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Hybrid 2D nanodevice (Graphene/h-BN): Selecting $NO_x$ gas through the device interface†

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Recent experiments demonstrate a highly controlled synthesis of in-plane graphene/hexagonal boron nitride heterostructures, thereby paving the way for the development of a new category of hybrid 2D nanodevices with different interfaces. Herein, by employing a combination of density functional theory (DFT) and the non-equilibrium Green's function formalism (NEGF), gas sensor capability of a G-hBN, containing two different interfaces, is assessed by exposing it to $NO$, $NO_2$, $NH_3$, and $CO_2$ gases molecules. Our results show that there exists a strongest interaction between $NO_x$ molecules and G-hBN, meanwhile, NO and NO$_2$ molecules are more reactive in the interface regions than the pristine ones. In addition, the electron transport properties reveal that for a distinct two-port voltage applied, the gas molecule $NO$ ($NO_2$) can be detected by high changes in the conductance and $CO_2$($NH_3$) has insignificant signal conductance, leading to selectivity of gas. The proposed device bring out as a great potential candidate to be applied as $NO_x$ gas selector through the 2D nanodevice interface.

1 Introduction

Semiconducting metal oxide based gas sensors have been widely used and explored due to their relatively easy fabrication, high sensitivity towards many types of gases, low cost and energetic consumption. Nevertheless, a high temperature operation, long recovery time, low stability, and poor selectivity emerge as the main limiting factors for a practical use of this class of sensors in applications. In view of those drawbacks and motivated by the increasing interest for highly sensitivity, selectivity, cheap and stable gas sensors, new materials have been investigated. Some progress has been made through the investigations of low dimensional materials such as semiconducting nanowires, graphene sheets, graphene nanotubes, and transition metal dichalcogenide monolayers. In general, the high surface-to-volume ratio of 2D materials results in extremely sensitive electronic structure towards potential adsorbates and is an essential component in sensing applications.

As an example, it has been demonstrated that a graphene based gas sensor has a high capability in detecting the presence of a single molecule adsorbed on it. Following this result, the efficiency of a graphene gas sensor in detecting different types of gases has been intensively explored both experimentally and theoretically. The combination of graphene with hexagonal boron nitride (hBN) to form two-dimensional lateral heterostructures has been demonstrated through its synthesis based on a high control over the shape and size of the two different structures. In particular, it has been found that a graphene nanoroad embedded in hBN can sustain the outstanding transport properties of pristine graphene. For this material, it has been theoretically demonstrated that the electric current flux can be steered and stimulated to be confined to graphene stripe interfaces by adjusting the applied gate voltage. This system becomes a promising candidate as a highly sensitive and selective gas sensor architecture through combining the control over the electric current flow with the enhanced chemical reactivity of certain sites at the interface.

Motivated by all these studies, in this work, we investigate the gas sensing properties of a two-dimensional graphene/boron ni-
Fig. 1 (a) Top view of the proposed G-hBN sensing device corresponding to the electronic transport setup. The device is made of the scattering region and the (left/right) electrodes. The numbers indicate the adsorption sites (1 to 6) for the gas molecules. The different interfaces are denoted by the labels ‘CB’ and ‘CN’, respectively (see text). (b) The full relaxed geometry of the NO, NO$_2$, NH$_3$ and CO$_2$ molecules adsorbed on their most energetically stable position on the device. For each molecule, a top and a side view of the adsorption configuration is shown. The different atomic species are colored according to the scheme in the legend. The same color coding will be used throughout.

