectric phononic crystal. *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.* **73**, (2006).
12. Khelif, A., Aoubiza, B., Mohammadi, S., Adibi, A. & Laude, V. Complete band gaps in two-dimensional phononic crystal slabs. *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.* **74**, (2006).
  13. Mohammadi, S., Eftekhari, A. A., Khelif, A., Hunt, W. D. & Adibi, A. Evidence of large high frequency complete phononic band gaps in silicon phononic crystal plates. *Appl. Phys. Lett.* **92**, 221905 (2008).
  14. Li, F., Liu, J. & Wu, Y. The investigation of point defect modes of phononic crystal for high Q resonance. *J. Appl. Phys.* **109**, 124907 (2011).
  15. Yu, P. L. *et al.* A phononic bandgap shield for high-Q membrane microresonators. *Appl. Phys. Lett.* **104**, 023510 (2014).
  16. Tsaturyan, Y., Barg, A., Polzik, E. S. & Schliesser, A. Ultracoherent nanomechanical resonators via soft clamping and dissipation dilution. *Nat. Nanotechnol.* **12**, 776–783 (2017).
  17. Ghadimi, A. H., Wilson, D. J. & Kippenberg, T. J. Radiation and Internal Loss Engineering of High-Stress Silicon Nitride Nanobeams. *Nano Lett.* **17**, 3501–3505 (2017).
  18. Engelsen, N. J. *et al.* Elastic Strain Engineering for Ultralow Mechanical Dissipation. in *International Conference on Optical MEMS and Nanophotonics 2018-July*, (2018).
  19. Imboden, M. & Mohanty, P. Dissipation in nanoelectromechanical systems. *Physics Reports* **534**, 89–146 (2014).
  20. Guo, J., Norte, R. & Gröblacher, S. Feedback Cooling of a Room Temperature Mechanical Oscillator close to its Motional Ground State. *Phys. Rev. Lett.* **123**, 223602 (2019).
  21. Delić, U. *et al.* Cooling of a levitated nanoparticle to the motional quantum ground state. *Science (80-. ).* **367**, 892–895 (2020).
  22. Thomas, R. A. *et al.* Entanglement between distant macroscopic mechanical and spin systems. *Nat. Phys.* 1–6 (2020). doi:10.1038/s41567-020-1031-5

23. Huang, H. *et al.* Topologically protected zero refraction of elastic waves in pseudospin-Hall phononic crystals. *Commun. Phys.* **3**, (2020).
24. Tian, Z. *et al.* Dispersion tuning and route reconfiguration of acoustic waves in valley topological phononic crystals. *Nat. Commun.* **11**, (2020).
25. Lin, S. C. S. & Huang, T. J. Tunable phononic crystals with anisotropic inclusions. *Phys. Rev. B - Condens. Matter Mater. Phys.* **83**, 174303 (2011).
26. Chen, Z. G. & Wu, Y. Tunable Topological Phononic Crystals. *Phys. Rev. Appl.* **5**, (2016).
27. Lin, S. C. S. & Huang, T. J. Tunable phononic crystals with anisotropic inclusions. *Phys. Rev. B - Condens. Matter Mater. Phys.* **83**, 174303 (2011).
28. Bian, Z., Peng, W. & Song, J. Thermal tuning of band structures in a one-dimensional phononic crystal. *J. Appl. Mech. Trans. ASME* **81**, (2014).
29. Jim, K. L., Leung, C. W., Lau, S. T., Choy, S. H. & Chan, H. L. W. Thermal tuning of phononic bandstructure in ferroelectric ceramic/epoxy phononic crystal. *Appl. Phys. Lett.* **94**, 193501 (2009).
30. Robillard, J. F. *et al.* Tunable magnetoelastic phononic crystals. *Appl. Phys. Lett.* **95**, 124104 (2009).
31. Bou Matar, O. *et al.* Band gap tunability of magneto-elastic phononic crystal. *J. Appl. Phys.* **111**, 054901 (2012).
32. Huang, Y., Zhang, C. L. & Chen, W. Q. Tuning band structures of two-dimensional phononic crystals with biasing fields. *J. Appl. Mech. Trans. ASME* **81**, 1–8 (2014).
33. Cha, J. & Daraio, C. Electrical tuning of elastic wave propagation in nanomechanical lattices at MHz frequencies. *Nature Nanotechnology* **13**, 1016–1020 (2018).
34. Jang, J. H., Ullal, C. K., Gorishnyy, T., Tsukruk, V. V. & Thomas, E. L. Mechanically tunable three-dimensional elastomeric network/air structures via interference lithography. *Nano Lett.* **6**, 740–743 (2006).
35. Hatanaka, D., Bachtold, A. & Yamaguchi, H. Electrostatically Induced Phononic Crystal. *Phys. Rev. Appl.* **11**, 1 (2019).
36. Midtvedt, D., Isacsson, A. & Croy, A. Nonlinear phononics using atomically thin

