Nonlinear phononics using atomically thin membranes

Daniel Midtvedt1,†, Andreas Isacsson1 & Alexander Croy1,‡

Phononic crystals and acoustic metamaterials are used to tailor phonon and sound propagation properties by utilizing artificial, periodic structures. Analogous to photonic crystals, phononic band gaps can be created, which influence wave propagation and, more generally, allow engineering of the acoustic properties of a system. Beyond that, nonlinear phenomena in periodic structures have been extensively studied in photonic crystals and atomic Bose-Einstein condensates in optical lattices. However, creating nonlinear phononic crystals or nonlinear acoustic metamaterials remains challenging and only few examples have been demonstrated. Here, we show that atomically thin and periodically pinned membranes support coupled localized modes with nonlinear dynamics. The proposed system provides a platform for investigating nonlinear phononics.

1 Department of Applied Physics, Chalmers University of Technology, 5-41296 Göteborg, Sweden. † Present address: Max Planck Institute for the Physics of Complex Systems, Nothnitzer Str. 38, 01187 Dresden, Germany. Correspondence and requests for materials should be addressed to D.M. (email: midtvedt@pks.mpg.de) or to A.C. (email: croy@pks.mpg.de).
Nonlinear photonic\textsuperscript{1} and optomechanical crystals\textsuperscript{2–4}, as well as atomic Bose–Einstein condensates in optical lattices\textsuperscript{5–7} are well-known examples for which nonlinear phenomena in periodic structures have been intensively studied\textsuperscript{8}. All these systems exploit the wide tunability of system properties offered by artificial periodic structures. Similarly, phononic crystals can be engineered to have tailored phonon propagation properties\textsuperscript{9–11}. However, nonlinear effects in phononic crystals are, in general, much harder to obtain\textsuperscript{12}.

The basic building block of a phononic crystal consists of two coupled localized phonon modes, a phononic dimer. From studies of Bose–Einstein condensates\textsuperscript{13–15} and nanomechanical resonators\textsuperscript{16–19}, it is known that the presence of nonlinearities leads to a rich dynamical behaviour even for such a simple system. Moreover, coherent manipulation of phonons in coupled resonators has recently been demonstrated in the linear ‘Rabi’ regime\textsuperscript{20,21}. Accessing the nonlinear regime and the ability to scale the system beyond dimers will be crucial for realizing nonlinear phononics. Nanomechanical systems that exhibit a combination of strong nonlinearities and a high level of control provide an excellent platform for nonlinear phononic studies of Bose–Einstein condensates\textsuperscript{13–15} and nanomechanical systems that exhibit a regime\textsuperscript{20,21}. Accessing the nonlinear regime and the ability to scale the system beyond dimers will be crucial for realizing nonlinear phononics. Nanomechanical systems that exhibit a combination of strong nonlinearities and a high level of control provide an excellent platform for nonlinear phononic many-body effects. Due to the high frequencies of the modes and the low mass of the membrane, our system is an ideal candidate for studies in the quantum regime\textsuperscript{29}.

**Results**

**Pinned graphene membrane.** To realize the phononic crystal, we consider a graphene membrane, which is deposited on top of a square lattice of cylindrical pillars with radii \(a_s\) and lattice vectors \(a_1\) and \(a_2\) (see Fig. 1). Due to the large adhesion energy, the graphene membrane is essentially pinned at the pillars\textsuperscript{30,31}, but free to move otherwise. This situation may be described using a continuum mechanics model\textsuperscript{28,30}. The Lagrangian density of a graphene sheet subject to a pinning potential \(V(r)\) is

\[
\mathcal{L} = \frac{1}{2} \rho \dot{w}^2 - \frac{\kappa}{2} (\nabla^2 w)^2 - \frac{1}{2} \sigma_{ij} \partial_j \dot{u}_i - \frac{1}{2} V(r) w^2 \tag{1}
\]

where \(w(r)\) is the out-of-plane deformation, \(\rho\) is the mass density, \(\kappa\) is the bending rigidity, \(\sigma_{ij}\) is the stress and \(\epsilon_{ij}\) is the strain tensor\textsuperscript{22}. Here, we have used the out-of-plane approximation\textsuperscript{28}, which is valid for large pre-strain typically found for suspended graphene sheets\textsuperscript{33}. For isotropic pre-strain, \(\epsilon_{ij} = \delta_{ij} \epsilon_0 + \partial_i \omega \partial_j \omega / 2\), the Lagrangian density can be further simplified. The pre-tension \(T_0 = \kappa (a_s + 2\mu)\) determines the unit of frequency and \(\lambda\) and \(\mu\) are the Lamé parameters of the membrane. The pinning potential is given by \(V(r) = V_0 \sum_{nm} \Theta(|r - r_{nm}| - a_s)\), where \(V_0\) is the pinning strength and \(r_{nm} = n \mathbf{a}_1 + m \mathbf{a}_2\). Due to the periodicity of the pinning lattice, the system supports frequency bands analogous to the electronic bands in periodic solids. See Supplementary Fig. 1a for a calculation of the lowest lying frequency bands in the present system.

