Graphene membrane-based NEMS for study of interface interaction

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1. Introduction

A set of physical phenomena in bilayer and few layer graphene systems originate from relative rotation or displacement of the layers. Band gap and quantum transport depend on stacking of layers in trilayer graphene [1] and relative orientation of layers in bilayer graphene [2,3]. Superconductivity in twisted graphene bilayers have been recently reported [4]. Dynamic phenomena based on relative rotation or displacement of graphene layers include atomic-scale slip-stick motion of a graphene flake attached to a STM tip [5,6], rotation-assisted diffusion of a graphene flake [7], self-retracting motion of the layers at their telescopic extension [8,9] and motion of stacking dislocation by electric field [10].

In spite of the importance of interaction between graphene layers for fundamental physics and applications, only few works have been devoted even to determination of graphene-graphene binding energy. Namely, the following values of the binding energy have been obtained: $-43 \pm 5$ meV/atom based on heat of wetting data [11], $-52 \pm 5$ meV/atom using thermal desorption spectroscopy of polyaromatic hydrocarbons from graphite surface [12], $-35 \pm 10$ meV/atom [13], and $-31 \pm 2$ meV/atom [14] based on models which describe the balance between elastic and interlayer interaction energies to reproduce the geometry of cross-section of collapsed multilayer carbon nanotubes and multilayer graphene flake above a step of a graphite surface, respectively. Thus the considerable scatter of the obtained values of the graphene-graphene binding energy have been revealed by experimental studies using different techniques. The comprehensive first-principle calculations which give $\pm 56 \pm 6$ meV/atom by quantum Monte Carlo method [15] and $-36$ [16] and $-48$ meV/atom [17] using random phase approximation (RPA) should be also mentioned whereas numerous methods based on density functional theory with corrections for van der Waals interactions give the values from $-47$ to $-83$ meV/atom [18]. As for graphene-metal interaction so far as we know no experimental values on forces and energies of interface interaction between graphene and metals whereas calculations of graphene-metal interface energetics using RPA are available [19,20]. Thus elaboration of new techniques which intend to measure physical quantities related with energetics of interface interaction is very actual task.

Particularly the importance of accurate measurements of energetic characteristics of graphene-graphene and graphene-metal interface interaction is related with use of such characteristics as reference data for fitting of parameters of classical interatomic potentials. We consider this problem on example of 6–12 Lennard-Jones (LJ) potential [21] which is well-accepted for description of interaction energy $W_{LJ}$ between atoms of nonpolar molecules and has the form

$$W_{LJ} = 4\varepsilon \left( \frac{\sigma}{r} \right)^{12} - 2\varepsilon \left( \frac{\sigma}{r} \right)^{6}$$
A.I. SidhLo, et al

neighbor graphene layers and between atoms of graphene layer and membrane-based NEMS for measurement of characteristics of interface in-membrane based memory cell have been also considered [26]. Here we monstrates availability of graphene membrane-based NEMS. Graphene pressure [31,32] and mass [33] sensors and microphones [34] de-

Thus the direct measurements of characteristics of interaction between graphene layers in simulation of a carbon nanoscroll formation and possibility of the NEMS implementation is discussed in Section 2. Section 3 is devoted to calculations of operation characteristics of the NEMS on example of graphene-graphene interaction. Conclusions are summarized and possibility of the NEMS implementation is discussed in Section 4.
be performed by transmission electron microscopy (TEM). The schemes of experiment where manipulations with nanometer-size objects using a nanomanipulator are controlled by TEM imaging are available about 20 years [35].

3. Calculated characteristics of NEMS

For a simple model, we consider the NEMS based on a circular multilayer graphene membrane which has a clamped edge and a sample with a circular flat surface covered by graphene or graphite. The operational characteristics of the NEMS are determined by the following fixed sizes: the membrane diameter \( L \), the sample diameter \( l \), and the membrane thickness \( h \) (see Fig. 1c). Along with these fixed sizes there are two sizes which are changing during the NEMS operation: the distance \( d_0 \) between surface of the sample and center of the flat membrane (which is tuned by the nanomanipulator) and the distance \( d \) between surface of the sample and the bent membrane. Note that both distances \( d_0 \) and \( d \) can be measured in principle. The sticking distances \( d_{os} \) and \( d_s \) and the detachment distances \( d_{ot} \) and \( d_d \) correspond to the abrupt transitions between states 1 and 2 and between states 2 and 1, respectively.

Let us obtain the expressions which relate fixed sizes of the NEMS, \( L \), \( l \), and \( h \), the sticking distances \( d_{os} \) and \( d_s \) or the detachment distances \( d_{ot} \) and \( d_d \) and parameters of classical potential describing interaction between atoms of the membrane and the sample surfaces. For this purpose we consider energy balance of the NEMS on the example of the LJ potential (1). The total energy \( W \) of the NEMS is the sum of the van der Waals interaction energy \( W_{vdw} \) between the membrane and the sample and the elastic energy \( W_e \) of the membrane, \( W = W_{vdw} + W_e \).