- membranes. *Nat. Commun.* **5**, (2014).
37. Wang, Y., Lee, J., Zheng, X. Q., Xie, Y. & Feng, P. X. L. Hexagonal Boron Nitride Phononic Crystal Waveguides. *ACS Photonics* **6**, 3225–3232 (2019).
  38. Lee, C., Wei, X., Kysar, J. W. & Hone, J. Measurement of the elastic properties and intrinsic strength of monolayer graphene. *Science (80-. )*. **321**, 385–388 (2008).
  39. Nicholl, R. J. T. *et al.* The effect of intrinsic crumpling on the mechanics of free-standing graphene. *Nat. Commun.* **6**, 8789 (2015).
  40. Singh, R., Nicholl, R. J. T., Bolotin, K. I. & Ghosh, S. Motion Transduction with Thermo-mechanically Squeezed Graphene Resonator Modes. *Nano Lett.* **18**, 6719–6724 (2018).
  41. Bolotin, K. I. *et al.* Ultrahigh electron mobility in suspended graphene. *Solid State Commun.* **146**, 351–355 (2008).
  42. Deinhart, V. fib-o-mat. Available at: <https://pypi.org/project/fibomat/>.
  43. Lemme, M. C. *et al.* Etching of graphene devices with a helium ion beam. *ACS Nano* **3**, 2674–2676 (2009).
  44. Archanjo, B. S. *et al.* Graphene nanoribbon superlattices fabricated via He ion lithography. *Appl. Phys. Lett.* **104**, 193114 (2014).
  45. Fox, D. *et al.* Helium ion microscopy of graphene: Beam damage, image quality and edge contrast. *Nanotechnology* **24**, 335702 (2013).
  46. Mohammadi, S. *et al.* Complete phononic bandgaps and bandgap maps in two-dimensional silicon phononic crystal plates. *Electron. Lett.* **43**, 898–899 (2007).
  47. Capelle, T., Tsaturyan, Y., Barg, A. & Schliesser, A. Polarimetric analysis of stress anisotropy in nanomechanical silicon nitride resonators. *Appl. Phys. Lett.* **110**, 181106 (2017).
  48. Mohiuddin, T. M. G. *et al.* Uniaxial strain in graphene by Raman spectroscopy: G peak splitting, Grüneisen parameters, and sample orientation. *Phys. Rev. B - Condens. Matter Mater. Phys.* **79**, 1–8 (2009).
  49. Mueller, N. S. *et al.* Evaluating arbitrary strain configurations and doping in graphene with Raman spectroscopy. *2D Mater.* **5**, (2018).

