2D Diamond Superstructures in Interlayer-Bonded Twisted Bilayer Graphene: Thermomechanical Properties from Atomic-Scale Simulations

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The thermomechanical properties of graphene derivatives and metamaterials fabricated through chemical functionalization and defect engineering of graphene sheets can be tuned by tailoring the structural features of these 2D materials [1]. Such exceptional properties make these materials particularly appealing for numerous technological applications. Here, we report a systematic computational study on the thermomechanical behavior of superstructures of 2D diamond nanocrystals embedded in interlayer-bonded twisted bilayer graphene (TBG) with commensurate bilayers. These superstructures are formed by patterned chemical functionalization, with hydrogenation chosen for this study, which induces interlayer covalent C-C bonding in TBG. The generated 2D diamond superstructures have the periodicity of the TBG Moiré pattern, with diamond nanocrystal sizes that can be varied by varying the extent and pattern of hydrogenation. These structures are fully defined by specifying parameters that include the commensurate bilayer's twist angle, the interlayer bond pattern and density, and the concentration of sp^3-bonded C atoms that serves as a metric of the nanodiamond fraction in the superstructures [2].

We report results for the mechanical behavior of these graphene-diamond nanocomposite superstructures based on molecular-dynamics (MD) simulations. We have conducted uniaxial straining tests and report the dependence on the diamond fraction of the elastic properties, ultimate tensile strength, and fracture strain of the 2D diamond superstructures, demonstrating their remarkable mechanical strength. We find that a brittle-to-ductile transition occurs in these superstructures with increasing the 2D diamond concentration beyond a critical level [2]. The underlying ductile fracture mechanism is mediated by void formation, growth, and coalescence, in contrast to the typical brittle fracture of graphene. Furthermore, we have analyzed the response of these superstructures to nanoindentation loading [3]. We find that superstructures with a less-than critical 2D diamond concentration exhibit a strongly nonlinear inelastic response to indentation up to the onset of fracture mediated by a non-dissipative and non-recoverable stiffening effect that results in large hysteresis loops in indentation loading/unloading cycles. Finally, we report results for the lattice thermal conductivity of these superstructures. We find that the thermal conductivity is reduced significantly with increasing the 2D diamond fraction, which also makes these 2D materials very promising for thermal management applications.

References:

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