Scheffler (TS) and the Grimme-D3 approaches, as well as with the experimental results. For small molecules such as CO$_2$, we have also obtained a small deviation (< 0.01 eV) from experimental results. For the pseudopotentials the Projector Augmented Wave (PAW) method was used. The plane-wave cutoff energy was set at 550 eV. We have also considered periodic boundary conditions and Brillouin zone sampling at Γ point within the Monkhorst-Pack scheme. By applying periodic boundary conditions the hybrid system is formed by alternated graphene and h-BN nanoroads, both of them are ~13 Å wide. These widths assures that the system possesses a metallic character, as the energy band gap is of the order of the thermal fluctuation at room temperature, and the interaction between the graphene nanorod and its periodic image becomes negligible. All calculations were performed using a convergence criterion of $10^{-5}$ eV for the total energies, and 0.01 eV/Å for the residual atomic forces in each atoms component. The final relaxed atomic structure is shown in Fig.1(a) with dimensions of 26.2 Å × 20.0 Å × 30.3 Å.

In order to treat the electronic transport problem we employed the non-equilibrium Green’s function (NEGF) formalism using the DFT Hamiltonian as implemented in Transiesta. The heterostucture is split into three parts: a finite scattering region between two semi-infinite leads (left(L) and right(R) electrode) as depicted in Fig.1.a. In principle a solution for such a system cannot be found, as due to the full semi-infinite leads the matrix is infinite. However, this can be overcome through the definition of Green’s function for the total system as follows:

$$\mathcal{G}(E,V) = \left[ E \times S_L - H_S \rho \right] - \Sigma_L(E,V) - \Sigma_R(E,V) \right]^{-1}$$

where the contributions from the L(R) leads are given through the self energies $\Sigma_L(R)$, $S_L$ and $H_S$ are the overlap and Hamiltonian matrix elements of the scattering region. The goal is to obtain the energy-resolved transmission function $T(E)$, representing the probability of an incoming electron from L lead entering the scattering region and reaching the R lead. The transmission coefficient is given by:

$$T(E) = Tr \left[ \Gamma_L(E,V) \mathcal{G}(E,V) \Gamma_R(E,V) \mathcal{G}^\dagger(E,V) \right]$$

where the coupling matrices are defined as $\Gamma_\alpha = \Gamma \left[ \Sigma_\alpha - \Sigma_\alpha^\dagger \right]$, with $\alpha \equiv \{L,R\}$, and $\mathcal{G}$ ($\mathcal{G}^\dagger$) the retarded (advanced) Green’s function. For zero-bias calculations, the chemical potentials are the same for both leads ($\mu_L = \mu_R$) and the electrical current is zero by construction. The proposed nanodevice is composed by a hybrid two-dimensional graphene/boron nitride heterostructure referred as ‘G-hBN’ in the following. We probe the sensing properties by exploring some representative gas molecules such as NO, NO$_2$, NH$_3$ and CO$_2$ placed on top of device. Fig.1a, represents a graphene nanoroad width (~13 Å) forming an in-plane heterostructure with hexagonal boron nitride. The very small mismatch between their lattice parameters (< 2%) allows a lateral connection between the two domains without out-of-plane buckling. Experiments also suggest that the zigzag graphene/hBN interface type is energetically most favorable. Based on previous experimental and theoretical results, the

2 Methodology

Quantum mechanical calculations were performed using the density functional theory (DFT) as implemented in VASP and SIESTA. The former methodology was applied for atomic relaxation using the exchange-correlation potential based on a generalized gradient approximation (GGA-PBE). Dispersion interactions are accounted for through a vdw-DF scheme as implemented through the optB86b-vdW functional. We have chosen the vdw-DF correction since for graphite systems, adsorption energy compares well with other methods such as the Tkatchenko-Scheffler (TS) and the Grimme-D3 approaches, as well as with the experimental results. For small molecules such as CO$_2$, we have also obtained a small deviation (< 0.01 eV) from experimental results. For the pseudopotentials the Projector Augmented Wave (PAW) method was used. The plane-wave cutoff energy was set at 550 eV. We have also considered periodic boundary conditions and Brillouin zone sampling at Γ point within the Monkhorst-Pack scheme. By applying periodic boundary conditions the hybrid system is formed by alternated graphene and h-BN nanoroads, both of them are ~13 Å wide. These widths assures that the system possesses a metallic character, as the energy band gap is of the order of the thermal fluctuation at room temperature, and the interaction between the graphene nanorod and its periodic image becomes negligible. All calculations were performed using a convergence criterion of $10^{-5}$ eV for the total energies, and 0.01 eV/Å for the residual atomic forces in each atoms component. The final relaxed atomic structure is shown in Fig.1(a) with dimensions of 26.2 Å × 20.0 Å × 30.3 Å.

