## **Supplementary Material**

## 1. The amplitude of applied shear strain during LCF test loading

LCF has two fundamental characteristics [1]: plastic deformation in each cycle, and finite endurance. Moreover, it has been demonstrated that the plastic flow in GCuNL is mainly determined by copper layers [2]. Thus, before LCF simulations, it is necessary to investigate the mechanical behavior of copper under large strain. The stress-strain curves of (100)- and (111)-copper under shear loading are illustrated in SFig 1. According to the simulation results, the amplitude of applied strain  $\varepsilon$  is set near the start point of plastic flow zone, where  $\varepsilon_{xy}$ =0.15 for shear LCF simulations. The flow stress of copper in plastic flow zone at 300 K is about 2.5 GPa, which is consistent with previous research [3-5].



SFig 1. The shear stress-strain curves of copper in the (100) and (111) planes under shear loading.

## 2. The evolution of dislocation in copper layer

The movies of dislocation evolution in (100)- and (111)-stacking GCuNL composites are uploaded as attached files "100-dislocation.avi" and "111-dislocation.avi", since the graphene chirality does not influence the dislocation evolution.

For (100)-stacking GCuNL composites, dislocations are generated near the graphenemetal interfaces, and then rotated and inclined between graphene layers. The length of these dislocations is completely limited by the repeat layer spacing. The movement of these dislocations is translation in the *xy*-plane under cyclic shear loading, and their positions are relatively "stable" in a cycle, leading to a stable shear stress during stage II.

For (111)-stacking GCuNL composites, parallel slip bands are generated in the *xz*-plane. Although the positions of these slip bands vary with LCF loading cycles, the percentage of atoms in slip bands does not change. The activation of dislocations, which is considered as ideal shear in copper [3, 6], cannot result in large fluctuation of shear stress in GCuNL composites.

## References

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