In the following we take the lattice spacing \(a\) to be the unit of length, \(\sqrt{\rho / T_0}\) the unit of time and \(\rho a^2\) the unit of mass.

**Coupled localized modes.** Considering two vacancies in the lattice, which are far apart, one finds two degenerate, localized modes. Moving the vacancies closer together, the degeneracy is lifted and one obtains a symmetric and an antisymmetric mode at frequencies \(\Omega_{\pm}\). Figure 2a,b shows the mode shapes for \(\sigma = 1/5\). The frequency splitting indicates that the localized modes leak into the region of the neighbouring vacancy, which leads to an intermode interaction.

Figure 2c,d shows the centre frequency \((\Omega_+ + \Omega_-) / 2\) and the frequency difference \(\Omega_+ - \Omega_-\) of a dimer for different pillar radii. The centre frequency increases with increasing \(\sigma\), since the size of the suspended region shrinks. The frequency difference on the other hand decreases, because the overlap of the localized modes vanishes. For an estimate of the scaling behaviour, one can consider the suspended region to be approximately a square of size \(\xi \times \xi\). From a dimensional analysis one finds, that the frequency scales as \(\Omega \propto 1/\xi\). The length \(\xi\) can be taken as a linear function of \(\sigma\), as shown in Fig. 2c.

The interaction strength is calculated from the linearized equation of motion obtained from equation (1) by making the Ansatz \(w(r) = \psi_1(r) q_1 + \psi_2(r) q_2\). The localized mode shapes \(\psi_1(r)\) are expressed by linear combinations of the exact dimer eigenmodes (Wannier basis) or estimated by using mode shapes of the single-vacancy modes (tight-binding approximation)\textsuperscript{34}.

In both cases, one obtains the generalized eigenvalue problem

\[
\Omega^2 \sum_m S_{mn} q_m = \sum_m V_{nm} q_m \quad (m, n) = (L, R),
\]

where the overlap matrix \(S_{nm} = \langle \psi_n | \psi_m \rangle\), the interaction matrix \(V_{nm} = \langle \nabla \psi_m | \nabla \psi_n \rangle + \langle \psi_n | V \psi_m \rangle\) and angular brackets denote spatial integration. The diagonal terms \(\Omega^2 = V_{nn}\) give the individual mode frequencies and the off-diagonal elements \(\lambda_{nm} = V_{nm}\) are the linear coupling constants. In the tight-binding approximation, the overlap matrix \(S_{nm}\) is not diagonal, but the values on the diagonal dominate. The effective interaction matrix is then obtained by multiplying \(V_{nm}\) by \(S_{nn}\) from the left. From Fig. 2c,d, we find a good agreement between the tight binding and Wannier calculations for the chosen values of \(\sigma\). This implies that an array of such defect modes support frequency bands which are well described by the tight-binding method (see Supplementary Fig. 1c and Supplementary Note 1).

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**Figure 1 | Schematic illustration of the pinning-lattice setup.** Cylindrical pillars with diameter \(2a_s\) are arranged in a periodic lattice with lattice spacing \(a\). The suspended graphene membrane is pinned at the top of the pillars. By removing pillars, localized modes can be created.
Nonlinear effects. One consequence of the membrane being atomically thin is that geometrically induced nonlinear effects become appreciable already for moderate displacements. The respective contribution to the stretching energy in equation (1) is

$$E_{\text{nl}} = \frac{1}{8a} \left( (\partial_x w)^2 + (\partial_y w)^2 \right) .$$  \hspace{1cm} (3)

This expression accounts for the energy required to accommodate for the area increase associated with a transverse deflection of the membrane.