50. Chen, C. *et al.* Performance of monolayer graphene nanomechanical resonators with electrical readout. *Nat. Nanotechnol.* **4**, 861–867 (2009).
51. Tsaturyan, Y. Ultracoherent soft-clamped mechanical resonators for quantum cavity optomechanics. (University of Copenhagen, 2019).
52. Alemán, B. *et al.* Transfer-free batch fabrication of large- Area suspended graphene membranes. *ACS Nano* **4**, 4762–4768 (2010).
53. Van Der Zande, A. M. *et al.* Large-scale arrays of single-layer graphene resonators. *Nano Lett.* **10**, 4869–4873 (2010).
54. Bunch, J. S. *et al.* Electromechanical resonators from graphene sheets. *Science (80-. )*. **315**, 490–493 (2007).
55. Asmar, N. H. *Partial Differential Equations with Fourier Series and Boundary Value Problems. Foreign language books* (2004).
56. Waitz, R., Nöbner, S., Hertkorn, M., Schecker, O. & Scheer, E. Mode shape and dispersion relation of bending waves in thin silicon membranes. *Phys. Rev. B - Condens. Matter Mater. Phys.* **85**, (2012).
57. Vasseur, J. O. *et al.* Experimental and theoretical evidence for the existence of absolute acoustic band gaps in two-dimensional solid phononic crystals. *Phys. Rev. Lett.* **86**, 3012–3015 (2001).
58. Hsiao, F. L. *et al.* Complete band gaps and deaf bands of triangular and honeycomb water-steel phononic crystals. *J. Appl. Phys.* **101**, 44903 (2007).
59. Thongrattanasiri, S., Koppens, F. H. L. & García De Abajo, F. J. Complete optical absorption in periodically patterned graphene. *Phys. Rev. Lett.* **108**, 047401 (2012).
60. Eichenfield, M., Chan, J., Camacho, R. M., Vahala, K. J. & Painter, O. Optomechanical crystals. *Nature* **462**, 78–82 (2009).
61. Chen, C. *et al.* Graphene mechanical oscillators with tunable frequency. *Nat Nanotechnol* **8**, 923–927 (2013).

