

DFT Study of Electronic Transport Properties in Supported Armchair Graphene Nanoribbons

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INTRODUCTION

Armchair graphene nanoribbons (AGNR) hold great promise in nano-electronics because of the capability of opening a semiconducting gap in narrow ribbons[1]. However the effective use of AGNR in devices may be limited by structural and chemical modifications from a variety of sources including the support material, edge effects, width variability, doping and defects which all change the trends in the bandgap scaling. In this work we use density functional theory (DFT)[2] simulations to investigate how structural and chemical variability in AGNRs influence electron transport through changes in the band-structure and phonon modes as a function of the type of edge functionalization (H-,H₂-,O-, and OH-) and support material (h-BN, SiO₂, and HfO₂).

EDGE FUNCTIONALIZATION AND SUPPORT MATERIALS

Theoretical calculations predict that the band gap of hydrogen-terminated ribbons of width less than ~15nm is sensitive to the number of atomic layers[3]. The oscillatory dependence of the gap on the number of atomic layers along the ribbon width is attributed to the aromaticity of the graphene: This can be understood through the spatial distribution of the Clar-resonance structures (Clar sextets[4]) that become more localized because of the formation of the edge states, as illustrated in Fig. 1. Chemical functionalization of the edges, defects at the edges and in the bulk of the ribbon modify the aromaticity of the ribbons, and therefore the bandstructure. For AGNRs terminated by single hydrogen atoms the band gap oscillates as a function of the number of carbon atoms along the ribbon width, N, as: Eg(3N+1) > Eg(3N) > Eg(3N+2), as shown in Fig. 2a. *Ab initio* thermodynamic calculation[5] can be used in conjunction with the DFT simulations to look at the stability of different functional groups on the

edges as a function of temperature and partial pressure of surrounding gases. Figure 3 shows a plot the surface free energy as a function of hydrogen chemical potential. As can be seen, terminating the edge carbon atoms with two hydrogen atoms is more thermodynamically stable except at extremely low hydrogen partial pressures. The trend in the band gap scaling changes for ribbons terminated by double hydrogen atoms as: Eg(3N)>Eg(3N+2)>Eg(3N+1), as shown in Fig. 2b. Under actual environmental conditions a combination of single- and double-hydrogen terminations will exist making it difficult to see this scaling trend experimentally, consistent with what has been observed. An analysis of ribbons with various terminations, including oxygen atoms, hydroxyls, and various concentrations of single- and double-hydrogen atoms will be presented. In addition, the low field mobility and ballistic conductance in AGNRs with various surface terminations will be calculated using the Empirical Pseudopotential Method (EP)[6], benchmarked to the DFT results.

The substrate material used to support graphene-based devices can affect electronic transport significantly, often degrading the mobility[5]. In this work we consider three different substrates: h-BN, SiO₂ and HfO₂. Graphene supported by h-BN and SiO₂ (modeled as (001) α -quartz) is shown in Fig 4. Investigations of the coupling between the graphene or AGNR flexural modes and the polar modes of the substrates and its effect on the carrier mobility will be discussed.

REFERENCES

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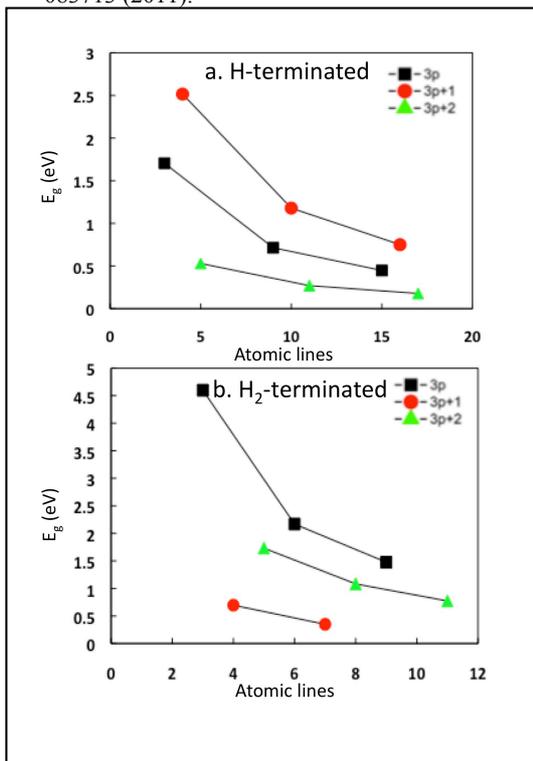


Fig. 1. Band gap of a) H-terminated and b) H₂-terminated edges of AGNR as a function of the number of atoms along the ribbon width.

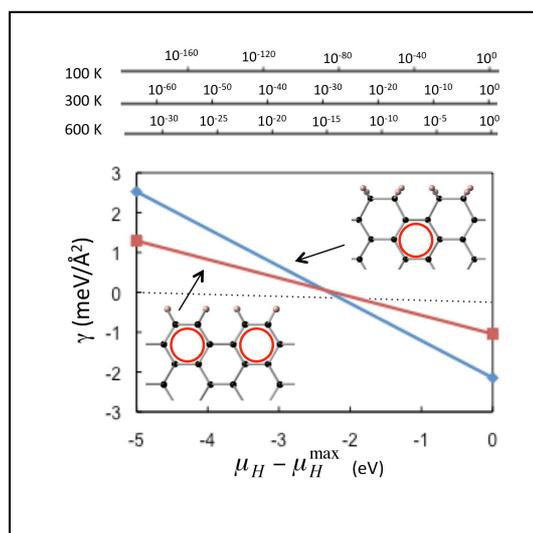


Fig. 2. Edge free energy of AGNR terminated with a single hydrogen atom (red line) and a double hydrogen atom (blue line) as a function of hydrogen chemical potential. The structure with the lower free energy is the most stable. The partial pressures of hydrogen for various temperatures are given on the top axis.

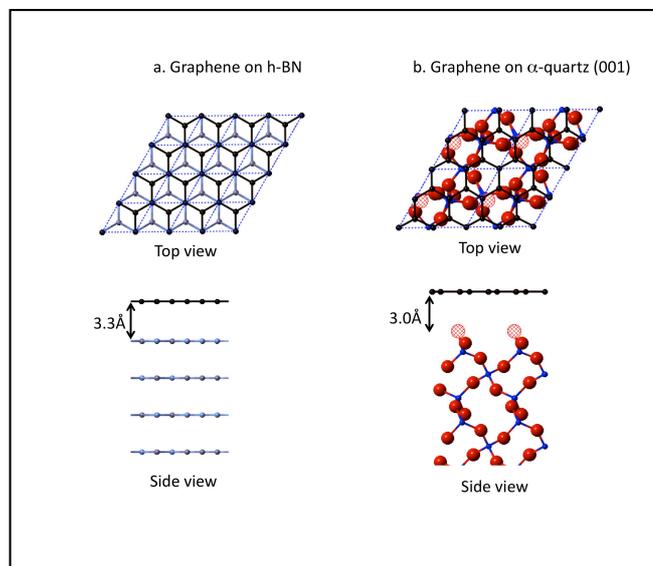


Fig. 3. Graphene supported by a) h-BN and b) α -quartz.