Control of interlayer interactions in multilayer graphene via nanodiamond insertion

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Abstract

Monolayer graphene exhibits unique electronic properties with a linear band dispersion, whereas multilayer graphene loses this linearity due to strong interlayer interactions. In this study, we experimentally constructed multilayer graphene structures in which nanodiamonds were inserted as nanoscale spacers to control the interlayer interaction. Raman spectroscopy (increase in the G'/G ratio and splitting of the G peak) confirmed the suppression of interlayer coupling, and atomic force microscopy revealed an expansion of the interlayer spacing. Furthermore, the combination of molecular dynamics simulations and a continuum model showed that the areal density and diameter of nanodiamonds determine the interlayer distance and the final morphology, and that the configuration with the lowest system energy is ultimately selected.

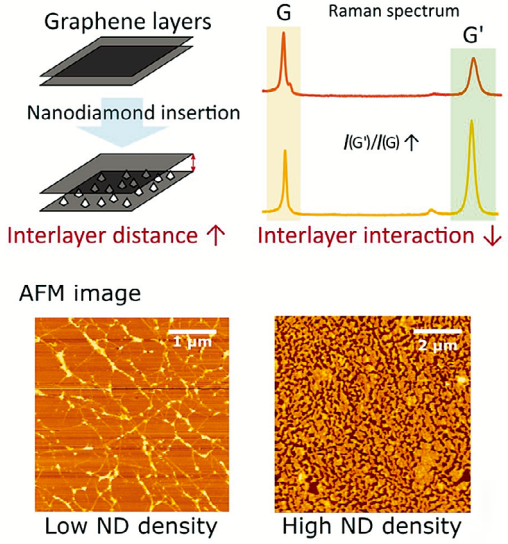
Background & Results

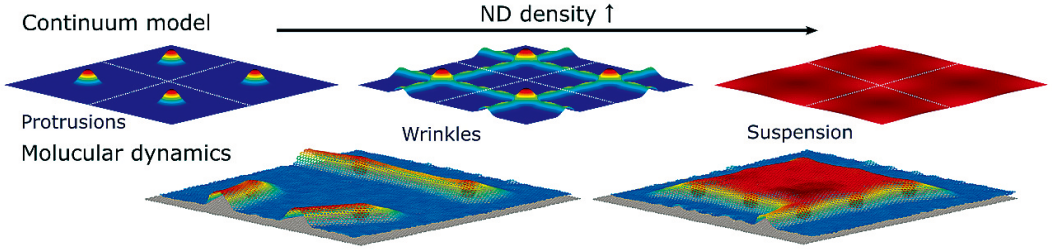
Monolayer graphene has a linear band structure, but multilayer graphene naturally adopts AB stacking as the energetically stable configuration, which enhances interlayer interactions and alters the electronic structure from that of the monolayer. To obtain monolayer-like characteristics at larger scales, active control of interlayer interactions in multilayers is critical. In this work, we inserted nanodiamonds between graphene layers as artificial "nanoscale spacers," experimentally constructing multilayer graphene structures with expanded interlayer spacing and suppressed interlayer interaction. Alternating stacking of graphene and nanodiamonds was achieved by combining transfer of monolayer graphene with spin coating of nanodiamonds. Raman spectroscopy (higher G'/G ratio and G-peak splitting) clearly indicated weakened interlayer coupling. AFM visualized the formation of suspended upper graphene and confirmed the increased spacing. These results serve as direct experimental evidence that interlayer interactions can be controlled by nanodiamond insertion.

Molecular dynamics simulations further clarified that nanodiamond areal density and particle diameter determine the interlayer distance and the final morphology: larger and denser nanodiamonds yield a larger spacing and facilitate suspension of the upper layer. Moreover, a continuum model showed that, between two possible stable morphologies (suspension and adhesion), the configuration with lower system energy is selected. The model predicts the existence of a critical nanodiamond density, consistent with experimental observations. In summary, this study presents principles and design guidelines for nanoscale structural control toward property engineering of multilayer graphene.

Significance of the research and Future perspective

This study demonstrates that inserting nanoscale spacers enables us to design the interlayer architecture of multilayer graphene and recover monolayer-like properties even in a multilayer system. This provides a pathway to reconcile large-volume scalability with desirable properties for graphene applications. Future work may integrate spacer selection, spatial arrangement control, and external stimuli to tailor optical response, electrical transport, and nonlinear response, thereby expanding application opportunities.





Patent	
Treatise	Ding, Mingda; Inoue, Taiki; Enriquez, John Isaac et al. Experimental and theoretical investigation of nanodiamond insertion on the interlayer interaction in multilayer stacking graphene. Carbon. 2024, 229, 119464. doi: 10.1016/j.carbon.2024.119464 Ding, Mingda; Inoue, Taiki; Enriquez, John Isaac et al. Reduction of interlayer interaction in multilayer stacking graphene with carbon nanotube insertion: Insights from experiment and simulation. The Journal of Physical Chemistry C. 2023, 127, 23768. doi: 10.1021/acs.jpcc.3c06132
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