MULTIMODE BLACK PHOSPHORUS NANOMECHANICAL RESONATORS WITH INTRINSIC MECHANICAL ANISOTROPY AND ELECTRICAL TUNABILITY

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ABSTRACT

We report on the design and experimental demonstration of the first electrically tunable multimode resonant nano-electromechanical systems (NEMS) enabled by thin black phosphorus (P) crystals with intrinsic strong mechanical anisotropy. Combined with finite element modeling, we show that mechanical anisotropy in the black P crystal is clearly manifested in the mode sequence, frequency spacing, and mode shapes in black P NEMS resonators, which differ from those of isotropic resonators. We further experimentally demonstrate a multimode black P NEMS resonator. The multimode resonances measured and spatially mapped in black P nanoresonators, show good agreement with the design, and provide experimental evidences for the effects of the crystal's intrinsic mechanical anisotropy on the multimode resonances. We also demonstrate the ability of tuning the multimode responses by electrical gating. The findings in this study shall help open new possibilities for future realization of on-chip electromechanical functions that can efficiently utilize the mechanical anisotropy at truly nanoscale.

INTRODUCTION

Black P has rapidly emerged as a new elemental, direct-bandgap, atomic-layer semiconducting crystal. In addition to its attractive electronic and optical properties, it is also particularly interesting for enabling new two-dimensional (2D)NEMS devices owing to its unique in-plane mechanical anisotropy (Fig. 1) that is unavailable in other atomic-layer crystals demonstrated to date [1]. In particular, it is predicted that its in-plane elastic moduli in orthogonal directions can differ by nearly 500% [2]. In comparison, the Young's modulus of silicon, the most-studied anisotropic MEMS material, only varies up to 50% with crystal orientation [3].

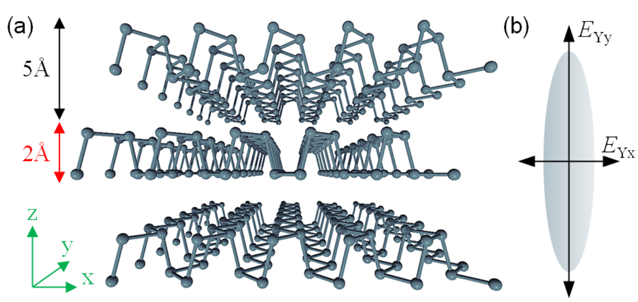


Figure 1: Crystal structure and mechanical anisotropy of black phosphorus (P), a new atomic-layer material. (a) Illustration of black P crystal consisting of vertically-stacked corrugated layers of P atoms. (b) Anisotropic in-plane elastic moduli of black P along armchair (E_{Yx}) and zigzag (E_{Yy}) directions.

To date, elastic properties of 2D materials are measured using nanoindentation method, in which a sharp indentation tip deflects a suspended 2D flake of known geometry and clamping conditions, and the Young's modulus is inferred from the load-deflection curves [4]. For black P, however, the strong anisotropy in elastic properties makes it less straightforward to directly apply the nanoindentation method. Theoretical predictions for its Young's moduli [2,5-7] (summarized in Table 1) range from 15.1GPa to 44.0GPa in armchair axis (E_{Yx}) and from 49.9GPa to 166.0GPa along zigzag axis (E_{Yy}); which present new complications and challenges toward deterministically designing devices with specific resonance responses.

Table 1: Theoretical calculations of Young's moduli of black P.

Work	E_{Yy} (GPa)	E_{Yx} (GPa)	E_{Yy}/E_{Yx}
Appl. Phys. Lett. 2014 [2]	*166.0*	*44.0*	3.77
arXiv 2014 [5]	*92.7*	*44.0*	2.11
Sci. Rep. 2014 [6]	*49.9*	*15.1*	3.30
J. Phys. D. 2014 [7]	*106.4*	*41.3*	2.58

DESIGN AND EXPERIMENT PROCEDURES

Design of Black Phosphorus (P) NEMS Resonators

A close examination of the results in the literature shows that while the absolute values in E_{Yx} and E_{Yy} differ significantly from study to study, the ratio of the elastic moduli in the two crystal orientations (E_{Yy}/E_{Yx}), a measure of the degree of anisotropy, differs by a factor of less than 2 among these studies (from 2.11 to 3.77). We therefore use E_{Yy}/E_{Yx} as a design parameter in this study instead of sweeping two independent parameters (E_{Yy} and E_{Yx}). In addition, we present the simulated frequency spacing results in form of frequency ratios (between a particular mode and the fundamental), which only depends on the ratio E_{Yy}/E_{Yx} and is insensitive to device diameter (d), thickness (t) and the absolute values of E_{Yy} and E_{Yx}.

