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Hexagonal-boron nitride substrates for electroburnt graphene nanojunctions

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HIGHLIGHTS

- Geometrical structures at the last stages of nanogap formation are obtained.
- The electrical conductance of such structures is predicted to be only marginally affected by the presence of hBN.
- The junctions exhibit a counterintuitive increase in electrical conductance just before the gap forms.

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ABSTRACT

We examine the effect of a hexagonal boron nitride (hBN) substrate on electron transport through graphene nanojunctions just before gap formation. Junctions in vacuum and on hBN are formed using classical molecular dynamics to create initial structures, followed by relaxation using density functional theory. We find that the hBN only slightly reduces the current through the junctions at low biases. Furthermore due to quantum interference at the last moments of breaking, the current though a single carbon filament spanning the gap is found to be higher than the current through two filaments spanning the gap in parallel. This feature is present both in the presence of absence of hBN.

1. Introduction

It is a pleasure to write this short paper in memory of Marcus Büttiker. The Lambert group has used Landauer–Büttiker formulae for more than 30 years, starting with disordered systems in the early 1980's [1,2] when the validity of such formulae was hotly debated. Since that time the group has applied these formulae to a range of problems in Andreev scattering [3], phonon transport [4,5], tunnelling through single molecules and chains [6,7], molecular spintronics [8] and most recently though nanopores leading to new strategies for DNA sequencing [9,10]. This paper is the latest in this long line and uses the Landauer–Büttiker formula to evaluate the electronic properties of electroburnt graphene nanoelectrodes.

The high thermal and mechanical stability of graphene, combined with its zero band gap and two-dimensional lattice [11], make it an ideal material for use as nano-electrodes [10,12–14]. Recently electroburnt graphene nanojunctions [14] have attracted increasing scientific interest, because their sub-nanometer gaps are sufficiently small to be spanned by single molecules. Indeed molecules with planar anchor groups bind particularly strongly to the surface of the graphene via π–π and van der Waals interactions and form stable electrode–molecule–electrode junctions [15]. Electroburnt graphene nanaojunctions can be grown on a silicon oxide substrate with a buried gate electrode, which provides a versatile three terminal platform for exploring and tuning electron transport through single molecules. However in such junctions, the thickness of the oxide barrier between the gate and molecule means that the electrostatic coupling of the gate electrode to the molecule is inefficient. Therefore strategies to increase the coupling are needed.

In this paper, our aim is to study the properties of graphene electroburnt junctions formed on an insulating hexagonal-boron nitride (hBN) substrate, which would allow a gate electrode beneath the hBN layer to be located much closer to the graphene electrode gap, thereby increasing the gating efficiency. Even in the absence of a molecule, electroburnt graphene junctions on silicon oxide or free-standing in vacuum exhibit unexpected quantum

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interference effects at the last moment of burning, just before the gap forms. In this paper, our aim is to examine electron transport through graphene nanojunctions and to determine if such effects persist in junctions formed on hBN.

Using the method described in [14], we used the molecular dynamics code *LAMMPS* [16] to generate 42 different initial examples of graphene electrodes near the moment of breaking. In each case the electrodes were covalently connected by a series of randomly generated carbon filaments or a constricted neck of graphene. Geometry relaxation was then carried out for each structure using the density-functional-theory (DFT) code *SIESTA* [17] with the same parameters as described in [14]. Next the transmission probability $T(E)$ of the electrons with energy E passing through the junction was calculated using our *GOLLUM* transport code [18] and finally the Landauer–Büttiker formula [19], was used to compute the electrical conductance and current–voltage relation for each structure.

2. Result and discussion

Fig. 1 shows an example of a nanojunction on free standing graphene (Fig. 1a) and on a hBN substrate (Fig. 1b). After geometry relaxation, the graphene is approximately A–B stacked on the hBN with a slight lattice mismatch [20]. The separation between the two layers is calculated to be approximately 3.4 Å in agreement with reported experimental [21] and theoretical values [22]. The substrate size is chosen to be larger than graphene to avoid the effect of hBN edges on the current. The corresponding transmission coefficient and current through the device is shown in Fig. 1c and d respectively. The transmission coefficient around the Fermi energy is almost the same in the absence and presence of hBN although at higher energies new resonances due to the weak coupling between carbon and boron or nitrogen atoms appear. Due to the fact that the transmission is only slightly changed around the neutrality point, the low-bias current is barely affected by the hBN. At higher voltages, the presence of additional transmission resonances around $E_F = -0.8$ V could cause the current in the presence of hBN to increase.

