Supporting Information

Nonlinear Mode Coupling and One-to-One Internal Resonances in a Monolayer WS$_2$ Nanoresonator

S. Shiva P. Nathamgari$^{1, 2}$, Siyan Dong$^{1, 2}$, Lior Medina$^1$, Nicolaie Moldovan$^3$, Daniel Rosenmann$^4$, Ralu Divan$^4$, Daniel Lopez$^4$, Lincoln J. Lauhon$^5$, and Horacio D. Espinosa$^{1, 2, *}$

*corresponding author: espinosa@northwestern.edu, Phone: 847-467-5989

$^1$Department of Mechanical Engineering, Northwestern University, Evanston, IL-60208, United States

$^2$Theoretical and Applied Mechanics Program, Northwestern University, Evanston, IL-60208, United States

$^3$Alcorix Co., Plainfield, IL-60544, United States

$^4$Center for Nanoscale Materials, Argonne National Laboratories, Argonne, IL-60439, United States

$^5$Department of Materials Science and Engineering, Northwestern University, Evanston, IL-60208, United States

S1. Device Fabrication

A schematic of the fabrication methodology is shown in Figure S1. Atomically thin WS$_2$ flakes were first mechanically exfoliated onto a Si/SiO$_2$ substrate from a bulk-crystal (2D Semiconductors) using the widely reported scotch-tape method. A thin layer of polymethyl methacrylate (PMMA) was then spin-coated onto the flakes. The flake/PMMA stack was released using KOH wet-etching and transferred to a hydrogen silsesquioxane (HSQ) coated Si/SiO$_2$ wafer. The HSQ/PMMA bilayer was patterned using e-beam lithography to define the clamping electrodes, followed by Au-deposition using e-beam evaporation and lift-off. The unexposed HSQ underneath the flake was removed using a developer, followed by critical point drying to release the flake.
**Figure S1.** Schematic of the fabrication methodology. Mechanically exfoliated flakes from a bulk-crystal were transferred to a Si substrate coated with HSQ using KOH wet-etching (step 1) and PMMA transfer (step 2). The PMMA, HSQ stack was then patterned using e-beam lithography to define the clamping regions (step 3), followed by Au-deposition (step 4), lift-off (step 5) and critical point drying (step 6).

**S2. Responsivity of the interferometer**

The responsivity of the interferometer can be calculated using a thin-film interference model, as shown schematically in Figure S2a. The intensity of light reflected from the optical cavity ($I_r$), is given by

$$
\frac{I_r}{I_0} = \frac{\left| r_1 e^{i(\phi_1+\phi_2)} + r_2 e^{-i(\phi_1-\phi_2)} + r_3 e^{-i(\phi_1+\phi_2)} + r_1 r_2 r_3 e^{-i(\phi_1-\phi_2)} \right|^2 \left| e^{i(\phi_1+\phi_2)} + r_1 r_2 e^{-i(\phi_1-\phi_2)} + r_1 r_3 e^{-i(\phi_1+\phi_2)} + r_2 r_3 e^{-i(\phi_1-\phi_2)} \right|^2}{\left| e^{i(\phi_1+\phi_2)} + r_1 r_2 e^{-i(\phi_1-\phi_2)} + r_1 r_3 e^{-i(\phi_1+\phi_2)} + r_2 r_3 e^{-i(\phi_1-\phi_2)} \right|^2}
$$

(1)
where \((I_0)\) is the intensity of the incident beam, \(r_1\), \(r_2\) and \(r_3\) are reflection coefficients at the vacuum-resonator, resonator-vacuum and vacuum-substrate interface. The reflection coefficients \(r_i\) \((i=1, 2, 3)\) can be related to the refractive indices as

\[
\begin{align*}
    r_1 &= \frac{n_v - n_r}{n_v + n_r} \\
    r_2 &= \frac{n_r - n_v}{n_r + n_v} \\
    r_3 &= \frac{n_v - n_{Si}}{n_v + n_{Si}}
\end{align*}
\]

Denoting the resonator thickness by \(d_r\) and the cavity depth by \(d_v\), the phase shifts due to each are given by

\[
\begin{align*}
    \phi_1 &= \frac{2\pi n_r d_r}{\lambda} \\
    \phi_2 &= \frac{2\pi n_v d_v}{\lambda}
\end{align*}
\]

\(\lambda\) is the wavelength of the probe laser (632.8 nm). Using refractive index values \(n_{Si} = 3.881 - 0.0019i\), \(n_r = 5.6104 - 0.7293i\) and \(n_v = 1\), the reflectance of the cavity has been calculated for a monolayer resonator \(d_r = 0.67\) nm and various values of cavity-depths (see Figure S2b). In our fabrication methodology, the thickness of the spin-coated HSQ layer determines the cavity depth. We chose a value of 375 nm that yielded a responsivity of \(3\times10^{-3}/\text{nm}\) (data point shown as a circle in Figure S2b).
Figure S2. A) Schematic of the thin-film interference model used to calculate the cavity’s reflectance. Solid arrows indicate incident beam, dotted lines indicate reflected beams. B) Plot showing the reflectance of the cavity for various depth values. A cavity depth of 375 nm yields a responsivity of (-) 3x10^{-3}/nm.

