Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Materials

The adhesion energy measured by a stress accumulation-peeling mechanism in the exfoliation of graphite

Minggang Xia[a, b, c, *], Chunping Liang[a, c], Zhaofang Cheng[a, c], Ruixue Hu[a, c], Shiru Liu[a, c]

[a] Department of Photoelectronic Information Science and Engineering, School of Science, Xi'an Jiaotong University, 710049, People's Republic of China

[b] Laboratory of Nanostructure and Physics Properties, Shaanxi Province Key Laboratory of Quantum Information and Optoelectronic Quantum Devices, School of Science, Xi'an Jiaotong University, 710049, People's Republic of China

[c] MOE Key Laboratory for Nonequilibrium Synthesis and Modulation of Condensed Matter, School of Science, Xi'an Jiaotong University, 710049, People's Republic of China

* Corresponding Author (M. X.) E-mail: xiamg@mail.xjtu.edu.cn

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1. Calculation of Graphite Adhesion Energy

Here, the graphite adhesion energy is defined as the energy expended when two pieces of graphite are detached far away each other. In order to calculate the graphite adhesion energy, we should figure out the van de Waals interaction between two pieces of graphite. Therefore, we chose Lennard-Jones potential $U(d)=-A/d^6+B/d^{12}$, where d is the distance between carbon and carbon atoms, A=15.54 eV $Å^6$, and B=2.40×10^4 eV $Å^{12}$. Although there are two different stacking styles for graphene layers, the relative positions of atoms on up and bottom layers keep invariable, seen in Fig. 1a in the text. Therefore, we chose two atoms in one unit on the top layer graphene and assume that each atom is located on the origin of coordinates. Such that, there are two cases, seen in Fig. 1S. In the first case, the projection position of the atom in the top layer is just at the center of hexagon in the bottom layer, seen B_2 atom in Fig.1a. The second one, the projection position of another atom in the top layer is just at the atom in the bottom layer, seen A_2 atom in Fig.1a. Then, we should calculate the distance between each atom in the unit in top layer and each atom on the bottom layer. In order to give a general expression, we express the position vector for each atom by the lattice basic vector $\vec{a}_1$ and $\vec{a}_2$. For the first case, the position vectors can be written as:

$$\begin{aligned} \left|\vec{b}_{A'}\right| &= \frac{a}{2}\sqrt{3(1+n_1+n_2)^2+(3n_1-3n_2+1)^2} \\ \left|\vec{b}_{B'}\right| &= \frac{a}{2}\sqrt{3(1+n_1+n_2)^2+(3n_1-3n_2-1)^2} \end{aligned} \tag{1}$$

where a is the bond length, $\vec{a}_1=\frac{\sqrt{3}}{2}a\vec{i}+\frac{3}{2}a\vec{j}$ and $\vec{a}_2=\frac{\sqrt{3}}{2}a\vec{i}-\frac{3}{2}a\vec{j}$.

For the second case, the position vectors can be written as:

$$\begin{aligned}\left|\vec{b}_{A''}\right| &= \frac{a}{2}\sqrt{3(n_1+n_2)^2+(3n_1-3n_2)^2} \\ \left|\vec{b}_{B''}\right| &= \frac{a}{2}\sqrt{3(n_1+n_2)^2+(3n_1-3n_2+1)^2}\end{aligned} \tag{2}$$

where $\vec{a}_1 = 0$ and $\vec{a}_2 = a\vec{j}$.

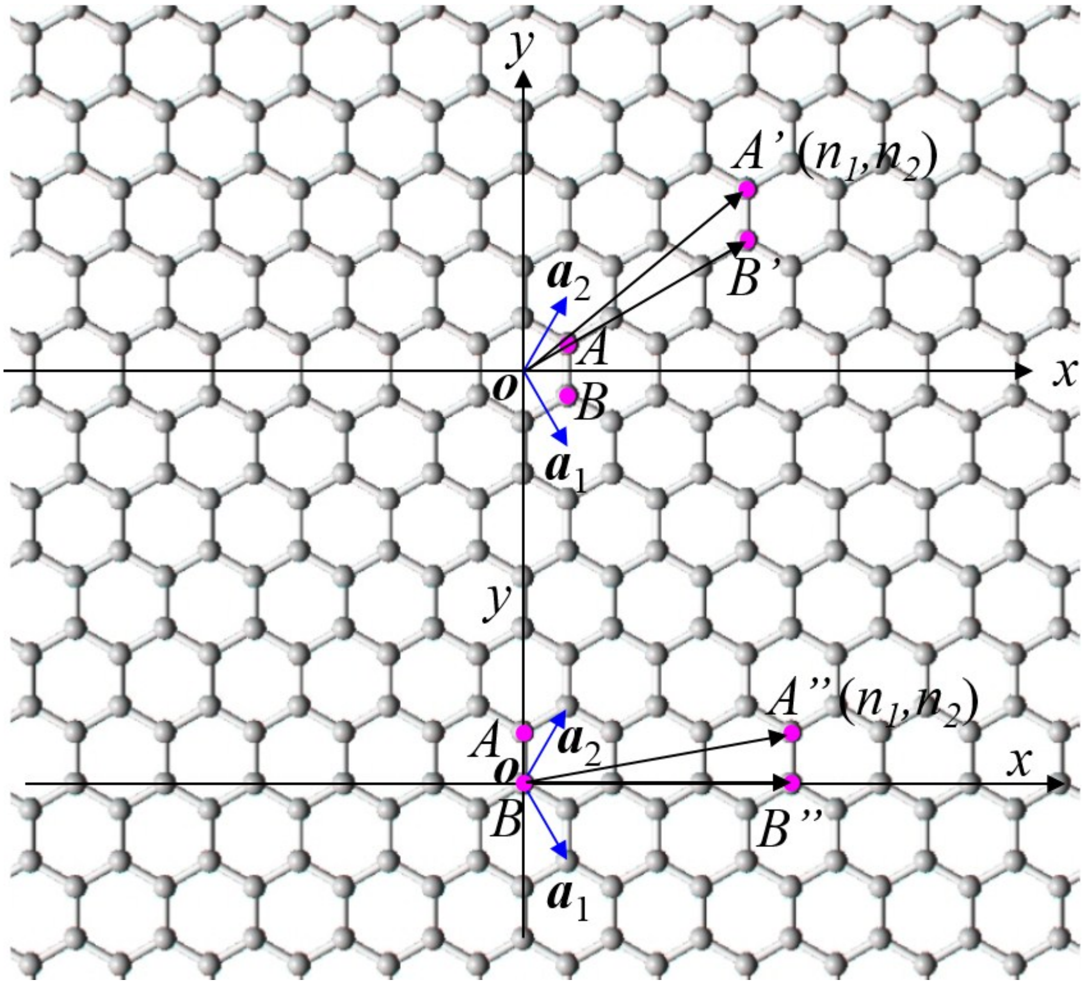

Fig.S1 Atom displace vectors in bottom layer. The atoms B and A in top layer are located at the origin of coordinates, which are top and bottom Cartesian coordinates.

