

# PHONONIC FREQUENCY COMB GENERATION VIA 1:1 MODE COUPLING IN MoS<sub>2</sub> 2D NANOELECTROMECHANICAL RESONATORS

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## ABSTRACT

We report on the experimental demonstration of phononic frequency comb (PFC) generation in atomically thin molybdenum disulfide (MoS<sub>2</sub>) resonant nanoelectromechanical systems (NEMS) vibrating near ~50 MHz in the very high frequency (VHF) band. PFC patterns are generated by tuning two close-by resonances with a gate voltage ( $V_g$ ), and driving them at an anticrossing point, where the two modes are coupled through 1:1 internal resonance. The PFC characteristics are tunable by varying RF driving voltage ( $v_{\text{drv}}$ ) and  $V_g$ . We find a threshold of PFC generation at  $v_{\text{drv}} = 550\text{mV}$  and  $V_g = -6.4\text{V}$ , with relatively wide comb teeth spacing around 2.44 to 2.65MHz. At  $V_g = -7\text{V}$ , the comb teeth spacing is reduced to 0.63MHz, exhibiting strong tunability of PFC.

## KEYWORDS

Phononic frequency comb (PFC), internal resonance, mode coupling, frequency tuning, nanoelectromechanical systems (NEMS), two-dimensional (2D) materials.

## INTRODUCTION

Phononic frequency combs (PFCs) are attractive and intriguing as new devices for mechanical frequency conversion, sensing, and precision measurement, following the immense success and impact of their pioneering optical counterparts (OFCs, Nobel Prize in Physics, 2005). A few PFCs have been demonstrated lately by coupling two or more mechanical resonance modes and transferring vibration energy back and forth among coupled modes. To achieve strong coupling between the resonance modes, special conditions such as 1:2 and 1:3 internal resonances [1,2], where nonlinear dynamics facilitates coherent energy transfer between coupled modes, have been employed. Internal resonances can be established when the frequency ratio of two modes is made close to an integer number [1,2,3]. Accordingly, for building PFC and achieving mode coupling conditions, it is desirable to employ devices with wide frequency tunability and rich nonlinearity.

The advent of two-dimensional (2D) materials such as graphene and molybdenum disulfide (MoS<sub>2</sub>) has enabled the creation of atomically thin NEMS resonators [4,5,6]. Thanks to their ultrathin suspended structures, these devices are operating in the membrane regime [4,5,6], where their resonance frequencies depend on strain, enabling ultrawide range of frequency tuning, up to  $\Delta f/f \sim 430\%$  [6]. Frequency tuning in 2D NEMS has been widely achieved via electrostatic forces induced by a gate voltage  $V_g$ , and the resonance frequency can be adjusted to lower or higher values through capacitive softening (at low  $|V_g|$ ) or stiffening (at high  $|V_g|$ , stretching dominates) effects

[5,6]. The exceptional frequency tunability of 2D NEMS resonators [5,6] shall facilitate accessing internal resonance conditions and mode coupling, thus they are well suited for building novel, tunable PFCs. In this work, we employ an electrostatically tunable four-layer (4L) MoS<sub>2</sub> NEMS resonator and demonstrate PFC generation using coupled resonance modes via 1:1 internal resonance. We attain the 1:1 internal resonance by tuning the resonance frequencies of two modes that are close-by initially. By introducing a single-frequency driving signal around the anticrossing point of coupled modes, a PFC is generated, and the intriguing characteristics and tunability of PFC patterns are investigated under different driving conditions.

