Tunneling properties of vertical heterostructures of multilayer hexagonal boron nitride and graphene

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We use first-principle density functional theory (DFT) to study the transport properties of single and double barrier heterostructures realized by stacking multilayer h-BN or BC$_2$N, and graphene films between graphite leads. The heterostructures are lattice matched. The considered single barrier systems consist of layers of up to five h-BN or BC$_2$N monoatomic layers (Bernal stacking) between graphite electrodes. The transmission probability of an h-BN barrier exhibits two unusual behaviors: it is very low also in a classically allowed energy region, due to a crystal momentum mismatch between states in graphite and in the dielectric layer, and it is only weakly dependent on energy in the h-BN gap, because the imaginary part of the crystal momentum of h-BN is almost independent of energy. The double barrier structures consist of h-BN films separated by up to three graphene layers. We show that already five layers of h-BN strongly suppress the transmission between graphite leads, and that resonant tunneling cannot be observed because the energy dispersion relation cannot be decoupled in a vertical and a transversal component.

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Mobility of suspended graphene can be extremely high, as demonstrated by experiments and theory. However, use of graphene in solid state devices typically requires deposition or growth on a dielectric substrate, which can strongly suppress mobility, due to electron coupling with dielectric-layer phonon modes, as demonstrated in the case of SiO$_2$ or HfO$_2$, the most commonly used dielectric materials.

Hexagonal boron nitride (h-BN) has been recently investigated as a promising dielectric for graphene, while boron nitride domains has been suggested as a way to engineer graphene nanoribbon transport properties.

h-BN has hexagonal geometry and a lattice constant closely matching that of graphene, and the electronic interaction of h-BN with a single graphite sheet or with bilayer graphene is very weak. As a result, graphene deposited on an h-BN substrate maintains its electronic and transport properties.

From this perspective, understanding transport along stacked graphene/h-BN structure can provide relevant information regarding the leakage current in graphene based Field Effect Transistors exploiting h-BN as gate dielectric or the performance of recently proposed vertical devices.

In this work, with ab-initio method, we calculate the quantum transport in single and double barriers consisting of h-BN and BCN layers between semi infinite graphite leads, using the formalism introduced by Choi et al., using a plane wave basis set in the local density approximation (LDA). A 35 Ry wave function cutoff has been considered, the Brillouin zone has been sampled using a $30 \times 30 \times 30$ Monkhorst-Pack grid. Atomic positions have been relaxed using a conjugate gradient algorithm until all components of all forces was smaller than 0.01 eV/Å. While the electronic minimization has been performed with a tolerance of $10^{-7}$ eV. It has been shown that LDA provides a reliable description of the geometry and the electronic structure in the presence of weak interactions between h-BN layers or between h-BN and graphene layers. On the other hand, the LDA representation for the exchange-correlation potential cannot correctly describe the ex-
cited states in organic systems and leads to overestimation of conductance. However, following other works on this subject, we expect that these effects have only minor consequences, and thus our conclusions should not be affected by this choice. The transmission probabilities have been calculated with the PWCOND module of Quantum Espresso. The transport properties are studied in the framework of Landauer formalism, where the ballistic conductance is given by $G = G_0 T(E)$, where $G_0$ is the quantum conductance $G_0 = 2e^2/h$, and $T(E)$ is the total transmission at the energy $E$.

$T(E)$ is obtained as
\[
T(E) = \sum_{k_{||}} \sum_i \sum_j T_{i,j}(k_{||}, E)
\]  
(1)

where $T_{i,j}(k_{||}, E)$ is the probability that an electron with energy $E$ and transversal momentum $k_{||}$ incoming from the $i$-th Bloch state is transmitted to the outgoing $j$-th state of the other electrode. Sums runs on both spins. The first sum is performed over $k_{||}$ belonging to the two-dimensional Brillouin zone (2D-BZ) of the supercell.

The transmission probability of a single h-BN barrier is shown in Fig. 2 for a different number of layers. For a given number $n$ of single h-BN layers, the transmission probability is almost independent of energy, in an energy range of about 3 eV starting from the Fermi level. This behavior can be explained by the structure of the complex bands in bulk h-BN shown in Fig. 3 in the energy interval considered, in the symmetry point K on the plane parallel to the layer interface (where the graphite states with real momentum reside), h-BN has a dispersion relationship according to which the imaginary wave vector is almost independent of energy, and therefore $T$ as well. Of course $T$ decays exponentially with increasing $n$, as can be verified by plotting the average value $\langle T \rangle$ in the energy range between the Fermi energy value and 3.4 eV above as a function of the number of layers, as in the inset of Fig. 2. The fitting exponential is
\[
\langle T \rangle \approx e^{-3.5n}
\]

and five h-BN layers strongly suppress the transmission. The dip in correspondence of $E - E_{\text{Fermi}} = 0$ eV is due to the null density of states in graphite leads. In addition, we also note that for $E - E_{\text{Fermi}} < -3$ eV, the transmission probability is suppressed even in a classically allowed region, probably because of a mismatch between propagating states in the h-BN barrier and in graphite.