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G-hBN architecture shown in Fig.1a is expected to be experimentally feasible using high atomic precision techniques to control the nanoroad width.

3 Results and discussion

Fig.1a shows the device composed by three regions: CN, CB interfaces and graphene. In order to look into the influence of these regions, we have chosen six positions, named by 1 to 6, on the material on top of which we place the molecules. The energetic stability of the target molecule on G-hBN device is assessed through the adsorption energy:

\[ E_{ads} = E_{[gas/G-hBN]} - E_{[gas]} - E_{[G-hBN]} \]  \( (3) \)

where \( E_{[gas/G-hBN]} \) is the total energy of the fully relaxed molecule on G-hBN, \( E_{[gas]} \) and \( E_{[G-hBN]} \) is the total energy of each separate component, gas and device, respectively. Fig.1b shows the most stable geometry of each gas molecule on the device after the full relaxation. The adsorption energy are depicted in Fig.2 for three different regions.

Inspection in Fig.2 reveals considerable adsorption energy variations with respect to the molecule and region of adsorption. As a first observation, the NH$_3$ and CO$_2$ molecules have almost the same adsorption energy (\( E_{ads} \approx -0.33 \text{ eV/molecule} \)) for all explored positions and regions on top of the G-hBN device. The CB and CN interfaces have almost no influence on the adsorption energy of these two molecule. Contrary to these, the NO and NO$_2$ molecules show a strong variation in their adsorption energy with respect to their position on the sensor. Overall, for these two molecules, the adsorption energy is higher on top of the graphene part of the material, indicating a weaker adsorption on graphene and hBN regions compared to the interfaces.\(^{22,48}\) On the later, the NO$_x$ molecules are much stronger adsorbed. The molecule NO shows the lowest adsorption energy on top of the CB interface at position 2 (\( E_{ads} = -1.00 \text{ eV/molecule} \)) whereas NO$_2$ is stronger adsorbed on top of the CN interface at position 6 (\( E_{ads} = -2.02 \text{ eV/molecule} \)). Our results show that all gas molecules prefer to interact with the interface regions, either the CB or the CN interface. Overall, the analysis on the energetics underlines that the G-hBN ultra-thin layer is more reactive towards the NO and NO$_2$ gases than the NH$_3$ and CO$_2$ ones. This strongly indicates the higher selectivity of our proposed device for NO$_x$ gases detection.

Table 1 shows the shortest atom-to-atom distance (D) between the G-hBN material and the gas molecule. As expected, we can note a correlation between adsorption energy with the distance. In addition, for example the NO$_2$ shows the lowest adsorption energy as well as the largest charge transfer as indicated through Bader Charge analysis.\(^{49}\) For CO$_2$ and NH$_3$ the results are similar comparing their adsorption, distance and charge transfer; lower adsorption energy, biggest distance as well as negligible charge transfer.

Next, we focus on the electronic transport properties, transmission curves, for the most energetic stable configurations for each target molecule, Fig.1b. The transmission curves for the pristine sensor and the device with target molecules are shown in Fig.3. As can be seen, the presence of the CO$_2$ and NH$_3$ molecules even at the site with the strongest adsorption does not show significantly influence on the transmission, thus the conductance, across the device. A small deviation with respect to the reference curve...
(pristine device) is only observed at an energy close to 0.1 eV, reachable by gating the device. As opposed to CO₂ and NH₃, the NO₂ molecules considered here, significantly affect the electronic transmission across the G-hBN device. The two molecules reveal a strong influence at different energies. Specifically, the transmission change for NO is significant at negative energies, below the Fermi level, while the transmission for NO₂ is strongly affected at positive energies (above the Fermi level). The result shows the possibility to detect NO(NO₂) controlling the gate voltage.

