Supplemental Material

Origin of anisotropic negative Poisson's ratio in graphene

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1. The complex behavior of NPR in graphene along zigzag direction

For the small variation of NPR curve for *zigzag case* (Fig. 3) in the main text, we would like to provide a detailed explanation as below: For the small peaks around ~37% and ~62% for zigzag case, the reason can be clearly understood intuitively from the evolution trends of b_2 and $b_1\cos(\theta)$. Specifically, with the strain increasing, both b_1 and θ increase [Fig. 4(a)], while the b_2 decreases [Fig. 4(b)]. As for the lattice constant in armchair direction [$2(b_2+b_1 \cos(\theta))$], the generally faster increase of b_1 than θ leads to the increase of its projection [$b_1\cos(\theta)$)] as shown in Fig. 4(b). There appear small peaks for the applied strains of ~37% and ~62%, respectively, which *match well* with the small variations along the overall curve for zigzag case (Fig. 3). The complex changes in $b_1\cos(\theta)$ not only overwhelms the decreased b_2 at large strains (Fig. 4(b)), leading to the overall increased lattice constant along the armchair direction and thus the NPR, but also lead to the small variation (at ~37% and ~62%) appeared in overall NPR curves. Note that above nonlinear structure evolutions are dominated by the nonlinear functions of $\cos(\theta)$ or $\sin(\theta)$ with the gradually increased strain, which directly correspond to the complex variations of Poisson ration for the zigzag case.

2. Phonon dispersions of graphene under tensile strain

We further provide a quantitative analysis of whether the predicted NPR in graphene can be realized based on above calculated results. Fig. S1 gives the phonon dispersions of graphene under tensile strains along zigzag and armchair directions, respectively. As shown in the Fig. S1, it is verified that the graphene under strain (0~20%) along armchair direction is thermodynamically stabile based on the calculated phonon dispersions with no imaginary frequency, which indicates that the predicted NPR in graphene under strain 18% along armchair direction is realizable. While for the strains along zigzag direction, the thermal stability of graphene can maintain as strains increased up to 25%, which is smaller than the threshold of NPR appearance in the zigzag direction. As for the imaginary frequency appeared under further larger strains, the possible reason might be lie in that the phonon dispersions are obtained by force calculation in absolute 0 K, and only the harmonic properties are considered in the calculations. If the effect of finite temperature or high-order anharmonicity beyond the harmonicity are fully considered, the part of imaginary frequency could be probably eliminated, which, however, are far beyond the scope of study and could be conducted in future works.

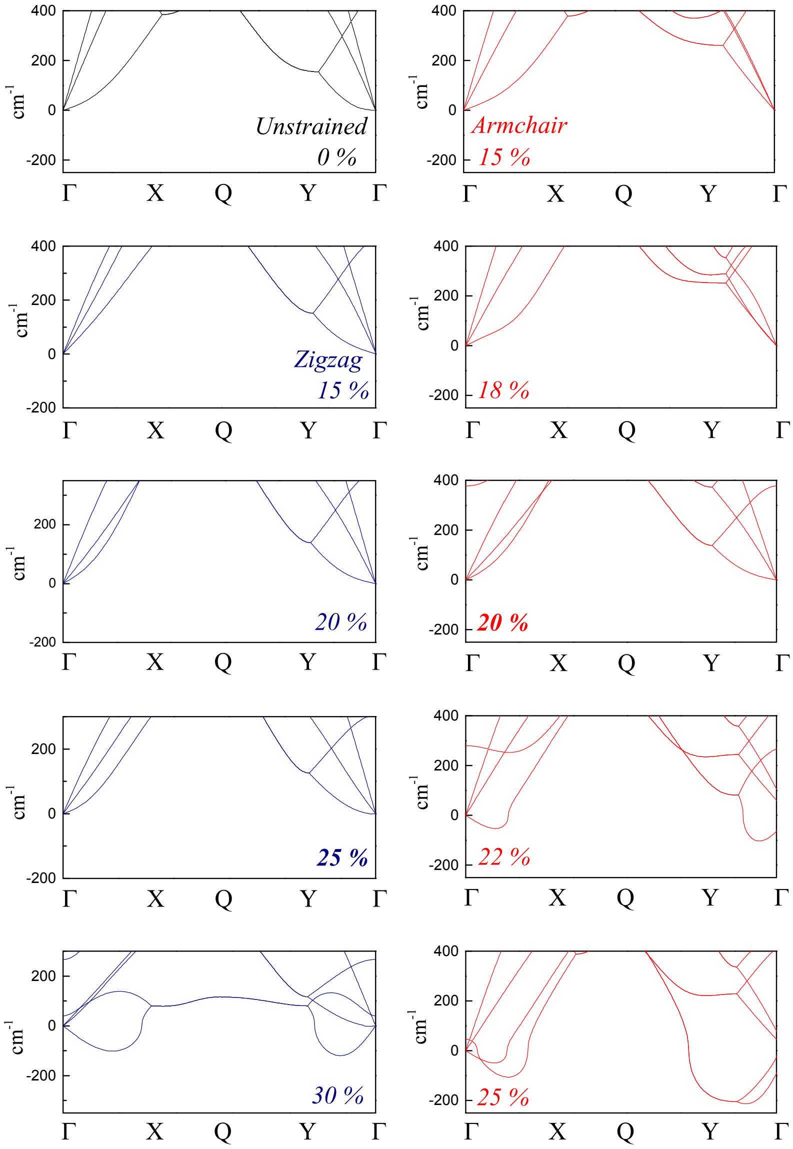

Figure S1: Phonon dispersions of graphene under tensile strains along zigzag and armchair directions, respectively. The *k* point labels Γ, X, Q, Y correspond to (0,0,0), (0.5,0,0), (0.5,0.5,0), (0,0.5,0), respectively, in fractions of the reciprocal lattice vectors. For the zigzag case, there is no imaginary frequency of phonon dispersion as the strain increased to 25%, while for the armchair case, the mode around Γ point has become imaginary as the strain increased to 22%.