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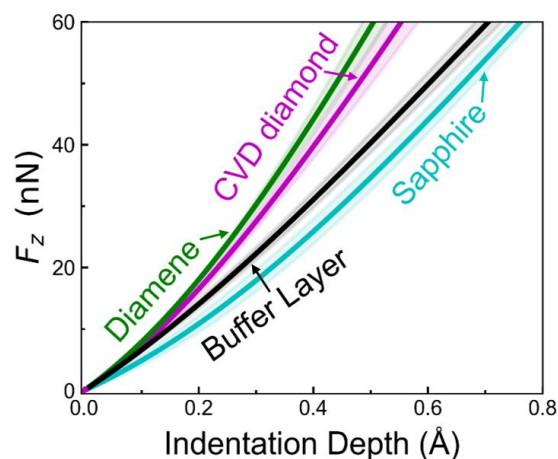
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# Pressure-induced formation of a new ultra-hard, ultra-thin diamond-like structure: from graphene to *diamene*.

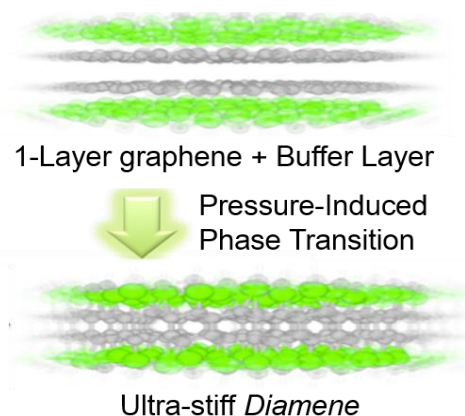
The study of the nanomechanical behavior of atomically thin graphene has led to the discovery of fascinating novel mechanical properties, such as its outstanding in-plane stiffness and out-of-plane flexibility, as well as unique frictional and wear characteristics at the nanoscale. Here we further explore the elastic properties of supported 2D films of graphene by the use of modulated Å-Indentation, a technique capable of achieving sub-Å indentations depths during force-indentation measurements. We experimentally demonstrate that at room temperature and under localized indentation pressure, a single layer of graphene on top of a carbon interface layer (buffer layer), both epitaxially grown on the Si-face of a SiC(0001) substrate, exhibits transverse stiffness superior to that of CVD bulk diamond (Fig.1), is resistant to perforation by a diamond indenter and shows a reversible change in electrical conductivity. Density functional theory (DFT) calculations indicate that the 1-layer graphene film undergoes a pressure-induced reversible phase transformation to a new ultra-stiff, ultra-hard diamond-like structure, named *diamene*, followed by  $sp^2$ -to- $sp^3$  chemical bond transitions. Furthermore, we find that the formation of ultra-stiff *diamene* is exclusive of 1-layer epitaxial graphene plus buffer layer films grown on SiC(0001), and it is not observed in the buffer layer alone, nor in thicker epitaxial graphene (2-layer or more) nor in exfoliated graphene films of any thickness on SiO<sub>2</sub> substrate.

## References

- [1] Gao, Yang, et al. *Nature Nanotechnology* 13.2 (2018): 133.
- [2] Cellini, Filippo, et al. *FlatChem* 10 (2018): 8-13.



**Figure 1:** Experimental force vs indentation curves showing that *diamene* (1-layer graphene + buffer layer) exhibits stiffness larger than CVD bulk diamond.



**Figure 2:** Illustration of the pressure-induced phase transition of the 1-layer graphene + buffer layer film to an ultra-stiff diamond-like structure (*diamene*).