# Figures

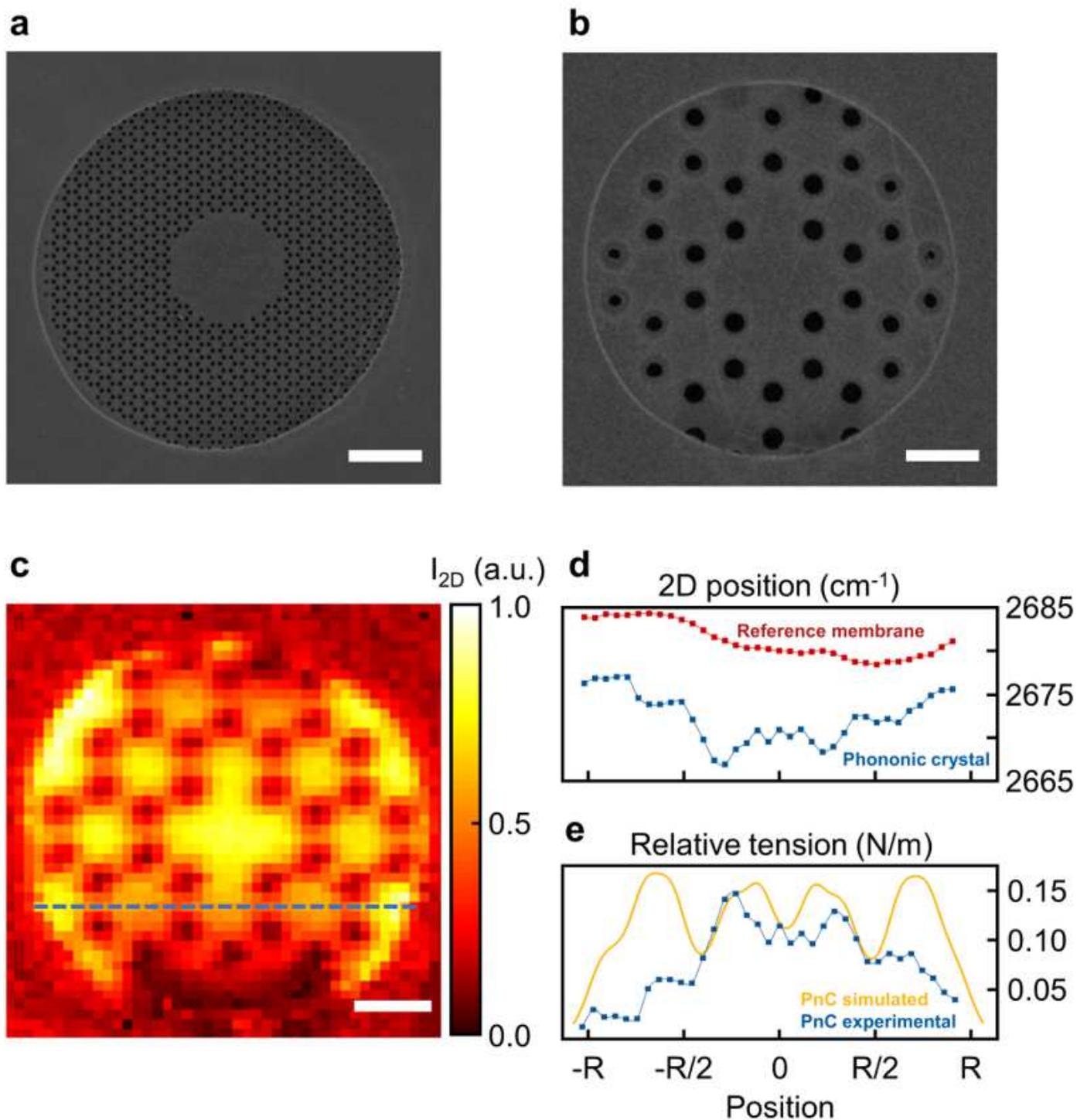
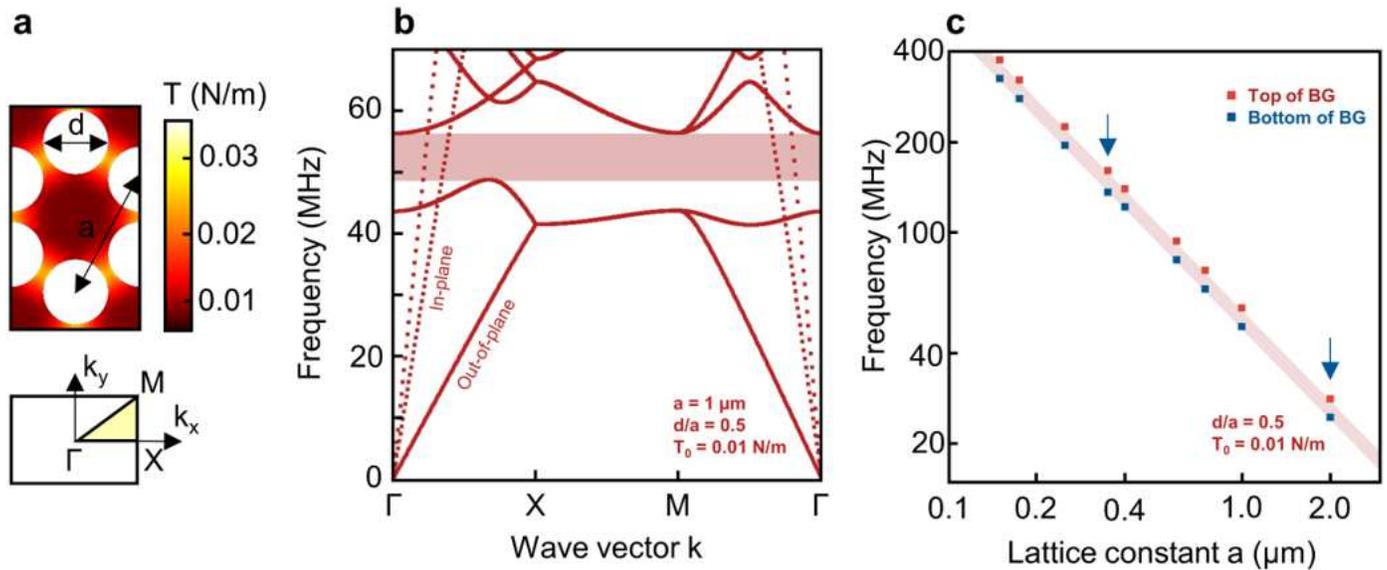