From (1) and (2), we can calculate the van de Waals interaction energy by Lennard-Jones potential. One hundred and fifty layers of graphene in two detached pieces of graphite are involved in the calculation. We obtain the adhesion energy of graphene about 0.29 J/m^2 by Matlab program written by us, which is a bit less than the molecular dynamic (MD) result 0.354 J/m^2 [1]. The difference is related to the coefficients A and B in Lennard-Jones potential.

2. Matlab Code for Calculating the Adhesion Energy of Graphite

```
clear
clc
a=1.42e-10;  % unit is m
c0=3.4e-10;  % unit is m
ceV=1.6e-19; % 1eV=1.6e-19 J
A=(15.54e-60)*ceV; % unit is J*m^6
B=(24000e-120)*ceV; % unit is J*m^12
r0_0=c0;
W=0.0;
N0=150;
N=200;
S=((3*sqrt(3)/2)*a^2);

for i=1:1:N0
   c=c0*i;
   Wj=0.0;
for n1=-N:1:N
   for n2=-N:1:N
      % A and B atoms with different (x,y)
      rA0=a*sqrt(3*(1+n1+n2)^2+(3*n2-3*n1+1)^2)/2;
      rA=sqrt(rA0^2+c^2);
      rB0=a*sqrt(3*(1+n1+n2)^2+(3*n2-3*n1-1)^2)/2;
      rB=sqrt(rB0^2+c^2);

      % A and B atoms with same (x,y)
      rA1=a*sqrt(3*(n1+n2)^2+(3*n2-3*n1)^2)/2;
      rAB=sqrt(rA1^2+c^2);
      rB1=a*sqrt(3*(n1+n2)^2+(3*n2-3*n1-1)^2)/2;
      rBA=sqrt(rB1^2+c^2);

      Wj=Wj+(A/(rA^6)-B/(rA^12))+(A/(rB^6)-B/(rB^12))+(A/(rAB^6)-
B/(rAB^12))+(A/(rBA^6)-B/(rBA^12));
```

```
   end
end
W=Wj/S+W;
end
fid=fopen('F:\Graphite20181009.txt', 'at');
fprintf(fid, '%6.4e %6.4e %6.4e %6.4e\n',rA,rAB,c,W);
fclose(fid);
```

3. Theory Basis and Measurement Principle of Adhesion Energy

In our case, we can’t use the method in reference [2] based on two reasons as follow. *First*, the adhesion energy (~10^{-1} J/m^2) of graphite [3-11] is less two orders than the total energy (~10^{1} J/m^2) [2] stored in the peeling arm, and at the same time accompanying a viscoelastic energy change in our whole test process. *Second*, the peeling arm in our case is a tape, consisting of a piece of thin plastic paper (~40 μm) and glue (~10 μm), which is different from the single layer peeling arm in reference [2].

The work done by external force equals to the total energy change including the five terms, (i) the elastic energy by bending peeling arm, (ii) the viscoelastic energy of glue, (iii) the kinetic energy of peeling front, (iv) the adhesion energy and (v) the tearing energy of graphite. In our test process, the energy conservation should be satisfied. So, we can figure out the adhesive energy G_a from an energy conservation argument [2],

$$G_a = \frac{1}{b}\left(\frac{dW_{ext}}{dx} - \frac{dW_b}{dx} - \frac{dW_{vis}}{dx} - \frac{dW_{tear}}{dx} - \frac{dE_K}{dx} \right), \tag{3}$$

in which dx is the infinitesimal length along the peeling front, b is the width of peeling front, dW_{ext} is the work by an external force, dW_b is the elastic energy by bending the peeling arm, dW_{vis} is the viscoelastic energy stored in the stretched glue, and dW_{tear} is the tearing energy of graphite slice. dE_K is the kinetic energy of the peeling front.

In the test process, the peeling arm is always in elastic strain rage because the first reason presented above. In addition, we can find easily that there are two approximately repeated steps in the whole test process, the stress accumulation (the tape bend) process and the peeling process according to the movie, seen in ***the Movies 1*** and ***2***. Therefore, in order to figure out the adhesion energy of graphite, we present the process of energy transfer as follow. The energy transfer is at first from the work done dW_{ext} by external force to the elastic energy dW_b by bending peeling arm and the viscoelastic energy dW_{vis} of glue, and then to the kinetic energy dE_K of the peeling front, the adhesion energy G_abdx and the tearing energy dW_{tear} of graphite slice. The previous process is just the stress accumulation process, while the latter refers to the peeling process.

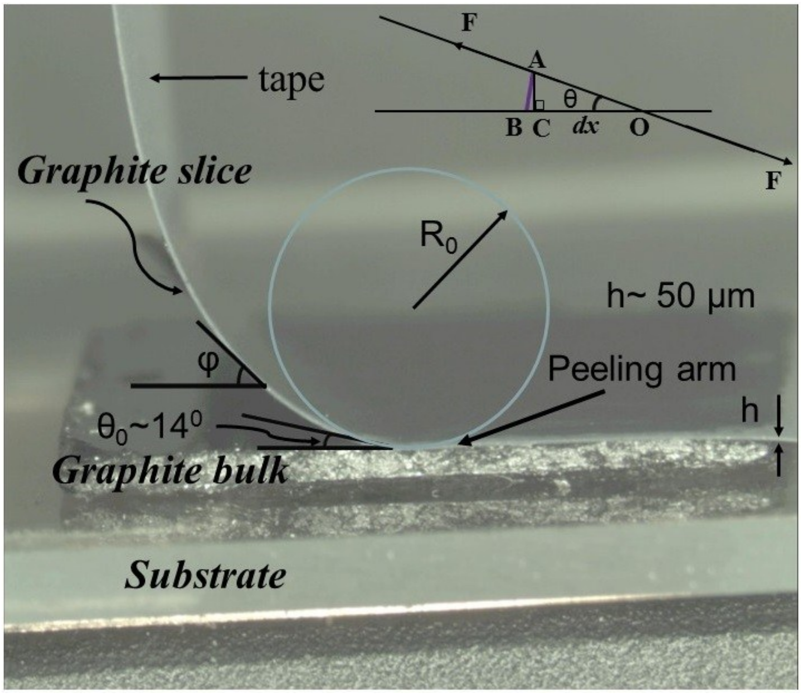

Fig. S2. A schematic diagram for showing the geometric construction in the process of peeling graphite slice.

