Supplementary Information

The consistent behavior of negative Poisson's ratio with interlayer interactions

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1. On the fundamental mechanism

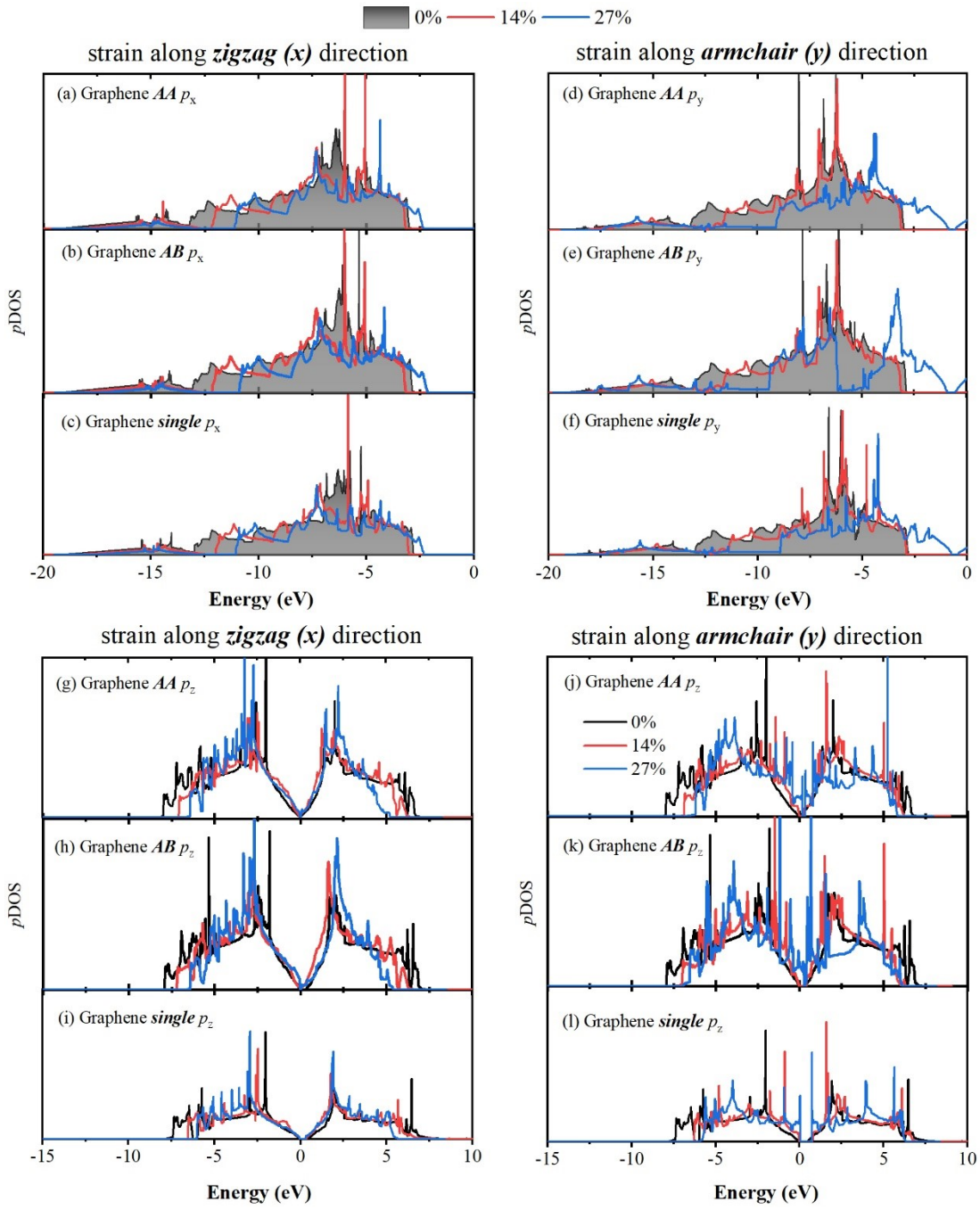

Figure S1: (a, b, c) The evolution of p_x when strain is applied along the zigzag direction (x). (d, e, f) The evolution of p_y when the strain is applied along the armchair direction (y). (g, h, i) The evolution of p_z when the strain is applied along the zigzag direction (x). (j, k, l) The evolution of p_z when the strain is applied along the armchair direction (y). (a, d, g, j), (b, e, h, k), and (c, f, i, l) illustrate the situation of AA bilayer, AB bilayer and single-lawyer graphene, respectively.