S3. Raman Spectroscopy

A He-Ne laser (632.8 nm peak wavelength) was used as the excitation source. The laser power was kept to < 450 µW to minimize sample heating. The laser was focused on the sample to a spot size of < 2 µm. The reflected light from the nanoresonator was filtered using a Rayleigh filter (efficient to <150 cm^{-1}), dispersed using a spectrograph (1800 grooves/mm) and measured on a Peltier-cooled detector, with a spectral resolution of < 1 cm^{-1}. The peak locations were identified by fitting the measured data to a multi-peak, Lorentzian line-shape, as shown in Figure S3. The peak location of Si (520.7 cm^{-1}) was used to account for any drift in the spectrograph. The peak locations of the E_{12g}, A_{1g} phonon modes were 351 cm^{-1} and 417.5 cm^{-1} respectively. A spacing of 66.5 cm^{-1} between the two modes confirmed that it was monolayer thick.³
**Figure S3:** Raman spectroscopy measurements confirm the nanoresonator to be monolayer thick. The measured data points (black, solid line) were fit to a multi-peak, Lorentzian line-shape to extract the peak locations of the $E_{2g}^1$, $A_g^1$ and Si phonon modes.

**S4. Mode-shapes obtained using modal analysis and estimation of critical buckling load**

We used ABAQUS to perform modal analysis, both with and without the inclusion of prestress. For the former case, the plate was prestressed by inducing an in-plane displacement (of different magnitudes) along one of the fixed boundaries, while the value of the membrane load was extracted from the opposite boundary as reaction forces. The nodal forces were averaged against the boundary length to determine the membrane load. Shell elements (S4R) were employed to model the nanoresonator as a thin, flat plate. The exact geometry as obtained from an SEM image was used in the calculations. 200 elements were used in the analysis after performing a convergence study. The different material parameters used in the analysis are listed in Table S1.

**Table S1:** List of material parameters used in modal analysis

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value used</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young’s modulus ($E_Y^4$)</td>
<td>270 GPa</td>
</tr>
<tr>
<td>Poisson’s Ratio ($\nu$)</td>
<td>0.3</td>
</tr>
<tr>
<td>Density ($\rho$)</td>
<td>7500 kg m$^{-3}$</td>
</tr>
<tr>
<td>Thickness$^5$</td>
<td>0.34 nm</td>
</tr>
</tbody>
</table>
Figure S4. Mode-shapes of the WS$_2$ nanoresonator obtained from the modal analysis. The top row shows modes 1-4 (from left to right), whereas the bottom row shows modes 5-8 (from left to right). In the color bar, red and blue correspond to opposite directions of motion in the out of plane direction.

We evaluated the critical buckling load in the following manner. The buckling eigenmodes were first extracted using linear analysis in ABAQUS (Abaqus 6.14 Online Documentation). Nonlinear buckling analysis was then performed by adding an imperfection to the plate in the form of the first buckling eigenmode. By varying the peak amplitude of the imperfection from -0.1 to 0.1 nm, the load ($N_{xx}$) vs out of plane displacement ($w_m$) curve was computed (see Figure S5a). Here, $w_m$ refers to the out of plane displacement of the plate’s centroid. As the imperfection was made asymptotically smaller, the equilibrium curve approached the bifurcation point, which allowed the estimation of the critical buckling load $N_{cr} \sim \text{16.74 } \mu\text{Nm}^{-1}$. As explained in the main text, by imposing a prestress of $\sigma_0 \sim \text{45 } \mu\text{Nm}^{-1}$, the predicted frequency for the fundamental mode matched the measured value of 1.04 MHz. In Figure S5b, the measured resonance spectrum (driving voltage 1.5 V$_{p-p}$ for modes 1-6 and 2.0 V$_{p-p}$ for modes 7-8) is presented along with the predicted values for
modes 1-4 (shown as vertical dashed lines). The discrepancy between the predicted and observed frequencies may be due to an inhomogeneous strain, imperfections and floppy/edge modes due to free-edge boundary conditions.\textsuperscript{6-8}

**Figure S5.** a) Membrane load ($N_{xx}$) as a function of the out of plane displacement of the plate’s centroid ($w_m$) for various positive (+ve) and negative (-ve) imperfection amplitudes. The value of $N_{xx}$ corresponding to the smallest, non-zero imperfection is taken as the buckling load. The arrows point in the direction of decreasing imperfection amplitude. The ideal (no imperfection) curve is shown in red. b) The measured resonance spectrum of the WS$_2$ nanoresonator is compared with the predicted frequencies (vertical dashed lines) from modal analysis using a prestress value $\sigma_0 \sim 45 \mu\text{Nm}^{-1}$. Modes 1-6 were acquired with a driving voltage of 1.5 V$_{p-p}$ whereas modes 7-8 were acquired with 2 V$_{p-p}$ actuation voltage for a better signal to noise ratio.

**S5. Accounting for spurious resonances from PZT disc**

In order to eliminate spurious resonances that originate from the PZT disc, we measured the reflectance from the gold electrodes (blue dot in the SEM image in Figure S6’s inset) as the driving frequency was varied. Unlike cavity-interferometric measurements on the sample, here, changes in the power of the reflected light due to the motion of the gold electrodes were measured. If $r$ is the reflectance coefficient of Au and $m$ is the modulation coefficient of the incident beam with power $P_i$, then the photodetector measures a time-averaged power given by $P_{det} = P_i r^2 m^2$. The
modulation coefficient has a frequency dependence and has local maxima at the resonance frequencies of the PZT disc. In Figure S6, the red curve displays the resonance spectrum from the WS$_2$ resonator for an actuation drive of 3 V$_{p-p}$ measured using cavity-interferometry; whereas the blue curve is the reflectance measurement on the gold electrode. Spurious resonances due to the motion of the PZT appear as common peaks in both spectra and are indicated by arrows. The reflectance measurement has additional peaks (circled in Figure S6), which do not appear in the interferometric measurements, presumably because both the PZT and the nanoresonator are in phase and get displaced by approximately the same distance.