Now, the most important question is how to separate the adhesion energy from two other terms. The tearing energy term can be excluded by peeling almost whole piece of graphite slice that is almost no tearing. This condition is also available if the graphite is peeled wholly in a certain period of peeling process. Such that, only the kinetic energy should be excluded in our case. According to later analysis, we can know that the kinetic energy of the peeling front comes from the viscoelastic energy stored in the glue. A photograph image before peeling is shown in Fig. S2.

According to the test process presented above, there is still a lot residual elastic energy stored in the peeling arm after peeling, including to the elastic energy by bending a tape and the viscoelastic energy of glue. In order to figure out the perpendicular component of stretch force to bulk graphite, we zoom-in the peeling front at the top-right panel in Fig. S2. From the geometry, we can calculate the perpendicular component of stretch force $F_{\perp} = F\sin\theta$ and the work for peeling graphite $dW_{\perp} = F\sin^2\theta dx$ when the peeling front moves a distance dx $(dx = OB = OA)$. The force F and peeling angle θ vary as the place of the peeling front. Thus, when the peeling front moves Δx, we can write the work for peeling graphite and the dissipation energy along peeling front as

$$\Delta W_{\perp} = \int_{x_0}^{x_0+\Delta x} F\sin^2\theta dx\,, \tag{4}$$

and

$$\Delta W_{\mathrm{P}} = \int_{x_0}^{x_0+\Delta x} F\cos^2\theta dx\,, \tag{5}$$

where the x_0 and x are just the coordinates of peeling front as $\theta = \theta_0$ and $\theta = \theta_{\min}$, respectively. The dissipation energy along peeling front is provided by the viscoelastic energy stored in the glue. The total work ΔW dissipated in the peeling process can be written as

$$\Delta W = \int_{x_0}^{x_0+\Delta x} F dx \, . \tag{6}$$

And the work $\varDelta W_{\mathrm{ext}}$ by the external force can be written as

$$\Delta W_{ext} = \Delta W_f = \int_{l_0}^{l_0+\Delta l} F dl \, . \tag{7}$$

The adhesion energy G_a of graphite equals to the work of peeling graphite slice per area, so we have

$$G_a = \frac{\Delta W_\perp}{b\Delta x} = \frac{\int_{x_0}^{x_0+\Delta x} F \sin^2 \theta dx}{b\Delta x} \, . \tag{8}$$

In the whole process, the external stretch force equals to the stretch force in the glue, shown in the inset of Fig. S2. In our test, we keep the velocity v_l of the external stretch force invariant, so $dl = v_l dt$. We assume that the velocity of the peeling front in the peeling process is a constant, then we have $dx = v_x dt$. According to the Eqs. (6) and (7), we can obtain the relation between the work $\varDelta W_f$ by external force and the total work $\varDelta W$ dissipated in the peeling process as follow:

$$\frac{\Delta W_f}{\Delta W} = \frac{\Delta l}{\Delta x} \tag{9}$$

Moreover, Eq. (9) also can be obtained from another way. We *first* assume that the tape length is invariable. Then we can obtain $dx = dl + ds$ according to geometry of the system, in which dl and ds are the displacement of the external stretch force and the arc length variation when the peeling front moves dx. *Afterwards*, we assume that $dl = dx$ according to the quickly moving of peeling front in the peeling process. And then we have $dx = ds$. *After that*, we can obtain $ds = \pi dR / 2$ when the curved tape can be considered as a part of arc from the geometry in Fig. S2. *Finally*, we have $dl \propto dx$. To sum up, we can also obtain the Eq. (9) according to the Eqs. (6) and (7).

By substituting Eq. (9) into Eq. (8), we can obtain

$$G_a = \frac{\Delta W_f \int_{x_0}^{x_0+\Delta x} F\sin^2\theta dx}{b\Delta l\Delta W} = \frac{1}{b\Delta l}\frac{\int_{l_0}^{l_0+\Delta l} Fdl \int_{x_0}^{x_0+\Delta x} F\sin^2\theta dx}{\int_{x_0}^{x_0+\Delta x} Fdx}, \tag{10}$$

Meanwhile the last term can be written as:

$$\frac{\int_{l_0}^{l_0+\Delta l} Fdl \int_{x_0}^{x_0+\Delta x} F\sin^2\theta dx}{\int_{x_0}^{x_0+\Delta x} Fdx} = \Delta W_f - \int_{l_0}^{l_0+\Delta l} Fdl \frac{\int_{x_0}^{x_0+\Delta x} F\cos^2\theta dx}{\int_{x_0}^{x_0+\Delta x} Fdx} \tag{11}$$

According to assumption about the constant velocity of peeling front, we can obtain that the dependence of *dl* on θ is the same as that of *dx*. Thus, Eq. (11) can be simplified as

$$\Delta W_f - \int_{l_0}^{l_0+\Delta l} Fdl \frac{\int_{x_0}^{x_0+\Delta x} F\cos^2\theta dx}{\int_{x_0}^{x_0+\Delta x} Fdx} = \Delta W_f - \int_{l_0}^{l_0+\Delta l} F\cos^2\theta dl \tag{12}$$

By substituting Eqs. (11) and (12) into Eq. (10), we can obtain,

$$G_a = \frac{\Delta W_f \int_{x_0}^{x_0+\Delta x} F\sin^2\theta dx}{b\Delta l\Delta W} = \frac{1}{b\Delta l}(\Delta W_f - \int_{l_0}^{l_0+\Delta l} F_{\mathrm{P}}\cos\theta dl)\ , \tag{13}$$

and that

$$\Delta W_f - F_{\min}\Delta l = \int_{l_0}^{l_0+\Delta l} Fdl - F_{\min}\Delta l = \int_0^{\Delta l} fdl \tag{14}$$

At the last moment of the peeling process, the external stretch force is the minimum locally when the external stretch force equals to the viscoelastic force along horizontal direction, $F_{\|}$. In the case, the last term in brackets in Eq. (13) is equivalent to the work by the minimum stretch force along curve, $F_{min}\Delta l$. Therefore, the adhesion energy of graphite can be written as

$$G_a = \frac{\Delta W_f - F_{\min}\Delta l}{b\Delta l}, \tag{15}$$

where F_{min} is the minimum stretch force detected by stretch force sensor.

