Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary information

Disorder Limits the Coherent Phonon Transport in Two-Dimensional Phononic Crystal Structure

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I. Unit cell configuration

The CNPnC structures with different porosities are respectively composed by each type of the holey unit cells (Figure S1b-e). The unit cells are constructed by ordered removing the carbon atoms in pristine C_3N (Figure S1a), while simultaneously ensuring that the hole edge atoms are nitrogen and the edge pattern is zigzag. These intrinsic requirements limit only four clusters of carbon atoms (six carbon atoms in each cluster) which are denoted as yellow color in Figure S1a can be removed. It is noted that the yellow atoms distributing in the up and down boundary are regards as one cluster due to the periodic boundary condition. In our study, the porosity is defined as $N_R/N_P \times 100\%$, where N_R and N_P are the number of removed atoms and the total number of atoms in the pristine C_3N (all atoms in unit cell before removal), respectively. Figure S1b-e denote the unit cells of the CNPnCs with different porosity ranging from 6.25% to 25%. The D-C_3N structure, on the other hand, is constructed by randomly placing the same number of the specific holes (hole pattern) in each block (dashed box in Figure S2) to ensure the same porosity as that of the CNPnC structure. Figure S2 shows a schematic figure of both CNPnC and D-C_3N for the porosity fixed as 12.5%, in which the structures are divided into multiple segments (denoted by the red dashed box) in length direction. In each segment, there are 12 possible positions for placing the hole pattern, in order to create the specific zigzag holes. As shown in Figure S2a, the CNPnC is created by periodic arrangement of 6 vertical holes in each segment. On the other hand, 6 holes are randomly placed among 12 possible positions in each segment to construct the D-C_3N structure (shown in Figure S2b).

II. Molecular dynamics simulations

To establish a temperature gradient, the atoms at two ends (1 nm) are coupled with Langevin heat bath at temperature T_L and T_R, respectively (shown in Figure S2). In our simulations, we set $T_L = T_0 (1 + \Delta)$ and $T_R = T_0 (1 - \Delta)$, where T_0 is the average temperature and Δ is the normalized temperature difference. To study the temperature effect on the coherent phonons' transportation, we vary T_0 from 300 K to 800 K and fix $\Delta = 0.03$.

In our simulations, after the system reaching the steady state, the cumulative energy ΔE added/subtracted to the heat source/sink region are recorded for 5 ns. The time step is set as 0.5 fs. By applying the linear fitting to the raw data of the cumulative energy ΔE, the energy change per unit time ($\Delta E/\Delta t$) is obtained, which is used to calculate the heat flux $J = \Delta E/(\Delta t \cdot S)$. The results presented here are averaged over 3 independent simulations with different initial conditions, and the error bar is obtained from the standard deviation of different runs.

III. The spectral phonon transmission coefficient

The spectrum phonon transmission coefficient $T(\omega)$ is calculated as[1]

$$T(\omega) = \frac{q(\omega)}{k_B \Delta T}, \tag{S1}$$

where k_B is the Boltzmann parameter and ΔT is the temperature difference between the two Langevin thermostats. Here $q(\omega)$ is the frequency dependent heat current across the imaginary cross-section, which can be calculated as[2]

$$q(\omega) = \frac{2}{t_s} Re \sum_{i \in L} \sum_{j \in R} \left\langle \mathbf{F}_{ij}(\omega) \cdot \mathbf{v}_i(\omega)^* \right\rangle, \quad \text{(S2)}$$

where t_s is the simulation time, and $\mathbf{F}_{ij}$ is the inter-atomic force on atom i due to atom j. Here, "L" and "R" denotes the left and right segment, respectively, which are located at two sides of the imaginary cross-section.

IV. Phonon dispersion relation calculation

The phonon dispersions of CNPnCs with different porosities (shown in Figure S3) along the transverse direction, from **Γ** (0, 0, 0) to **X** (1, 0, 0), are calculated by lattice dynamics implemented in GULP[3]. Obviously, the phonon dispersions become less folded and flattened in CNPnC with the larger porosity, thus the group velocity are significantly increased as the porosity increases, leading to the increase of thermal conductivity.

