Simulation of Semiconductor Processes and Devices 2016
Edited by E. Bär, J. Lorenz, and P. Pichler

Effect of Rotational Misalignment on Interlayer Coupling in a Graphene/hBN/Graphene van der Waal's heterostructure

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***Abstract*—We simulate the effects of rotational misalignment of the tunnel barrier layer between aligned channel layers in a monolayer-graphene/hBN/monolayer-graphene system. Through use of density functional theory (DFT) methods, we demonstrate a reduction in tunneling current due to weakened coupling across the rotationally misaligned interfaces between the channel layers and the tunnel barrier.**

Keywords—graphene, tunneling current, commensurate rotation, ITFET, interlayer coupling, heterostructures.

I. Introduction

Novel multi-layer device concepts such as interlayer resonant tunnel FETs employ channel/tunnel-barrier/channel geometries [1–3]. However, during layer-by-layer exfoliation of these multi-layer materials, rotational misalignment is the norm and may substantially affect device properties. Consider for the sake of illustration that the tunneling currents I could be approximated in the form $I \approx \eta e^{-\kappa t}$ near flat band, where η and κ are constants and t is the tunnel barrier thickness. Misalignment between the channel and tunnel barrier band structures, in principle, could alter the evanescence coefficient κ. However, extrapolation of experimental results [2,3] consistent with the observed value of κ (corresponding to about an order of magnitude reduction per hBN layer) in a bilayer graphene/multilayer hexagonal boron nitride (hBN)/bilayer graphene system, even with the *graphene bilayers* aligned to each other (confirmed by resonant interlayer tunneling) but not to the hBN tunnel barrier, to a few *and even zero layers* still leads to a prediction of weak interlayer tunneling. Thus, this result cannot be explained by the evanescence coefficient alone.

In this work we consider, through use of density functional theory (DFT) methods, instead, a reduction in tunneling due to weakened coupling ***across the rotationally misaligned interface*** *between the channel layers and the tunnel barrier*. This weakened coupling, localized to the two hBN/graphene interfaces with mutually aligned hBN layers within the tunnel barrier as to be expected via the employed hBN exfoliation technique, would affect the lead coefficient η, qualitatively consistent with the extrapolated weak tunneling in the few hBN layer limit. As a prototypical system, we simulate the effects of rotational misalignment of the tunnel barrier layer between aligned channel layers in a monolayer (ML)-graphene/hBN/monolayer-graphene system, where this system is used largely to minimize the atom count in our DFT simulations. However, these results may be relevant to all such van der Waal's heterostructures.

II. Computational Details

For illustrative purposes, in the weak coupling limit—e.g., short of the interlayer current approaching to the lead-limited value—the current I varies as

$$I = \frac{8\pi e}{\hbar}\sum_{\mu\nu}\left|M_{\mu\nu}\right|^2[f(E_\mu) - f(E_\nu)]\delta(E_\mu - E_\nu) \qquad (1)$$

within a first-order Bardeen transfer Hamiltonian approximation where the coupling parameter $M_{\mu\nu}$ is obtained from the overlap of the opposite channel layer wavefunctions across the interlayer tunnel barrier [4, 5]. This interlayer coupling also produces a symmetric/antisymmetric splitting of the otherwise degenerate band structures in the opposite layers under flat band conditions, which is only proportional to $M_{\mu\nu}$ in the weak coupling limit. Thus, by calculating the energy splitting due to coupling through a given tunnel barrier, we also can infer the still greater effect on interlayer tunneling currents.

