Enhancement of thermoelectric properties via resonant electronic transport in graphene nanoribbons

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Besides the fascinating discoveries on graphene electronic properties [1, 2], much attention has been recently focused on thermal [3] and thermoelectric [4] properties of graphene structures. The promising results for two-dimensional graphene [5] have suggested also exploring thermoelectric properties of one-dimensional structures as the graphene nanoribbons (GNRs). Recently, it has been predicted that the thermoelectric figure of merit *ZT* can exceed unity in honeycomb chains of carbon atoms [6] and also in long rough edge GNRs [7]. However, detailed information on the potential of short GNRs to provide with a high factor of merit is still missing.

In this work a new way to enhance the figure of merit *ZT* in GNRs is proposed. The dependence of electronic and thermal transport on GNR edge orientation is explored and exploited to suggest a GNR structure with higher *ZT* values compared to the perfect armchair (AGNRs) and zigzag (ZGNRs) nanoribbons. The suggested structure is built by alternating different edge orientations in the ribbons (Mixed-GNR) as presented in Figure 1.a. In the figure are shown a perfect AGNR and a ZGNR with their elementary cells and a MGNR generated by alternating AGNR and ZGNR portions. The enhancement of thermoelectric properties in MGNR is connected to two main phenomena: the resonant tunneling of electrons and the reduction of thermal conductance.

In the analyzed ribbons, the figure of merit *ZT*, defined by the expression $T\,G_e\,S^2 / \kappa$ is evaluated by solving both the electron and phonon transport equations. The simulation of electron transport provides the thermopower *S*, the electronic conductance G_e and the electron contribution to the thermal conductance κ, while the simulation of phonon transport gives access to the phonon contribution to the thermal conductance. Charge transport simulations in the GNRs are developed on the basis of the non-equilibrium Green's functions (NEGF formamlism) and an atomistic tight-binding Hamiltonian is used to model the ribbons [8, 9]. In recent works the NEGF formalism has been successfully applied also to the phonon transport problem. In most of these works, a force constant model (FCM) limited to the fourth nearest-neighbors [10,7] was considered for phonons. Here, we use a similar method with a fifth nearest-neighbors FCM phonon Hamiltonian [11].

To present the enhancement of thermoelectric properties in MGNR the influence of GNR edge orientation on electronic, thermal and thermoelectric properties is investigated. We compare for instance 18-AGNR, 20-AGNR, 18-ZGNR and two 18-MGNRs with different elementary cells. The main simulation results may be summarized as follows: the electronic conductance, the thermal conductance, the thermopower and the figure of merit, calculated at room temperature, are shown in figure 1.b, 1.c, 1.d and 1.e respectively. Electron transport properties of the MGNR are strongly dependent on the fraction *n* of armchair edges and the fraction *m* of zigzag edges (MGNR (*n,m*)). As expected from an NNTB calculation, the ZGNRs have a metallic behavior and the AGNRs have a band gap strongly dependent on the number of dimmers in the elementary cell. In MGNR, the presence of armchair edges induces a band gap opening, and the presence of zigzag edges induces edge-localized electron states. So the armchair edge sections can be seen as barriers for the localized zigzag edge states. This interpretation is confirmed by the resonant oscillation of the electronic conductance and the positive values of the thermopower, which is a classical behavior of multi-barrier channels [12]. If the armchair edge section is narrow, MGNR (1, 1) for instance, the band gap is small and the resonant tunneling effect is not important. If the fraction of armchair edges over the zigzag ones is increased, as shown for an

MGNR (4,1), the width of the gap is enhanced and the resonant phenomenon becomes dominant. To explain this resonant tunneling effect, the atomic LDOS is plotted in Fig. 2 for the chemical potential of 0.3 eV. The figure shows clearly the regions of strong electron density localized at the zigzag edges separated by depleted armchair zones. In addition to this resonant tunneling effect, MGNRs exhibit very low thermal conductance compared to perfect AGNRs or ZGNRs. The phenomenon is probably due to the mismatch of phonon modes between AGNR and ZGNR portions. The resonant tunneling transport provides a rather high value of *S*, which induces a high *ZT* value of more than 0.1, boosted by the low thermal conductance. It is expected that further optimization of GNR structures with appropriate width may result in ZT values exceeding unity at room temperature.