## DEVICE FABRICATION

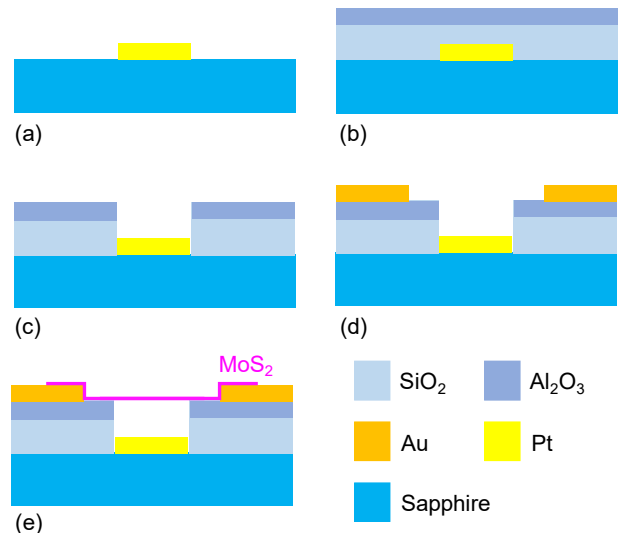


Figure 1: Fabrication process of the MoS<sub>2</sub> 2D NEMS resonator. (a) Creation of local gates on a 4-inch sapphire substrate. (b) Deposition of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> dielectric layers on the sapphire substrate with local gates. (c) Patterning of microtrenches using RIE. (d) Deposition of source and drain electrodes. (e) Dry transfer of MoS<sub>2</sub> flakes onto prepatterned cavities.

We design and fabricate electrostatically tunable circular drumhead 2D NEMS resonators with local gates [7]. A 4-inch insulating sapphire wafer is employed as a substrate to minimize undesirable RF signal crosstalk. First, we pattern local gates (23nm Pt on 2nm Ti) for electrostatic control (Fig. 1a). Next, 300nm SiO<sub>2</sub> and 20nm Al<sub>2</sub>O<sub>3</sub> dielectric layers are grown on top of the local gates using plasma enhanced chemical vapor deposition (PECVD) and atomic layer deposition (ALD), respectively, to ensure good electrical insulation between the local gates and NEMS devices (Fig. 1b). Afterwards,

we pattern microcavities using reactive ion etching (RIE) (Fig. 1c). Subsequently, we deposit source and drain electrodes (30nm Au on 5nm Ti) and contact pads (200nm Au on 5nm Ti) (Fig. 1d). Finally, we dry transfer MoS<sub>2</sub> flakes onto the microtrenches with various diameters (Fig. 1e) to make circular drumhead 2D NEMS resonators.

## RESULTS AND DISCUSSIONS

After device fabrication, the number of layers of the suspended MoS<sub>2</sub> flake is confirmed by Raman and photoluminescence (PL) measurements. Figure 2 shows measured Raman and PL spectra obtained from the suspended region of the device. The device shows a separation of  $E_{1g}^1$  and  $A_{1g}$  Raman peaks of 24.3cm<sup>-1</sup>, indicating clear signatures of four-layer (4L) MoS<sub>2</sub> (Fig. 2a) [8]. The measured PL spectrum exhibits pronounced indirect band transition around ~900nm (Fig. 2b) [9], also verifying the suspended MoS<sub>2</sub> membrane is 4L. We also observe a sharp PL peak at ~694nm, which is attributed to emission from the sapphire substrate.

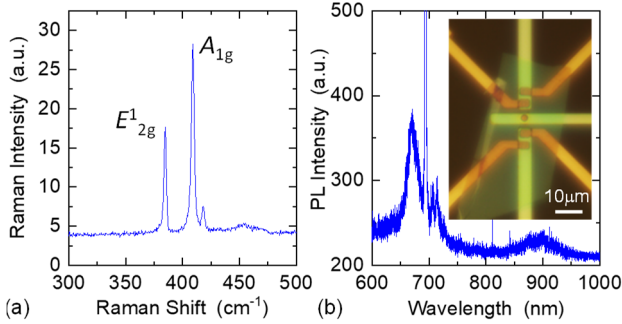


Figure 2: Raman and photoluminescence (PL) signatures of a 4L MoS<sub>2</sub> NEMS resonator. Measured (a) Raman and (b) PL spectra from the suspended region, verifying 4L MoS<sub>2</sub>. Inset: optical microscope image of the device with a diameter of 2μm and thickness of 2.8nm.