We use finite element modeling (FEM, via COMSOL) to calculate the expected multimode responses of circular black P drumhead resonators. To focus on the effects of mechanical anisotropy, we fix most device parameters (which do not affect the frequency ratios), and only vary E_{Yy} (while fixing E_{Yx}=44.0GPa) in order to sweep E_{Yy}/E_{Yx} values. Table 2 shows the simulation parameters used in the device design. For comparison, we also show results from an isotropic case in which $E_{Yy}=E_{Yx}$=44.0GPa.

Table 2: FEM (COMSOL) simulation parameters of multimode black phosphorus (P) drumhead resonators.

Description	Expression	Value
Device Diameter	d	10μm
Flake Thickness	t	95nm
Material Density	ρ	2690kg/m^3
Poisson's Ratio	ν	0.17 [2]
Elastic Modulus along Armchair Axis	E_{Yx}	44.0GPa [2,5]
Range of Elastic Modulus along Zigzag Axis	E_{Yy}	44.0GPa to 176.0GPa
Range of Elastic Moduli Ratio	E_{Yy}/E_{Yx}	1, 2 to 4

Figure 2 shows the results from the FEM simulations. In presenting the results, we label every mode by using (m,n) to describe its mode index and shape, where m and n are the numbers of antinodes (*i.e.*, 1 + number of nodal lines) along the armchair and zigzag crystal orientations, respectively.

There are two important reasons for using this naming system, which are associated with the simulation results: (i) the mode sequence and frequency spacing clearly depends on the elastic moduli ratio E_{Yy}/E_{Yx}, thus the modes cannot be simply identified using their sequence in the frequency spectrum or frequency ratios; instead, for each mode the mode shape itself is a good identifier; (ii) the mode shapes of a black P NEMS resonator clearly differ from those of an isotropic crystalline counterpart, therefore the convention used in describing the mode shapes for a circular disk resonator cannot be applied to such anisotropic resonators.

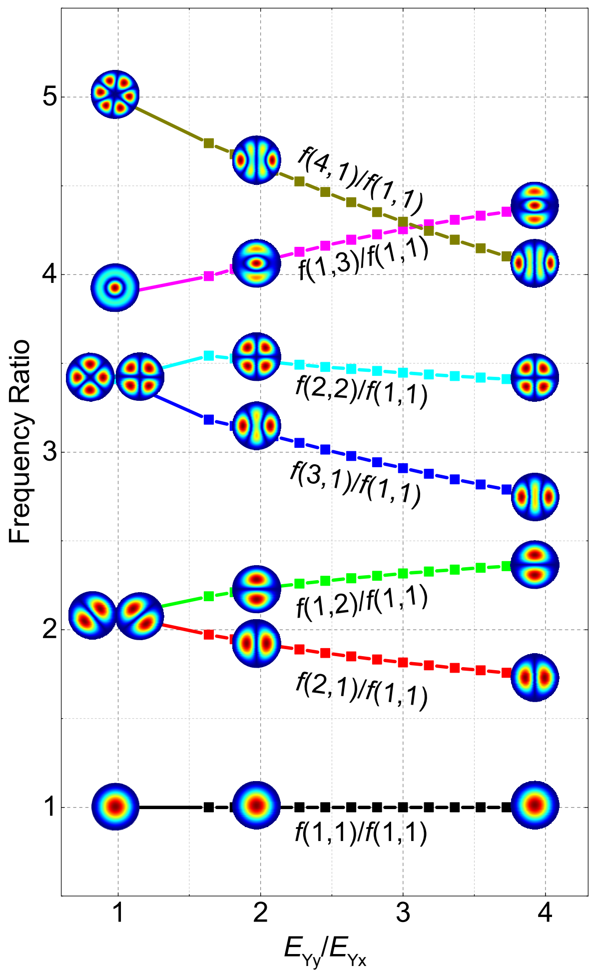


Figure 2: Effect of mechanical anisotropy on the multimode resonances in black P resonators (FEM simulation results). The frequency ratios of every mode to the fundamental f(m,n)/f(1,1) are plotted as functions of E_{Yy}/E_{Yx}, together with the simulated mode shapes and their evolutions with E_{Yy}/E_{Yx}. Note that the mode sequence depends on the E_{Yy}/E_{Yx} value.

Based on the above observations and considerations, it is important that one measures both the resonance frequencies and mode shapes in black P multimode resonators in order to correctly characterize such devices and thus verify the device designs. Therefore the device structures and experimental systems shall facilitate spectral and spatially resolved multimode resonance measurements.

