Crack propagation in graphene monolayer under tear loading

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Crack propagation in graphene monolayer under tear loading is investigated via an energy-based analytical model and molecular dynamics (MD) simulations. The classical mechanics-based model describes steady-state crack propagation velocity as a function of applied stress, lateral dimension and loading geometry, as well as the critical stress and critical size for initiating steady crack propagation. MD simulations reveal that cracks propagate along the zigzag direction but yield different “fracture surface” roughnesses for different loading geometries. MD simulations and the predictions of the analytical model are in excellent agreement. Our findings lead to an improved fundamental understanding of the mode-III crack of monolayer graphene necessary for the design and fabrication of graphene-based devices.

1 Introduction

As the strongest material ever known, one of the biggest concerns for graphene is fracture. Understanding graphene fracture is key to designing nanodevices and composites. Furthermore, understanding graphene in particular as a thin sheet or a two-dimensional material bears important implications for engineering applications of membrane structures in general. Recent studies have shown that the fracture of graphene cannot be simply described by conventional theories, and can be affected by non-continuum effects, multi-scale effects, and nonlinear mechanical properties. For instance, Yin et al. claimed that the Griffith criterion remains valid for cracks down to 10 nm but overestimates the strength of shorter cracks in tensile fracture of graphene. Cao et al. found that the fracture toughness of graphene can be improved by functional groups. Understanding and tailoring fracture are important purposes in engineering applications of membrane structures including graphene. In particular, crack evolution is a key step to reveal fracture mechanics of graphene. Moreover, graphene is an ideal model material to investigate fracture mechanics with atomic resolution. For atomistic simulations, the in-plane dimensions can be adequately large (e.g., up to micrometers) in order to minimize the system size effects on both the fracture modes and loading without significantly increasing the computational cost.

Thus, revealing fundamental mechanisms of graphene fracture can also provide a valuable insight into the dynamic fracture of other 2D and quasi-2D materials. In particular, there are serious concerns about the critical dimensions below which conventional continuum theories break down.

Understanding how cracks propagate (e.g., velocity and trajectory) in materials has been a central question in dynamic fracture. There are three basic ways of applying a force to enable crack propagation: opening (mode I), sliding (mode II), and tearing (mode III). Most studies have focused on mode-I fracture (tensile fracture). Besides tensile fracture, other fracture modes in a graphene monolayer are also of interest. Fracture dynamics of a graphene monolayer is still under-explored, especially for crack propagation. In general, cracks propagate along the zigzag direction in pristine graphene following the path of the minimal fracture surface energy. The crack propagation velocity generally cannot exceed the Rayleigh-wave speed, while the Rayleigh-wave speed is not well defined for free-standing graphene due to its out-of-plane displacement. For the Griffith cracks, Le et al. found that the stable crack propagation velocity increases with the applied pre-strain. Although considerable efforts have already been dedicated to crack evolution and self-folding mechanics in graphene to investigate the fundamental mechanisms of dynamic fracture, an analytical model to describe the crack propagation velocity is still lacking, despite its importance in engineering applications. For example, dynamic fracture of a graphene monolayer under tear loading has rarely been investigated, despite its importance in applications such as nanotripping and nanosensors. As we are aware, the relationship between energy and crack propagation of the graphene monolayer under tear loading has been rarely reported.
In this work, we develop an energy-based model to describe the steady-state propagation velocity of mode-III cracks in graphene, as a function of applied stress, lateral dimension, and loading geometry. Molecular dynamics (MD) simulations are then utilized to characterize fracture dynamics under tear loading and to examine the validity of our model. Two chiralities (zigzag and armchair) are explored and their similarities and differences are analyzed. The effects of applied stress and lateral dimension are examined with MD simulations which show excellent agreement with the analytical model. Furthermore, our energy-based analysis indicates that the propagation velocity of a crack tip is affected both by local debonding power and the far-field inertial effect.

2 Analytical models and simulation details

A mode-III crack in 2D is a common tearing mode which occurs when shear stress is out of the sample plane. Fig. 1 shows the loading geometry for the classical mechanics-based model analysis. An edge notch (EN) is placed along the x-axis on the left edge of graphene. Here, we define the armchair/zigzag loading when the EN is parallel to the armchair/zigzag direction. A pair of constant stresses are applied to two loading segments (upper loading and lower loading) near the left edge. The two opposite stresses create shear near the EN and initiate crack propagation towards graphene interior. To simplify the following discussion of the mechanical model, only one crack tip and half of the sample along the y-axis are considered.

