

Adhesion of graphene on muscovite mica: an *ab initio* study

A.N. Rudenko¹, F.J. Keil¹, M.I. Katsnelson², and A.I. Lichtenstein³

¹Institute of Chemical Reaction Engineering, Hamburg University of Technology,
Eißenendorfer Str. 38, D-21073 Hamburg, Germany

²Institute for Molecules and Materials, Radboud University Nijmegen,
Heijendaalseweg 135, 6525 AJ Nijmegen, The Netherlands

³Institute of Theoretical Physics, University of Hamburg, Jungiusstraße 9, D-20355 Hamburg, Germany
rudenko@tu-harburg.de

Graphene is at the focus of many research activities worldwide. Its remarkable electronic properties make this material a promising candidate for a large variety of electronic applications. Although graphene is treated as a free-standing sheet, practically it is usually deposited on different substrates owing to the peculiarities of preparation techniques. The role of substrates and their effect on the electronic properties of graphene are actively debated, but are still not clearly understood.

Muscovite mica [$\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$] is an appealing substrate for use in graphene-based devices owing to its relative atomic smoothness and good dielectric properties. Experimental studies show that graphene monolayers display an exceedingly flat structure being placed on mica, which is several times smoother than graphene on the most widely used dielectric (SiO_2) [1]. So far, the absence of theoretical investigations of this system was not allowing to establish unambiguously the nature of interfacial interactions between the mica surface and graphene.

In our work, we investigate the adhesion and electronic properties of graphene on a muscovite mica surface using the density functional theory (DFT) with van der Waals interactions taken into account. By assuming an atomic disorder on the mica surface we examine different possibilities for its surface structure: (i) electroneutral surface with uniform distribution of K^+ ions; (ii) electropositive structure with double K^+ coverage; and (iii) electronegative structure in the absence of K^+ ions (see Fig. 1).

We found that irregularities in the local structure of cleaved mica surface provide different mechanisms for the mica-graphene binding. By assuming electroneutrality for both surfaces, the binding is mainly of vdW nature, barely exceeding thermal energy per carbon atom at room temperature. In contrast, if potassium atoms are non uniformly distributed on mica, the different regions of the surface give rise to *n*- or *p*-type doping of graphene. In turn, an additional interaction arises between the surfaces, significantly increasing the adhesion. In Fig. 2 the densities of states (DOS) for all three considered cases are shown displaying the Fermi level shifts in the electropositive and electronegative cases.

A non uniform distribution of potassium atoms over the mica surface also provide a mechanism for variations of graphene height on mica. An estimation of maximum height variation shows an agreement with topographic data of atomic force microscopy (AFM) [1]. The upper limit of experimentally observed height variations corresponds to the calculated variations of the distance between graphene and the different types of mica surfaces (1.5 Å) with probability around 99%. This correspondence between theoretical results and experimental data allows us to conclude that the irregularity of potassium distribution on the substrate plays a major role in the formation of the graphene corrugations on mica.

In addition, it is important that the typical shape of a graphene electronic structure remains unchanged while graphene is deposited on mica (Fig. 2). This makes mica a potential candidate for its use as a substrate for graphene-based electronics, in spite of the fact that induced charge impurities may somewhat restrict the unique transport properties of graphene.

References

[1] C. H. Lui, L. Liu, K. F. Mak, G. W. Flynn, and T. F. Heinz, *Nature*, **462** (2009) 339.

Figures

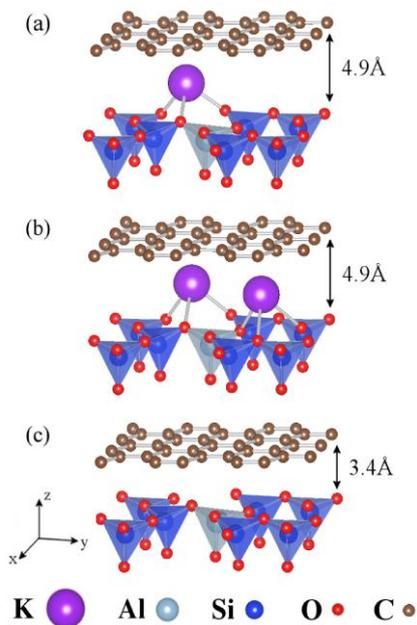


Fig. 1. Equilibrium structure of graphene supported on a (a) neutral mica surface, (b) positive mica surface, and (c) negative mica surface. Only the topmost tetrahedral layer of mica is shown.

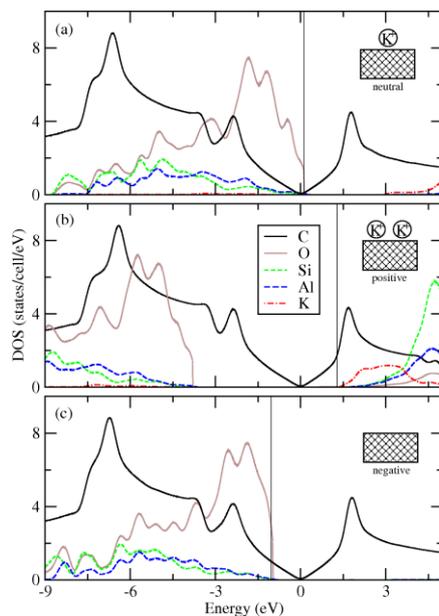


Fig. 2. Projected electronic density of states for graphene deposited on the (a) neutral mica surface, (b) positive mica surface, and (c) negative mica surface. Oxygen DOS is reduced by a factor of 5 for clarity. Zero energy corresponds to the Dirac point of graphene. The vertical line accentuates the Fermi level.