Acknowledgments

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References

[1] A.K. Geim and K.S. Novoselov, Nature materials, **6** (2007) 183-191
[2] A.H.C. Neto et al., Rev. Mod. Phys., **81** (2009) 109-162
[3] A.A. Balandin et al. Nano Letters, **8** (2008) 902-907
[4] Y.M. Zuev, W. Chang and P. Kim, Phys. Rev. Lett., **102** (2009) 096807
[5] D. Dragoman and M. Dragoman, Appl. Phys. Lett., **91** (2007) 203116
[6] X. Ni, G. Liang, J.S. Wang, and B. Li, Appl. Phys. Lett., **95** (2009) 192114
[7] H. Sevinçli and G. Cuniberti, Phys. Rev. B, **81** (2010) 113401
[8] G. Fiori and G. Iannaccone, Electron Device Letters, **28** (2007) 760-762
[9] P. Zhao, J. Chauhan and J.Guo, Nano Letters, **9** (2009) 684-688
[10] Y. Chen et al., J. Phys. Condens. Matter, **22** (2010) 372202
[11] M. Mohr et al., Phys. Rev. B, American Physical Society, **76** (2007) 035439
[12] G. D. Guttman, E. Ben-Jacob and D. J. Bergman, Phys. Rev. B, **51** (1995) 17758-17766

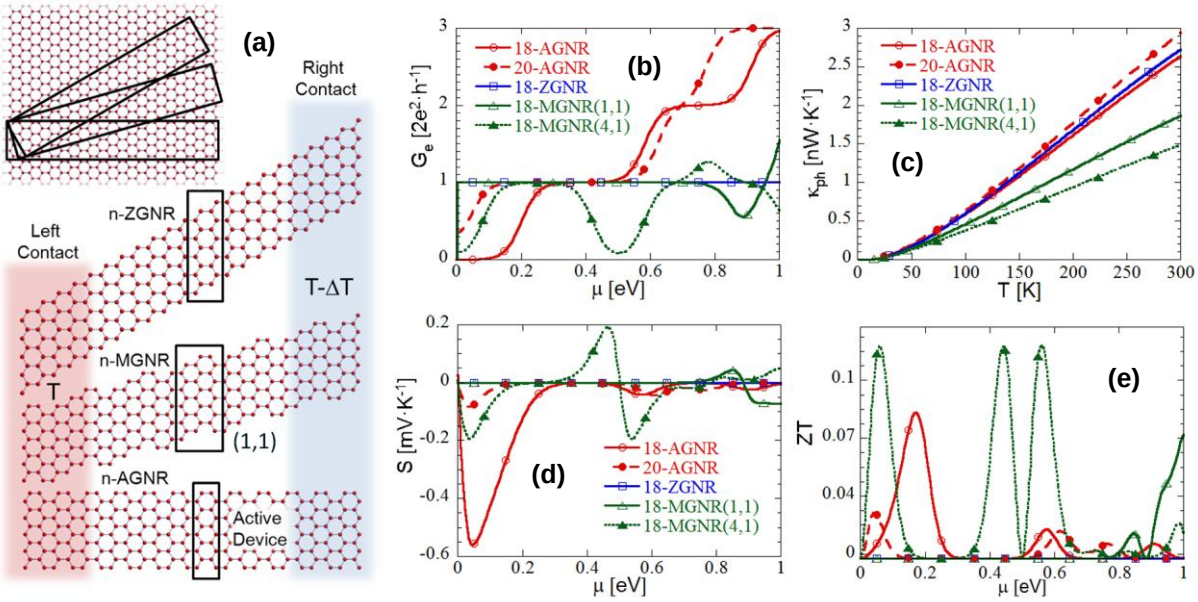

Figure 1. (a) Different edge orientations considered in simulations (b) Electronic conductance G_e as a function of the chemical potential μ, (c) phonon contribution to thermal conductance as a function of the temperature, (d) Seebeck coefficient and (e) factor of merit ZT as a function of chemical potential for a 18-AGNR, 20-AGNR, 18-ZGNR, 18-MGNR with chirality (1, 1) and 18-MGNR with chirality (4, 1).

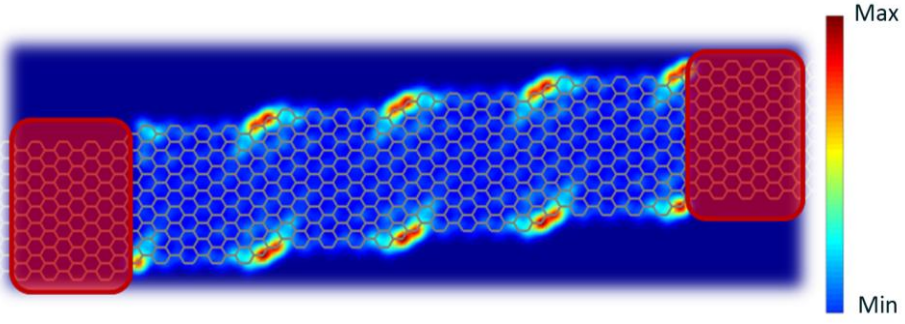

Figure 2. Atomic LDOS of the 18-MGNR with chirality (4,1) calculated at the chemical potential of 0.3 eV.