Next, we investigate the NO₂ gas molecules on top of the two different interfaces (CB and CN) and their influence on the transport properties. Fig.4a)–b) represents how the presence of either NO or NO₂ on top of the device changes the transmission. For this we consider the positions 2 and 6, where the device is more reactive (see Fig.2). In case of NO in Fig.4a) a strong deviation from the transmission of the pristine device is evident for the whole energy range presented. There is also a large difference in the transmission related to the positioning of NO on two interfaces at positions 2 and 6. The similar behavior is observed for NO₂ in panel b) below the Fermi level. For positive energies (i.e. gating voltages), the differences in the transmission curves corresponding to the two positions for the NO₂ are quite smaller close to the Fermi level. However, a noticeable transmission difference is observed above ≈0.1 eV for the two sites considered, allowing to identify in which interface the NO₂ molecule is adsorbed. The same can be done for the NO case in the entire energy range considered. Overall, regardless the NO₂ adsorption region on the device, the proposed sensor architecture is able to electrically discriminate between the two molecules.

In order to better understand the detection mechanism in the device, we turn to the analysis of the projected electronic density of states (PDOS) for the G–hBN device and the corresponding electronic transmission curves. Fig.5a)–b) show the respective results for pristine G–hBN, i.e. without the presence of the gas molecules. A ballistic behavior in the transmission can be clearly seen through the two electron channels below and above the Fermi energy. Inspection of the PDOS also shows that the CB (CN) atoms and carbon central atoms (C) mainly contribute to left(right) plateaus. Nevertheless, there is a minority contribution of CN (CB) atoms on the left (right) transmission channel. One can note that the contribution to the DOS of the CN interface is always higher than that of the CB interface.

Fig.5 c)–f) shows an interesting finding when the molecule is adsorbed on the CB (CN) interface. In these cases, the PDOS shows a significant decrease compared to the pristine material. An overall difference in those cases, though, is the fact that the equivalent (in the pristine material) contributions of the CB and the CN interface change. Interestingly, the PDOS contribution of a particular interface is lowered when a gas molecule is placed on top of it and is enhanced for the opposite interface further away from the molecule. In addition with molecule adsorption, the interface PDOS peaks are slightly displaced towards the Fermi level, leading to an enhancement of the total PDOS at that energy for the combined system. As an example, in Fig.5 c) the PDOS contribution is higher at the CN interface compared to CB interface (which NO is placed). This fact is due to the molecule hybridization with C central atoms. In the opposite case (Fig.5 e), when NO is positioned on top of CN interface, CB contribution is bigger than CN peak due interface, because in the later case there are atoms hybridization with gas. Similar trends can be seen for the other molecule, NO₂ (Fig.5 d) and f). The contribution of the carbon atoms to the PDOS shows a small deviation from the uniform parabolic like curve in the pristine case.

In order to explore the spatial behavior of the electrons in the material/molecule system, we turn to the charge density redistribution. This is based on the interaction between the gas molecules and the G-hBN device and is given through:

\[ \Delta \rho(\vec{r}) = \rho_{\text{gas}/G-hBN}(\vec{r}) - \rho_{\text{gas}}(\vec{r}) + \rho_{G-hBN}(\vec{r}) \]

where \( \rho_{\text{gas}/G-hBN}, \rho_{\text{gas}} \) and \( \rho_{G-hBN} \) are the charge densities of the combined gas/material system, the isolated gas molecule and the isolated material, respectively. The charge densities redistribution are position dependent and can be visualized as shown in Fig.6.