![Figure S6](image)

**Figure S6:** Measurements are made both on the WS$_2$ nanoresonator (red curve) and the PZT disc (blue curve) to eliminate any parasitic resonances from the latter. Such resonances appear as common peaks in both measurements and are indicated here by arrows. Inset shows an SEM image in which the red dot is the region where the WS$_2$ resonance spectrum was recorded, while the blue dot corresponds to the region where PZT motion was measured.

**S6. Weakly nonlinear behavior in modes 1, 2**

The measured frequency response data for mode 1 are fit to a Duffing model with additional higher order nonlinear terms to account for the mixed behavior, i.e.

$$\frac{d^2x}{dt^2} + (\gamma + \eta x^2) \frac{dx}{dt} + \omega_0^2x + \alpha x^3 + \beta x^4 + \delta x^5 = \frac{F}{m_e} \cos(\omega t)$$

(7)
The nonlinear terms effectively modify the resonance frequency $\omega_e$ and damping $\gamma_e$. As shown in ref.9, the fits are performed by expressing $\omega_e, \gamma_e$ as polynomial functions of the peak-amplitude $x_0$, viz. $\omega_e = \omega_0^2 + A_1 x_0^2 + A_2 x_0^3 + A_3 x_0^4$ and $\gamma_e = \gamma + B_1 x_0^2$. The measured frequency response data for mode 2 is fit to a standard Lorentzian line-shape.

**Figure S7:** The measured data for modes 1, 2 (shown as circles in a, c) are fit to the line-shapes described in the text. The fits are shown using solid lines. From bottom to top, the drive voltages are 0.5, 0.75, 1.5 and 2 V p-p. (b), (d): The extracted linewidth for different actuation voltages are plotted in subfigure (b) for mode 1 and (d) for mode 2. The case corresponding to zero voltage is
the thermo-mechanical resonance. The error bars for the driven resonances are within the data points.

**S7. Derivation of the reduced order (RO) model from Föppl–von Kármán plate theory**

In this section, we derive the nonlinearity coupled, ordinary differential equations (in time) for the modal co-ordinates starting from a Föppl–von Kármán plate model for the WS\(_2\) nanoresonator.

For simplicity, we assume a rectangular geometry \((2\hat{a} \times 2\hat{b})\) for the plate with thickness \(\hat{d}\) and fix the origin at its center. As boundary conditions, we take two opposite edges to be fixed \((\hat{x} = \pm \hat{a})\) and the remaining two edges to be free \((\hat{y} = \pm \hat{b})\). Further, we assume an isotropic elastic modulus\(^{10}\), neglect rotary and in-plane inertial terms\(^{11}\) and impose an initial imperfection in the plate in the form of an out of plane deformation profile \(\tilde{w}_0\). We employ the following normalization scheme where variables with an accent are the actual variables and those without are the normalized variables, viz.

\[
\begin{align*}
a &\equiv \frac{\hat{a}}{\hat{d}}, & b &\equiv \frac{\hat{b}}{\hat{d}}, & x &\equiv \frac{\hat{x}}{\hat{a}}, & y &\equiv \frac{\hat{y}}{\hat{b}}, & t &\equiv \hat{t} \sqrt{\frac{D}{\hat{a}^4 \rho \hat{d}}} , & w_0 &\equiv \frac{\tilde{w}_0}{\hat{d}}, \\
u &\equiv \frac{\hat{u}}{\hat{d}}, & v &\equiv \frac{\hat{v}}{\hat{d}}, & w &\equiv \frac{\tilde{w}}{\hat{d}}, & N_{ij} &\equiv \frac{\hat{a}^2}{D} \tilde{N}_{ij}, & M_{ij} &\equiv \frac{\hat{a}^2}{D \hat{d}} \tilde{M}_{ij} 
\end{align*}
\]

\(u, v, w\) represent the normalized displacement in \(x, y\) and \(z\) directions, \(\tilde{w}_0\) is the initial imperfection in the plate, \(\nu\) is Poisson’s ratio, \(\alpha\) is the aspect ratio \(\frac{\hat{a}}{\hat{b}}\). \(N_{ij}\) denotes the normalized membrane load obtained by integrating the stresses through the thickness, i.e. \(\int_{-d}^{d} \sigma_{ij} dz\), whereas \(M_{ij}\) is the bending moment. The governing equations can then be summarized as follows:

\[
\begin{align*}
\left( a \frac{\partial^2 u}{\partial x^2} + b \frac{\partial^2 v}{\partial x \partial y} - \frac{\partial w_0}{\partial x} \frac{\partial^2 w_0}{\partial x^2} + \frac{\rho}{\hat{a}^4 \rho \hat{d}^2} \frac{\partial^2 w_0}{\partial x^2 \partial y} \right) + \nu \alpha^2 \left( b \frac{\partial^2 v}{\partial x \partial y} + \frac{\partial w_0}{\partial x} \frac{\partial^2 w_0}{\partial x \partial y} - \frac{\rho}{\hat{a}^4 \rho \hat{d}^2} \frac{\partial^2 w_0}{\partial x \partial y} \frac{\partial^2 w_0}{\partial x^2} \right) + \alpha \frac{1-v}{2a^2} \left( \frac{1}{b} \frac{\partial^2 u}{\partial y^2} + \frac{1}{a} \frac{\partial^2 w_0}{\partial x^2} \right) + \\
\frac{1}{ab} \left( \frac{\partial^2 w_0}{\partial x \partial y} + \frac{\partial w_0}{\partial x} \frac{\partial^2 w_0}{\partial x \partial y} - \frac{\rho}{\hat{a}^4 \rho \hat{d}^2} \frac{\partial^2 w_0}{\partial x \partial y} \frac{\partial^2 w_0}{\partial x^2} \right) &= 0
\end{align*}
\]

\[
\begin{align*}
\alpha \left( b \frac{\partial^2 v}{\partial y^2} + \frac{\partial w_0}{\partial y} \frac{\partial^2 w_0}{\partial y^2} - \frac{\rho}{\hat{a}^4 \rho \hat{d}^2} \frac{\partial^2 w_0}{\partial x \partial y} \frac{\partial^2 w_0}{\partial y^2} \right) + \nu \left( a \frac{\partial u}{\partial x \partial y} + \frac{\partial w_0}{\partial x} \frac{\partial^2 w_0}{\partial x \partial y} - \frac{\rho}{\hat{a}^4 \rho \hat{d}^2} \frac{\partial^2 w_0}{\partial x \partial y} \frac{\partial^2 w_0}{\partial x^2} \right) + \frac{1-v}{2a^2} \left( \frac{1}{b} \frac{\partial^2 v}{\partial x \partial y} + \frac{1}{a} \frac{\partial^2 w_0}{\partial x^2} \right) + \\
\frac{1}{ab} \left( \frac{\partial^2 w_0}{\partial x \partial y} + \frac{\partial w_0}{\partial x} \frac{\partial^2 w_0}{\partial x \partial y} - \frac{\rho}{\hat{a}^4 \rho \hat{d}^2} \frac{\partial^2 w_0}{\partial x \partial y} \frac{\partial^2 w_0}{\partial x^2} \right) &= 0
\end{align*}
\]
\[
\frac{\partial^2 w}{\partial t^2} - \left( \frac{\partial^2}{\partial x^2} + \alpha^2 \frac{\partial^2}{\partial y^2} \right) \left( \frac{\partial^2 (w-w_0)}{\partial x^2} + \alpha^2 \frac{\partial^2 (w-w_0)}{\partial y^2} \right) = \frac{\partial}{\partial x} \left( \bar{N}_{xx} + N_{xx} \right) \frac{\partial w}{\partial x} + \alpha N_{xy} \frac{\partial w}{\partial y} + \nabla^2 \left( \frac{\partial w}{\partial x} \right) \left( \frac{\partial w}{\partial y} \right) \right)
\]

Equations (9-10) are the in-plane equilibrium equations, whereas equation (11) is the out of plane equilibrium equation with non-linear, von-Karman strains implicit in \( N_{ij} \). \( \bar{N}_{xx} \) is the intrinsic prestress in the plate. The terms originating from the imperfection \( \bar{w}_0 \) have been highlighted in bold in all three equations. The boundary conditions in terms of the normalized variables are

\[ u = v = w = 0, \quad \frac{\partial w}{\partial x} = 0, @ x = \pm 1 \quad \text{and} \quad N_{yy} = N_{xy} = M_{xy} = M_{yy} = 0, @ y = \pm 1 \] (12)

Next, we use the Galerkin decomposition in terms of modal co-ordinates and mode-shape functions for the displacements \( u, v \) and \( w \)

\[
\begin{align*}
    u(x, y, t) & \approx \sum_{i=1}^{n} p_i(t) \Pi_i(x, y), & v(x, y, t) & \approx \sum_{i=1}^{n} e_i(t) H_i(x, y) \\
    w(x, y, t) & \approx \sum_{i=1}^{n} q_i(t) \Phi_i(x, y), & w_0(x, y) & \approx \sum_{i=1}^{n} q_i^{(0)} \Phi_i(x, y)
\end{align*}
\] (13)

For conciseness, we use indicial notation in the subsequent portion. By using this decomposition in the governing equations (9), (10) and (11), followed by integration in the spatial co-ordinates \( x, y \) and enforcing the essential boundary conditions (12), we arrive at the following set of ordinary differential equations for the modal co-ordinates \( p_i, e_i \) and \( q_i \), namely

\[
\begin{align*}
-\alpha \left( p_i l_{ik}^{\Pi xx} + \nu a e_i l_{ik}^{\Pi xy} \right) + \left( q_i q_j - q_i^{(0)} q_j^{(0)} \right) a_{ij}^{(1)} + \frac{1 - \nu}{2 ab} \left( \frac{p_i l_{ik}^{\Pi yx}}{b} - \frac{e_i l_{ik}^{H y y x}}{a} \right) + \frac{1}{ab} \left( q_i q_j - q_i^{(0)} q_j^{(0)} \right) b_{ij}^{(1)} = 0
\end{align*}
\] (14)