In Fig. 1 the transmission probability as a function of energy is shown for a single barrier of BC$_2$N of $n$ atomic layers between graphite leads. The atomic structure is shown in the inset. A qualitatively similar behavior as h-BN is observed, with an exponential decay of the transmission probability in a narrow range around Fermi energy. BC$_2$N has a gap of 1.6 eV.

Fig. 2 shows the transmission probability as a function of energy for some single and double barrier structures. All these systems are symmetric, with two identical barriers separated by one, two or three graphene layers. For clarity, hereinafter, we have indicated the single barrier systems with the acronym SB followed by the number of h-BN layer (e.g. in Fig. 1, SB5). In the same way, the double barrier systems are related with DB and three numbers, namely the number of h-BN layers on the left, the number of graphene layers in the central region, and the number of h-BN layers on the right (e.g. in Fig. 1, DB2|1|2). By comparing the transmission of the single h-BN barrier (SB1) with that of the double h-BN barrier (DB1|1|1) (Fig. 4), the noteworthy aspect is that the insertion of a single graphene sheet has a noticeable effect on transport even in the direction perpendicular to the plane (Fig. 5). While the insertion of even a single BN (SB1) sheet strongly suppresses the transmittance (particularly in the zone near the Fermi energy), the presence of a triple layer (DB1|1|1) yields almost the same behavior as the single-BN sheet. In addition, DB1|1|1 has a
FIG. 4. Transmission probability as a function of energy for a single BC$_2$N barrier of $n$ atomic layers between graphite leads. The inset shows the atomic structure of the BC$_2$N$^n$ isomer studied.

FIG. 5. Transmission probability as a function of the energy (eV) for different systems; a: SB1 (solid black line), SB2 (dashed red line), DB1|1|1 (dotted green line). b: DB1|1|1 (solid black line), DB1|2|1 (dashed red line), DB1|3|1 (dotted green line).

FIG. 6. a) Transmission probability as a function of the energy (eV) for DB2|1|2; b) and c) Transmission probability as a function of energy for DB2|1|2 system with step sampling $2 \times 10^{-3}$ and $4 \times 10^{-5}$.

FIG. 7. Transmission probability of the double barrier system DB2|1|2 as a function of energy for selected $k_\|$: a) (0.28000;0.28867); b) (0.31000;0.32909); c) (0.28000;0.33486); in unit $\frac{2\pi}{a}$ with $a=2.503$ Å

much higher transmittance than SB2, despite the identical number of h-BN layers. In Fig. 6b, we report the transmittance of double-barrier systems in which two h-BN layers are separated by one, two or three graphene layers.

It is also important to notice that no resonant tunneling occurs in the double barrier structures, in the whole energy range explored, as we have verified with a very dense energy sampling of transmission probability, down to steps of $10^{-3}$ eV, as shown in Fig. 6 referred to (DB2|1|2) case. The reason is due to the fact that the energy dispersion relation in graphene cannot be written as the sum of a longitudinal and a transversal component. Therefore, for each transversal wave vector $k_\|$, one observes resonances in $T(E)$ with unity transmission probability peaks, as shown in Fig. 7 at different energies. When the sum over $k_\|$ resonance do not occur at the same energy and are therefore washed out.

I. CONCLUSION

In this paper we have investigated the tunneling probability of vertical heterostructures consisting of single and
double layers of h-BN and BC$_2$N separated by graphene sheets and connected to graphite leads. These structures are interesting from the point of view of graphene electronics, also because few devices have been proposed in which transport occurs in the direction perpendicular to graphene planes,\textsuperscript{15,26,27} As expected, the transmission probability is exponentially dependent on the number of layers, and is already strongly suppressed by a single monolayer of h-BN or BC$_2$N. In addition, due to the energy dispersion relationship in the energy gap, for which the imaginary wave vector is almost constant as a function of energy, the transmission probability has only small dependence on energy. We have also shown that resonant tunneling is completely suppressed by the peculiar energy dispersion relation of graphene, that cannot be decomposed in the sum of a longitudinal and a transversal component.

II. ACKNOWLEDGMENT

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