To get insight into the fundamental mechanism, we further study the evolution of orbital

projected density of states (*p*DOS). It is well known that the C-C σ bonds come from the hybridized C- p_x /p_y orbitals, and the solo C- p_z orbital forms the π bonds and electronic Dirac cone.[1] Thus, we study p_x, p_y and p_z for bilayer and single-layer graphene when strain is applied. As shown in Figure S1, it has almost same value and trend in some hybridized orbitals. When strain is applied along zigzag direction (x), p_x orbital decreased slightly then increased near the valance band maximum (VBM), which is almost same trend as p_y when strain is applied along the armchair direction (y). That's probably because the direction of orbitals is same as the direction of uniform strain applied. Moreover, p_z barely change near VBM during stretching. Thus, the different responses in orbital perpendicular to the direction of the strain applied cause different responses in NPR when strain is applied along zigzag and armchair direction.

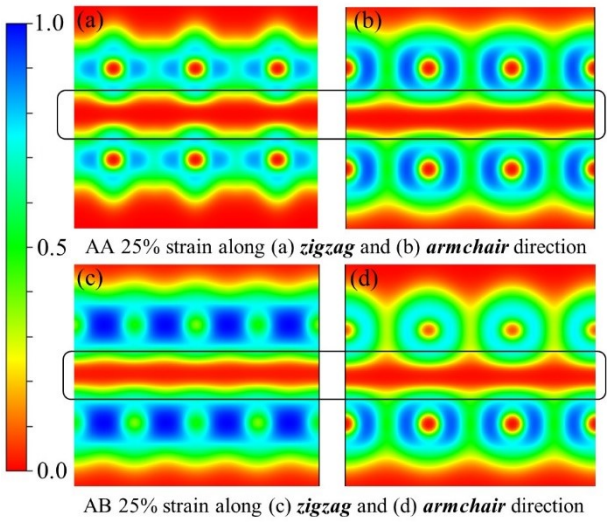

Figure S2: The evolution of the electronic localization functions (ELF) of (a, b) AA and (c, d) AB bilayer graphene when 25% strain is applied along (a, c) zigzag and (b, d) armchair direction, respectively. The areas framed by the black edges represent the interlayer position of AA and AB bilayer graphene.

As shown in Figure S2, there is low density of electrons in the interlayer position of AA and AB bilayer graphene probably because the electrons are restricted in in-plane area.[2] Thus, the interlayer interaction is weak and then have little effect on in-plane NPR, casing the consistency of bilayer and single-layer graphene. Furthermore, low density of electrons doesn't mean no electrons in the interlayer position of bilayer graphene. And these electrons unrestricted in 2D plane will slightly decrease the in-plane geometry variation on bilayer graphene.