On the other hand, the adhesion energy of graphite is a far less than the total energy stored in the peeling arm, so the peeling angle θ changes a little in the peeling process, and then we have $\theta \approx \theta_0$ and $\theta \approx \theta_{min}$. In the case, we can obtain the work for peeling graphite slice $\Delta W_{\perp} = \Delta W \sin^2\theta_0$. Then, the adhesion energy of graphite can be written as

$$G_a = \frac{\Delta W \sin^2\theta_0}{b\Delta x}. \tag{16}$$

Actually, adhesion energy of graphite does not relate to the total work ΔW dissipated in the peeling process because ΔW is in direct proportion to the Δx according to Eq. (6). Then, Eq. (16) can be rewritten as

$$G_a = \frac{\Delta W_f \sin^2\theta_0}{b\Delta l} = \frac{\Delta W_f - \Delta W_f \cos^2\theta_0}{b\Delta l}. \tag{17}$$

The term $\Delta W_f \cos^2\theta_0 = F_{||}\,\Delta l\,\cos\theta_0$, which is just the work by the minimum stretch force along curve. Therefore, Eq. (13) is just equivalent to Eq. (17) when $\theta=\theta_0$.

Hereto, we presented the adhesion energy of graphite relating to the work by the external force. Hereafter, we should give the work by the external force for the viscoelastic glue. For viscoelastic composite, the relationship between the stress f and the strain s is quadric, $f = cl^2$ (c is a constant, and l is the displacement) [2, 12-14]. The work by viscoelastic force f can be written as

$$W_f = c(l_2^3 - l_1^3)/3 = (f_2 l_2 - f_1 l_1)/3 = (W_{f2} - W_{f1})/3\,, \tag{18}$$

in a peeling process according to the work definition $W_f = \int f(l)dl$. The work by viscoelastic force f can be obtained by integrating the stretch force along the displacement, seen in Fig. S3.

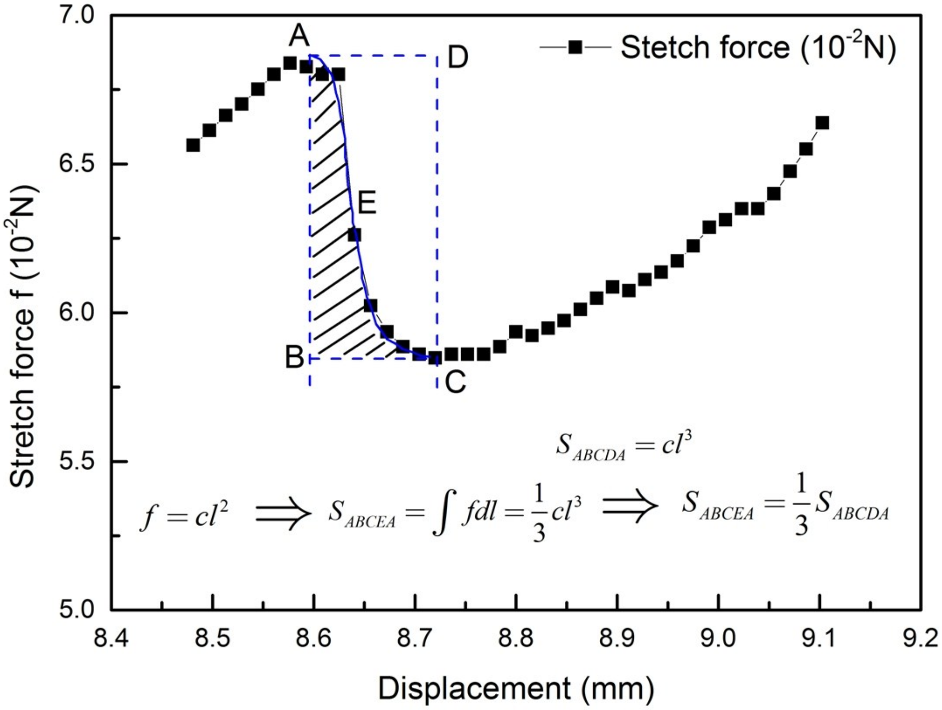

Fig. S3 An example for showing to calculate the work by viscoelastic force from the stretch force vs. displacement curve. S$_{\mathrm{ABCDA}}$ is the change in the energy ΔE of the system. S$_{\mathrm{ABCEA}}$ is just $G_a b \Delta s$. The data is selected from the segment between 8.5 and 9.1 mm in Fig. S6(b).

Therefore, we can figure out

$$G_a = \frac{F_{\max} - F_{\min}}{3b}\,, \tag{19}$$

in which the adhesion energy of graphite slice equals to the one third of the difference between the local maximum and the minimum force divided by *b*, seen in Fig. S3.

Furthermore, we can use the result about a thin layer peeling off a body in Reference [15], from which the peeling energy (that is G_a) is half of the change in the energy of the system. Considering our case, we can figure out the change in the energy *ΔE* of the system according to the curve of external stretch force *F* vs. its displacement *Δl*. In our system, the viscoelastic force is assumed as the inner force in the system. Therefore, we can write clearly

$$\Delta E = F_{\max}\Delta l - \int F dl = \frac{2}{3}(F_{\max} - F_{\min})\Delta l \,, \tag{20}$$

in which the integral in the Eq. (20) is completed with the help of the work by viscoelastic force. So we have

$$G_a = \frac{\Delta E}{2}\frac{1}{b\Delta l} = \frac{F_{\max} - F_{\min}}{3b} \,, \tag{21}$$

in which F_{max} and F_{min} are the maximum and minimum external stretch force in one circle, respectively. Eq. (21) is the same as Eq. (19). To date, we obtain the adhesion energy expression from two models, separately.

4. The Lower Velocity to Peel Four Graphite Slices.

The arithmetic average of the external force difference value (F_{max}-F_{min}) can be obtained by

$$\overline{F} = \sum_{i=1}^{n}(F_{\max} - F_{\min})_i / n \,, \tag{22}$$

in which n is the number of (F_{max}-F_{min}). Its standard deviation of arithmetic mean can be written as

$$\Delta F=\sqrt{\frac{\sum_{i=1}^{n}[(F_{\max}-F_{\min})_i-\overline{F}]^2}{n(n-1)}}\,, \tag{23}$$

Therefore, we have the external stretch force as

$$F=\overline{F}\pm\Delta F\,, \tag{24}$$

and the adhesion energy can be written as

$$G_a=(\overline{F}\pm\Delta F)/3b\,, \tag{25}$$

The all data above the external stretch force and the adhesion energy are obtained by Eqs. (22)-(25) in Figures in text and the ***Supporting Materials***.