The elastic energy \( W_d \) of the circular membrane with a clamped edge which is loaded in the center has the form [36]:

\[
W_d = 16 \pi D \frac{2}{L^2} (d_0 - d)^2, \tag{2}
\]

where \( d_0 - d \) is the deflection of the membrane center and \( D \) is the membrane flexural rigidity

\[
D = \frac{E h^3}{12(1 - \nu^2)}, \tag{3}
\]

where \( E \) and \( \nu \) are the Young's modulus and the Poisson's ratio, respectively, \( h \) is the membrane thickness. According to the measurements of the flexural rigidity of the multilayer graphene membranes, formula (3) is applicable for membranes with thickness \( h \) exceeding 3 nm [37] (about 10 graphene layers) with the values of elastic constants \( E = 0.92 \text{TPa}, \nu = 0.16 \) measured for graphite [38]. Here we use these values of the Young's modulus and the Poisson's ratio.

Here we consider the case where \( d \ll l \) and \( l \ll L \). In this case the van der Waals interaction energy \( W_{vdw} \) between the parallel membrane and the sample located at the distance \( d \) from the membrane center is given by Ref. [26]:

\[
W_{vdw} = \frac{\pi \sigma_{12}^2}{S_a} \left( \frac{1}{5d^{10}} - \frac{1}{2d^4} \right), \tag{4}
\]

where \( S_a \) is the area per one atom of graphene layer, \( S_a = 2.619 \text{Å}^2 \). The parameters of the LJ potential \( \epsilon = 2.757 \text{meV} \) and \( \sigma = 3.393 \text{Å} \) are fitted [22] to reproduce the interlayer binding energy, equilibrium interlayer spacing and c-axis compressibility of graphite are used at calculations below.

Thus the total energy of the NEMS takes the form

\[
W = W_{vdw} + W_d = \frac{\pi \sigma_{12}^2}{S_a} \left( \frac{1}{5d^{10}} - \frac{1}{2d^4} \right) + \frac{32 \pi D (d_0 - d)^2}{L^2} = \frac{a}{5d^{10}} - \frac{b}{2d^4} + c(d_0 - d)^2, \tag{5}
\]

where

\[
a = \frac{\pi^2 \sigma_{12}^2}{S_a}, \quad b = \frac{\pi^2 \sigma_{12}^2}{S_a}, \quad c = \frac{32 \pi D}{L^2}.
\]

The dependences of both components of the total energy \( W \) of the NEMS, the elastic energy \( W_d(d_0) \) and the van der Waals interaction energy \( W_{vdw}(d) \), on the distance \( d \) between the surfaces of the bent membrane and the sample at the fixed distance \( d_0 \) between the surfaces of the flat membrane and the sample are shown in Fig. 2a. The bistability of the NEMS has its origin in presence of the single minimum in dependences of both energies \( W_d(d) \) and \( W_{vdw}(d) \) at different distances \( d \). To show loss of the bistability of the NEMS with the change of the fixed distance \( d_0 \) between the surfaces of the flat membrane and the sample the dependences \( W(d) \) of the total energy \( W \) of the NEMS on the distance \( d \) between the surfaces of the bent membrane and the sample are shown in Fig. 2b for three fixed distances \( d_0 \). Namely, line 3 corresponds to some distance \( d_{os} \) where the system is bistable (that is two minima in the dependence \( W(d) \) are present) and line 2 corresponds to sticking distance \( d_{os} < d_{os} \) where the second minimum in the dependence \( W(d) \) is vanished and the abrupt transition from state 1 to state 2 occurs. This means that sticking of the membrane takes place when the sample is moved toward the membrane by the nanomanipulator. Analogously the abrupt transition from state 2 to state 1 (detachment of the membrane) occurs at backward motion of the sample. (Line 1 corresponds to distance \( d_0 < d_{os} \) where the dependence \( W(d) \) also has only the first minimum.) The loss of the bistability of the NEMS, that is the sticking and detachment of the membrane at forward and backward motion of the sample, respectively, means hysteresis in the dependences of the total potential energy \( W(d_0) \) and the distance \( d(d_0) \) between the surfaces of the bent membrane and the sample on the distance \( d_0 \) between the surfaces of the flat membrane and the sample (see Fig. 3).

For the bistable NEMS the function \( W(d) \) has two minima at \( d_{min1} \) and \( d_{min2} \) which correspond to the stable states 1 and 2, respectively, the maximum \( d_{max} \) between these minima and two inflection points at \( d_{in1} \) and \( d_{in2} \) between \( d_{min1} \) and \( d_{max} \) and between \( d_{min2} \) and \( d_{max} \), respectively. For the sticking distance \( d_{os} \) between the surface of the
As discussed above the distances $d$ are also roots of the equation $s d$ where $a$, $b$, and $c$ are defined by Eq. (6).

