Supplementary Information:

Dynamics of topological phonon edge states in polymer chains and supramolecular lattices on surfaces

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Supplementary Methods

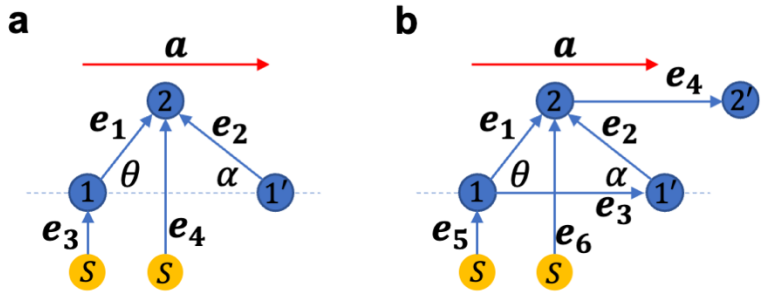
We used the dynamical matrix approach[1] to describe the linear response of our simplified mechanical systems. A *d*-dimensional frame consisting of N point masses connected by N_C springs is fully described by the compatibility matrix C, relating the Nd-dimensional vector of sites displacements $\boldsymbol{u}$ to the N_C-dimensional vector of bond elongations $\boldsymbol{e}$:

$$\mathrm{C} \cdot \boldsymbol{u} = \boldsymbol{e} \qquad (1)$$

The normal modes of the system are the eigenvalues of the dynamical matrix $\mathbf{D}$, which is given by:

$$\mathrm{D} = \mathrm{M}^{-1}\mathrm{C}^{*}\,\kappa\,\mathrm{C}\,, \qquad (2)$$

where κ is the $N_C \times N_C$ diagonal matrix of spring constants. C^{*} denotes the conjugate transpose matrix of C. M^{-1} is the inverse of the diagonal matrix of masses. Bond elongations are obtained using, $\boldsymbol{e_n} = \hat{e}_n \cdot \left(\boldsymbol{u}_j - \boldsymbol{u}_i\right)$, where $\hat{e}_n$ is the unit vector pointing from site i to j in the undistorted lattice.



Supplementary Schema 1: **a.** aSSH model showing the elongation vectors $\boldsymbol{e_n}$ for the unit cell characterized by the unit cell vector $\boldsymbol{a}$ vector. **b.** daSSH model showing the elongation vectors $\boldsymbol{e_n}$ for the unit cell characterized by the unit cell vector $\boldsymbol{a}$ vector. The substrates mases *S* are fixed. We assume all masses are equal to m.

For the aSSH case:

$$\boldsymbol{e_1} = \hat{e}_1 \cdot (\boldsymbol{u}_1 - \boldsymbol{u}_2)$$

$$\boldsymbol{e_2} = \hat{e}_2 \cdot \big(\boldsymbol{u}_2 - \boldsymbol{u}_1(\boldsymbol{r} + \boldsymbol{a})\big)$$

$$\boldsymbol{e_3} = \hat{e}_3 \cdot \boldsymbol{u}_1$$

$$\boldsymbol{e_4} = \hat{e}_4 \cdot \boldsymbol{u}_2$$

In this way we can construct the connectivity matrix:

$$\begin{pmatrix} e_1 \\ e_2 \\ e_3 \\ e_4 \end{pmatrix} = \begin{pmatrix} \hat{e}_{1x} & -\hat{e}_{1x} & \hat{e}_{1y} & -\hat{e}_{1y} \\ -e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\hat{e}_{2x} & \hat{e}_{2x} & e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\hat{e}_{2y} & -\hat{e}_{2y} \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} u_{1x} \\ u_{2x} \\ u_{1y} \\ u_{2y} \end{pmatrix} \quad (3)$$

where $\boldsymbol{a}$ is the unit vector. In terms of the angles of Scheme 1 our connectivity matrix reads:

$$\mathrm{C} = \begin{pmatrix} \cos\theta & -\cos\theta & \sin\theta & -\sin\theta \\ -e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\cos\alpha & \cos\alpha & e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\sin\alpha & -\sin\alpha \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \quad (4)$$

We consider the case where all masses are equal to $m = 1$. Then equation 2 allow us to construct the dynamical matrix:

$$\mathrm{D}_{\mathrm{aSSH}} = \begin{pmatrix} \cos\theta & -e^{-i\boldsymbol{k}\cdot\boldsymbol{a}}\cos\alpha & 0 & 0 \\ -\cos\theta & \cos\alpha & 0 & 0 \\ \sin\theta & e^{-i\boldsymbol{k}\cdot\boldsymbol{a}}\sin\alpha & 1 & 0 \\ -\sin\theta & -\sin\alpha & 0 & 1 \end{pmatrix} \begin{pmatrix} \kappa_1 & 0 & 0 & 0 \\ 0 & \kappa_2 & 0 & 0 \\ 0 & 0 & \kappa_3 & 0 \\ 0 & 0 & 0 & \kappa_4 \end{pmatrix} \begin{pmatrix} \cos\theta & -\cos\theta & \sin\theta & -\sin\theta \\ -e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\cos\alpha & \cos\alpha & e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\sin\alpha & -\sin\alpha \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$$

$$\mathrm{D}_{aSSH} = \begin{pmatrix} D_{11} & D_{12} & D_{13} & D_{14} \\ D_{21} & D_{22} & D_{23} & D_{24} \\ D_{31} & D_{32} & D_{33} & D_{34} \\ D_{41} & D_{42} & D_{43} & D_{44} \end{pmatrix} \quad (5)$$

