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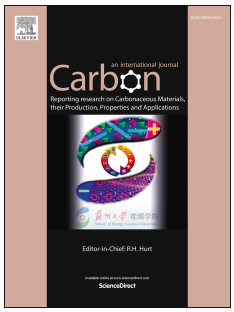

Thermal transport in nanocrystalline graphene investigated by approach-to-equilibrium molecular dynamics simulations

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Thermal transport in nanocrystalline graphene investigated by approach-to-equilibrium molecular dynamics simulations

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Abstract

Approach-to-equilibrium molecular dynamics simulations have been used to study thermal transport in nanocrystalline graphene. Nanostructured graphene has been created using an iterative process for grain growth from initial seeds with random crystallographic orientations. The resulting cells have been characterized by the grain size distribution, by the number of atoms in each grain and the number of atoms in the grain boundary. Introduction of nanograins with a radius of gyration of 1 nm has led to a significant reduction in the thermal conductivity to 3% of the one in single crystalline graphene. Analysis of the vibrational density of states has revealed a general reduction of the vibrational intensities, in particular of the mode at 53 THz. This mode is additionally found to shift to lower frequencies with decreasing grain size. Vibrations of grain boundary atoms have led to the evolution of a mode at 30 THz. Thermal conductivity has been evaluated as a function of the grain size with increasing size up to 14 nm and it has been shown to follow an inverse rational function. The grain size dependent thermal conductivity can be described by a function assuming a connection in series of conducting elements and resistances for thermal transport.

1. Introduction

Graphene is a material of great interest from a technological point of view owing to its high carrier mobility and optical transparency. Furthermore, it provides excellent physical strength and is chemically inert towards many environmentally ubiquitous species [1–4]. Such properties paved the way for graphene's career in applications such as field effective transistors, Schottky junction diodes or as transparent and flexible displays in various electronic devices [5–9].

Miniaturization of electronic devices is an important topic in recent research. A major issue for such devices is the required heat dissipation which becomes more and more important with decreasing dimensions. Crystalline graphene is a promising material in this regard. It inheres both high electronic carrier mobility and excellent thermal conductivity which additionally provides the function as heat dissipator.

Fabrication of single-crystalline graphene sheets, however, is not trivial. Common fabrication methods include epitaxial film growth and chemical vapor deposition (CVD), where the latter is promising in particular for large-scale production. In all methods, defects are introduced in the crystalline structure resulting from limitations of the kinetic properties in the growth process and defects of the substrates. These defects limit the size of single-crystalline domains rendering the graphene sheets rather polycrystalline. The introduction of grain boundaries and defects in such polycrystalline graphene can drastically change electronic and thermal transport properties of the system. In fact, previous experimental measurements of the thermal conductivity in graphene resulted in values ranging from 600 to 10000 W/mK [10]. This wide range in the value of thermal conductivity demonstrates its high sensitivity to various process conditions, sample size, and structural features including defects and grain boundaries.

Characterization and control of the structural properties of polycrystalline graphene, such as the grain size, and their effect on the electrical and thermal transport properties are thus of major interest in research of graphene in electronic and thermoelectric devices. The thermal conductivity of graphene is in particular interesting due to its 2D nature. The differences to 3D graphite have been discussed recently [11] as well as the length dependent thermal conductivity of 2D graphene [12–15]. Regarding the effect of environmental parameters, a significant difference in the thermal conductivity has been found for suspended ($\sim$1600 W/mK) [16] and SiO_2 supported (600 W/mK) [17] graphene. Furthermore, thermal conductivity of CVD produced graphene has been shown to be reduced by 30% when wrinkles are introduced in the structure [18]. A systematic investigation of the size effect of single-crystalline domains on the thermal conductivity of graphene based on experiments, however, is still matter of investigation.

Due to the complexity of generating trustworthy polycrystalline simulation cells, little research has been done so far on the theoretical investigation of the properties of

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