To estimate the influence of the energy (equation (3)) for the dimer, we use the Wannier basis, that is, linear combinations of the eigenmodes for $w$. The energy is written as

$$E_{\text{nl}} = \frac{1}{8a} \left( D_{\text{LR}} q_1^4 + D_{\text{RR}} q_2^4 + Q_{\text{LR}} q_1^2 q_2^2 + C_{\text{LR}} q_1 q_2^2 + C_{\text{RR}} q_1^2 q_2 \right),$$  \hspace{1cm} (4)

where $D_{\text{LR}}$, $C_{\text{LR}}$, and $Q_{\text{LR}}$ are the Duffing, the cubic coupling and the the quadratic coupling constants, respectively (see Methods section).

Figure 2e,f shows the behaviour of $D_{\text{LR}}$, $C_{\text{LR}}$ and $Q_{\text{LR}}$ as functions of $\sigma$. The Duffing constant increases with increasing $\sigma$ due to the shrinking suspension area. The nonlinear coupling constants, $C_{\text{LR}}$ and $Q_{\text{LR}}$, decrease analogous to the frequency splitting because the overlap of the localized modes vanishes. Using the same square-membrane approximation as for the centre frequency, one finds the scaling $D_{\text{LR}} \propto 1/\sigma^6$, which is in good agreement with the numerical data.

Dynamics in rotating wave approximation. Since the time scales associated with $\Omega_\perp$ and the nonlinearities are well separated, it is useful to consider the dynamics of the coupled oscillator system in the rotating wave approximation (RWA). To this end, we define $q_m = \sqrt{1/2\Omega_0} (a_m e^{-i\omega t} + a_m^* e^{i\omega t})$ and $\dot{q}_m = -i\sqrt{\Omega_0}/2 (a_m e^{-i\omega t} - a_m^* e^{i\omega t})$, where $\Omega_0$ is a reference frequency. The slowly changing amplitudes $a_m$ obey the equations of motion $\ddot{a}_m = -i\partial H/\partial a_m^*$ (see also Supplementary Note 2)35. For a symmetric system, that is, $D = D_{\text{LR}}$, $C = C_{\text{LR}}$, $Q = Q_{\text{LR}}$ and $\lambda = \lambda_{\text{LR}}$, the Hamiltonian $H$ in RWA is given by

$$H \approx \frac{\delta \Omega_0}{2} |a_t|^2 + \frac{\delta \Omega_2}{2} |a_t|^2 + \frac{U}{2} (|a_t|^4 + |a_L|^4)$$

$$+ J + 3W \left( |a_t|^2 + |a_L|^2 \right) (a_t a_t^* + a_L a_L^*)$$

$$+ K \left[ |a_t|^4 + \frac{1}{4} (a_t^2 a_t^2 + a_L^2 a_L^2) \right] ,$$  \hspace{1cm} (5)

where $\delta \Omega_0 = (\Omega_0^2 - \Omega_{\text{ref}}^2)/\Omega_0$ is a small detuning from the reference frequency, and we have defined $J = \lambda_0/\Omega_0$, $U = (3D)/(8\Omega_0^2)$, $K = Q/(8\Omega_0^2)$ and $W = (3C)/(32\Omega_0^2)$ to characterize ‘hopping’, on-site interactions and interaction tunnelling, respectively. The values of $J$, $U$, $K$ and $W$ following from Fig. 2 are shown in Supplementary Fig. 2.

Equation (5) corresponds to the classical analogue of the improved two-mode model36 for Bose particles in a double-well potential. The Hamiltonian (equation (5)) can be expressed in terms of only two variables. Setting $a_n = |a_t|^2 e^{i\phi n}$, we define the ‘population imbalance’ $z = (|a_t|^2 - |a_L|^2)/Z^2$ and the relative
phase $\Delta \phi = \phi_R - \phi_L$. Note that the ’total population’ $Z^2 = |a_R|^2 + |a_L|^2$ is conserved. We further set the reference frequency to the average frequency, $\Omega^2 = (\Omega^2 + \Omega^2)/2$, which maps equation (5) onto the Hamiltonian for a rigid rotor:

$$
H_{\text{eff}} = \frac{U_{\text{eff}}}{2} + J_{\text{eff}} \sqrt{1 - z^2} \cos(\Delta \phi) + \frac{K_{\text{eff}}}{2} (1 - z^2) \cos(2\Delta \phi) - \mu_{\text{eff}} z,
$$