Device Fabrication

Black P NEMS resonators are fabricated using a dry-transfer method that minimizes device exposure to the ambient or contaminations [8] to preserve the high quality of the black P crystals, as illustrated in Fig. 3. AFM measurements (for determining flake thickness) are performed after all electrical and optical measurements, again to minimize the exposure to the ambient prior to the resonance measurements. The resulting device (Fig. 4b) has a 95nm-thick (Fig. 4c) black P crystal suspended over a 10μm-diameter, 290nm-deep circular microtrench (fully clamped), and is contacted with Au/Cr electrodes. We choose the circular drumhead structure in order to best reveal the effect of intrinsic mechanical anisotropy of black P crystal on the multimode resonances [1].

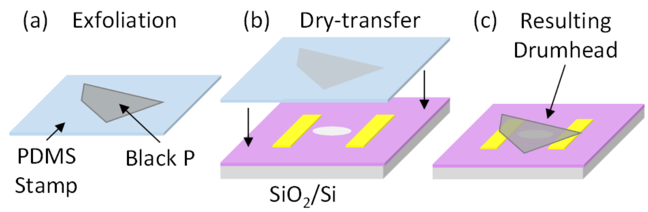


Figure 3: Schematic illustration of device fabrication. (a) Exfoliation of black P onto PDMS stamp. (b) Dry transfer on to pre-patterned substrate. (c) The suspended device structure.

Measurement System

We drive the resonator electrostatically and detect the device motion with a laser interferometry, which facilitates spatial mapping of the resonance mode shapes through scanning spectromicroscopy measurements [9].

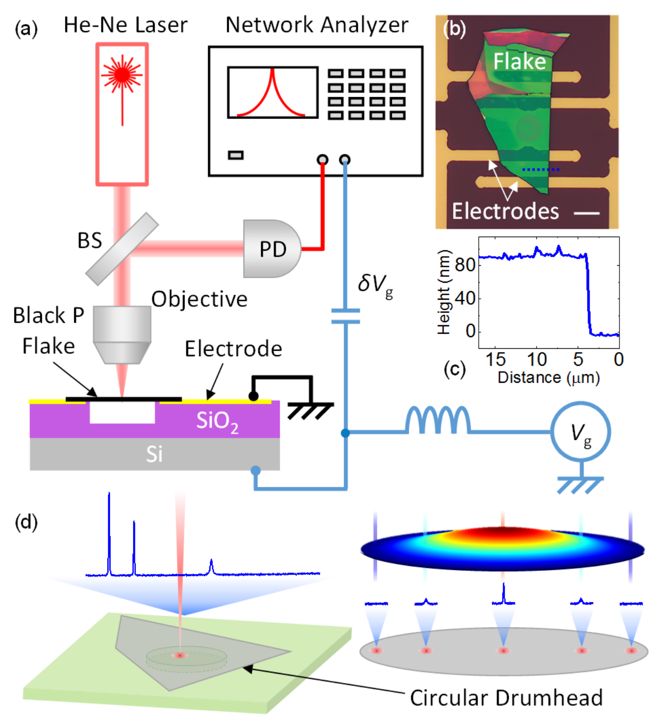


Figure 4: Measurement system and device structure. (a) Schematic illustration of custom-built laser interferometry system. Flexural vibrations of the Black P NEMS resonator are electrically

excited and interferometrically detected using a He-Ne laser. The chip is positioned in a vacuum chamber on a motorized XYZ stage (not shown). PD: Photodetector. BS: Beam splitter. (b) Microscopic image showing structural details of a 10μm-diameter circular black P drumhead resonator with electrodes. Scale bar: 10μm. (c) AFM height trace is measured along the dotted line in (b), showing a ~95nm flake thickness in the suspended region. (d) Schematic of the microspectroscopy measurement (a complete spectrum is obtained from one location), and spectromicroscopy measurement, where the laser scans across the device surface and the measured responses from all the locations are collected to reconstruct the mode shape.

Figure 4 schematically illustrates the laser interferometry system with high displacement sensitivity that is capable of detecting high-order/multiple mechanical resonances of 2D NEMS resonators. The flexural vibrations are actuated by applying an AC drive on top of a DC polarization (V_g+δV_g) at a back gate, and detected by a He-Ne laser focused on the suspended area. The multiple reflections from the flake/vacuum and vacuum/Si interfaces create optical interference that is modulated by the device motion and read out by a low-noise photodetector. The device is measured in moderate vacuum (~10mTorr, vacuum chamber not shown) and the on-device laser power is ~600μW to minimize heating or degradation of the black P crystal.