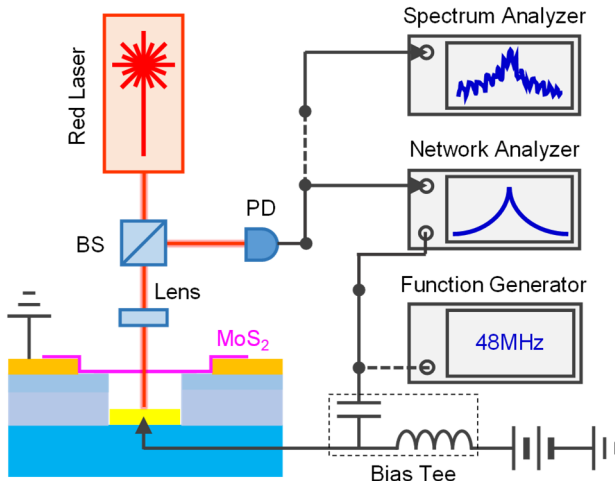


Figure 3: Schematic of laser optical interferometry system. A 633nm red laser light is focused on a suspended MoS<sub>2</sub> membrane. Three measurement configurations are available to detect undriven thermomechanical noise, electrostatically driven resonance, and PFC.

We employ a customized laser interferometry system to characterize the 4L MoS<sub>2</sub> NEMS resonator. A 633nm He-Ne laser is focused onto the suspended MoS<sub>2</sub> circular drumhead using a 50× microscope objective. To avoid any

excess laser heating, the laser power is limited to below 300μW on the device. The reflected light modulated by the resonance motion is converted to an electronic signal using a photodetector and measured by using a spectrum analyzer for undriven thermomechanical noise. A network analyzer is used to measure the electrostatically driven resonances. PFC patterns are generated by applying a single-frequency driving signal from a function generator, and measured by using the spectrum analyzer. All the measurements are performed with the MoS<sub>2</sub> device in moderate vacuum ( $p \approx 10$ mTorr) and at room temperature ( $T \approx 300$ K).

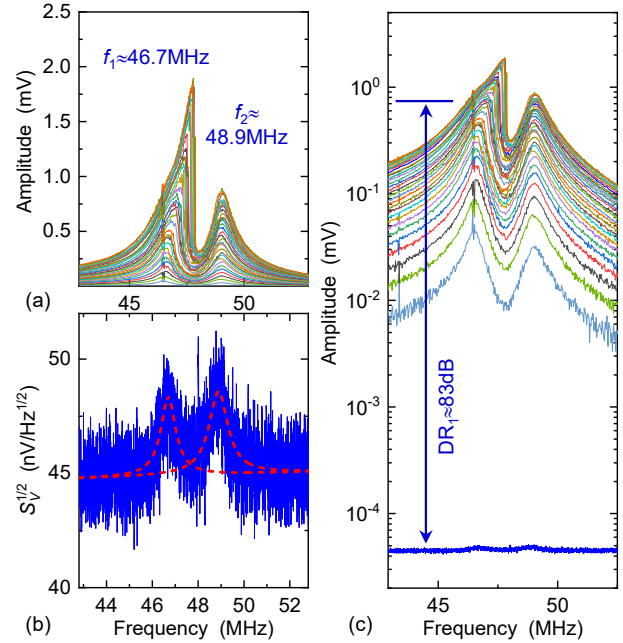


Figure 4: Nonlinearity and DR of the 4L MoS<sub>2</sub> device at  $V_g = -3V$ . (a) Driven resonance at varying RF drive  $v_{drv} = 10\text{--}300\text{mV}$  with a 10mV step. (b) Measured thermomechanical noise of the 1<sup>st</sup> and 2<sup>nd</sup> modes. Red dashed lines are fitting to a simple harmonic resonator model. (c) Log plot of (a) and (b), showing  $DR_1 = 83\text{dB}$ .