At a steady state, a crack tip propagates at a constant velocity, \( v_f \). The applied stresses are relatively small so that \( v_f \) is sufficiently low to avoid dynamic fracture instabilities. Within a time increment \( \Delta t \), the moving crack tip leaves behind two newly separated graphene segments moving upward or downward, at a velocity of \( \pm v_f \). Here we consider only one of the two segments with \( v_f \). For this segment, the work due to the applied stress (\( dW \)) transforms into kinetic energy (\( dE_k \)) and potential energy (\( dE_p \))

\[
\begin{align*}
\text{d}W &= \text{d}E_k + \text{d}E_p, \\
\text{d}W &= \sigma L h |v_f| \text{d}t,
\end{align*}
\]

with

\[
\begin{align*}
\text{d}W &= \sigma L h |v_f| \text{d}t,
\end{align*}
\]

where \( \sigma \) is the stress applied on the unit area in the yz-plane, \( L \) is the half length of the y-dimension, \( h = 3.35 \) Å is the thickness of the graphene monolayer, and \( v_f \) is the z-component of the velocity of the fractured segment under consideration (\( v_f \)).

The corresponding kinetic energy increment \( dE_k \) is

\[
dE_k = \frac{1}{2} \rho L h |v_f| |v_f|^2 \text{d}t,
\]

where \( \rho = 2.33 \) g cm\(^{-3} \) is the density of graphene and \( v_f = |v_f| \). The potential energy increment \( dE_p \) due to C–C bond breaking as the crack tip advances is

\[
dE_p = \hbar v_f \delta \text{d}t,
\]

where \( v_f = |v_f| \) and \( \delta \) denotes the fracture surface energy (per unit area). As the bond density along the zigzag direction (0.413 Å\(^{-1} \)) is smaller than that along the armchair direction (0.48 Å\(^{-1} \)), the crack tip propagates in the former direction due to the lower energy cost in breaking the C–C bond. \( \delta \) can be calculated with bond density and the cleavage energy for one single C–C bond. It follows that \( \delta \approx 10 \) J m\(^{-2} \) in our simulations, close to previous results.\(^{13,32} \)

At the steady stage, \( |v_f| = v_f / \sqrt{2} \) can be obtained from geometry relations. \( v_f = v_f^z \) is assumed for small tension in graphene. Thus, \( v_f \) and \( v_f \) are related via

\[
v_f = 2 v_f^z.
\]

Here \( z \) is a geometric factor, and \( z = \sqrt{1/2} \) and \( \sqrt{2/3} \) for the zigzag and armchair directions, respectively.

Combining the above equations, we obtain \( v_f \) as a function of loading stress (\( \sigma \)) and lateral graphene dimension (\( L \)):

\[
v_f = z \left[ \frac{2(\sigma L - \sqrt{2} \sigma \delta)}{L \rho} \right]^{1/2},
\]

The above equation states that steady-state crack propagation velocity in graphene under tear loading is anisotropic, and can be quantitatively determined from the applied stress, lateral graphene dimension, and loading geometry. The critical stress to initiate a crack is obtained with \( v_f = 0 \), as

\[
\sigma_c = \sqrt{2} \sigma / L.
\]

In the limiting case of \( L \to \infty \), \( \sigma_c \to 0 \), and we have

\[
v_f(lim) = z \sqrt{2} \sigma / \rho.
\]

In this situation, \( dE_p \) becomes a small quantity compared to \( dW \) and \( dE_k \) and \( \delta \) does not impact \( v_f \) any longer. At the same time, a small \( \sigma \) value is enough to generate steady propagation of the tip. Accordingly, the critical size to initiate a crack under a certain \( \sigma \) can also be obtained by setting \( v_f = 0 \) (eqn (6))

\[
L_c = \sqrt{2} \sigma / \sigma_c.
\]

Thus, based on the energy analysis with considering the non-continuum effects of C–C bond breaking, the analytical model to describe the relationship between loading stress and the crack propagation velocity is obtained. The propagation velocity
of the crack tip $v_t$ is simultaneously influenced by the loading stress $\sigma$ and the lateral graphene dimension $L$, while the critical stress $\sigma_c$ is determined by $L$.

