GRAPHENE AND 2DM VIRTUAL CONFERENCE & EXPO

Graphene online 2020 October 19-23

On the Vibrational and Thermal Properties of Amorphous Graphene: an atomistic investigation

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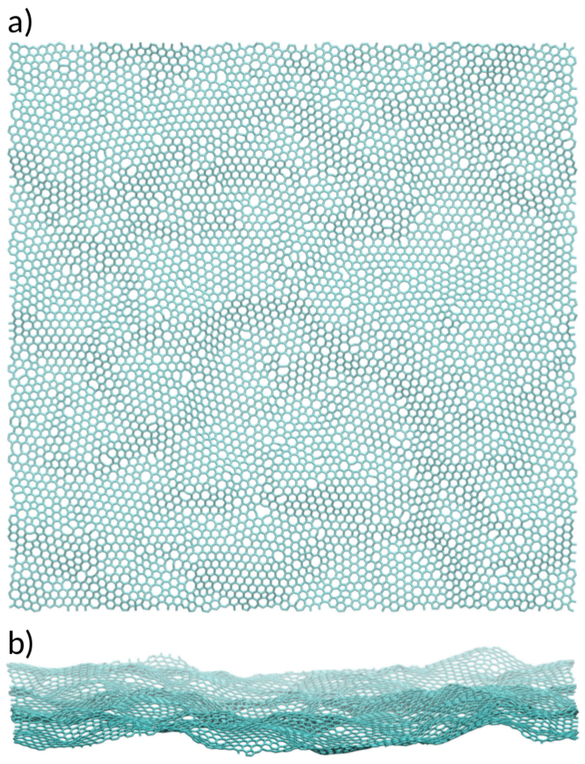
Introduction

The recent demonstrated possibility to synthesize wafer-scale two-dimensional amorphous carbon monolayers, structurally dominated by sp2 hybridization has initiated a new platform of low-dimensional materials to explore as alternative forms of membranes with enhanced chemical reactivity which could serve as coating materials [1,2]. Furthermore, their physical properties could pave the way for their use for permeation and diffusion barriers in magnetic recording devices and flexible electronics. It is clear that the properties of amorphous graphene (a-G) are a consequence of both the nature and the amount of disorder in the atomic configuration of the material. Being the amount of disorder mostly related to the synthesis parameters in the fabrication process, it represents a tunable parameter which can be ideally used to control the quality of the material and its physical properties. This poses the need for a systematic investigation of the properties of a-G as a function of the degree of amorphousness. While the impact of the level of randomness induced by topological disorder on the electronic properties of a-G has already been extensively investigated, thermal properties have received a relatively minor attention. In this study we employ classical molecular dynamics to provide a systematic analysis of the vibrational modes in a-G samples as a function of the degree of amorphousness; the effect of structural disorder on the density of states, the participation ratio, and the intrinsic character of the vibrational modes is given. Finally, the contribution of each single mode to the overall thermal conductivity is calculated, highlighting the most effective vibrations responsible for heat transport in a-G.