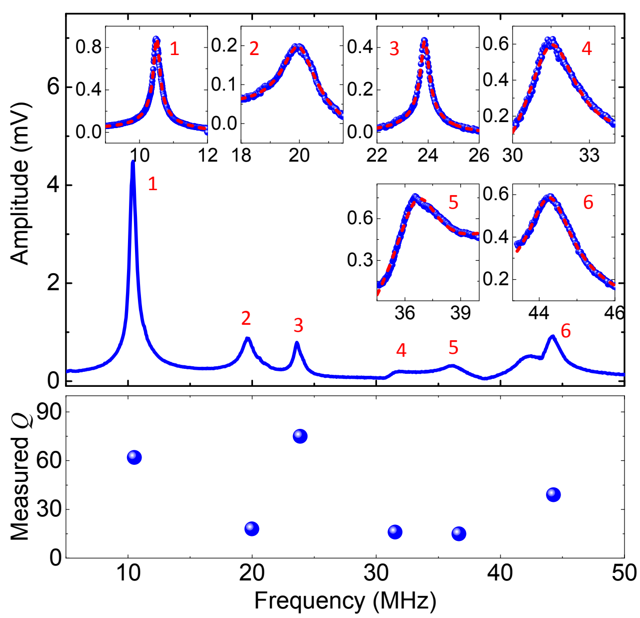


Figure 5: Measured multimode resonance characteristics of the 10μm black P drumhead resonator. A total of 6 flexural modes up to 45MHz (HF/VHF bands) are detected in vacuum at room temperature, and the insets show zoom-in measurements of each mode and fitting to a finite-Q simple harmonic resonator model to extract both resonance frequency (f) and quality (Q) factor. The bottom plot shows Q vs. f for all the 6 modes.

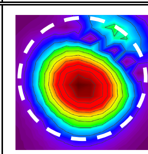
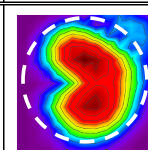
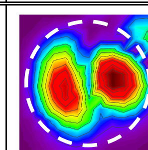
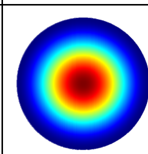
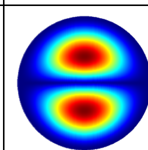
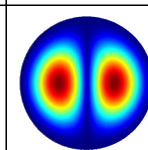
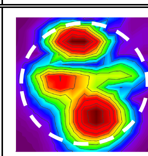
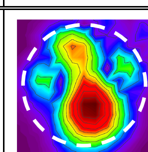
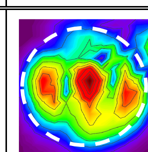
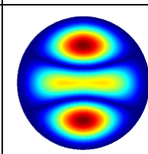
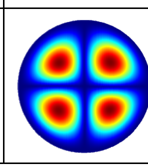
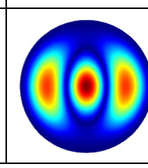
The mode shapes are spatially mapped by employing the scanning spectromicroscopy technique [9], as illustrated in Fig. 4d. The vacuum chamber is mounted on a programed XYZ translational stage (not shown). We first define a 16μm×16μm mapping area X_i∈(-8μm, 8μm), Y_j∈(-8μm, 8μm) that encloses the 10μm-diameter circular suspended region, with the origin (0,0) located at the device center. The steps along both X and Y directions are set to be 1μm. At each position (X_i, Y_j), we excite one particular mechanical resonance $f(m,n)$ and measure the frequency response. The resulting signal amplitude (A_{ij}) is used to determine the device displacement at position (X_i, Y_j) for the given mode, and is then plotted in a 2D color map (A_{ij} as a function of X_i, Y_j) for visualizing the corresponding mode shape.

RESULTS AND DISCUSSIONS

Multimode Resonances

The measured frequency response over a wide frequency range (from 1MHz to 50MHz) is shown in Fig. 5. We identify 6 resonant modes which we investigate in detail. Measurement with a zoomed-in narrower frequency range is then performed for each mode; and the data are fitted to a damped driven harmonic resonator model (Fig. 5 insets). From the fitting results we extract the resonance frequencies (f) and qualify (Q) factors for each mode. These modes have frequency range of 10 to 45MHz (in HF/VHF bands), and Q factors from 15 to 75.

Table 3: Comparison between spatially-mapped and simulated mode shapes of the multimode responses.

