

Two-Dimensional Thermal Transport in Graphene: Intrinsic vs. Extrinsic Effects

Alexander A. Balandin

Nano-Device Laboratory, Department of Electrical Engineering and Materials Science and Engineering Program, Bourns College of Engineering, University of California, Riverside, CA 92521 U.S.A.

balandin@ee.ucr.edu

Recent years witnessed a rapid growth of interest of the scientific and engineering communities to thermal properties of materials. The increasing importance of thermal properties is explained both by practical needs and fundamental science. Heat removal has become a crucial issue for continuing progress in electronic industry [1]. The knowledge of how well and how fast the material conducts heat becomes essential for design of the next generation of integrated circuits. From another side, efficient thermoelectric energy conversion requires finding materials, which simultaneously have high electrical conductivity but strongly suppressed thermal conductivity K . Material's ability to conduct heat is deeply rooted in its atomic structure, and knowledge of thermal properties can shed light on many other materials' characteristics. Thermal conductivity of materials changes when they are structured on a nanometer scale. The latter can happen because of the intrinsic effects, e.g. phonon dispersion and velocities of the low-dimensional crystal differ from those in bulk [2], or extrinsic effects such as increased phonon - rough boundary scattering when the feature size of nanostructure becomes comparable to that of the phonon mean free path (MFP) in a given material [3]. Other extrinsic effects on thermal conductivity include strain in the lattice and phonon scattering on lattice defects or impurities.

The discovery of graphene [4-5] further stimulated the interest to thermal properties because, for the first time, it became possible to study experimentally heat conduction in strictly 2D crystals. The heat conduction in 2D crystals is particularly intriguing because of the theoretically predicted logarithmic divergence of thermal conductivity K with the size of 2D crystal [6-7]. The K divergence in 2D crystals means that unlike in 3D bulk, the crystal lattice anharmonicity alone is not sufficient for restoring thermal equilibrium, and one needs to either limit the system size or introduce disorder to have the physically meaningful finite value of K . To elucidate the physics of heat conduction in graphene, it is important to determine whether thermal transport is mostly limited by the *intrinsic* properties, e.g. by the dimensionality of the lattice and its dynamics, or by the *extrinsic* effects, e.g. phonon scattering from rough interfaces, edges, defects and impurities or graphene – substrate interactions. Heat conduction in the suspended graphene will be closer to the intrinsic phonon transport regime while that in the encased graphene will be closer to the extrinsic transport regime. The first experimental study of the evolution of heat conduction in few-layer graphene (FLG), found that K of suspended uncapped FLG, which is the highest in the single-layer graphene (SLG), decreases with increasing number of the atomic planes n , and approaches the bulk graphite limit (see Figure 1a) [8]. For comparison, the thickness dependence of thermal conductivity of the encased disordered carbon films is also shown [9].

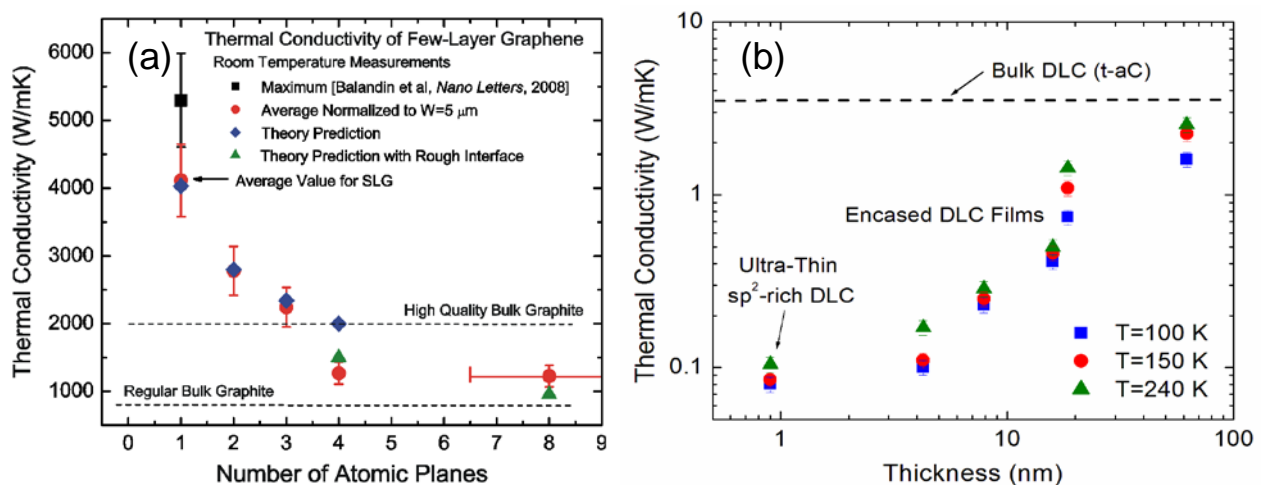


Figure 1: Thermal conductivity of the suspended FLG (a) and in the encased ultra-thin diamond-like carbon films (b) as function of thickness. The figure (a) is adapted from Ref. [8] while the figure (b) is based on the data reported in Ref. [9].