\[
\begin{align*}
\alpha \left( \alpha^2 b e_i l_{ik}^{H y y} - \nu a p_i l_{ik}^{H x y} \right) + \left( q_i q_j - q_i^{(0)} q_j^{(0)} \right) a_{ij}^{(2)} - \frac{1 - \nu}{2 ab} \left( \frac{1}{b} p_i l_{ik}^{H x y x} + \frac{1}{a} e_i l_{ik}^{H x x} \right) + \frac{1}{ab} \left( q_i q_j - q_i^{(0)} q_j^{(0)} \right) b_{ij}^{(2)} = 0
\end{align*}
\] (15)
The different coefficients multiplying the modal co-ordinates in equations (14), (15) and (16) are functions of the mode-shapes (and their derivatives) and have been tabulated below. Here, we would like to show the origin of the cubic coupling terms in the RO model. We restrict the number of modes \( n=2 \), corresponding to modes 7, 8 in Figure S5, with corresponding modal co-ordinates \( q_1, q_2 \) and mode-shape functions \( \phi_1, \phi_2 \) for the out of plane displacement \( w \). Equations (14), (15) are linear in \( p_i, q_i \); for \( n=2 \), they represent a 4x4 system of linear equations in \( p_1, q_1, p_2, q_2 \). Consequently, their solution can be expressed as

\[
p_i = \lambda_{ijl} q_j q_l - \lambda_{ijl}^{(0)} q_j^{(0)} q_l^{(0)}, \quad e_i = \mu_{ijl} q_j q_l - \mu_{ijl}^{(0)} q_j^{(0)} q_l^{(0)}, \quad i = 1, 2
\]  

(17)

Consider the first coupling term in equation (16), i.e.

\[
ap_i q_m s_{imk}^{u,x} = a \left[ \lambda_{ijl} q_j q_l - \lambda_{ijl}^{(0)} q_j^{(0)} q_l^{(0)} \right] q_m s_{imk}^{u,x} = a \left[ \lambda_{ijl} q_j q_l q_m s_{imk}^{u,x} - \lambda_{ijl}^{(0)} q_j^{(0)} q_l^{(0)} q_m s_{imk}^{u,x} \right]
\]  

(18)

The term \( \lambda_{ijl} q_j q_l q_m s_{imk}^{u,x} \) has four repeated indices \((i, j, k, m)\) and one free index \( (k) \), and when expanded fully, it results in eight terms (for \( n=2 \)) of the following form - \( q_1^3, q_1^2 q_2, q_1 q_2^2, q_2^3 \). Similarly, it can be shown that the following terms lead to cubic coupling terms - \( e_i q_m s_{imk}^{v,y} \), \( q_i q_j q_m z_{ijk}^x \), \( p_i q_j q_m s_{imk}^{v,x} \), \( e_i q_m s_{imk}^{v,x} \), \( q_i q_j q_m z_{ijk}^{xy} \), \( q_i q_j q_m z_{ijk}^{xy} \) and \( p_i q_m s_{imk}^{v,x} \). The initial imperfection \( w_0 \), however only leads to linear coupling terms such as \( q_i^{(0)} q_j^{(0)} q_m z_{ijk}^x \), \( q_i^{(0)} q_j^{(0)} q_m z_{ijk}^{xy} \) and \( q_i^{(0)} q_j^{(0)} q_m z_{ijk}^{xy} \).

Expressions for different coefficients in equation (14)

\[
a_{ik}^2 = \int_{-1}^{1} \int_{-1}^{1} \Pi_k \Pi_k \, dx \, dy
\]

\[
o_{ijk}^{(1)} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \phi_i}{\partial x} \frac{\partial \phi_j}{\partial x} \left( \frac{\partial \phi_l}{\partial x} + v \alpha^2 \frac{\partial \phi_l}{\partial x} \right) \Pi_k \, dx \, dy
\]

(19)

\[
i_{ik}^{\Pi,xx} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Pi_k}{\partial x} \frac{\partial \Pi_k}{\partial x} \, dx \, dy
\]

\[
i_{ik}^{\Pi,xy} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Pi_k}{\partial y} \frac{\partial \Pi_k}{\partial y} \, dx \, dy
\]

\[
i_{ik}^{\Pi,yy} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial^2 \Pi_k}{\partial y^2} \Pi_k \, dx \, dy
\]

(20)

\[
g_{ijk}^{(1)} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial}{\partial y} \left( \frac{\partial \phi_i}{\partial x} \frac{\partial \phi_j}{\partial y} \right) \Pi_k \, dx \, dy
\]

S12
Expressions for different coefficients in equation (15)

\[ l_{ik}^{H,yy} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial^2 H_l}{\partial y^2} H_k \, dx \, dy \quad l_{ik}^{H,xn,y} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial H_k}{\partial y} \frac{\partial \Pi_l}{\partial x} \, dx \, dy \]  

(22)

\[ a_{ijk}^{(2)} = \int_{-1}^{1} \int_{-1}^{1} \left( \frac{\partial}{\partial y} \left( \Phi_i \frac{\partial \Phi_j}{\partial y} \right) \right) H_k + \nu \frac{\partial}{\partial x} \left( \Phi_i \frac{\partial \Phi_j}{\partial y} \right) H_k \, dx \, dy \]  

(23)

Expressions for different coefficients in equation (16)

\[ f_k = \int_{-1}^{1} \int_{-1}^{1} \Phi_k \, dx \, dy \]  