V. Phonon wave packet simulation

For CNPnC and D-C_3N structure, the length ratio between the pristine C_3N and the CNPnC/D-C_3N region is 1:2. Periodic boundary conditions are used for all directions. Before the wave packet simulation, each structure is relaxed with NPT ensemble at 0.1 K. The wave packet was formed via linear combinations of the vibration normal modes as following

$$\mu_{il,\alpha} = A\varepsilon_{i\alpha,\lambda} e^{ik_0(x_l - x_0)} e^{-(x_l - x_0)^2/\eta^2}, \quad \text{(S3)}$$

where $\mu_{il,\alpha}$ is the αth displacement component of ith atom in the lth unit cell, A is the amplitude of the wave packet, and $\varepsilon_{i\alpha,\lambda}$ is the αth eigenvector component of eigen-mode λ for the ith atom. The wave packet has the wavevector k_0 and is centered around x_0 in the coordinate space. The parameter η is used to define the spatial width of the wave packet. To initialize velocities, we added time dependence to Equation S3 and differentiated it as

$$v_{il,\alpha} = -i\omega_{\lambda} u_{il,\alpha} , \quad \text{(S4)}$$

To investigate the low frequency phonons' transportation behaviors in the CNPnC and D-C_3N structure, we choose the transverse acoustic (TA) phonon mode of pristine C_3N as the excited wave packet with the wavevector $k_0 = 0.2*(2\pi/a)$, where a is the lattice constant. The corresponding frequency is 5.6 THz and the amplitude A = 0.01 Å. We have also tested other polarizations with the same wave vector and found qualitatively similar results as shown in Figure 3.

Reference

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2 K. Sääskilahti, J. Oksanen, J. Tulkki and S. Volz, *Phys. Rev. E*, 2016, **93**, 052141.

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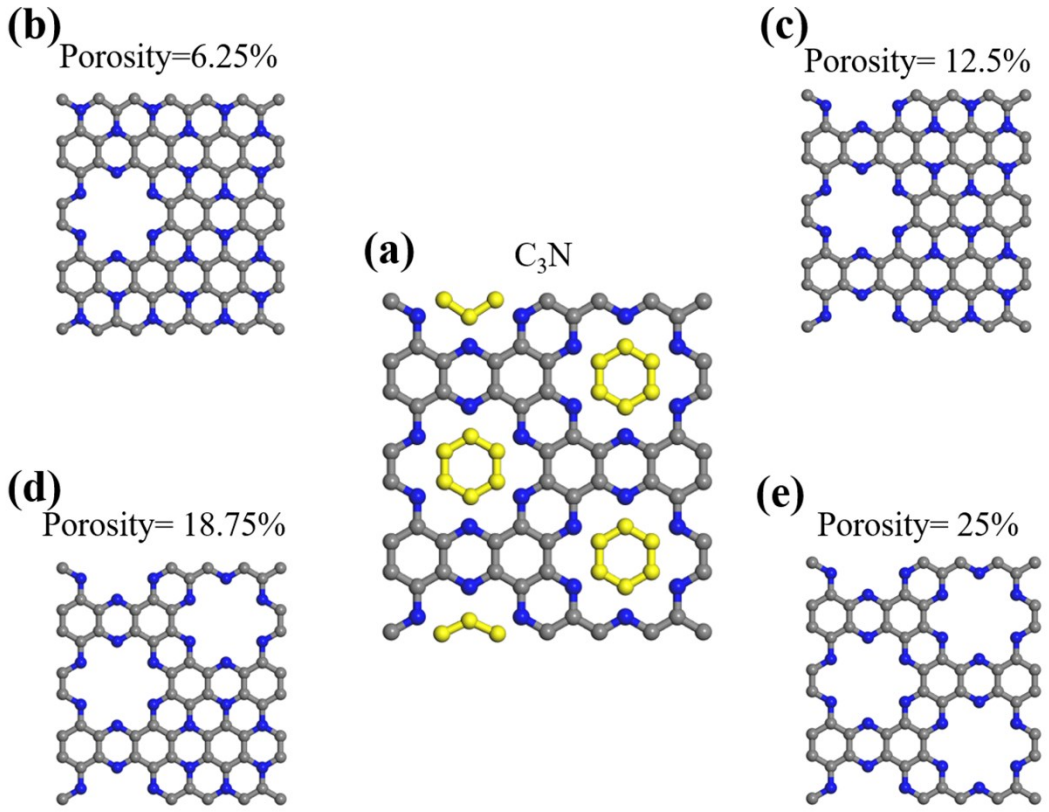

Figure S1. **Unit cell configuration for CNPnC structures.** (a) The schematic of the unit cell of the pristine C_3N. The yellow atoms denote that can be removed atoms. (b)-(e) The unit cells the CNPnCs with different porosity ranging from 6.25% to 25%.