The evolution of heat conduction in “free” suspended FLG was explained by considering the changes in the phonon Umklapp scattering. As n in FLG increases, the phonon dispersion changes and more phase-space states become available for the phonon scattering, leading to K decrease. The phonon scattering from the top and bottom boundaries in suspended FLG with a small thickness is limited if a constant n is maintained over the graphene layer length. The small thickness of FLG ($n < 4$) also means that phonons do not have transverse component in their group velocity farther weakening the phonon scattering from the top and bottom boundaries. In FLG films with $n > 4$ the boundary scattering may increase because of the non-zero transverse component of the phonon velocity and difficulties of maintaining constant n through the whole area of the thicker FLG flakes. These factors can lead to experimental K below the graphite limit of ~ 2000 W/mK. The bulk graphite K recovers for thicker films. The experimentally observed evolution of heat conduction in FLG with the number of atomic planes varying from $n=1$ to $n\sim 4$ [8] is in agreement with the recent calculations performed for the crystal lattices described by the Fermi-Pasta-Ulam Hamiltonians [7]. It was numerically confirmed that K diverges in 2D and 1D anharmonic lattices with the size of the system. At the same time, the anharmonicity is a sufficient condition to have a finite intrinsic K value in 3D crystals [7]. Moreover, it was shown that as the system dimensionality changes from 1D to 2D and 3D, the intrinsic K rapidly decreases from its maximum value in the low-dimensional system to the 3D bulk crystal value. The calculated thermal crossover behavior was consistent with the measured heat conduction evolution in suspended FLG [8].

The situation is entirely different for encased graphene where thermal transport is limited by the acoustic phonon scattering on the top and bottom boundaries and disorder, which is unavoidable when FLG is embedded between two layers of dielectric or other materials. An experimental study [10] found $K \approx 160$ W/mK for encased SLG at $T=310$ K. It increased to ~ 1000 W/mK for graphite films with the thickness $H \sim 8$ nm [10]. Thermal conduction in the encased FLG was limited by the rough boundary scattering and disorder penetration through graphene (the presence of the evaporated oxide on top of graphene is known to cause defects in the graphene layer). Correspondingly, the K dependence on FLG thickness H was similar to other material system where K is extrinsically limited and scales with H . In conventional crystalline thin films, where H is smaller than the phonon MFP, but still much larger than the lattice constant, K grows with H as until it reaches the bulk limit. A similar dependence is observed for encased ultra-thin films of diamond-like carbon (DLC) (Figure 1b). The overall values of K in the encased amorphous carbon films are much smaller than those for encased FLG, as expected for the disordered materials, but the $K(H)$ scaling trend is similar. In both, the encased FLG and ultra-thin films of DLC, the scaling may not follow exactly the same dependence as in crystalline films because of the influence of disorder and changes of the film material properties themselves. The described evolution of heat conduction in FLG and differences in the phonon transport regimes are important for graphene’s applications in electronics, e.g. high-frequency transistors, and thermal management, e.g. in thermal interface materials.

Acknowledgements

This work was supported, in part, by U.S. ONR award N00014-10-1-0224, SRC-DARPA FCRP Center on Functional Engineered Nano Architectonics (FENA) and DARPA Defense Microelectronics Activity (DMEA) award H94003-10-2-1003. The author thanks S. Roche, A. Geim and K. Novoselov for useful discussions.

References

- [1] C. Lin and K. Banerjee, IEEE Trans. Electron Dev., **55**, 245 (2008).
- [2] A. Balandin and K.L. Wang, Phys. Rev. B, **58**, 1544 (1998).
- [3] T. Borca-Tasciuc, *et al.*, Microscale Thermophys. Eng., **5**, 225 (2001).
- [4] K.S. Novoselov, A.K. Geim, *et al.*, Science, **306**, 666 (2004).
- [5] A.K. Geim and K.S. Novoselov, Nature Mater., **6**, 183 (2007).
- [6] For a review, see S. Lepri, *et al.*, Phys. Rep. **377**, 1 (2003) and references therein.
- [7] K. Saito and A. Dhar, Phys. Rev. Lett., **104**, 040601 (2010).
- [8] S. Ghosh, *et al.*, Nature Materials, **9**, 555 (2010).
- [9] A.A. Balandin, *et al.*, Appl. Phys. Lett., **93**, 043115 (2008).
- [10] W. Jang, *et al.*, Nano Lett., **10**, 3909 (2010).