Figure 1

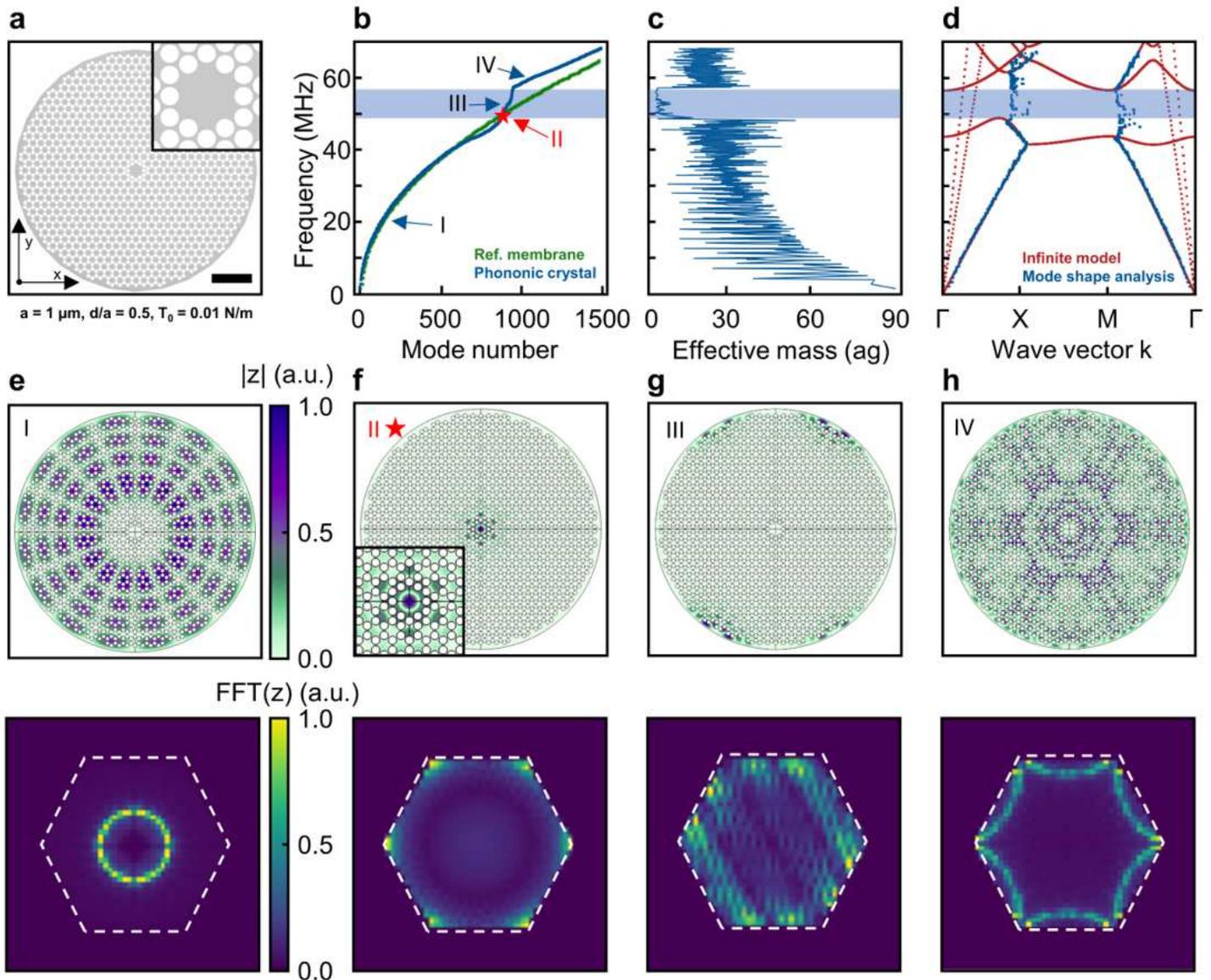
Graphene phononic crystals and tension redistribution. a,b, Helium ion micrographs of prototype monolayer graphene phononic crystal devices with lattice constants 350 nm and 2  $\mu\text{m}$ , respectively. The phononic pattern, a honeycomb lattice of holes with a defect in its centre allows us to localize a