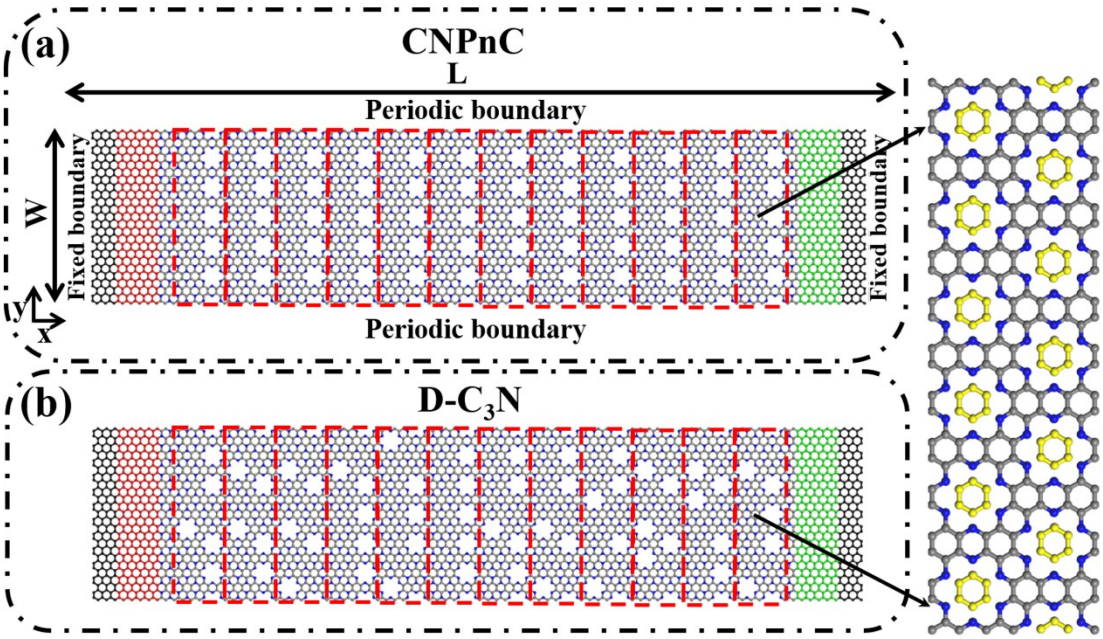

Figure S2. Schematic simulations set up of the CNPnC and D-C3N. (a) and (b) are the schematic simulations set up of the CNPnC and D-C_3N, respectively. Here, the porosity is fixed at 12.5%. Fixed boundary conditions (black atoms) are used along the length (L) direction, while periodic boundary conditions are used along the width (W) direction. The red and green cluster of atoms denote the high and low temperature, respectively. The inset on the right side highlights the 12 possible positions for placing the same hole pattern.

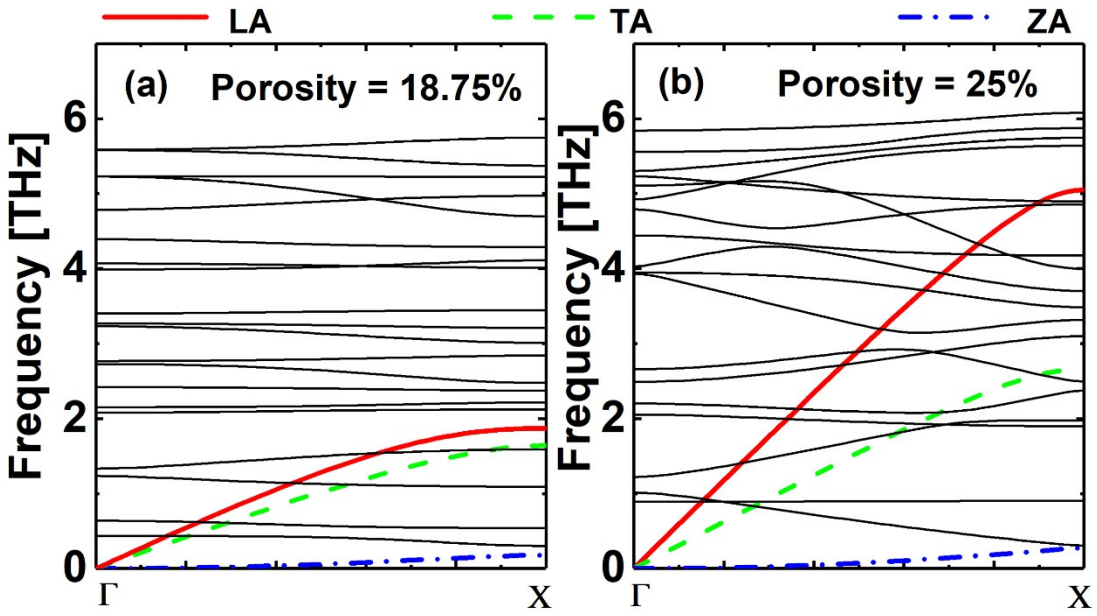

Figure S3. **Phonon dispersion relation for CNPnCs with different porosity.** (a), (b)Low frequency part of the phonon dispersion curves of CNPnCs along ΓX direction with porosity as 18.75% and 25%, respectively.