(25)

\[ m_{ik} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial^2 \Phi_i}{\partial x^2} \frac{\partial^2 \Phi_k}{\partial x^2} \, dx \, dy + 2a^2 \int_{-1}^{1} \int_{-1}^{1} \frac{\partial^2 \Phi_k}{\partial x^2} \frac{\partial^2 \Phi_i}{\partial y^2} \, dx \, dy + \alpha^4 \left( \int_{-1}^{1} \int_{-1}^{1} \frac{\partial}{\partial y} \left( \Phi_k \frac{\partial^2 \Phi_i}{\partial y^2} \right) \, dx \, dy \right) \]  

(26)

\[ a_{ik} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \Phi_k \, dx \, dy \quad a_{ijk}^{u} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \Pi_l \Phi_k \, dx \, dy \]  

(27)

\[ a_{ijk}^{v} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} H_j \Phi_k \, dx \, dy \]  

(28)

\[ r_{ik} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial x} \frac{\partial \Phi_k}{\partial x} \, dx \, dy - \alpha^2 \left( \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \frac{\partial \Phi_k}{\partial y} \bigg|_{y=-1} \, dx \right) \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \Phi_k \, dx \, dy \]  

(29)

\[ r_{ik}^{(1)} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial x} \frac{\partial \Phi_k}{\partial x} \, dx \, dy \]  

(30)

\[ s_{lmk}^{u,x} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Pi_l}{\partial x} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy \quad s_{lmk}^{v,y} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial H_l}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy \]  

(31)

\[ z_{ijm}^{u} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial x} \frac{\partial \Phi_j}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy + \alpha^2 \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \frac{\partial \Phi_j}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy \]  

(32)

\[ z_{ijm}^{u,x} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Pi_l}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy \quad z_{ijm}^{v,y} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial H_l}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy \]  

(33)

\[ z_{ijm}^{xy} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \frac{\partial \Phi_j}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k \, dx \, dy \]  

(34)

\[ z_{ijm}^{xy} = \int_{-1}^{1} \int_{-1}^{1} \frac{\partial \Phi_i}{\partial y} \frac{\partial \Phi_j}{\partial y^2} \Phi_k \, dx \, dy \]  

(35)
\[ s_{lmk}^{\nu,x} = \frac{1}{2} \int_{-1}^{1} \frac{\partial H_l}{\partial x} \frac{\partial^2 \Phi_m}{\partial y^2} \Phi_k dx dy \quad s_{lmk}^{\nu,y} = \frac{1}{2} \int_{-1}^{1} \frac{\partial H_l}{\partial y} \frac{\partial^2 \Phi_m}{\partial x^2} \Phi_k dx dy \] (36)

**S8. Perturbation Solution by the Method of Scales**

We employ a reduced order model that consists of two Duffing oscillators with nearly commensurate frequencies and weakly non-linear coupling terms, given by the following pair of differential equations\(^{12}\):

\[
\begin{align*}
\ddot{x} + \omega_1^2 x &= \epsilon [-\mu_1 \dot{x} + k_1 x^3 + a_1 x^2 y + b_1 x y^2 + c_1 y^3 + d_1 y + F\cos(\omega t)] \quad (37) \\
\ddot{y} + \omega_2^2 y &= \epsilon [-\mu_2 \dot{y} + k_2 y^3 + a_2 x^2 y + b_2 x y^2 + c_2 x^3 + d_2 x] \quad (38)
\end{align*}
\]

where \(x\) and \(y\) refer to the modal degrees of freedom, \(\epsilon\) is a small parameter. The closeness of the two frequencies can be expressed using a detuning parameter \(\sigma_1\), where \(\omega_2 = \omega_1 + \sigma_1 \epsilon\). We solve the model by using the method of scales. We seek solutions, \(x\) and \(y\) of the form

\[
\begin{align*}
x &= x_0 + \epsilon x_1 + O(\epsilon^2), \quad y &= y_0 + \epsilon y_1 + O(\epsilon^2) \quad (39)
\end{align*}
\]

Introducing fast and slow time scales \(T_0 = t, T_1 = \epsilon t\), and denoting their derivatives by \(D_0 = \frac{\partial}{\partial T_0}\) and \(D_1 = \frac{\partial}{\partial T_1}\), we can express the first and second time derivatives as

\[
\begin{align*}
\dot{x} &= (D_0 + \epsilon D_1)x, \quad \ddot{x} = (D_0 + \epsilon D_1)^2 x \sim (D_0^2 + 2\epsilon D_0 D_1)x \quad (40)
\end{align*}
\]

Using these expressions for the time scales and their derivatives, the governing equations (37) and (38) can be rewritten as

\[
\begin{align*}
(D_0^2 + 2\epsilon D_0 D_1)(x_0 + \epsilon x_1) + \omega_1^2 (x_0 + \epsilon x_1) &= \epsilon [-\mu_1 (D_0 + \epsilon D_1)(x_0 + \epsilon x_1) + k_1 (x_0 + \epsilon x_1)^3 + a_1 (x_0 + \epsilon x_1)^2 (y_0 + \epsilon y_1) + b_1 (x_0 + \epsilon x_1)(y_0 + \epsilon y_1)^2 + c_1 (y_0 + \epsilon y_1)^3 + d_1 (y_0 + \epsilon y_1) + F\cos(\omega t)] \quad (41) \\
(D_0^2 + 2\epsilon D_0 D_1)(y_0 + \epsilon y_1) + \omega_2^2 (y_0 + \epsilon y_1) &= \epsilon [-\mu_2 (D_0 + \epsilon D_1)(y_0 + \epsilon y_1) + k_2 (y_0 + \epsilon y_1)^3 + a_2 (x_0 + \epsilon x_1)^2 (y_0 + \epsilon y_1) + b_2 (x_0 + \epsilon x_1)(y_0 + \epsilon y_1)^2 + c_2 (x_0 + \epsilon x_1)^3 + d_2 (x_0 + \epsilon x_1)] \quad (42)
\end{align*}
\]