And the components:

$$D_{11} = D_{22} = \kappa_1 \cos^2\theta + \kappa_2 \cos^2\alpha$$

$$D_{12} = -\kappa_1 \cos^2\theta - \kappa_2 e^{-i\boldsymbol{k}\cdot\boldsymbol{a}} \cos^2\alpha$$

$$D_{13} = D_{24} = D_{31} = D_{42} = \kappa_1 \sin\theta\cos\theta - \kappa_2 \sin\alpha\cos\alpha$$

$$D_{14} = D_{32} = -\kappa_1 \sin\theta\cos\theta + \kappa_2 e^{-i\boldsymbol{k}\cdot\boldsymbol{a}} \sin\alpha\cos\alpha$$

$$D_{21} = -\kappa_1 \cos^2\theta - \kappa_2 e^{i\boldsymbol{k}\cdot\boldsymbol{a}} \cos^2\alpha$$

$$D_{23} = D_{41} = -\kappa_1 \sin\theta\cos\theta + \kappa_2 e^{i\boldsymbol{k}\cdot\boldsymbol{a}} \sin\alpha\cos\alpha$$

$$D_{33} = \kappa_1 \cos^2\theta + \kappa_2 \cos^2\alpha + \kappa_3$$

$$D_{34} = -\kappa_1 \sin^2\theta - \kappa_2 e^{-i\boldsymbol{k}\cdot\boldsymbol{a}} \sin^2\alpha$$

$$D_{43} = -\kappa_1 \sin^2\theta - \kappa_2 e^{i\boldsymbol{k}\cdot\boldsymbol{a}} \sin^2\alpha$$

$$D_{44} = \kappa_1 \cos^2\theta + \kappa_2 \cos^2\alpha + \kappa_4$$

Equation 5 can be can also written as a Hermitian matrix:

$$\mathrm{D}_{aSSH} = \begin{pmatrix} D_{11} & D_{12} & D_{13} & D_{14} \\ D_{12}^* & D_{11} & D_{14}^* & D_{13} \\ D_{13} & D_{14} & D_{11} + \kappa_3 & D_{34} \\ D_{14}^* & D_{13} & D_{34}^* & D_{11} + \kappa_4 \end{pmatrix} \quad (6)$$

Similarly, for the daSSH model, the connectivity matrix is given by:

$$\mathrm{C} = \begin{pmatrix} \cos\theta & -\cos\theta & \sin\theta & -\sin\theta & 0 & 0 \\ -e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\cos\alpha & \cos\alpha & e^{i\boldsymbol{k}\cdot\boldsymbol{a}}\sin\alpha & -\sin\alpha & 0 & 0 \\ 1 - e^{i\boldsymbol{k}\cdot\boldsymbol{a}} & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 - e^{i\boldsymbol{k}\cdot\boldsymbol{a}} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix} \quad (7)$$

And the dynamical Hermitian matrix:

$$\mathrm{D}_{daSSH} = \begin{pmatrix} D_{11} & D_{12} & D_{13} & D_{14} & 0 & 0 \\ D_{12}^* & D_{11} & D_{14}^* & D_{13} & 0 & 0 \\ D_{13} & D_{14} & D_{33} & D_{34} & 0 & 0 \\ D_{14}^* & D_{13} & D_{34}^* & D_{33} & 0 & 0 \\ 0 & 0 & 0 & 0 & \kappa_5 & 0 \\ 0 & 0 & 0 & 0 & 0 & \kappa_6 \end{pmatrix} \quad (8)$$

where the components are:

$$D_{11} = D_{22} = \kappa_1 \cos^2\theta + \kappa_2 \cos^2\alpha + 2\kappa_3(1 - \cos\boldsymbol{k}\cdot\boldsymbol{a})$$

$$D_{12} = -\kappa_1 \cos^2\theta - \kappa_2 e^{-i\boldsymbol{k}\cdot\boldsymbol{a}} \cos^2\alpha$$