We first calibrate the two resonance modes of the 2D NEMS resonator, by investigating their basic resonance characteristics, nonlinearity, and dynamic range (DR) [5]. We set  $V_g = -3V$  and measure undriven thermomechanical noise without applying  $v_{drv}$ . The resonance frequencies and quality ( $Q$ ) factors of the two modes are determined by fitting the data to a simple harmonic resonator model [4]:

$$S_{x,th}^{1/2}(\omega) = \left( \frac{4k_B T \omega_n}{M_{eff} Q (\omega^2 - \omega_n^2)^2 + \omega^2 \omega_n^2 / Q^2} \right)^{1/2}. \quad (1)$$

Here  $k_B$  is Boltzmann constant,  $M_{eff}$  is the effective mass,  $\omega_n$  is the angular resonance frequency of  $n^{\text{th}}$  mode. We find two resonance modes at  $f_1 \approx 46.7\text{MHz}$  with  $Q_1 \approx 60$ , and  $f_2 \approx 48.9\text{MHz}$  with  $Q_2 \approx 50$  (at  $V_g = -3V$ , red dashed lines in Fig. 4b). Next, both  $V_g$  and  $v_{drv}$  are applied to the gate using a bias tee, and RF drive  $v_{drv}$  is gradually increased to study nonlinearity of the resonance modes. For the 1<sup>st</sup> mode, clear bifurcation is observed under high  $v_{drv}$ , showing stiffening Duffing nonlinearity. DR is determined by a ratio of the amplitude at the onset of nonlinearity (*i.e.*, 1dB compression point) to the thermomechanical resonance,

and we obtain  $DR_1 \approx 83\text{dB}$  (Fig. 4c). Meanwhile, the amplitude of the 2<sup>nd</sup> mode increases linearly with respect to  $v_{\text{drv}}$ , and it does not exhibit clear Duffing nonlinearity even at  $v_{\text{drv}} = 300\text{mV}$ , leading to  $DR_2 \geq 85\text{dB}$ .

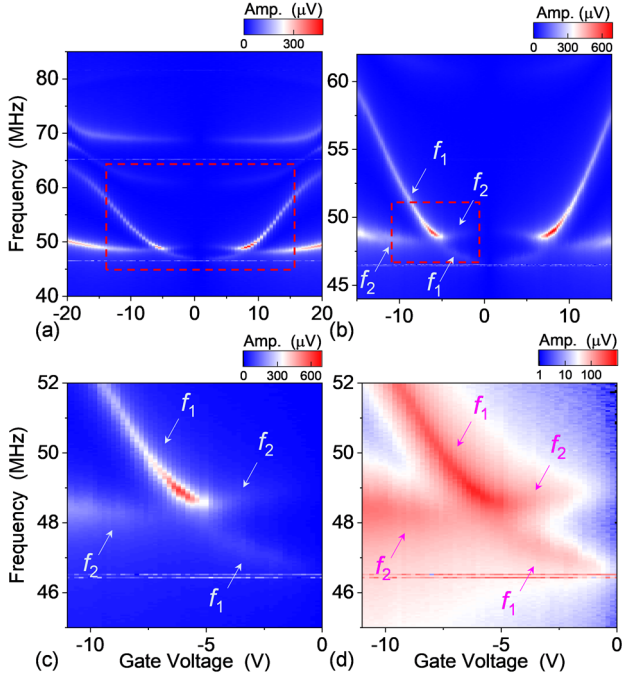


Figure 5: Frequency tuning behavior of the MoS<sub>2</sub> NEMS resonator. 2D color map is constructed by combining measured multimode resonances at varying  $V_g$ . The color represents the amplitude of the responses. (a) Frequency tuning map with varying  $V_g$  from  $-20\text{V}$  to  $20\text{V}$  with a  $0.5\text{V}$  step. (b) Frequency tuning curves in the red box of (a). The resonance curves are measured every  $0.2\text{V}$  step. (c) Zoom-in view of red box in (b). (d) Log color scale of data in (c), showing an anticrossing point of the two modes.