Fig.6 depicts the charge density redistribution for NO₂ molecules on CB and CN interfaces. It can be observed that in all cases the charge density is redistributed and partially located on the gas molecule due to the interaction between the counterparts. From the structural point of view, the molecule positions for NO(NO₂) at position 2-CB(6-CN) is lying on a graphene(hBN) site, and for NO(NO₂) at position 6-CN(2-CB) the molecule is located close to a C-B(C-N) part. We can note a charge concentration in a gas molecule for NO in position 6-CN and NO₂ for both positions. These results agree with the hints on the charge transfer from the material to the molecule as shown in Tab.1. For NO at position 2-CB we verify regions with negative and positive charge redistribution on the device and the effect is a quasi negligible charge transfer to the device with a the value showed in Tab.1. A part of this charge density is found between the gas molecule and the part of the layer closest to the molecule, underlining their significant interaction. These characteristics also point to a gas specific behavior in the electronic properties of the gas sensor nanodevice.
In the end, we attempt to quantify all the above results from our simulations in order to evaluate the sensitivity towards gas detection. All our findings point to distinct features in the case of the NO and NO$_2$ gas molecules. In the case of the former molecule, the features are much more pronounced. We define the sensitivity of our device as follows:

$$S(V_{\text{gate}}) = \left| 1 - \frac{g_{\text{gas}}}{g_0} \right| \times 100\%,$$

where we are using a specific gate voltage ($V_{\text{gate}}$) for the evaluation of the transmission, where $g_0$ is the reference conductance across the pristine G-hBN device and $g_{\text{gas}}$ the conductance of the device with gas molecule. From the sensitivity at a specific sampling gating voltage, we are able to evaluate how well the identity of a specific gas molecule can be resolved. The device sensitivity at two gate voltages according to the definition above and for all gas molecules is summarized in Fig.7. We have chosen for this, a positive and a negative voltage based on the findings in Fig.3. These had shown that NO can be better detected at negative energies (i.e. gate voltages), while NO$_2$ at positive energies. We can still confirm from Fig.7a) that at low energies a clear sensing of NO is possible. This is evident from the high sensitivity value for this molecule leading to a sensitivity over 40 %. At the same gate voltage, the device is less than 5 % sensitive in detecting NO$_2$. For the other two molecules, the device is not efficient. At the positive gate voltage, the situation is reversed. First, the device is 15 % sensitive in detecting NO, while the sensitivity rises close to 50 % for the NO$_2$ molecule. At the same gate voltage the device shows the same detection capability for identifying NH$_2$ and NO. The lowest sensitivity is found for CO$_2$. Note, that the results in the figure are given also as a function of the relevant adsorption energy. Accordingly, it is not the influence of the strength of the

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**Fig. 5** a) Transmission as a function of energy difference for the pristine device (G-hBN) and its b) respective density of states. In c) and e) the PDOS for the molecule NO adsorbed. By applying periodic boundary conditions the hybrid system is formed by alternated graphene and h-BN nanoroads, both of them are ∼13Å wide. These widths assures that the system possesses a metallic character, as the energy band gap is of the order of the thermal fluctuation at room temperature, and the interaction between the graphene nanorad and its periodic image becomes negligible [Nanotechnology 27 2016 365503]. on top of the CB interface at the position 2 and the CN interface at the position 6, respectively is shown. In d) and f) the PDOS for the gas molecule NO$_2$ at the same positions as in c) and e), respectively are given. The energy is shifted in all graphs, so that the Fermi level is set at 0 eV.