vibrational defect mode. Scale bar length is  $2\ \mu\text{m}$ . c, Intensity map of the Raman-active 2D mode of graphene for the device shown in (b). The periodic pattern is clearly visible. d, Raman 2D-mode position along a line cut (dashed line in (c)) for a PnC (blue) and reference membrane (red). The PnC shows a periodic variations of much larger amplitude compared to the fluctuation in the reference sample. e, Comparison of the relative tension extracted from Raman measurements (blue) to the simulated tension distribution (yellow) confirming the redistribution of tension upon patterning. The simulation includes spatial broadening due to the finite size of the laser spot.



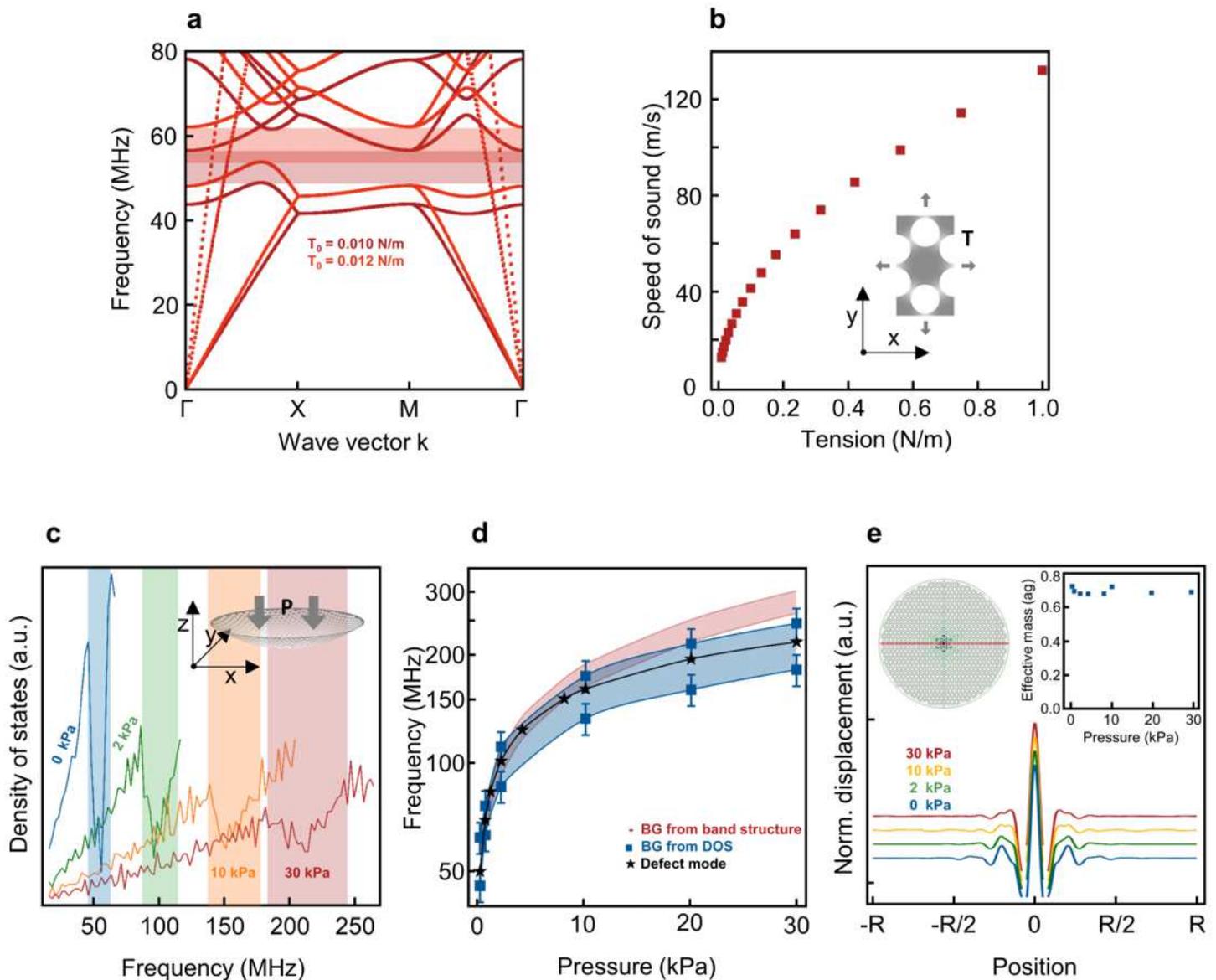
**Figure 2**

Band structure calculations of an infinite graphene phononic crystal. a, Unit cell of the honeycomb lattice with redistributed tension (top) and the corresponding first Brillouin zone (bottom). b, Phononic band structure for the unit cell shown in (a). In-plane modes are shown as dashed lines, out-of-plane modes as solid lines, and the corresponding quasi-bandgap region as red shaded area. c, Top (red) and bottom (blue) of the bandgap vs. lattice constant. The blue arrows indicate the lattice constant of the devices from Fig. 1a,b.



**Figure 3**

Finite size model of a graphene phononic crystal. a, Device geometry for the finite system simulations (scale bar is  $5 \mu\text{m}$ ). A central “defect” region is designed to localize one vibrational mode and decouple it from its environment b, The first 1500 simulated eigenfrequencies vs. mode number for a phononic crystal device (blue) and a circular membrane without patterning (green). The bandgap region from the infinite model is shown