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**Multifunctional Crumpling and Unfolding of Large-Area Graphene** XUANHE ZHAO, Duke University — Crumpled graphene films of atomic thickness are used in diverse applications including electronics, energy storage, composites, and biomedicine. While it is known that the degree of crumpling strongly affects the properties of graphene and the performance of graphene-based devices and materials, in existing technology it is not possible to fold and unfold crumpled graphene films in a controlled manner. Here we present a new approach, investigated by joint experiment, atomistic simulation and theory, to control reversible crumpling and unfolding of large-area graphene, achieved by harnessing mechanical instabilities of graphene adhered on highly pre-strained polymer substrate. By relaxing the pre-strain in the substrate in a particular order, we crumple graphene films into tailored self-organized hierarchical structures that mimic super-hydrophobic leaves. The degree of crumpling in graphene is controlled by stretching/relaxing the substrate. The reversible crumpling and unfolding of graphene films enables us to fabricate large-area conductive coatings and electrodes capable of giant stretchability, high transparency, super-hydrophobicity, and tunable wettability. We further demonstrate the use of novel graphene-polymer laminates as artificial muscles.

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