To validate the model for steady-state crack propagation velocity ([eqn (6)]) and reveal underlying mechanisms, we perform MD simulations with a large-scale atomic/molecular massively parallel simulator.\textsuperscript{33} An adaptive intermolecular reactive empirical bond order (AIREBO) potential with a cutoff of 2.0 Å for the reactive empirical bond order (REBO) part is used to describe the interaction between C atoms.\textsuperscript{34,35}

Fig. 1 shows the loading geometry for tearing graphene along the armchair or zigzag direction. Periodic boundary conditions are applied along the $y$-axis only. Given the periodic boundary conditions, two equivalent edge notches (ENs) along the $x$-axis are placed on the left edge of the graphene sample: one in the middle and the other at periodic boundaries along the $y$-axis. Two representative orientations, armchair and zigzag, are explored in our simulations. Prior to tear loading, the system is relaxed at 300 K and zero pressure with a constant pressure–temperature ensemble, and both ends along the $x$-axis are fixed during relaxation. For tear loading, a microcanonical ensemble is used; a pair of constant stresses are applied in opposite directions to the upper and lower portions of the sample left edge, corresponding downward loading (DL) and upward loading (UL), respectively. Only the right end of the graphene sample is fixed during tear simulations. The sample dimensions are $120 \times 126$ nm$^2$ and $120 \times 34$ nm$^2$ for the zigzag loading and $110 \times 70$ nm$^2$ for the armchair loading. The time step for integrating the equation of motion is 1 fs in all simulations.

### 3 Results and discussion

We carry out a series of MD simulations of tearing along both zigzag and armchair directions, and the results are presented in Fig. 2–6. Fig. 2(a) shows a typical snapshot during the zigzag loading with $\sigma = 1.91$ GPa and $L = 168$ Å viewed from different perspectives. After a short relaxation and incubation, a crack is initiated from the edge notch and propagates towards the positive $x$-axis at a steady velocity $v_t$; the crack forms along the zigzag direction, yielding “fracture surfaces” as smooth as those obtained before loading. This phenomenon is consistent with previous studies suggesting that the cleavage of armchair bonds is much easier in graphene.\textsuperscript{36} The divided graphene retains approximately its initial shape, indicating little energy dissipation in the form of potential energy except for bond breaking.

For the armchair loading with $\sigma = 0.79$ GPa (Fig. 2(b)), the crack is initiated at the edge notch and still propagates along the zigzag direction, but deviates from the edge notch orientation, since the notch is not oriented along the zigzag direction. The crack tip may take a random zigzag direction at a random location during its propagation, so the fracture surface can be irregular and thus much rougher than the initial notch.

In order to reveal the characteristics of crack propagation and examine the validity of our model, we perform quantitative analyses of both loading cases. Fig. 3 shows the $x$-coordinates of the crack tip and the $z$-coordinates of the upward loading segment (UL) at different times. The $y$- and $z$-coordinates of the crack tip essentially remain unchanged, and thus are not discussed. The crack tip is recognized via calculating the coordination numbers (CN) of C atoms, and is represented by the atom with $\text{CN} = 2$ and with the largest $x$-coordinate. After a short period of equilibration, both of the coordinates increase linearly, indicating that both of them move at a steady speed. The slope of the crack tip equals to $v_t = v_x$, and that of the UL curve is $v_{UL}$. These two curves become parallel at a later stage, demonstrating that they reach the same steady-state velocity, i.e., $v_t = v_{UL}$, which is consistent with our model ([eqn (5)]).

Considering the lateral size effect as indicated by eqn (6) and (7), we perform simulations at different $L$-values. For illustrative purpose, the $v_t(\sigma)$ curves for two different lateral sizes are shown and compared against the predictions by our model (Fig. 4(a)). The critical stress for initiating crack propagation, $\sigma_c$, is 0.60 GPa for $L = 168$ Å and 0.16 GPa for $L = 629$ Å as obtained from eqn (7); no steady crack propagation occurs for $\sigma < \sigma_c$. Our model

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**Fig. 2** Atomic configurations for (a) the zigzag loading with $\sigma = 1.91$ GPa and $L = 168$ Å at $t = 45$ ps, and (b) the armchair loading with $\sigma = 0.79$ GPa and $L = 350$ Å at $t = 140$ ps.
predictions for \( v_t(s) \) and \( s_c \) agree well with direct simulations for all the cases. As expected, \( v_t \) increases with increasing \( s \) at a given \( L \), since the work done by the applied stress increases. In addition, \( v_t \) for \( L = 629 \, \text{Å} \) is larger than that for \( L = 168 \, \text{Å} \) at the same \( s \), and this difference is more pronounced at small \( s \). This can be explained by the energy distribution. As indicated by eqn (2) and (4), the energy spent on breaking the C–C bonds remains unchanged, while the energy input is proportional to \( L \). In the case of small \( L \), a larger applied stress is needed to overcome the energy barrier to break C–C bonds, resulting in a larger \( s_c \) value (Fig. 4(a)). At the same \( s \) above \( s_c \), a larger portion of the input energy is spent on breaking C–C bonds, so the kinetic energy increase is smaller in small \( L \) cases. This effect is much more marked near \( s_c \).