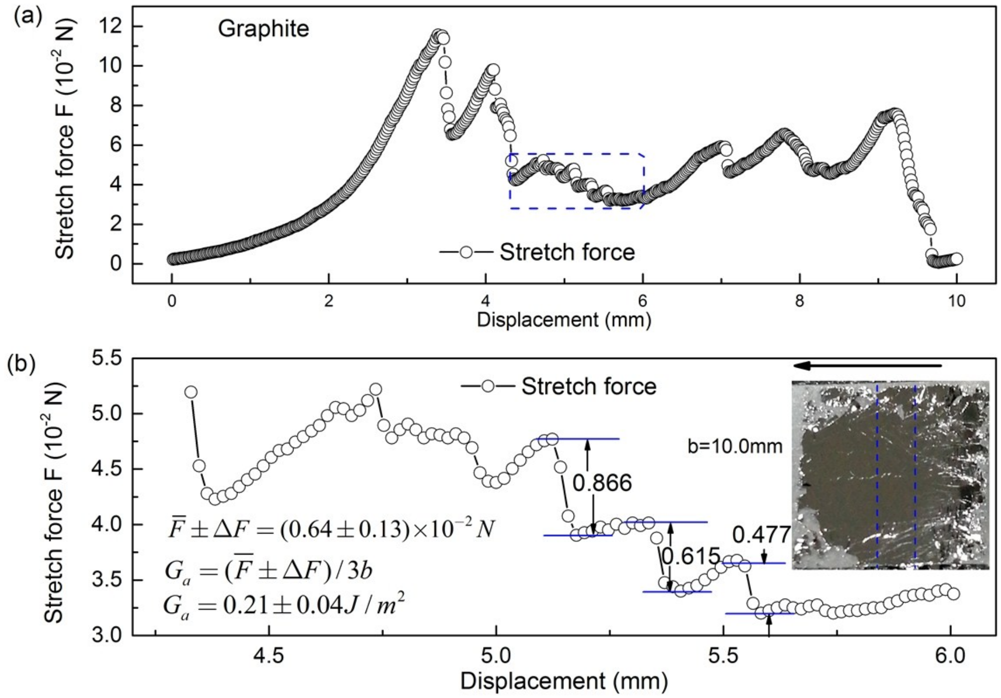

Fig. S4. The stretch force vs. the stretch displacement for a piece of graphite slice peeling from bulk graphite. (a) is plotted for showing all of data. (b) is zoomed in the blue dashed square in (a) to figure out the adhesion energy by the “stress accumulation-peeling” mechanism. The arrows in (a) for indicating stretch direction. The insets in (b) is the graphite slice after peeling from the bulk graphite. The blue dashed square corresponds to the peeling process between two blue vertical dashed lines on the inset, graphite slice. Less data leads to larger deviations. The stretch average velocity is about 0.084 mm/s.

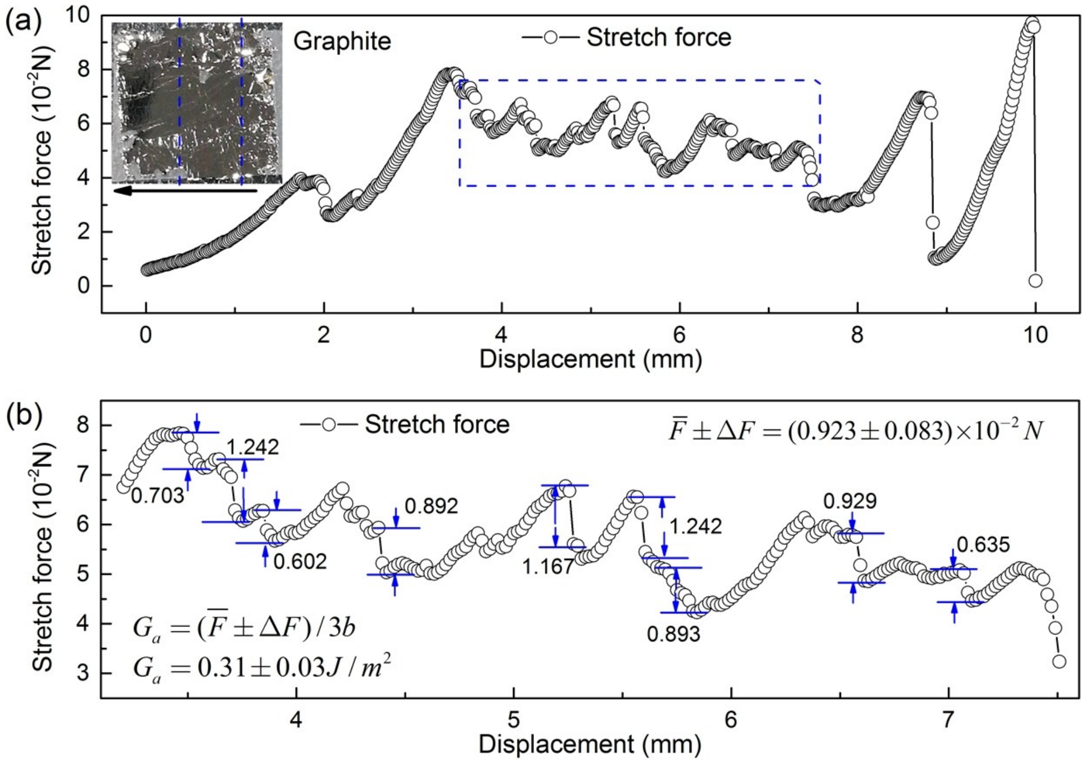

Fig. S5. The stretch force vs. the stretch displacement for another piece of graphite slice peeling from bulk graphite. The captions are the same as Fig. S4. The stretch average velocity is about 0.084 mm/s.