2. Verification of the structural stability

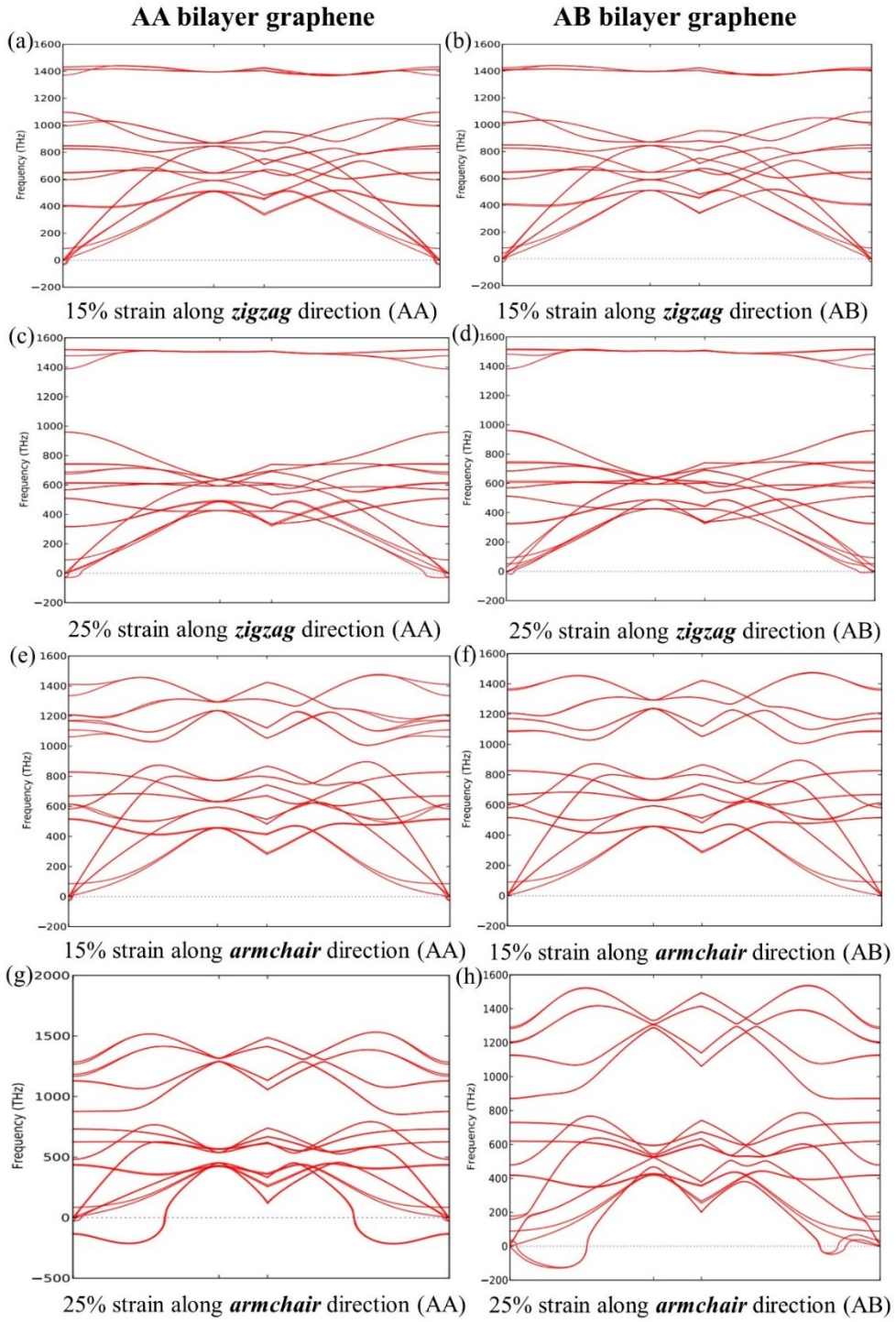

Figure S3: Phonon dispersions of (a, c, e, g) AA and (b, d, f, h) AB bilayer graphene when strain ((a, b, e, f) under 15% strain, (c, d, g, h) under 25% strain) is applied along (a, b, c, d) zigzag or (e, f, g, h) armchair direction, respectively.

We use phonon dispersions to verify the structural stability for AA and AB bilayer and single-layer graphene. As for single-layer graphene, it is the strongest material ever measured and it can sustain a large strain (≥25%)[3,4]. However, the mode has become imaginary in single-layer graphene when 25% strain is applied along armchair direction[1], as same as Fig. S3(g, h) in bilayer graphene under the same strain condition. Fig. S3 shows that AA and AB bilayer is stable when strain is applied along zigzag direction while the mode has become imaginary when strain near 25% is applied along armchair direction, probably because bilayer graphene structure break down near 30% strain is applied along armchair direction.

Reference

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