(6)

where $U_{\text{eff}} = (U - K)Z^2/2$, $J_{\text{eff}} = (J + WZ^2)Z^2$, $K_{\text{eff}} = KZ^4/4$ and $\mu_{\text{eff}} = (\Omega^2 - \Omega^2)Z^2/4 \Omega_D$. The last term is due to detuning of the individual mode frequencies and tilts the double-well potential defined by this Hamiltonian. In the present system, $K_{\text{eff}} < \max(U_{\text{eff}}, J_{\text{eff}})$ (see Supplementary Fig. 2), and the second to last term in equation (6) does not crucially influence the dynamics. Taking $\mu_{\text{eff}} \to 0$, the corresponding equations of motion are

$$
\dot{z} = J_{\text{eff}} \sqrt{1 - z^2} \sin(\Delta \phi),
$$

(7a)

$$
\dot{\Delta \phi} = U_{\text{eff}} z - J_{\text{eff}} \frac{z}{\sqrt{1 - z^2}} \cos(\Delta \phi) - \mu_{\text{eff}}. \quad (7b)
$$

For atoms in optical lattices, equation (6) is usually obtained in the semiclassical limit, for example by using the Gutzwiller Ansatz. Here, we derived the Hamiltonian (equation (6)) within a completely classical description employing the RWA (see also Supplementary Note 2). As a consequence, the total population $Z$ used above has the unit of an action and is proportional, but not equal to the total number of phonons.

For $\mu_{\text{eff}} = 0$, the system undergoes a pitch-fork bifurcation at a critical value of the ratio $U_{\text{eff}}/J_{\text{eff}}$, with qualitatively different dynamics on each side of the bifurcation. This defines two regimes, denoted in the literature as the ‘Rabi’ regime for $U_{\text{eff}}/J_{\text{eff}} < 1$ and ‘Josephson’ regime for $U_{\text{eff}}/J_{\text{eff}} > 1$ (ref. 15). The Rabi regime is characterized by oscillation of $z$ with vanishing temporal mean. In the Josephson regime, there exist self-trapped fixed points at $z = \pm z_0$ and $\Delta \phi = 0$, corresponding to either of the localized modes being predominantly excited. At even larger ratios ($U_{\text{eff}}/J_{\text{eff}} > 2$), running phase modes appear (Fig. 3b-d).

Note the different dependence of $U_{\text{eff}}$ and $J_{\text{eff}}$ on the vibrational amplitudes, entering through the parameter $Z$. This implies that the ratio can be tuned either through changes in the system itself (through the pillar radii), or by changing the vibrational amplitude of the dimer. In Fig. 3a, the ratio $U_{\text{eff}}/J_{\text{eff}}$ is shown as a function of the pillar radius and the vibrational amplitude scaled by the pre-strain. The dashed line corresponds to the transition between the two regimes. At a fixed pillar size, the system can be tuned into either regime through the vibrational amplitude.

**Frequency tuning.** External forces applied to the membrane will lead to a static deformation of the membrane. Due to the non-linearities of the system, this can be used to tune the frequencies and couplings. Considering small oscillations around the static deformation, the equations of motion are linearized, which leads to the following renormalized frequencies and linear couplings:

$$
\Omega_n \to \Omega_n^2 + \frac{3}{2d_e} \delta_n^2 \Omega_n^2 + \frac{1}{4d_e} Q_{nm} \delta_m^2 + \frac{3}{4d_e} C_{nm} \delta_m \delta_n, \quad (8a)
$$

$$
\lambda_{nm} \to \lambda_{nm} + \frac{1}{2d_e} Q_{nm} \delta_m \delta_n + \frac{3}{8d_e} C_{nm} (\delta_m^2 + \delta_n^2), \quad (8b)
$$

where $\delta_n$ are the static deflections. Assuming that the latter can be tuned independently for each oscillator, for instance via local back gates, this allows for complete control over the frequencies

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**Figure 3 | Dynamical regimes of equation (6).** (a) Ratio of nonlinear to linear interaction $U_{\text{eff}}/J_{\text{eff}}$ as a function of ‘total population’ $Z$ and pillar radius $\sigma$. By tuning the vibrational amplitude of the system, both the ‘Rabi’ (blue) and ‘Josephson’ (red) regimes are accessible. The dashed line corresponds to $U_{\text{eff}}/J_{\text{eff}} = 1$, while the dashed-dotted line corresponds to $U_{\text{eff}}/J_{\text{eff}} = 2$. (b-d) Phase space plots of corresponding to the different regimes for tilt $\mu_{\text{eff}} = 0$. (b) At low values of the ratio ($U_{\text{eff}}/J_{\text{eff}} = 1/10$), the system undergoes Rabi-like oscillations with vanishing temporal mean of the population imbalance $z$. (c) For values of the ratio $1 < U_{\text{eff}}/J_{\text{eff}} < 2$ (here $U_{\text{eff}}/J_{\text{eff}} = 3/2$), the system develops two elliptic fixed points with $z$ having non vanishing temporal mean. (d) For higher ratios (here $U_{\text{eff}}/J_{\text{eff}} = 3$) running phase modes appear.
A natural extension of the dimer configuration considered in the Results is a periodic array of defect modes. Analogous to the dimer, the energy of such a system can be expressed in terms of the amplitudes of localized (Wannier) modes. In general, the resulting Hamiltonian in RWA is given by