Mode (*m*,*n*)	**(1,1)**	**(2,1)**	**(1,2)**
Experiment			
Simulation			
Mode (*m*,*n*)	**(3,1)**	**(2,2)**	**(1,3)**
Experiment			
Simulation			

These 6 modes are then carefully mapped using the spectromicroscopy techniques to resolve the mode shapes. The results are summarized in Table 3. From the measurements we clearly identify each measured mode shape with (m,n), finding very good agreement with simulations. Specifically, we observe clear difference between the measured mode shapes and those expected from a circular drumhead resonator made of isotropic material, which is characterized by nodal circles and nodal diameters. In contrast, in black P resonators, except for the few lowest modes, nodal lines do not take the form of circles or diameters, but as multiple curves that does not necessarily cross the center of the circle. The spatially resolved measurements clearly show that the intrinsic mechanical anisotropy is manifested in the multimode resonant responses in black P NEMS.

Frequency Spacing and Scaling

We further examine the frequency spacing and scaling for multimode black P resonators, with the results from both measurement and simulation shown in Table 4. We represent the frequency spacing by using the $f(m,n)/f(1,1)$ ratio, the frequency of each mode normalized to the fundamental resonance.

The experimental values show good agreement with the predictions from the FEM simulations. Specifically, the frequency ratios from all the 6 modes (in the form of $f(m,n)/f(1,1)$) all fall within the expected range as in the device design (see Table 4). This shows that the measured elastic moduli ratio in black P crystal, E_{Yy}/E_{Yx}, resides between the values 2 and 4, as predicted by the theoretical calculations (listed in Table 1).

Table 4: Comparison between spatially-mapped and simulated multimode frequency ratios.

$f(m,n)$/f(1,1)	E_{Yy}/E_{Yx}=2	Experiment	E_{Yy}/E_{Yx}=4
$f(1,1)/f(1,1)$	*1.000*	*1.000*	*1.000*
$f(2,1)/f(1,1)$	*1.922*	*1.896*	*1.739*
$f(1,2)/f(1,1)$	*2.231*	*2.271*	*2.371*
$f(3,1)/f(1,1)$	*3.109*	*2.999*	*2.749*
$f(2,2)/f(1,1)$	*3.512*	*3.488*	*3.399*
$f(1,3)/f(1,1)$	*4.071*	*4.213*	*4.384*

Tuning of Multimode Resonances

We further explore electrostatic gate tuning of the multimode resonances in the black P NEMS resonator. We measure the multimode resonances of the device between 1MHz and 50MHz, and perform the frequency sweep for each DC gate voltage (V_g) from -30V to 30V (with 1V step). Figure 6 shows that all the resonant modes exhibit clear gate tuning effect. Note that the measurement is taken with the laser position optimized for detecting the fundamental mode (1,1), and thus the signal for the higher modes are less visible using the same scale.

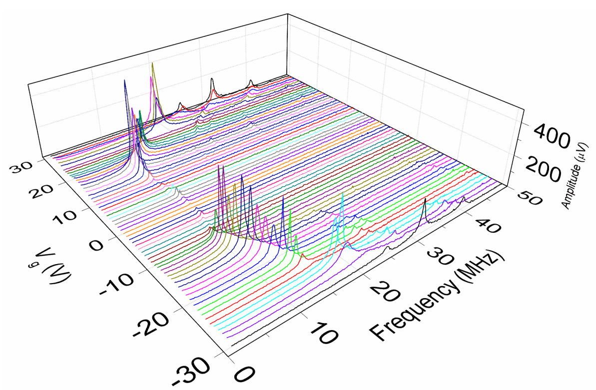


Figure 6: Measured gate tuning of the black P drumhead NEMS resonator, shown in a 3D-plot of multimode frequency response curves under V_g from -30V to+30V.

From the data in Fig. 6, we clearly observe two competing effects. In low V_g range ($|V_g|$<15V), frequency decreases as V_g increases, as the device is dominated by capacitive softening effect [10]; at larger $|V_g|$ values, electrostatic tensioning effect [11] becomes clearly visible and dominant as the resonance frequency increases with the magnitude of V_g. We find that f can be tuned up by 163% (from 9.45 MHz to 24.9MHz for the fundamental mode) in this gate voltage range.

CONCLUSIONS

In summary, we have designed and fabricated the first multimode circular drumhead-shaped black P NEMS resonator. Using spatial mapping techniques in addition to the conventional spectral measurements, we successfully identify 6 resonant modes by directly visualizing their mode shapes. The measurements confirm that the black P resonator exhibit clear effects from mechanical anisotropy upon its multimode resonances, and show very good agreement with the expected device performance from the design. By varying the gate voltage, we observe strong tuning of the multimode resonances. The results demonstrate the power of using spatially-resolved multimode measurements to determine the device's material properties, and elucidate the mode sequence in black P NEMS resonators, which can lead to new resonant sensors or transducers that leverage the unique mechanical anisotropy in black P.

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