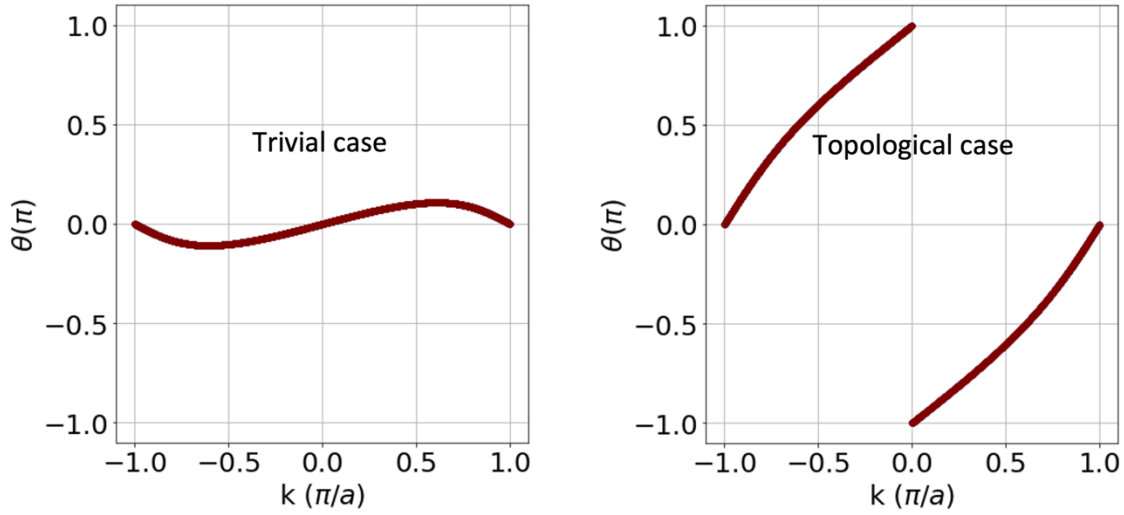
$$D_{13} = \kappa_1 \sin\theta\cos\theta - \kappa_2 \sin\alpha\cos\alpha$$

$$D_{14} = -\kappa_1 \sin\theta\cos\theta + \kappa_2 e^{-i\boldsymbol{k}\cdot\boldsymbol{a}} \sin\alpha\cos\alpha$$

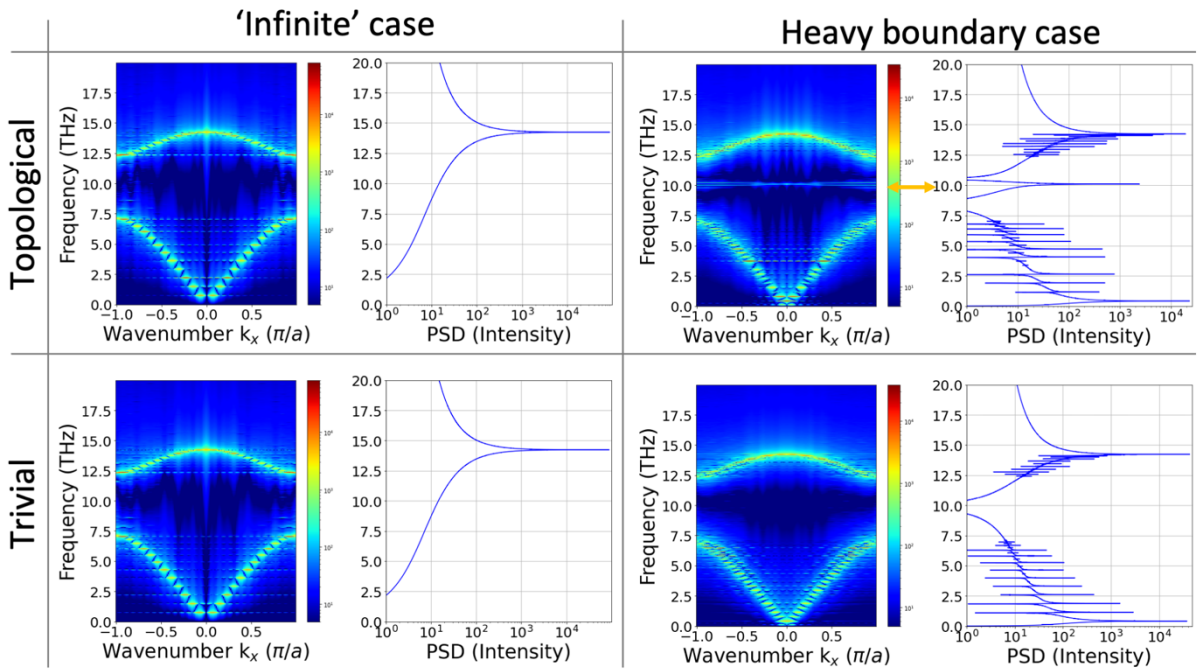
$$D_{33} = \kappa_1 \sin^2\theta + \kappa_2 \sin^2\alpha$$

$$D_{34} = -\kappa_1 \sin^2\theta - \kappa_2 e^{-i\boldsymbol{k}\cdot\boldsymbol{a}} \sin^2\alpha$$