$$H = \sum_n \left( \frac{\Delta E_n}{2} |a_n|^2 + \sum_{m,n} \sum_{k,l} J_{mn,kl} a^*_n a^*_m a_k a_l + \sum_{m,n} W_{mn} |a_n|^2 |a_m|^2 \right),$$

where $J_{mn}$ and $W_{mn}$ denote ‘hopping’ and interaction constants, respectively.

To actuate the motion of the oscillators, external pressure may be applied to the membrane. In nanoelectromechanical systems, this is typically achieved by the electrostatic interaction with a metallic back gate. This leads to the pressure $P_L \propto -G(r)V^2_g$ with $V_g$ denoting the gate voltage and $G(r)$ the shape of the gate. The force acting on each oscillator is given by $f^{(ext)}(r) \propto -\langle \psi_r |G|V^2_g \rangle$. A d.c. voltage leads to a static displacement $\tilde{q}_n > 0$ in equation (8), while an a.c. voltage can be used to drive the system.

The coupling of the membrane to its environment leads to dissipation of the mechanical motion. This has been extensively studied in the context of atomically thin membrane resonators, and quality factors of $Q > 10^4$ have been reported for graphene resonators at low temperatures. Phenomenologically, the dissipation can be described by a viscous damping term proportional to $Q^{-1} \tilde{q}_n$ in the equations of motion. In the Rabi regime, the oscillation frequency is given by $|f|$ and $Q \gg 1/|f|$ is required. We find for $\kappa = 1/5$, that $1/|f| \approx 65$ and $Q \approx 500$ would be sufficient to resolve the oscillations. In addition to the viscous damping, nonlinear dissipation has been observed in carbon-based resonators, which might give rise to interesting nonlinear dissipation effects.

Due to the high frequencies of the localized modes and the low mass of the membrane, our system is a promising candidate for studies in the quantum regime. The presence of intrinsic nonlinearities makes it attractive for creation and detection of non-classical states. More generally, realizing lattices of interacting flexural modes would pave the road for quantum many-body phononics.

**Methods**

**Nonlinear constants.** Using the Ansatz $w(t) = \psi_1(r)\phi_1 + \psi_2(r)\phi_2$ in equation (3) leads to the expression of the energy (equation (4)) in terms of the nonlinear constants

$$D_\alpha = \left\langle \left[ \left( \partial_\alpha \psi_1 \right)^2 + \left( \partial_\alpha \psi_2 \right)^2 \right] \right\rangle,$$

$$C_{\Delta \alpha} = 4 \left\langle \left[ \left( \partial_\alpha \psi_1 \right)^2 + \left( \partial_\alpha \psi_2 \right)^2 \right] \left[ \left( \partial_\beta \psi_1 \right) \left( \partial_\beta \psi_2 \right) + \left( \partial_\beta \psi_1 \right) \left( \partial_\beta \psi_2 \right) \right] \right\rangle,$$

$$C_{\Delta \beta} = 4 \left\langle \left[ \left( \beta \partial_\beta \psi_1 \right) \left( \beta \partial_\beta \psi_2 \right) \right] \left[ \left( \partial_\alpha \psi_1 \right) \left( \partial_\alpha \psi_2 \right) + \left( \partial_\alpha \psi_1 \right) \left( \partial_\alpha \psi_2 \right) \right] \right\rangle,$$

$$Q_{\Delta \alpha} = 2 \left\langle 4 \left( \partial_\alpha \psi_1 \right) \left( \partial_\alpha \psi_2 \right) \left( \partial_\beta \psi_1 \right) \left( \partial_\beta \psi_2 \right) + \left( \partial_\alpha \psi_1 \right)^2 \left( \partial_\alpha \psi_2 \right)^2 + 3 \left( \partial_\beta \psi_1 \right)^2 \left( \partial_\beta \psi_2 \right)^2 + \left( \partial_\beta \psi_1 \right)^2 \left( \partial_\beta \psi_2 \right)^2 \right\rangle.
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Author contributions

All authors contributed equally to the work presented in this paper.

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