in blue. c, Effective mass for each mode. The modes within the bandgap (blue) show a more than 100-fold decrease in effective mass compared to the fundamental mode. d, Band structure calculated from the finite model via mode-shape analysis (blue) along with the band structure from the infinite model (red). The low energy acoustic branches fit well, and the bandgap regions coincides with the simulated results from the infinite model (red). e-h Exemplary mode shapes in real (top) and reciprocal space (bottom) for: (e) a mode below the bandgap (I), (f) the defect mode (II), (g) another highly localized mode in the bandgap (III) and (h) a mode above the gap (IV).



**Figure 4**

Mechanically tunable graphene phononic crystal. a, Band structure for initial tension values  $T_0 = 0.010$  N/m (red) and  $T_0 = 0.012$  N/m (orange). The entire out-of-plane branch scales strongly with tension. The position and width of the bandgap are equally tension-dependent. b, Speed of sound for the out-of-plane modes extracted from (a) vs. tension. c, Density of states calculated from the finite model as a function of pressure applied to the suspended PnC ( $T_0 = 0.010$  N/m). d, Pressure dependence of resonance frequency of the central defect mode (stars), of the bandgap from infinite model (red shaded), and of the bandgap extracted from the density of states (blue squares). The defect mode remains within the bandgap even at high pressures. e, Line cut for the spatial profile of the defect mode at different pressures (vertically offset for clarity). Even at large applied loads, the mode shape remains localized and the effective mass (inset) stays constant.

## Supplementary Files

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