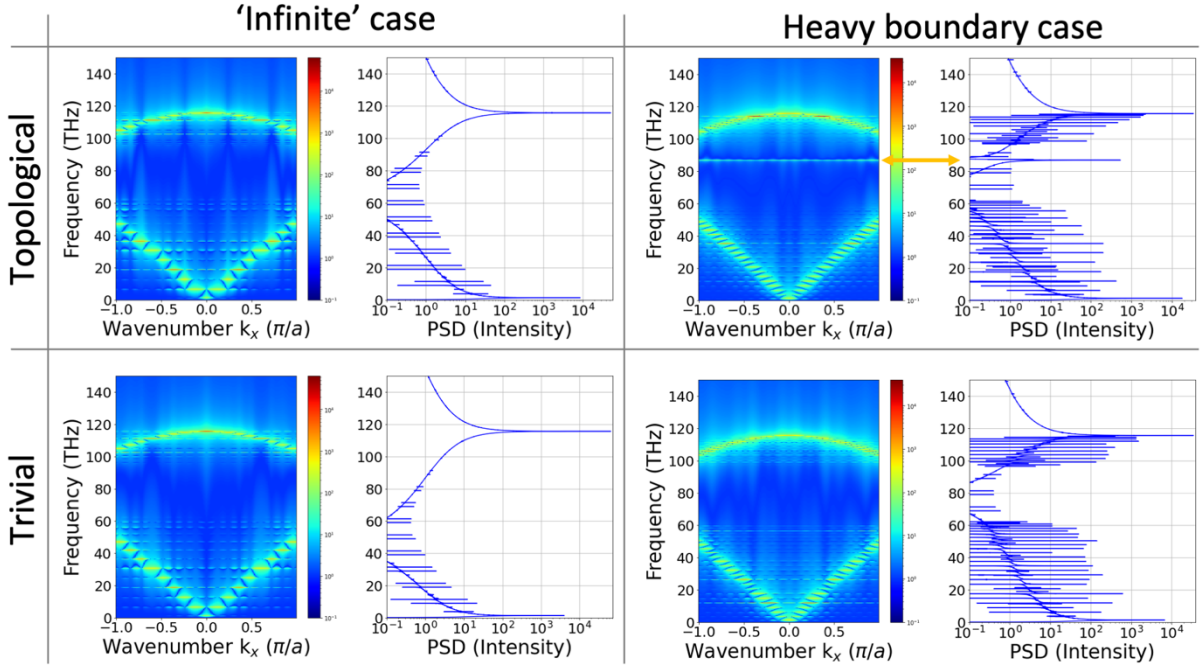
Supplementary Figures



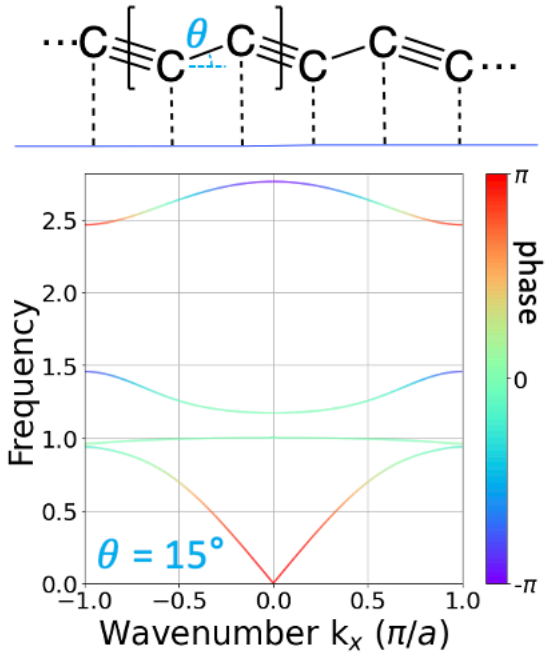
Supplementary Figure 1. Geometric phase for the trivial and topological pSSH model of Extended Data Fig. 1