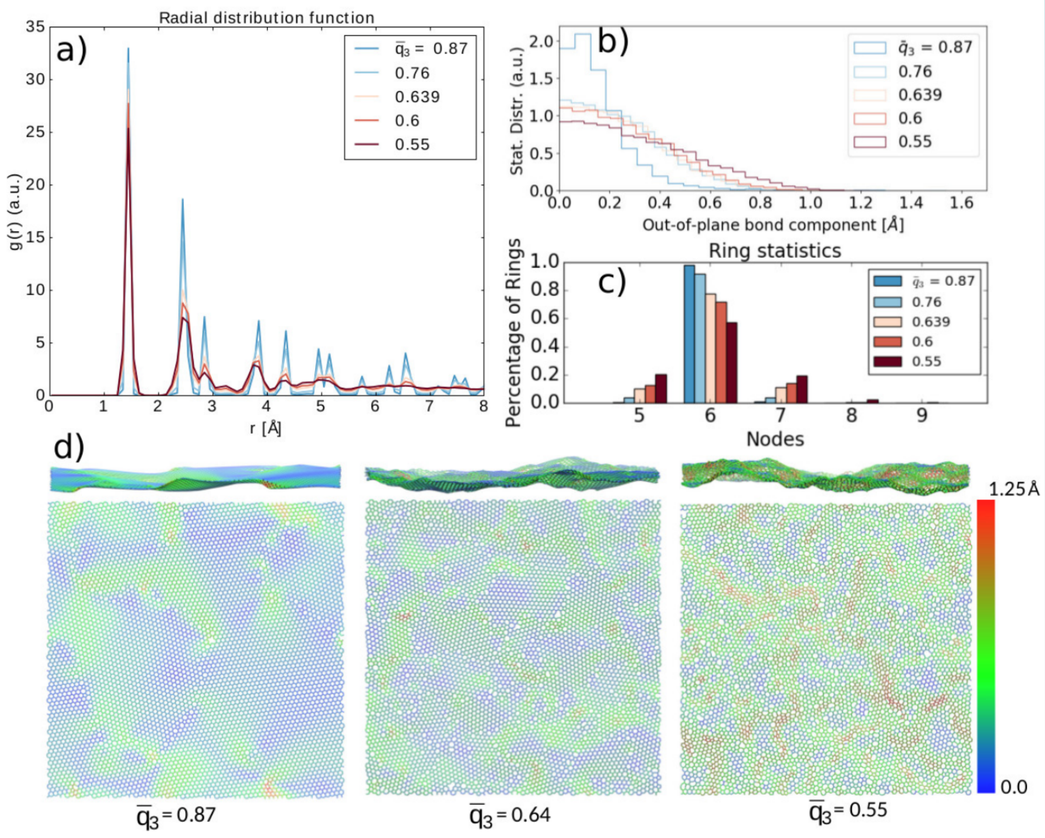
Amorphous Graphene: Structural Characterization

Amorphous Graphene Samples built via classical Molecular Dynamics: Structural Analysis