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**Fig. 6** The charge density redistribution at position 2-CB for the case of (a) NO, and (b) NO$_2$ adsorbed on the G-hBN sensor. Panels (c) and (d) depict the charge density redistribution at the position 6-CN for NO and NO$_2$, respectively. The axes are also shown and each panel includes a top and side view of part of the material and the molecule. The blue and red colors in the charge redistribution isosurfaces correspond to $\rho(\vec{r}) < 0$ and $\rho(\vec{r}) > 0$, respectively.
As representative molecules we have considered CO and hexagonal boron-nitride as a sensor for small gas molecules. The molecules are placed on top of the material at positions corresponding to their strongest interaction with the material (position 2 for CO, NH\textsubscript{3}, NO, and position 6 for NO\textsubscript{2}). The data in panels a) and b) correspond to a gating voltage of V\textsubscript{gut}=0.16 V and V\textsubscript{gut}=0.16 V, respectively.

**Fig. 7** Device sensitivity relevant to gas detection. The results are given for all gas molecules studied here with respect to their adsorption energy on the G-hBN sensor. The molecules are placed on top of the material at positions corresponding to their strongest interaction with the material (position 2 for CO\textsubscript{2}, NH\textsubscript{3}, NO, and position 6 for NO\textsubscript{2}). The data in panels a) and b) correspond to a gating voltage of V\textsubscript{gut}=0.16 V and V\textsubscript{gut}=0.16 V, respectively.

interaction, rather the type of the molecule that controls the sensitivity. At the same position, different molecules show a distinct interaction with the sensor, but may lead to the same sensitivity (see CO\textsubscript{2}, NH\textsubscript{3}, and NO at a positive gate voltage). In the same spirit, placing the target molecule on different positions also leads to distinct sensitivity factors. Overall, it should be made clear, that a thorough tuning of the device and its interactions with the gas molecules is essential in enhancing the gas detection potential of the G-hBN.

### 4 Conclusions

Based on quantum-mechanical simulations, we have proposed the use of a two-dimensional heterostructure made of graphene and hexagonal boron-nitride as a sensor for small gas molecules. As representative molecules we have considered CO\textsubscript{2}, NH\textsubscript{3}, NO\textsubscript{2}, and NO. We have first analyzed the interaction of these molecules with the G-hBN sensor by placing the molecules on top of the material at certain positions. We have found a much stronger interaction in the case of the NO\textsubscript{3}, which indicated a higher sensitivity towards sensing these molecules. Note, that these are better adsorbed on the different interfaces: NO on the CB interface, while NO\textsubscript{2} on the CN interface. The analysis on the electron charge redistribution showed a clear movement of the charge towards the molecule and varies with respect to the identity of the molecule and its placement on top of the G-hBN. The main results of our work involve the calculation of the electronic transmission of the combined gas/device system as compared to that of the pristine material. In the case of the CO\textsubscript{2} and NH\textsubscript{3} molecules, the electronic transmission is very close to that of the pristine device. Again, for the NO\textsubscript{3} molecules, the deviations are much stronger and detectable. The most distinct features were observed in the case of NO. Based on these, we could conclude with defining a gas detection sensitivity of the device. This clearly revealed the high potential of the G-hBN material in identifying the NO\textsubscript{3} molecules.

Interestingly, the detection of the NO gas is enhanced at lower gate voltages corresponding to electronic energies below the Fermi level. The opposite was found for the NO\textsubscript{2} gas, for which the device is more sensitive at positive gate voltages. In the end, we could show that a CO\textsubscript{2} or a NH\textsubscript{3} gas cannot be detected at a negative gate voltage, but a low detection is possible in the positive range. Note, that all results were obtained from *in vacuo* simulations, neglecting dynamics and the presence of other gas molecules. As a further step, we need to evaluate, how the trends provided here are modified by various factors. These are related to different concentrations of gas molecules and their mixtures, the presence of other impurities, the configurational variability of the molecules, possible defects on the material, etc. These aspects will possibly shift the trends we have observed here. Nevertheless, here, we have presented a proof of principles study to underline the high potential of the G-hBN ultra-thin material in sensing small gas molecules. This is an important step in further exploring the potential of this material as a gas sensor. It remains to be shown in which ways the exact microscopic details of such a material can be tuned in order to realize the sensing device.

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