The inflection points are determined by the condition $\frac{d^2 W}{dd^2} = 0$ which using Eq. (5) takes the form

$$\frac{11a}{d^3} = \frac{5b}{d^2} + c = 0.$$ (7)

The Eq. (7) does not contain distance $d_0$ between the surface of the sample and the center of the flat membrane. Therefore the values of the inflection points $d_{in1}$ and $d_{in2}$ do not change at moving of the sample by the nanomanipulator. Thus the inflection points determined by Eq. (7) are the sticking distances $d_s$ and the detachment distances $d_d$ between the surface of the sample and the bent membrane

$$d_{in1} = d_s = \left(\frac{5b + \sqrt{25b^2 - 44ac}}{2c}\right)^{1/6},$$

$$d_{in2} = d_d = \left(\frac{5b - \sqrt{25b^2 - 44ac}}{2c}\right)^{1/6}. $$ (8)

As discussed above the distances $d_s$ and $d_d$ are also roots of the equation $\frac{dW}{dd} = 0$ which takes the form

$$c(d - d_0) + \frac{b}{d^3} - \frac{a}{d^4} = 0.$$ (9)

where $a$, $b$, and $c$ are defined by Eq. (6).

The sticking distance $d_0$, and the detachment distance $d_{od}$ between surface of the sample and center of the flat membrane are found by substituting $d_s$ and $d_d$, respectively (which are determined by Eq. (8)), into Eq. (9). Thus, it yields

$$d_0 = d_s + \frac{b}{cd_s^2} = \frac{a}{cd_s^4}, \quad d_{od} = d_d + \frac{b}{cd_d^2} - \frac{a}{cd_d^4}.$$ (10)

The concept of the NEMS proposed in Section 2 is based on measurements of the sticking distance $d_s$ between the surface of the sample and the bent membrane while moving sample toward the membrane.

Fig. 3. Calculated dependences of the distance $d$ between the surfaces of the bent membrane and the sample (top panel) and the total potential energy $W$ of the NEMS (bottom panel) on the distance $d_0$ between the surfaces of the flat membrane and the sample at forward and backward motion of the sample (shown by arrows). In order to show the energy $W$ in a double logarithmic scale the minimum value $W_{min}$ of the energy $W$ (at the distance $d_0 = 0.34$ nm) is used as a reference.

sample and center of the flat membrane (where the transition from state 1 to state 2 takes place) the values of $d_{max}$, $d_{min1}$, and $d_{min2}$ coincide (see Fig. 2). Similarly, for the detachment distance $d_{od}$ the values of $d_{max}$, $d_{min2}$, and $d_{min2}$ coincide.

Let us use Eq. (11) to consider the range of the distances $d_s$ for the NEMS with the different membrane thicknesses and membrane and sample diameters (see Fig. 4). The calculated sticking distance $d_s$ ranges from 3 to 100 nm and from 1 to 30 nm for the NEMS based on 10-layer and 100-layer graphene membranes, respectively. Note that even for the smallest sticking distance $d_s = 0.5$ nm for considered sizes of the NEMS (which is obtained for sample diameter $l = 2$ nm and 10-layer membrane diameter $L = 200$ nm) the relative difference between values of $d_s$ calculated using Eqs. (10) and (11) is only 0.03. This relative difference decreases with the increase of the sticking distance. The distances near the upper limits of the calculated distances ranges which can be easily measurable by TEM correspond to the sample diameter $l = 1\mu m$ and membrane diameter $l = 10\mu m$.

4. Discussion and conclusions

We propose the NEMS designed to study interaction between graphene and surface of a sample. This NEMS is based on a multilayer graphene membrane which bends due to van der Waals attraction to surface of a sample. The performed analysis of the balance between

\[ d_s = \frac{1}{2} \left( \frac{5\pi A}{2D} \right)^{1/6} \left( \frac{L}{S_h} \right)^{1/3}, \]

\[ d_{od} = \frac{6}{5} d_s = \frac{3}{5} \left( \frac{5\pi A}{2D} \right)^{1/6} \left( \frac{L}{S_h} \right)^{1/3}. \]

(positioning the sample at the distance $d_{od}$ from the flat membrane).

Fig. 4. Calculated dependences of the sticking distance $d_s$ (in nm) according to Eq. (11) on the ratio of the membrane and sample diameters $L/l$ for NEMS with the different membrane thicknesses $h = 33.5$ nm (100 graphene layers; dashed lines) and $h = 3.35$ nm (10 graphene layers; solid lines) and different sample diameters $l = 0.1\mu m$ (lines 1 and 3) and $l = 1\mu m$ (lines 2 and 4).

Thus only the attractive term $-A/r^6 = -4\pi\rho^2/r^6$ with the single parameter $A$ can be taken into account in the LJ potential (1). In this case the sticking distance $d_s$ from Eq. (8) (formally at $a \to 0$) takes the form

with the sticking distance $d_{od}$ from Eq. (10) takes the form

The concept of the NEMS proposed in Section 2 is based on measurements of the sticking distance $d_s$ between the surface of the sample and the bent membrane while moving sample toward the membrane.
Such measurements can also be used in combination with other
A.I. Siahlo, et al.
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\section*{Data availability}

The raw data required to reproduce these findings are available to download [41].

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