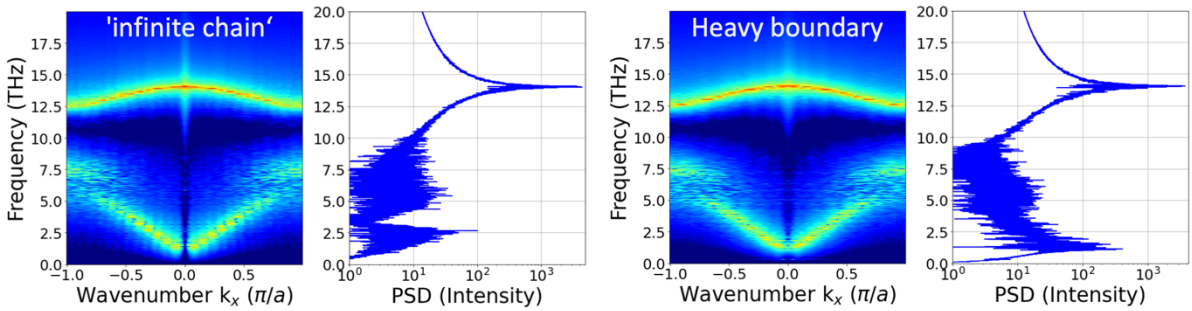
Supplementary Figure 2. 1D phonon Su-Schrieffer-Heeger (pSSH) model, consisting on 52 mass chain of alternating spring constants κ_1 and κ_2. For the topological (trivial) case κ_1 = 3000kJ/mol·nm^2 (1000kJ/mol·nm^2) and κ_2 = 1000kJ/mol·nm^2 (3000kJ/mol·nm^2). For the heavy boundary case the first and last mass are equal to 100au and the rest are 1au. For the infinite case all the masses are equal to 1au.



Supplementary Figure 3 1D polyyne model, consisting on 52 carbon atoms of alternating spring constants κ_s = 899560 kJ/mol·nm^2 and κ_t = 2127146 kJ/mol·nm^2, and equilibrium distances d_s = 1.362 Å and d_t = 1.200 Å. For the topological (trivial) case the bond next to the first and last atoms is κ_t (κ_s). For the heavy boundary case the first and last atoms have an atomic mass are equal to 36au and the rest are 12au. For the infinite case all the masses are equal to 36au.

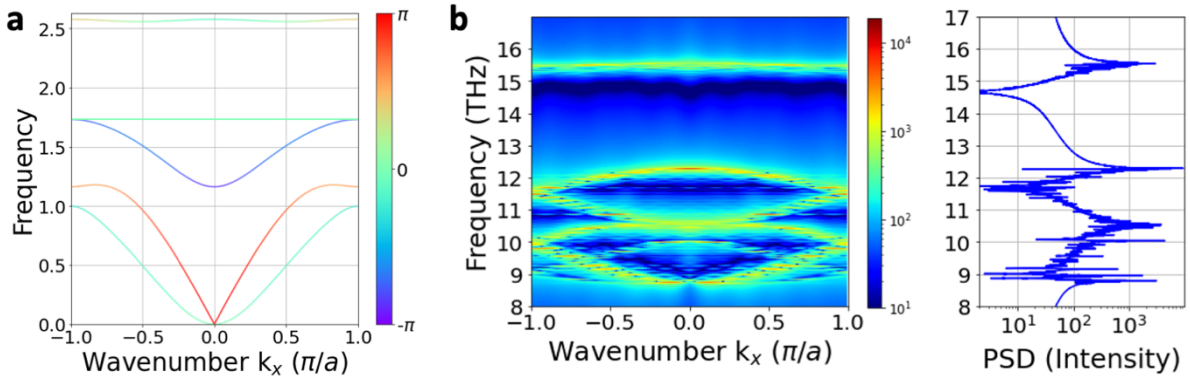


Supplementary Figure 4. aSSH model at equilibrium for a finite angle θ=15° between the masses and the substrate.

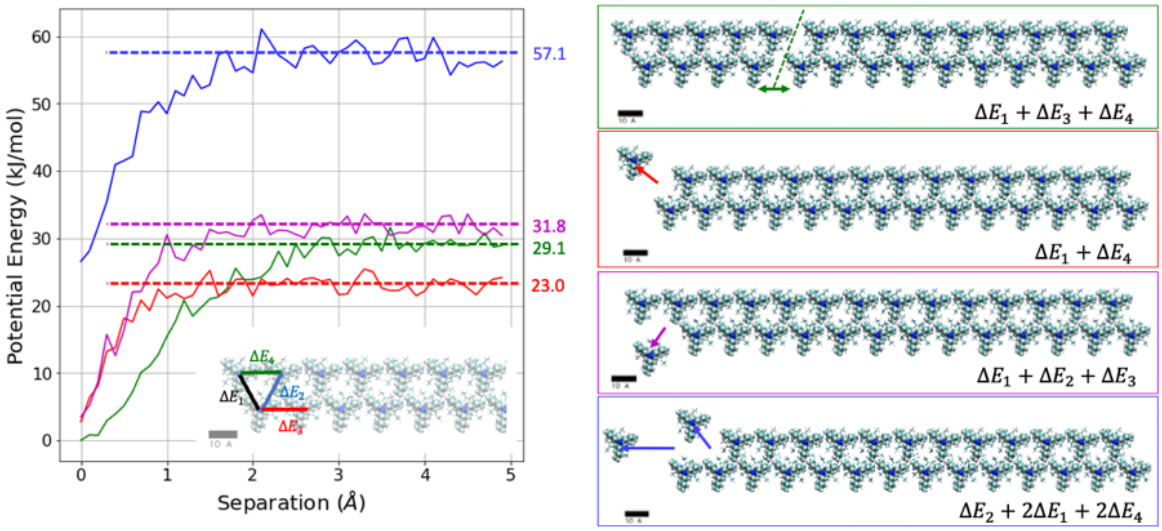


Supplementary Figure 5. Trivial case of aSSH model (Figure 2 of main text). The system consists of 52 mass chain of alternating spring constants κ_1 and κ_2. For the topological (trivial)