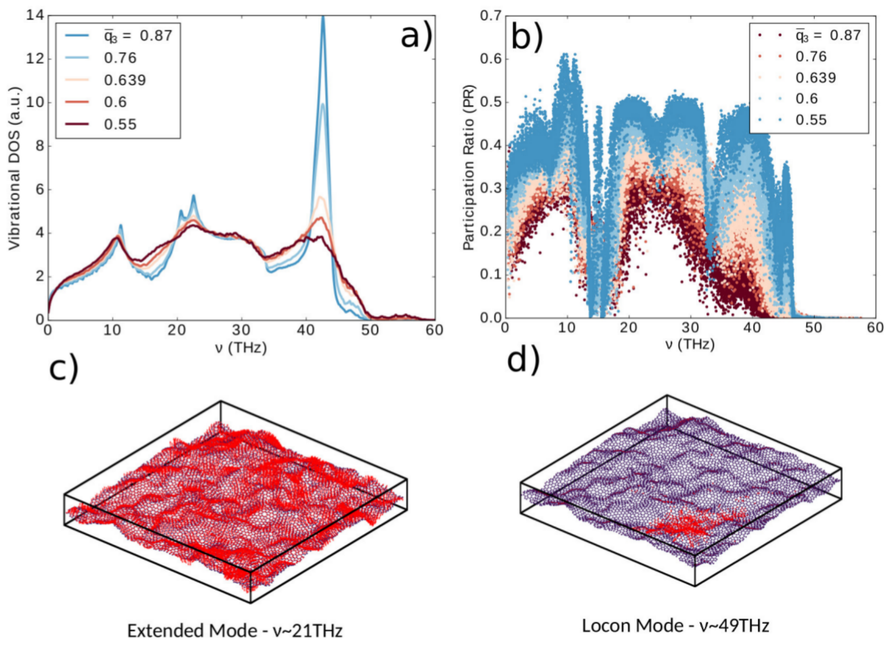


Sample Generation. To generate a-G stystems a crystalline graphene sample is gradually heated in the constant-volume, constant-temperature (NVT) ensemble with a rate of 50 K/ps up to a temperature of 10000 K at which the system is found in a two-dimensional liquid melt state. The system is further gradually cooled down from with different cooling rates in the range [50K/ps, 1000K/ps]. Depending on the cooling rate applied, amorphous systems are obtained differing in the amount of disorder or, equivalently, in the degree of amorphousness. Specifically, the faster the cooling process, the more disordered the final system.

The radial distribution functions g(r) (RDFs) of the samples as a function of the degree of amorphousness (a), quantified via the averaged triatic order parameter q3. **Smaller values of q3 denote more amorphous systems.** The same position of the first peak in the RDFs is found in any sample, suggesting that the average C-C bond length in the first coordination shell is similar in all the structures. There is a significant broadening in the second and third peaks of the RDF, denoting an increased disorder in the structure of a-G. a-G structures exhibit somestatic intrinsic ripples: specifically, strongly wrinkled structures correspond to the more amorphous ones.



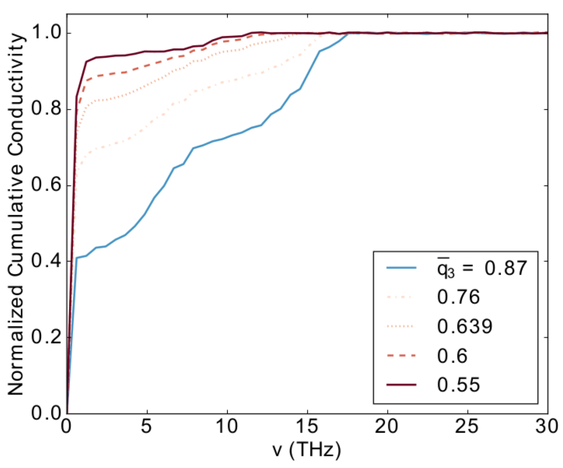
Impact of Disorder on Vibrational and Thermal Properties



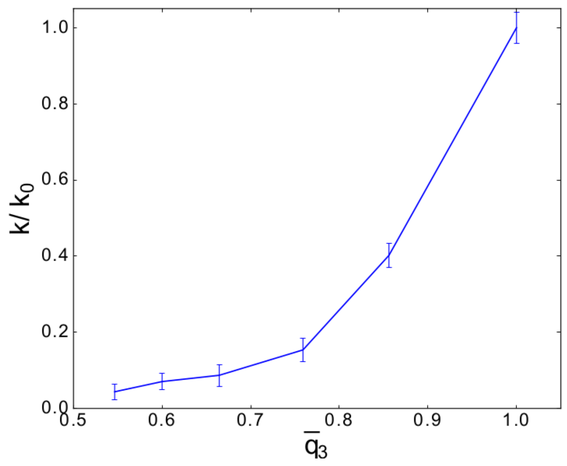
The VDOSs present peaks which are clearly reminiscent of the phonon bands of graphene. The higher the degree of amorphousness, the broader the peaks which eventually merge into a continuous VDOS without frequency gaps. Participation Ratio (PR) decreases with amorphousness -> stronger mode localization with increasing disorder.

Acknowledgments: "ModElling Charge and Heat trANsport in 2D-materIals based Composites - MECHANIC". Reference number: PCI2018-093120 funded by Ministerio de Ciencia, Innovación y Universidades

Vibrational and Thermal Analysis



k0: thermal conductivity of pristine graphene @ 300K



When indreasing disorder, mostly low-frequency extended modes contribute effectively to k. Thermal conductivity reduction of more than two orders of magnitude is found wrt to pristine graphene.

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References

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