To achieve a 1:1 internal resonance condition, we investigate electrostatic frequency tuning of the MoS<sub>2</sub> device by sweeping  $V_g$  from  $-20\text{V}$  to  $20\text{V}$  with fixed  $v_{\text{drv}} = 10\text{mV}$ . The electrostatic force induced by  $V_g$  stretches down the MoS<sub>2</sub> membrane toward the local gate, increasing tension and thus resonance frequency. Meantime, it also introduces capacitive softening effect, which decreases resonance frequency. Interplay between these two effects leads to ‘U’ or ‘W’ shaped frequency tuning curves, depending on initial device parameters such as built-in tension and gap between the suspended MoS<sub>2</sub> and the gate [5,6]. In this study, both frequency tuning behaviors are observed from the 4L MoS<sub>2</sub> NEMS resonator (Fig. 5a). As  $|V_g|$  changes from 0 to  $20\text{V}$ , the 1<sup>st</sup> resonance mode  $f_1$  exhibits the ‘U’ shaped tuning curve, and the resonance frequency increases from  $\sim 47\text{MHz}$  to  $\sim 64\text{MHz}$ , offering frequency tunability of  $\Delta f_1/f_1 \approx 36\%$ . While the resonance frequency of the 2<sup>nd</sup> mode is  $f_2 \sim 50\text{MHz}$  near  $V_g = 0\text{V}$ , which shifts to  $\sim 48\text{MHz}$  at  $|V_g| = 10\text{V}$ , and then moves back to  $\sim 50\text{MHz}$  at  $|V_g| = 20\text{V}$ , showing the ‘W’ shaped turning curve. The different tuning behaviors in the 1<sup>st</sup> and 2<sup>nd</sup> modes bring their resonances to an anticrossing point near  $V_g \approx -6$  to  $-7\text{V}$  (Fig. 5c & 5d), giving lower and upper frequency (energy) branches, and breaking continuity in the  $f_1$  and  $f_2$  tuning curves versus  $|V_g|$ . As  $|V_g|$  increases, the

lower (energy) branch assumes mode  $f_1$  and then  $f_2$ , while the higher (energy) branch switches from mode  $f_2$  to  $f_1$ .

Since energy transfer between coupled modes takes place in the nonlinear regime, we characterize the nonlinear dynamics of the MoS<sub>2</sub> resonator focusing on the 1:1 internal resonance conditions. Based on the frequency tuning maps shown in Fig. 5c and 5d,  $V_g$  is set to be  $-6.4\text{V}$ , and resonance characteristics are measured with varying  $v_{\text{drv}}$ . At this 1:1 internal resonance setting, we find  $f_1 \approx 47.3\text{MHz}$  with  $Q_1 \approx 100$  and  $f_2 \approx 48.1\text{MHz}$  with  $Q_2 \approx 50$  from the thermomechanical noise (Fig. 6b), showing a frequency splitting of  $0.8\text{MHz}$ . Next, we introduce  $v_{\text{drv}} = 5\text{mV}$  to  $800\text{mV}$  to the gate with a  $15\text{mV}$  step, to excite the resonance motion. Figure 6a shows evolution of the resonance with varying  $v_{\text{drv}}$ . As  $v_{\text{drv}}$  increases, the peak point shifts upward in frequency, showing an asymmetric resonance shape (i.e., stiffening Duffing nonlinearity). Even at very high  $v_{\text{drv}} = 800\text{mV}$ , the resonance appears only weakly nonlinear, and the onset of the nonlinearity could be much higher than the measured amplitude. By comparing the measured amplitude at  $v_{\text{drv}} = 800\text{mV}$  with the noise floor, we estimate a measured  $DR_1 \geq 102\text{dB}$ . The surprisingly high DR may be attributed to the interplay of the polarity of Duffing nonlinearity induced by stiffening and capacitive softening effects. Moreover, the results in Fig. 4c and Fig. 6c indicate the MoS<sub>2</sub> device possesses excellent tunability of DR, over  $19\text{dB}$ , by controlling  $V_g$ .