Equating equal powers of \(\epsilon\) (i.e. 0, 1) yields the following set of equations

\[
\begin{align*}
D_0^2 x_0 + \omega_1^2 x_0 &= 0, \quad D_0^2 y_0 + \omega_2^2 y_0 = 0 \quad (43) \\
D_0^2 x_1 + \omega_1^2 x_1 &= -2 D_0 D_1 x_0 - \mu_1 D_0 x_0 + k_1 x_0^3 + a_1 x_0^2 y_0 + b_1 x_0 y_0^2 + c_1 y_0^3 + d_1 y_0 + F\cos(\omega T_0) \quad (44)
\end{align*}
\]
\[ D_0^2 y_1 + \omega_0^2 y_1 = -2 D_0 D_1 y_0 - \mu_2 D_0 y_0 + k_2 y_0^3 + a_2 x_0^2 y_0 + b_2 x_0 y_0^2 + c_2 x_0^3 + d_2 x_0 \quad (45) \]

Seeking general solutions of the form \( x_0 = A(T_1)e^{i\omega_1 T_0} + \text{cc} \) and \( y_0 = B(T_1)e^{i\omega_2 T_0} + \text{cc} \), which satisfy equation (43) (\( \text{cc} \) denotes complex conjugate), equations (44) and (45) simplify to

\[ D_0^2 x_1 + \omega_0^2 x_1 = -2 i \omega_1 A' e^{i\omega_1 T_0} - i \mu_1 \omega_1 A e^{i\omega_1 T_0} + k_1 \left( A^3 e^{i\omega_1 T_0} + 3 |A|^2 A e^{i\omega_1 T_0} \right) + \]
\[ + a_1 \left( A^2 B e^{i(2 \omega_1 + \omega_2) T_0} + A^2 B e^{i(-2 \omega_1 - \omega_2) T_0} + 2 |A|^2 B e^{i\omega_2 T_0} \right) + b_1 \left( B^2 A e^{i(2 \omega_2 + \omega_1) T_0} + B^2 A e^{i(2 \omega_2 - \omega_1) T_0} + 2 |B|^2 A e^{i\omega_1 T_0} \right) + d_1 B e^{i\omega_2 T_0} + \frac{F}{2} e^{i\omega T_0} + \text{cc} \quad (46) \]

\[ D_0^2 y_1 + \omega_0^2 y_1 = -2 i \omega_2 B' e^{i\omega_2 T_0} - i \mu_2 \omega_2 B e^{i\omega_2 T_0} + k_2 \left( B^3 e^{i\omega_2 T_0} + 3 |B|^2 B e^{i\omega_2 T_0} \right) + \]
\[ + a_2 \left( A^2 B e^{i(2 \omega_1 + \omega_2) T_0} + A^2 B e^{i(-2 \omega_1 - \omega_2) T_0} + 2 |A|^2 B e^{i\omega_2 T_0} \right) + b_2 \left( B^2 A e^{i(2 \omega_2 + \omega_1) T_0} + B^2 A e^{i(2 \omega_2 - \omega_1) T_0} + 2 |B|^2 A e^{i\omega_1 T_0} \right) + d_2 B e^{i\omega_1 T_0} + \text{cc} \quad (47) \]

In equations (46) and (47), \( A' \) denotes differentiation with respect to the slow time scale \( T_1 \); also, the secular terms that lead to the solvability conditions have been highlighted in bold. Introducing a detuning parameter \( \sigma \) that captures the separation between the driving and resonance frequencies, i.e. \( \omega = \omega_1 + \epsilon \sigma \), the solvability conditions for equations (46) and (47) are

\[ -2 i \omega_1 A' - i \mu_1 \omega_1 A + 3 k_1 |A|^2 A + a_1 \left( A^2 B e^{-i\sigma_1 T_1} + 2 |A|^2 B e^{i\sigma_1 T_1} \right) + b_1 \left( B^2 A e^{i2\sigma_1 T_1} + 2 |B|^2 A e^{i\sigma_1 T_1} \right) + d_1 B e^{i\sigma_1 T_1} + \frac{F}{2} e^{i\sigma T_1} = 0 \quad (48) \]

\[ -2 i \omega_2 B' - i \mu_2 \omega_2 B + 3 k_2 |B|^2 B + a_2 \left( A^2 B e^{-i\sigma_1 T_1} + 2 |A|^2 B \right) + b_2 \left( B^2 A e^{i\sigma_1 T_1} + 2 |B|^2 A e^{i\sigma_1 T_1} \right) + c_2 |A|^2 A e^{-i\sigma_1 T_1} + d_2 B e^{-i\sigma_1 T_1} = 0 \quad (49) \]

