Comment on "Structural and Electronic Properties of *T* Graphene: A Two-Dimensional Carbon Allotrope with Tetrarings"

In a recent Letter, Y. Liu *et al.* [1] reported an *ab initio* calculation on the structural and electronic properties of *T* graphenes (TGs). The authors claimed to have found two new stable forms of a two-dimensional (2D) carbon allotrope with tetrarings. By analyzing equilibrium height difference in TG, they showed two distinct carbon allotropes: planar TG with zero height difference (Δz) and buckled TG with 0.55 Å height difference. Our detailed calculations show that the buckled TG is not a stable form as they suggested, and the maximum height difference of the fully relaxed buckled TG is 2 orders of magnitude smaller than their value, which means that the fully relaxed buckled TG cannot be distinguishable with planar TG.

We performed *ab initio* calculations Perdew-Burke-Ernzerhof generalized gradient approximation (PBE GGA) [2] on the density functional with a project-augumented-wave method as implemented in VASP (Vienna *ab initio* simulation package) [3]. The vacuum slabs of 15 Å were inserted and momentum space integration was performed using $18 \times 18 \times 8$ Monkhorst-Pack *k*-point mesh [4]. The electronic wave functions were expanded with plane waves up to a kinetic-energy cutoff of 660 eV as in Ref. [1] For a given lattice constant with fixed vacuum slab thickness, internal coordinates starting from buckled TG were fully optimized until the residual Hellmann-Feyman forces were $<10^{-4}$ eV/Å.

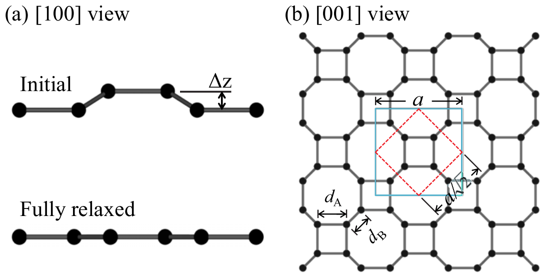

FIG. 1 (color online). (a) Initial and fully relaxed geometry of the buckled TG. (b) Unit cell of planar (red dotted line) and buckled (blue solid line) TG.

The equilibrium lattice constant for our calculation is ~4.875 Å, which is consistent with the previous calculation in Ref. [1]. However, height differences (Δz) of the fully relaxed TG are ~0.0002 Å, which is 250 times smaller than that of Y. Liu *et al* [1]. Such a small height difference in the fully relaxed TG suggests that the buckled TG is not a stable form and relaxed into the planar TG spontaneously, as shown in Fig. 1(a) [5]. Figure 1(b) depicts the unit cell of two different TGs. As one can clearly see, the buckling is related with a $\sqrt{2} \times \sqrt{2}$ reconstruction pattern of planar TG. Y. Liu *et al.* [1] reported the lattice constant of 3.42, 4.84 Å for planar and buckled TG. $\sqrt{2} \times 3.42$ is approximately ~4.836 Å, which is 99.9% of the lattice constant of their buckled TG (4.84 Å). If buckled TG has a height difference of 0.55 Å (which is ~10% of lattice constant), the bonding length of each carbon bonding might be enlarged ~10%, which is extremely larger for carbon bonding. Second, the buckled TG and the planar TG have a nearly similar carbon bonding character except buckling. It is hard to believe that similar carbon bonding produces two different stable structures with almost identical carbon connectivity. The third point is that the buckled TG is essentially a quasi 2D structure. Therefore, a phonon structure and an electronic structure should be examined in a full 3D brillouin zone, not in a 2D brillouin zone as they reported [1]. We also have tested the stability of buckled TG in local density approximation and Heyd-Scuseria-Ernzerhof range separated hybrid functional [5–7] and the main physical results are the same as PBE GGA. It should be noted that both previous paper by Y. Liu *et al.* [1] and our current calculation cannot completely rule out the possibility of existence of metastable buckled TG, in a larger finite system with long-range interaction and finite temperature. In conclusion, by using the fully relaxed calculation and the simple bonding arguments, we have shown that the proposed buckled TG [1] is not stable against spontaneous structural transformation into the planar TG.

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