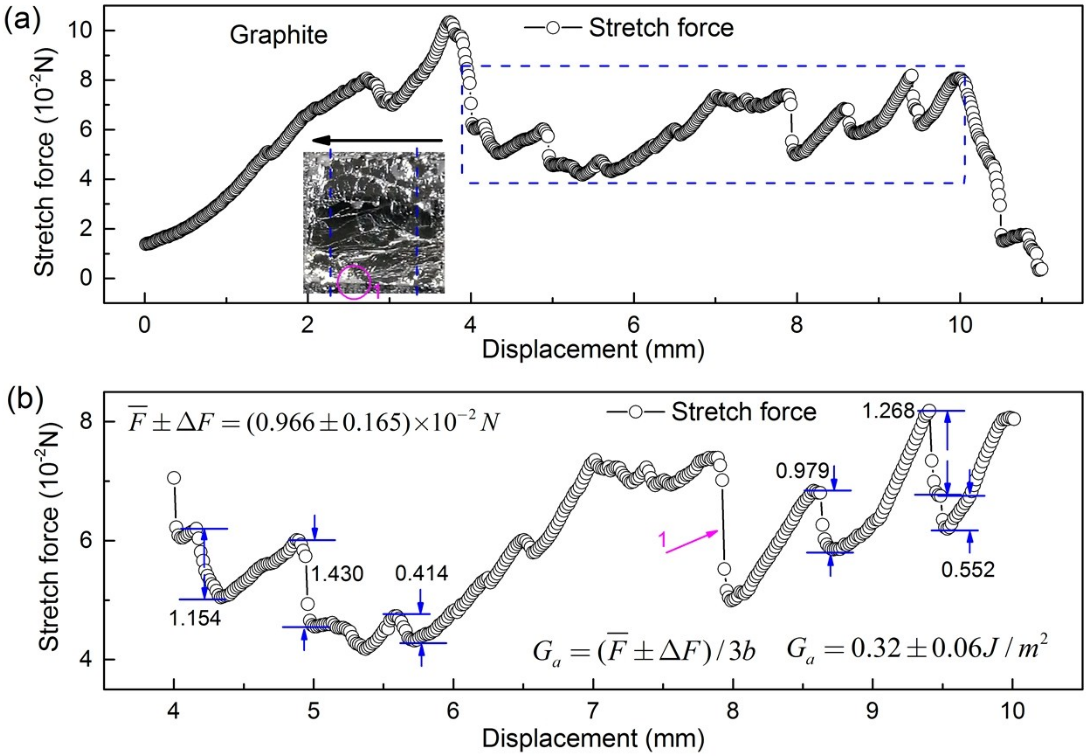

Fig. S6. The stretch force vs. the stretch displacement for another piece of graphite slice peeling from bulk graphite. The captions are the same as Fig. S4. There is a big hole in graphite marked by a circle with a label 1 in the inset of (a), and the corresponding the position also is denoted as an arrow in (b). The stretch average velocity is about 0.084 mm/s.

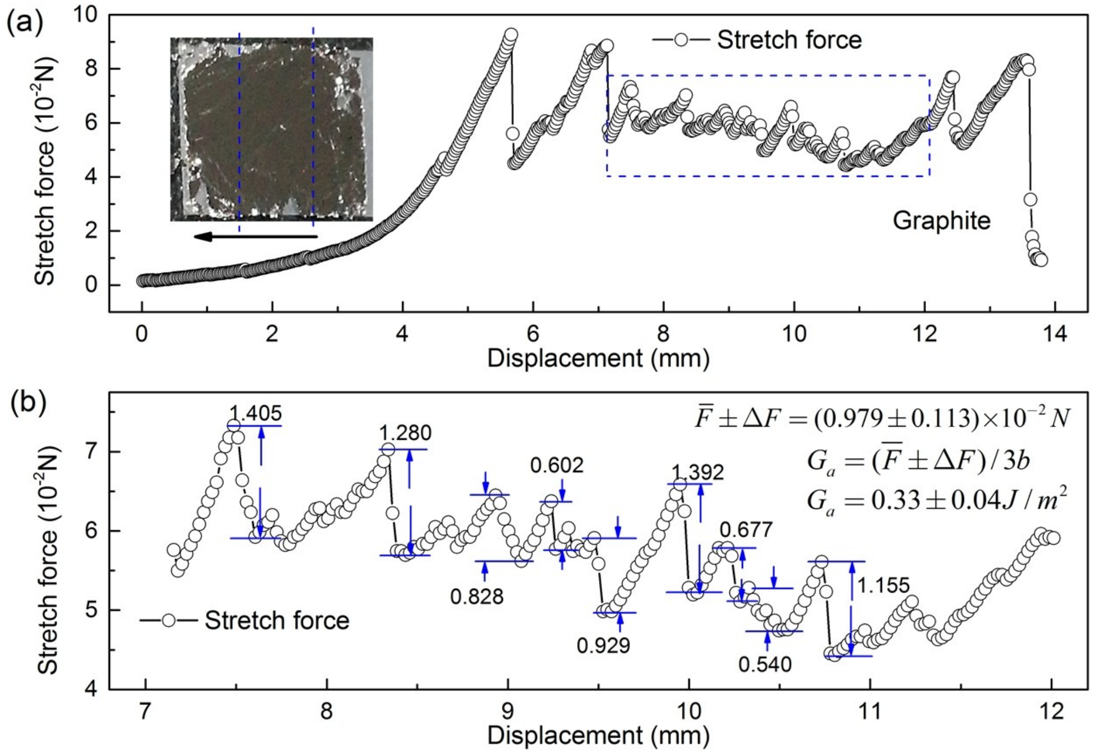

Fig. S7. The stretch force vs. the stretch displacement for a piece of graphite slice peeling from bulk graphite. The captions are the same as Fig. S4. The stretch average velocity is about 0.084 mm/s.