To create a junction with nano-meter size gap, a sufficiently-high bias voltage is applied to burn a pre-patterned constriction of graphene [14]. When the graphene begins to burn, feedback control is activated and the bias voltage is dropped. This process is repeated several times to break the junction. One complicating feature is the presence of the oxygen atoms around the burning site, which can affect the shape and size of the junctions. This oxygen could be supplied from air or in vacuum from the SiO_2 oxide substrate. The use of hBN could avoid this feature and help to form more reproducible junctions. Fig. 2 shows five examples of such a junctions created in the absence of the oxygen atoms. Structures 3, 4 and 5 are formed from successively narrower constrictions, which are stable after DFT relaxation. In contrast, a constriction formed from a single chain of hexagons without edge termination is not stable and forms two parallel carbon chains, as shown in s structure 2. The most narrow structure 1 is formed from a single carbon chain and has relaxed to form a series of single and triple bonds in agreement with the known properties of oligoynes [23–25]. Such chains have been recently observed experimentally using TEM on free standing graphene [26].

The conductances of devices 1–5 are shown in Fig. 2b. Junctions 1 and 2 of Fig. 2a are interesting, because the low-bias current (ie at voltages less than 0.5 V) of the single carbon chain (device 1) is higher than device 2 (two parallel carbon chains) in low bias voltages for a wide bias window. This is highly non-classical, since one would expect a higher conductance for two parallel resistors than for a single resistor. In our recent paper [14], we discussed the

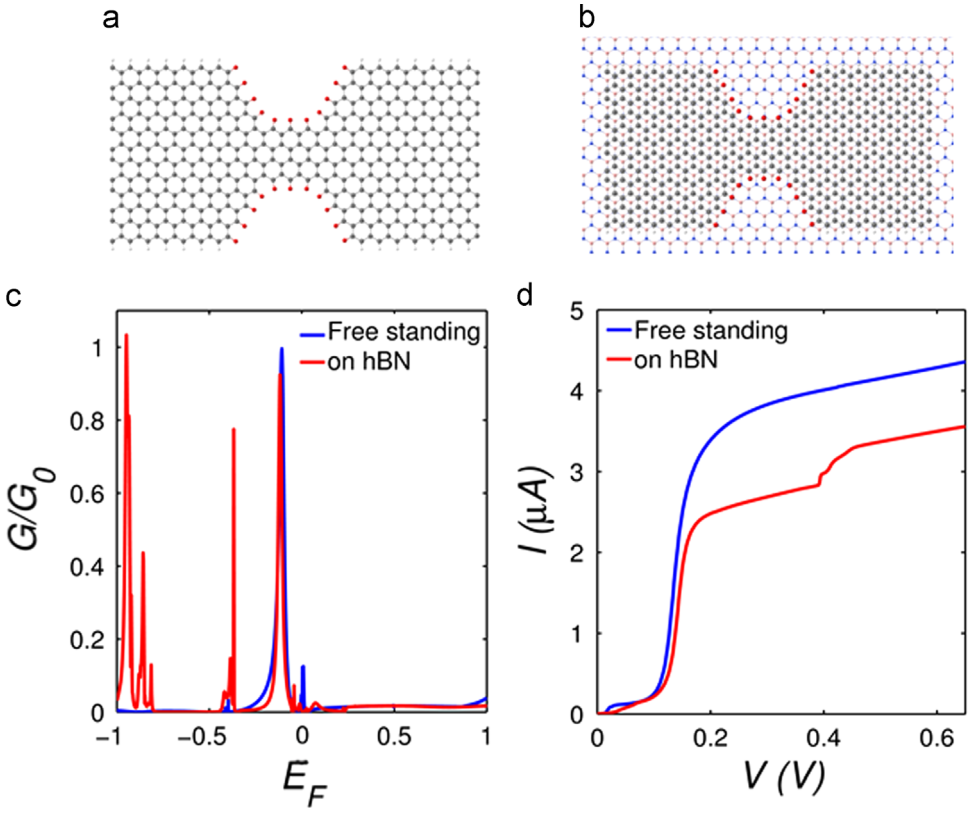

Fig. 1. Graphene nano-junctions (a) free standing graphene, (b) on hexagonal boron nitride substrate, (c) the conductance in different Fermi energies and (d) current–voltage characteristic.