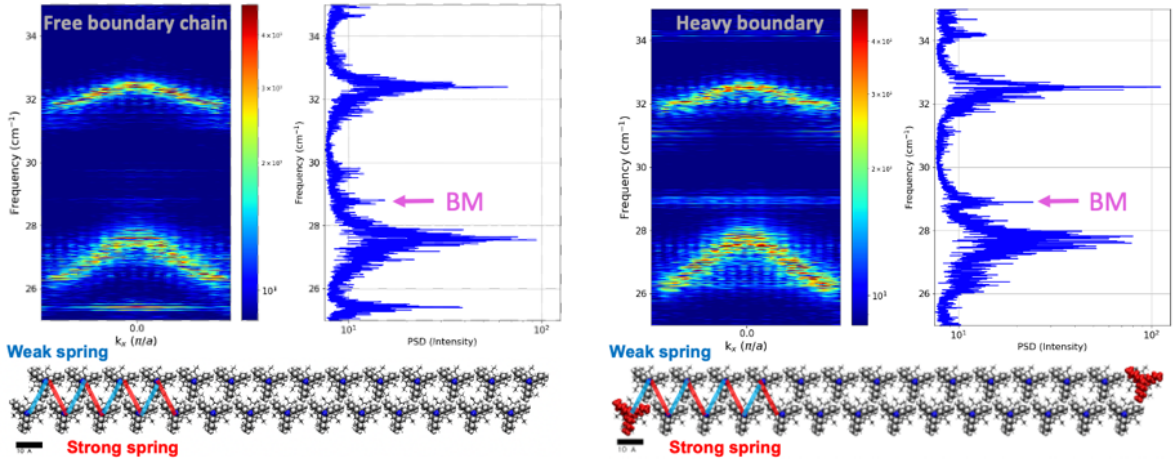
case κ_1 = 3000kJ/mol·nm (1000kJ/mol·nm) and κ_2 = 1000kJ/mol·nm (3000kJ/mol·nm), while κ_3 = 1000kJ/mol·nm for all masses. For the heavy boundary case, the first and last masses are equal to 100au and the rest are 1au. For the infinite case, all the masses are equal to 1au. The substrate is harmonically restricted in 3D by a κ_{3D} = 10^6 kJ/mol·nm in all three dimensions. The masses are restricted to move in a plane with a harmonic restriction of κ_y = 10^6 kJ/mol·nm.



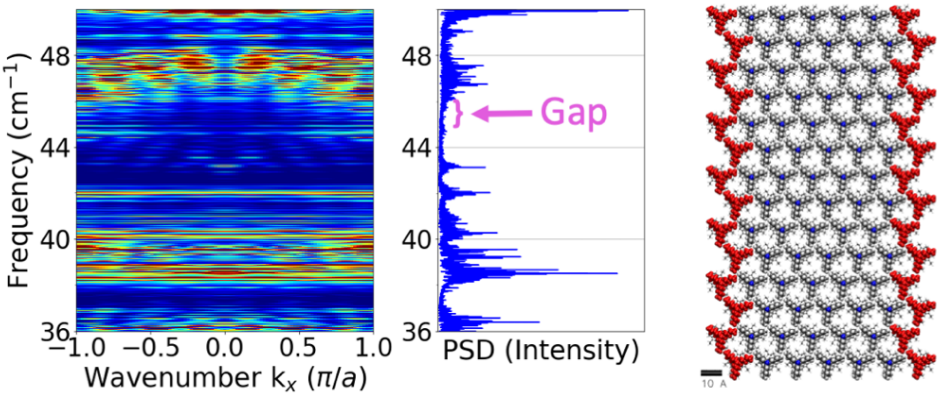
Supplementary Figure 6. Trivial case of daSSH model (Figure 4 of main text). **a.** The dynamical band structure on the left is generated using the parameters $\kappa_1 = 1$, $\kappa_2 = 3$, $\kappa_3 = 3$ and $\kappa_4 = \kappa_5 = 0.5$, $\theta = \alpha = 60°$ and $m_c = 1$. **b.** Molecular dynamics (MD) phonon band structure of the heavy boundary case for a system of 52 masses. Bond parameters: κ_1 = 1000kJ/mol·nm (3000kJ/mol·nm in Figure 4) and κ_2 = 3000kJ/mol·nm (1000kJ/mol·nm in Figure 4), κ_3 = 3000kJ/mol·nm, and $\kappa_5 = \kappa_6$ = 500kJ/mol·nm. The first and last masses are equal to 100au and the rest are 1au.