We performed DFT simulations to calculate the band splitting of aligned and misaligned systems. We began by relaxing the atomic structures, using the projector-augmented wave method with a plane-wave basis set as implemented in the Vienna *ab initio* simulation package (VASP). Graphene/hBN/graphene supercells were created using the Virtual NanoLab software with the hBN layer rotated with respect to the mutually aligned graphene layers. The lattice constants of graphene and hBN are well matched (to within 1.6%), and the considered rotational misalignments are necessarily commensurately rotated for our calculations. Therefore, lattice strain is minimal for the considered supercell structures. Figure 1 (a) and (b) show the commensurately rotated graphene/ML-hBN/graphene supercells with rotation angles, 21.79° and 13.17°, respectively [6] corresponding to two

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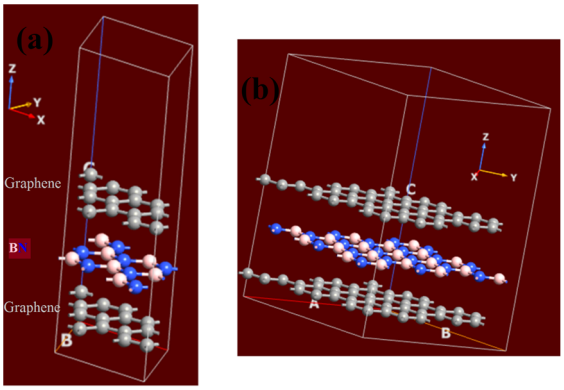

Fig. 1. Supercells of Graphene-ML hBN-Graphene with hBN layers commensurately rotated with respect to graphene layers at rotation angles of (a) 21.79° and (b) 13.17°. C atoms are colored slate gray, B atoms in light pink and N atoms in blue.

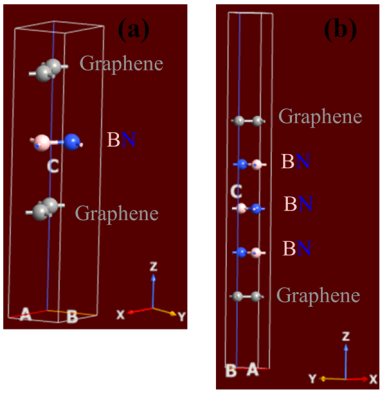

Fig. 2. Supercells of aligned system with (a) monolayer hBN and (b) trilayer hBN. C atoms of graphene are placed on top of B atoms of the hBN layer.

smallest supercells. Supercells with varying number of aligned hBN layers are shown in Fig. 2. In the case of the aligned system, the C atoms of graphene are placed on top of B atoms of the hBN layer, which is the energetically most stable configuration [7]. The local density approximation (LDA) was employed for the exchange-correlation potential as LDA can model materials well with the same orbital character at conduction and valence band edges. Large supercell sizes prevented the use of more computationally intensive hybrid functionals or GW methods in our calculations. Van der Waal's forces were modeled using the DFT-D2 scheme wherein a semi-empirical correction is added to the conventional Kohn-Sham DFT theory. A k-mesh grid of 7×7×3 for the sampling of the first Brillouin zone of the supercell was selected according to Monkhorst-Pack type meshes with the origin being at the Γ point for all calculations except the band structure calculation (where a fine k-space resolution along major axes was sampled). Atomistic relaxations were allowed to converge when the Hellmann-Feynman forces on the atoms were less than 0.001 eV/Å. The interlayer degeneracy splitting calculated in this way is only weakly **k**-dependent within a few hundred meV of the Dirac (**k**) point. For specificity, we calculated the splitting at ≈100 meV above the Dirac (**K**) point (avoiding the intersecting band structures about the Dirac point), as illustrated in Fig. 3.

III. Results

For reference, the interlayer energy splittings with hBN layer number for the fully-aligned ML-graphene/hBN/ML-graphene system calculated by the above procedure are listed in Table I for one to four layers of hBN, which is reasonably in line with the known zero hBN layer energy splitting limit of 370 meV represented by a Bernal stacked graphene bilayer [8]. For one, two or even more layers, such coupling essentially would short the graphene layers together in the large area systems, in contrast to the experimental results for rotationally misaligned systems [2,3].