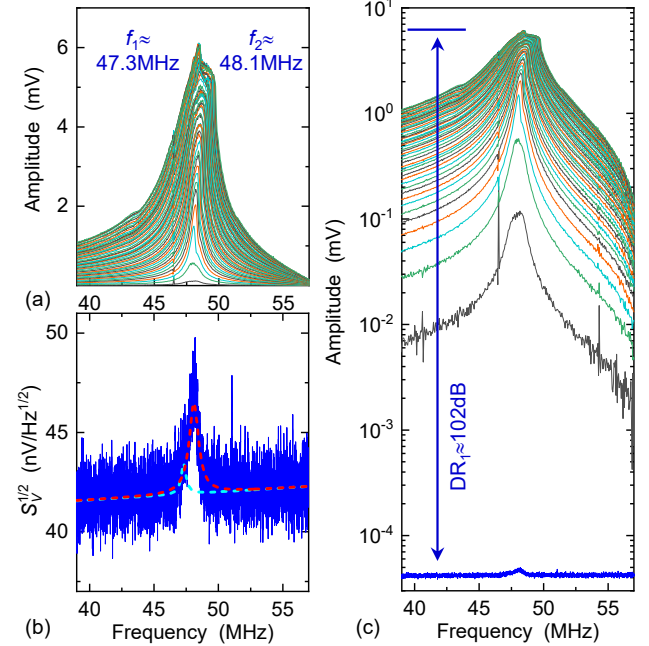


Figure 6: Nonlinearity and DR of the 4L MoS<sub>2</sub> resonator at  $V_g = -6.4\text{V}$ . (a) Evolution of the driven resonance for  $v_{\text{drv}} = 5\text{mV}$  to  $800\text{mV}$  with a  $15\text{mV}$  step. (b) Measured thermomechanical Brownian motion of the MoS<sub>2</sub> device. Cyan and red dashed lines are fitting to Eq. 1. (c) The amplitudes from (a) and (b) are plotted in a log scale to clearly show the measured DR of the resonance mode.

We now turn to focus on PFC generation in the MoS<sub>2</sub> NEMS resonator by employing the 1:1 internal resonance mode coupling (Fig. 7a). A single-frequency driving signal near the anticrossing ( $f_{\text{drv}} = 48.06\text{MHz}$ ) is applied to the gate with  $V_g = -6.4\text{V}$ , and PFC spectrum is monitored by



using the spectrum analyzer. No PFC pattern is generated when  $v_{\text{drv}} < 550\text{mV}$  (Fig. 7b). Upon  $v_{\text{drv}} > 550\text{mV}$ , equally spaced comb teeth appear near  $f_{\text{drv}}$ , showing a PFC. As the coupled modes start to be weakly nonlinear at  $v_{\text{drv}} = 550\text{mV}$  (Fig. 6a), the PFC generation appears to be related to nonlinearity of the coupled modes. Initially, the measured comb spacing is  $\Delta f = 2.53\text{MHz}$  at  $v_{\text{drv}} = 550\text{mV}$  (Fig. 7b); it increases to  $\Delta f = 2.65\text{MHz}$  at  $v_{\text{drv}} = 570\text{mV}$  (Fig. 7c) and then decreases to  $\Delta f = 2.44\text{MHz}$  at  $v_{\text{drv}} = 650\text{mV}$  (Fig. 7d). The ratio between PFC spacing and drive frequency,  $\Delta f/f_{\text{drv}} = 0.055$ , is much higher than those in PFCs with much larger MEMS resonators ( $\Delta f/f_{\text{drv}} \approx 10^{-6} \sim 10^{-3}$ ) [1,2]. In the range of  $v_{\text{drv}} > 700\text{mV}$ , the PFC pattern gradually disappears as the amplitudes of the comb teeth become smaller.