5. The Higher Velocity to Peel the Graphite Slice.

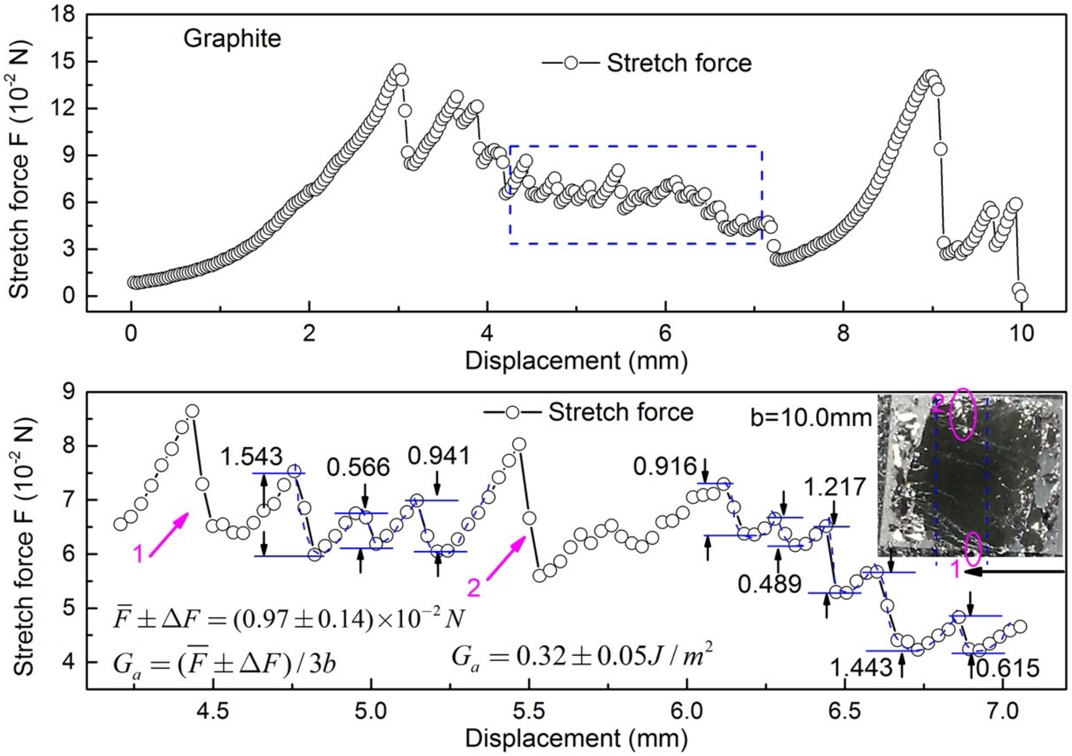

Fig. S8. The stretch force vs. the stretch displacement for a piece of graphite slice peeling from bulk graphite. (a) is plotted for showing all of data. (b) is zoomed in the blue dashed square in (a) for figuring out the adhesion energy by the “stress accumulation-peeling” mechanism. The inset in (b) is the graphite slice on tape after peeling from the bulk graphite. The arrows in (b) is for indicating stretch direction. There are some broken holes circled by two pink ellipses, indicated as number 1 and 2. The stretch force corresponding to the location 1 and 2 are indicated as two pink arrows along with number 1 and 2. The stretch average velocity is about 0.15 mm/s.

6. The evaluation of the Work for Tearing Graphite.

The graphite is usually teared at the peeling process. Maybe, we can estimate the tear energy of graphite slice. There is about twenty tear lines per millimeter, and 100 layers (~30 nm) are included in per tear line, seen in Figs. S9 and S10. The C-C bond energy is about 618 KJ/mol [16], then we can calculated the tear energy about 0.023 J/m^2, which is less than the adhesion energies measured by us 0.33±0.04, 0.32±0.05, 0.21±0.04, 0.31±0.03, 0.32±0.06 J/m^2. The cracks here are different from kink structures observed before [17], which can be verified by the different focal length of an optical microscope at the two sides of crack. The tear energy will be less than 0.023 J/m^2 if they are kink structures. If a piece of graphite with 300 layers (~100nm thick, seen in Figs. S11) is torn, then the tearing energy is about 0.07 J/m^2. The energy is a considerable large comparing with the adhesion energy of graphite slice. Furthermore, if the tearing direction is perpendicular to the peeling direction or a big tear (Figs. S11), the tearing energy is to be very large, even larger two orders (~2.3 J/m^2) than that of the first case, which just corresponds to the positions indicated by the arrow in Figs. S6 and S8. Therefore, we can make sure that the measured adhesion energy of graphite slice almost don't affected by a bit tear of the graphite slice if there is not obvious tear.

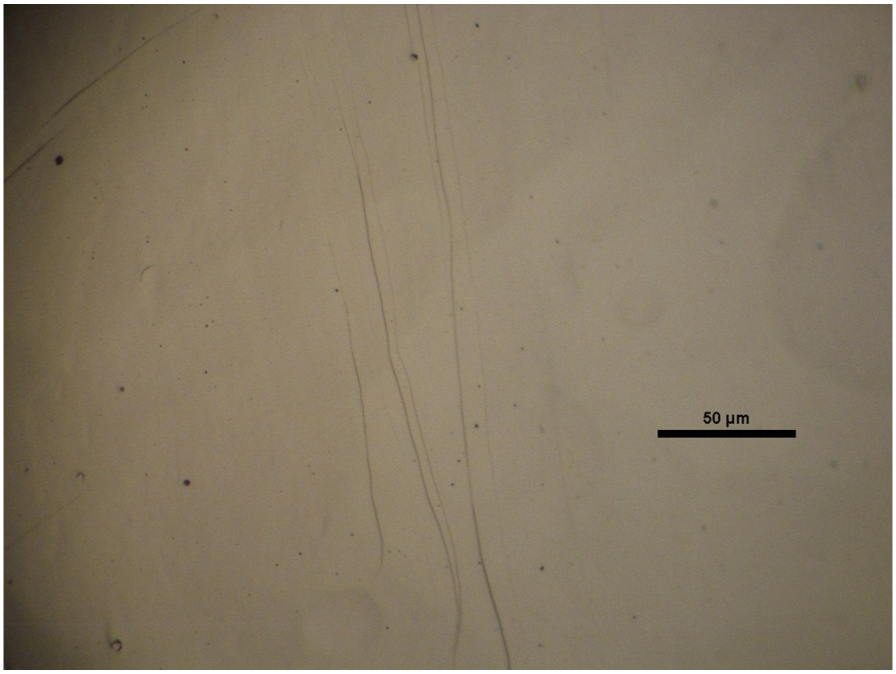

Fig. S9. A photograph for showing a bit cracks on a piece of graphite slice peeled from bulk graphite.