For ML-graphene/ML-hBN/ML-graphene system with the hBN layer misaligned by commensurate rotation with mutually aligned graphene layers, such as for a resonant tunneling devices, Table II *shows a substantial reduction in the energy splitting with layer misalignment*. As also shown, such a reduction in interlayer coupling due to this interface-localized effect would drop the interlayer current per unit area by factors of 8.7 and 25 for the two commensurate rotation angles of 21.79° and 13.17°, respectively, within the perturbation limit of Eq. (1). Thus, despite the growth in primitive unit cell size by factors of 7 and 19 respectively, the expected interlayer current *per primitive unit cell* for these two rotation angles would remain roughly constant and actually decrease somewhat to 0.80 and 0.76 times that for the aligned system. Thus, overall, the results of these simulations are consistent with significant reductions in the lead coefficient η.

Table I. Variation of interlayer energy splitting as function of number of hBN layers for aligned graphene/hBN/graphene heterostructures.

# of hBN layers	Band Splitting (meV)
1	69.1
2	20.4
3	2.24
4	0.485

Table II. Variation of interlayer energy splitting as function of commensurate rotation angle for graphene/ML hBN/graphene, along with number of atoms per unit cell and effect on perturbation current.

Rotation Angle (degrees)	Degree of Band Splitting (meV)	# of atoms in Supercell	Relative Current Drop (wrt aligned case)
0°	69.1	6	-
21.79°	23.4	42	8.7
13.17°	13.8	114	25

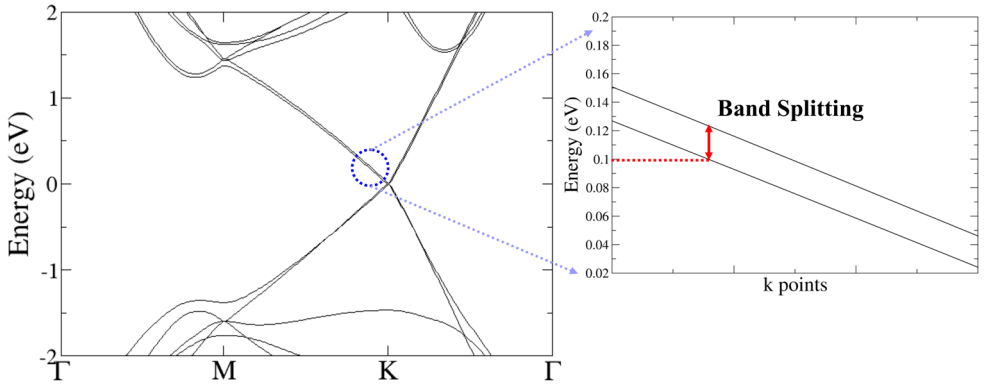

Fig. 3. Band structure of rotationally misaligned graphene/ML-hBN/graphene after relaxation. The (roughly constant) band splitting is measured ~100 meV above the Dirac (K) point.

We currently are working to extend these calculations to larger rotation angles and more hBN layers to understand the precise mechanism of this reduction in coupling (which may not be as simple as the number of spatially overlapping atoms) and to see if this specific trend of approximately conserving current continues to larger unit cells (and to thicker hBN barriers to look for any additional effects on evanescence), as well as to see if there also are rotational effects on the evanescence coefficient κ. It is quite likely, however, that we will not reach a saturation point for reducing the interlayer tunneling current per unit area with rotation given the DFT computational demands.

IV. Conclusion

In summary, the effects on interlayer coupling of rotational misalignment of the tunnel barrier layer between mutually aligned channel layers in resonant tunneling graphene/hBN/graphene systems has been considered through DFT simulation. We found a strong reduction in interlayer tunneling current due to misalignment that is consistent with the weak tunneling currents observed in experimental results that cannot be ascribed to the evanescence coefficient. These results may be relevant to all such van der Waal's heterostructures.

Acknowledgment

This work was supported through the Southwest Academy of Nanoelectronics (SWAN) by the Nanoelectronics Research Initiative (NRI) within the Semiconductor Research Corporation (SRC). The authors acknowledge the Texas Advanced Computing Center (TACC) at The University of Texas at Austin for providing high performance computing resources that have contributed to the research results reported within this paper.

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