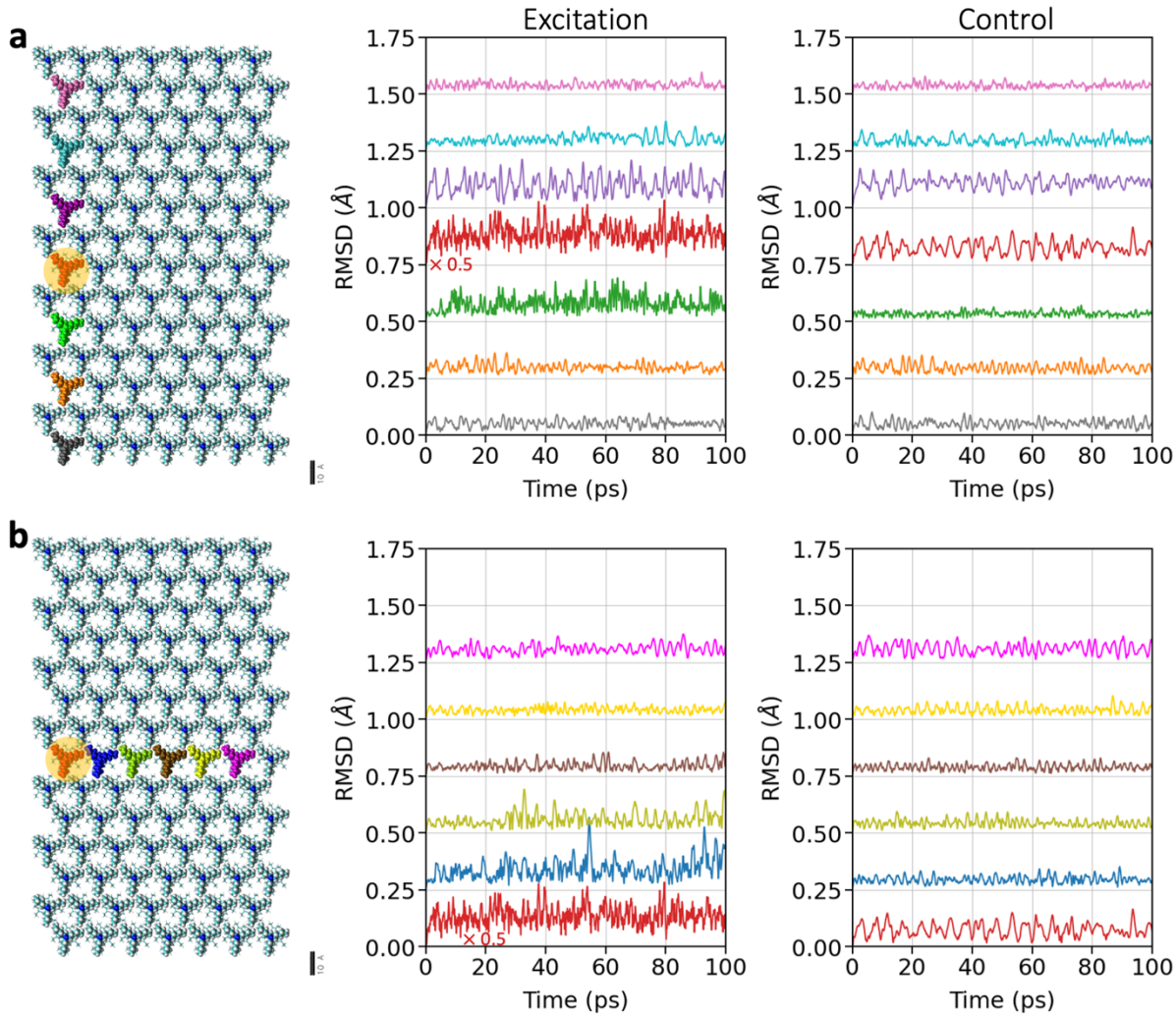
Supplementary Figure 7. Equivalent-spring constants from the realization of the daSSH model. From the dissociation energy plot, the interaction energies were calculated and equivalent springs constants determined. ΔE_1 = 14.73 kJ/mol, ΔE_2 = 10.99 kJ/mol, ΔE_3 = 6.04 kJ/mol, ΔE_4 = 8.33 kJ/mol. The atomistic top view in the inset starts with strong spring. The CHARMM potential energy was calculated every 0.1Å following the minimization algorithm L-BFGS.