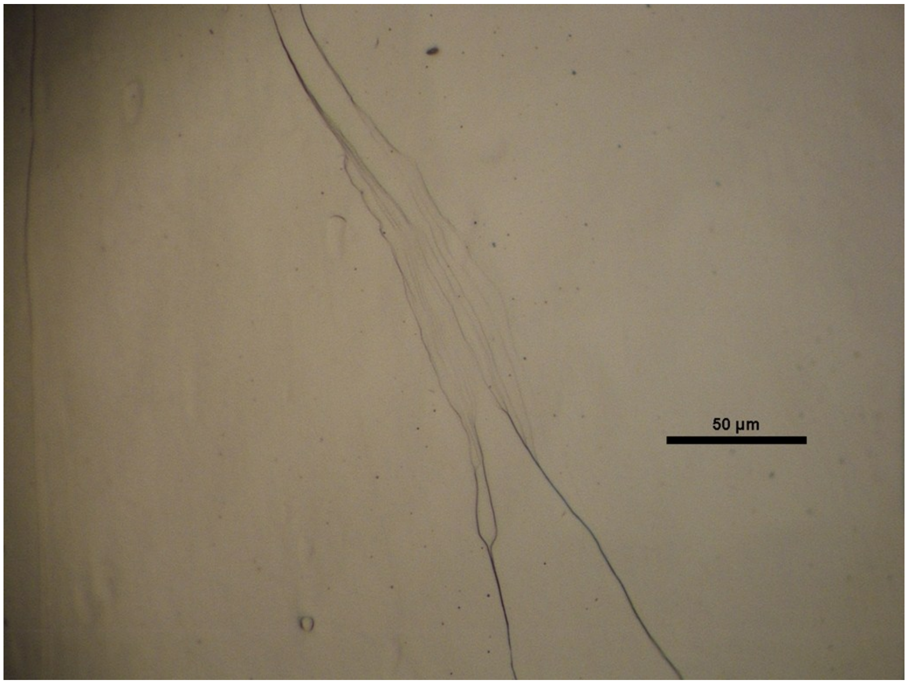

Fig. S10. A photograph for showing a bit cracks on another piece of graphite slice peeled from bulk graphite. The number of cracks is more than the previous sample.

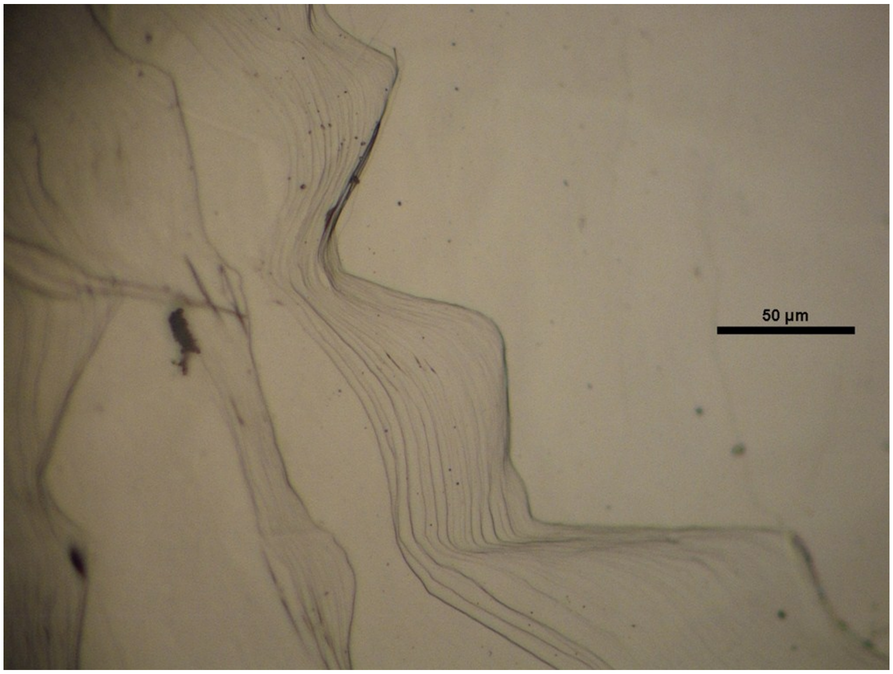

Fig. S11. A photograph for showing a big tear. Many layers are torn on another piece of graphite slice peeled from bulk graphite.

It should be noted that we don’t present any atomic force micrograph (AFM) image because the graphite slice peeled bulk graphite is stuck on a tape. The tape is too flexible to implement AFM scanning. Likewise, the bulk graphite is also too flexible. Therefore, we give the thickness of the crack graphite according to the experience from lots of the exfoliation experiment conducted before.

7. Comparison for Adhesion Energy of Graphite in Experiment

Table 1S. Reported experiment measurements of the graphite adhesion energy (in J/m^2).

Reference number	Research method	Size and type of Graphene	Adhesion energy (J/m^2)
[3]	TEM measurement	Graphite	0.21 (+0.09, − 0.06)

[4]	Thermal desorption	Aromatic molecules	0.32 ± 0.03
[5]	Friction by AFM	Graphite	0.19±0.01
[6]	SEM experiment	WMCN[a]-Graphene	0.200-0.360
[7]	Retraction by AFM	Graphene	0.307
[8]	Self-retraction	Graphite	**0.33**
[9]	Friction by AFM	Graphite	0.227±0.005
[10]	Blisters	Graphite	0.221±0.011
[11]	Contact-separation	Graphene	0.119 ± 0.003
In this work	stress accumulation-peeling measurement	Graphite	**0.31-0.34** (or 0.21)
In this work	Calculation	Graphite	**0.29**

[a] WMCN:Mutil-layer carbon nantubes

8. The Error Evaluation.

The error in the measurement includes in three terms, apparatus error E_{app}, measurement error E_{meas} and accidental error. The accidental error only can be excluded by selection of data in carefully and cannot be calculated. The apparatus error is about ±0.2 mV, which is about E_{app} =0.5%. The measurement error is about 0.4mV, which is about E_{meas} =1.0%. Such that, the total error is about E_t=1.2% according to the error synthesis formula:

$$E_t = \sqrt{E_{meas}^2 + E_{app}^2} \tag{26}$$

The error in theoretically 1.2% is less than the mean deviation 5%-20% in measurement. Therefore, the error mainly comes from the measurement process. The mean deviation of adhesion energy has been shown in all figures.

9. The Movie to Peel PDMS from Glass.

The movie to monitor the exfoliation process can be found in the *Movie 1*. From the movie, we can find that there is a peeling front, and its moving velocity is related to the stretch force. The stretch force increases when the moving velocity of peeling front is very slow, however the stretch force decreases quickly when the moving velocity becomes quicker. The stress accumulation and peeling processes are shown clearly in *Movie 1*. The movie is selected from the segment between 8.0 and 12.0 mm in Fig. 3(d). Actually, we have more than 8 movies for PDMS peeled from glass. Here, we only present one representative movie for convenience.

10. The Movie to Peel Graphite Slice from Bulk Graphite.

The real time movie to monitor the exfoliation process can be found in the *Movie 2*. The stress accumulation and peeling processes are shown clearly in *Movie 2*. The movie is selected from the segment between 8.5 and 9.1 mm in Fig. S6(b). Actually, we have more than 20 movies for graphite slices. Here, we only present one representative movie for convenience.

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