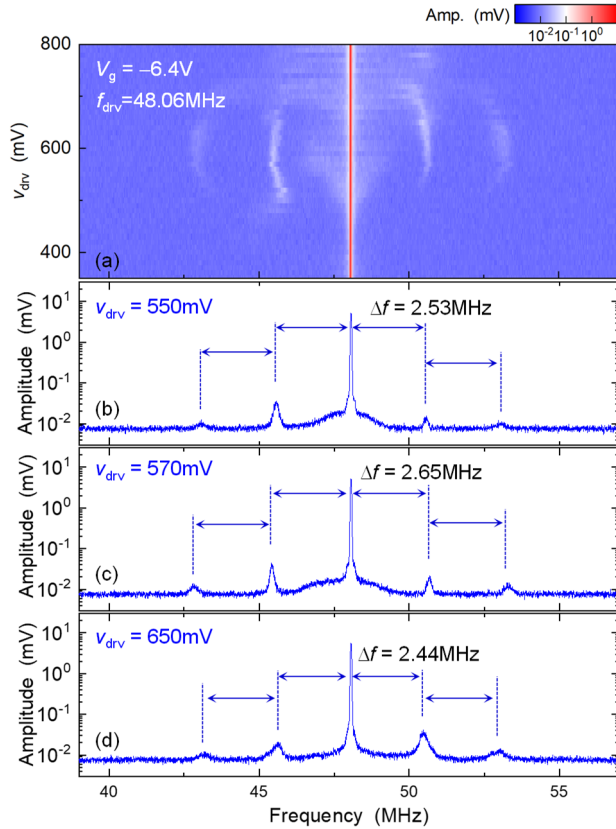


Figure 7: PFC generation in the 4L MoS<sub>2</sub> NEMS resonator. (a) 2D color plot of the PFC at  $V_g = -6.4\text{V}$ , and  $f_{\text{drv}} = 48.06\text{MHz}$  with  $v_{\text{drv}} = 370\text{mV}$  to  $800\text{mV}$ . Examples of the individual PFC spectral responses at (b)  $v_{\text{drv}} = 550\text{mV}$ , (c)  $v_{\text{drv}} = 570\text{mV}$ , and (d)  $v_{\text{drv}} = 650\text{mV}$ .

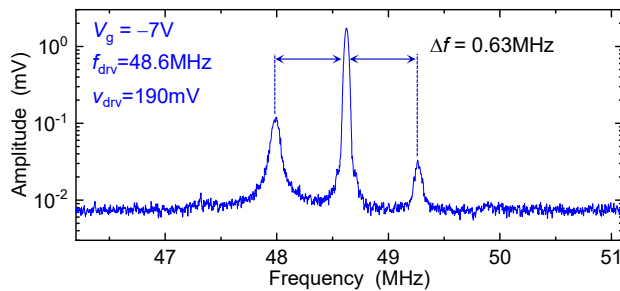


Figure 8: Measured PFC spectral response at  $V_g = -7\text{V}$ ,  $f_{\text{drv}} = 48.6\text{MHz}$ , and  $v_{\text{drv}} = 190\text{mV}$ .

We also investigate the characteristics of the PFC at different  $V_g$ . At  $V_g = -7\text{V}$  with  $f_{\text{drv}} = 48.6\text{MHz}$  and  $v_{\text{drv}} =$

$190\text{mV}$ , a PFC pattern emerges with  $\Delta f = 0.63\text{MHz}$  (Fig. 8). Compared with the PFC patterns demonstrated at  $V_g = -6.4\text{V}$ , this has narrower frequency spacing. This further shows the strong tunability of the PFC with different  $V_g$  and thus the resonance mode coupling conditions.

## CONCLUSION

In summary, we have demonstrated PFC generation in an atomically thin MoS<sub>2</sub> NEMS resonator. PFC responses are studied with various excitation and tuning parameters, including  $V_g$ ,  $f_{\text{drv}}$ , and  $v_{\text{drv}}$ . Comparison between the PFC responses and resonance behaviors, including frequency tuning and DR, suggests that the PFC generation is related to 1:1 mode coupling and nonlinearity induced by the input driving signal. The PFC demonstrated and the findings herein will be valuable for applications such as improving sensitivity of resonant sensors and stabilizing oscillators.

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