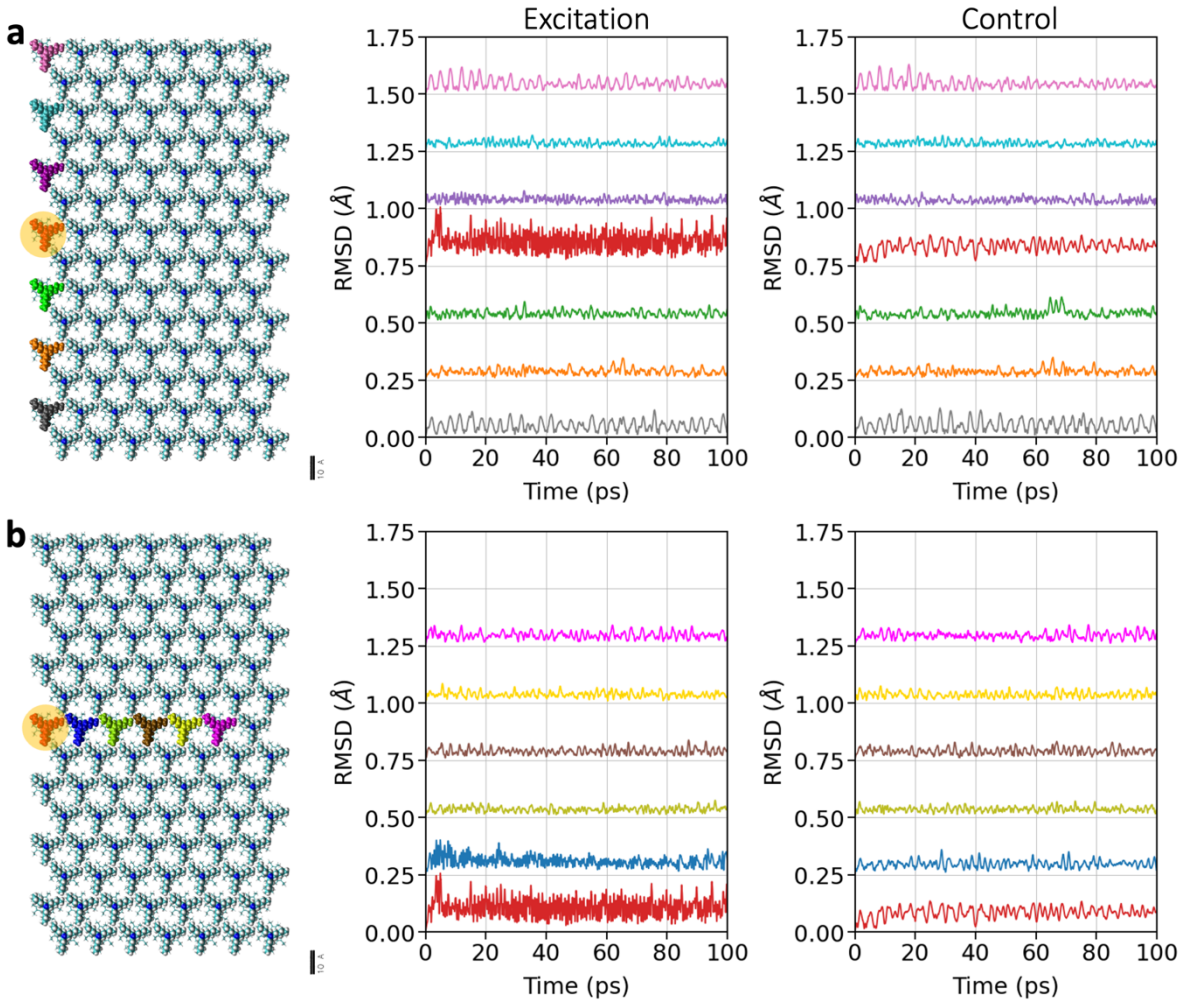
Supplementary Figure 8. Trivial case for the Figure 4 of the main text. In both cases, the molecular chain starts with a weak spring. We identified a boundary mode in both free boundary and heavy boundary.



Supplementary Figure 9. Trivial case for the Figure 5 of the main text. Here the heavy boundary is next to a weak spring in alternating rows of the ribbon. We identified a gap in the region of interest.



Supplementary Figure 10. Root mean square distance (RMSD) fluctuations on the coloured molecules as an effect of the excitation of the red molecule circled in yellow in the molecular diagrams. **a.** Along the boundary and **b.** along the bulk. Right panels show the control experiment using the same initial velocities as in the excitation. The MD simulation conditions are the same as in Figure 5 of the main text.



Supplementary Figure 11. RMSD fluctuations on the coloured molecules as an effect of the excitation of the red molecule circled in yellow in the molecular diagrams. **a.** Along the boundary and **b.** along the bulk. Right panels show the control experiment using the same initial velocities as in the excitation. The MD simulation conditions are the same as in Figure 6 of the main text.

References:

1. Lubensky, T. C., Kane, C. L., Mao, X., Souslov, A. & Sun, K. Phonons and elasticity in critically coordinated lattices. *Rep. Prog. Phys.* **78**, 073901 (2015).