Only those coupling terms that are proportional to \( x^2 y \) in equation (48) and \( xy^2 \) in equation (49) are retained. Using Cartesian notation for the amplitudes \( A = \frac{1}{2} \left( p_1(T_1) - q_1(T_1) \right) e^{i\phi_1(T_1)} \) and \( B = \frac{1}{2} \left( p_2(T_1) - q_2(T_1) \right) e^{i\phi_2(T_1)} \) and inserting them in the solvability conditions (48), (49), followed by separating the real and imaginary parts, we derive the following set of equations:

\[ p_1' = -\mu p_1 - \sigma q_1 + \frac{3 k_1}{8 \omega_1} (p_1^2 + q_1^2) q_1 + \frac{A}{8 \omega_1} (2(p_2^2 + q_2^2) q_1 - (p_2^2 - q_2^2) q_1 + 2 p_1 p_2 q_2) \quad (50) \]

\[ q_1' = -\mu q_1 + \sigma p_1 - \frac{3 k_1}{8 \omega_1} (p_1^2 + q_1^2) p_1 + \frac{F}{2 \omega_1} - \frac{A}{8 \omega_1} (2(p_2^2 + q_2^2) p_1 + (p_2^2 - q_2^2) p_1 + 2 p_2 q_1 q_2) \quad (51) \]
\( p_2' = -\mu p_2 - (\sigma - \sigma_1)q_2 + \frac{3k_2}{8\omega_2} (p_2^2 + q_2^2)q_2 + \frac{\Lambda}{8\omega_2} (2(p_1^2 + q_1^2))q_2 - (p_1^2 - q_1^2)q_2 + 2p_1p_2q_1) \) (52)

\( q_2' = -\mu q_2 + (\sigma - \sigma_1)p_2 - \frac{3k_2}{8\omega_2} (p_2^2 + q_2^2)p_2 - \frac{\Lambda}{8\omega_2} (2(p_1^2 + q_1^2)p_2 + (p_1^2 - q_1^2)p_2 + 2p_1q_1q_2) \) (53)

Bifurcation analysis was performed on the quadruple set of first order ODEs in \((p_1, q_1, p_2, q_2)\) space using the open-source software MatCont\(^1\)3.

**S9. Bifurcation analysis on mode 7**

Consider the simplified reduced order model given below

\[ \ddot{x} + \omega_1^2 x = \epsilon [-\mu_1 \dot{x} - k_1 x^3 - \Lambda x y^2 + F \cos(\omega t)] \] (54)

\[ \ddot{y} + \omega_2^2 y = \epsilon [-\mu_2 \dot{y} - k_2 y^3 - \Lambda x^2 y] \] (55)

We employed units of \(\mu V\) for the displacement to directly compare with the experimental data. The units of the linearized frequencies \(k_i\) and damping \(\mu_i\) remain unchanged; the units for the nonlinear coefficients \(k_i\) and \(\Lambda\) were adjusted accordingly (see Table S2). The value of the cubic nonlinear coefficient \(k_2\) was estimated from the data corresponding to the softening portion of mode 8 (Figure S8a). The same value was used for \(k_1\). The value of \(\Lambda\) was iteratively varied until the predicted frequency response matched with the measured data for an actuation voltage of 3 V\(_{p-p}\). Then, the value of \(\Lambda\) was fixed at \(-1e-1 \text{ s}^2/\mu \text{V}^2\) and the force term \(F\) was varied to match with the measured data for different applied voltages. The actuation force on the nanoresonator results from inertial coupling with the motion of the PZT disc. For a harmonic base excitation \(d = d_0 \sin \omega t\) of the PZT, the inertial force acting on the resonator is given by \(F = T m_r \omega^2 d_0\), where \(m_r\) is the mass of the nanoresonator and \(T\) is the transmission coefficient. The peak displacement \(d_0\) varies linearly with the applied voltage and hence, \(F \propto V\). Figure S8b shows a scatter plot with those values of \(F\) that resulted in the best fit for the measured data at different actuation voltages used in the experiments. The expected linear relation between \(F, V\) is reproduced. The bifurcation set for the two Hopf points shown in Figure 5d in the main text predicts that for force values < 5 \(\text{s}^2/\mu \text{V}\), the Hopf points disappear. This corresponds to an actuation voltage of \(~1.5 \text{ V}_{p-p}\), as estimated from the linear fit in Figure S8b.
Table S2. Values of different parameters used in the bifurcation analysis

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value used, units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\omega_1, \omega_2$</td>
<td>$2\pi \times (1.34, 1.36) \text{ MHz}$</td>
</tr>
<tr>
<td>$\mu_1 = \mu_2 = \mu$</td>
<td>0.32 MHz</td>
</tr>
<tr>
<td>$k_1, k_2$</td>
<td>$-2e-3 \text{ s}^2/\mu \text{ V}^2$</td>
</tr>
<tr>
<td>$\Lambda$</td>
<td>$-1e-1 \text{ s}^2/\mu \text{ V}^2$</td>
</tr>
<tr>
<td>$F$</td>
<td>$(10, 12, 15, 22) \text{ s}^2/\mu \text{ V}$</td>
</tr>
</tbody>
</table>

Figure S8. a) The softening portion in the measured data of mode 8 (indicated by the dotted lines) was used to estimate the cubic nonlinear term $k_2$. b) Drive force $F$ values that reproduced the measured frequency response curves for mode 7 at various actuation voltages are shown along with a linear fit (dashed line). The circular marker corresponds to the point on the bifurcation set where the Hopf bifurcations disappear.

References