As in the zigzag loading, we obtain the coordinates of the crack tip and the UL segment in the armchair case and the \( v_t(s) \) curve along with our model predictions (Fig. 4(b)). The former is consistent with that for zigzag loading and thus is not shown here. The phenomena are similar in both zigzag and armchair cases (except that the crack trajectory is more complicated in the armchair case), and the model predictions agree with direct simulations of \( v_t \) and \( s_c \).

Since the models for the zigzag and armchair loadings are similar and both of them agree well with direct simulations (Fig. 4(a) and (b)), we choose the zigzag loading as a representative...
case for its simplicity in the following discussions. As indicated by eqn (6), increasing $L$ or $\sigma$ leads to a faster $v_t$. However, their exact trends seem to be different. In order to reveal the effect of $L$ on $v_t$, we perform a series of simulations at different $L$ at the same $\sigma$ along the zigzag direction, and the results are shown in Fig. 5 together with the predictions by our model. The intersection of the predictions with the horizontal axis is $L_c$ for each loading stress. Our model predictions for $v_t(L)$ agree well with direct simulations which also show the absence of steady propagation for $L < L_c$. As expected, $v_t(L)$ increases with increasing $L$ for $L > L_c$, but the trend is quite different from $v_t(\sigma)$ (Fig. 4(a)). $v_t(L)$ increases sharply when $L$ just exceeds $L_c$ in all the cases explored. Then it increases slowly, approaching $v_t(\text{lim})$ when $L \gg L_c$. This observation indicates that $v_t$ is no longer sensitive to the system size but only determined by the applied stress at this stage. As inferred from eqn (8) and (9), $v_t(\text{lim})$ increases with increasing $\sigma$ at fixed $L$, and $L_c$ decreases with increasing $L$ at fixed $\sigma$.

Given this analytical model, we derive powers ($dW/dt$, $dE_p/dt$ and $dE_p/dt$) as a function of $\sigma$ for the total work, kinetic energy and potential energy for loading along the zigzag direction (Fig. 6). MD simulations (dots in Fig. 6) show excellent agreement with the analytical model (lines). At the initialization stage of fracture, the debonding power $dE_p/dt$ is higher than the kinetic power $dE_k/dt$. However, with increasing loading $\sigma$, the kinetic power increases rapidly at a higher rate than the debonding power, indicating a rapid increase in the portion of the inertial energy during dynamic fracture. According to eqn (4), the crack propagation velocity $v_t$ can also be given as:

$$v_t = \frac{1}{\rho c} \frac{dE_p}{dt}$$  \hspace{1cm} (10)

Thus, $v_t$ is proportional to $dE_p/dt$ (the local energy release rate) at the crack tip. Ideally, $v_t$ is only affected by the local debonding power, but not by far-field effects. However, in an actual process, the far-field inertial effect gradually constitutes the majority of the work done by the applied stress, and the growth rate of $v_t$ is limited. This is highly consistent with previous studies related to dynamic fracture.37–39

The far-field effects may be different for different $L$. Here, we define $\sigma_c$ as the stress when $dE_p/dt = dE_k/dt$ (the inset in Fig. 6), and plot $\sigma_t$ and $\sigma_c$ as a function of $L$ in Fig. 7. The whole $L-\sigma$ plane is divided into three regimes by these two curves: regime N with no steady crack propagation, regime P where $E_p$ is dominant (the majority of the input work is consumed by debonding the C–C bond), and regime K where $E_k$ is dominant (the majority of the input work is consumed due to the far-field inertial effect). Both $\sigma_t$ and $\sigma_c$ decrease with increasing $L$, as indicated by eqn (3), (4) and (7); the gap between $\sigma_t$ and $\sigma_c$ becomes smaller at large $L$ as a result of the energy concentration. Since the input work is uniformly distributed along the y-direction, it is easier for a small system to concentrate the input energy on the crack tip. In this regard, the far-field effect is much weaker for small $L$, resulting in a larger gap of regime P (Fig. 7).

Although this model is deduced mainly for mode-III cracks in graphene, it bears certain generality. With suitable modifications, our model could also be applied to mode-III cracks in other materials, and mode-I and II cracks, or even fracture processes.

4 Conclusions

We develop an energy-based model from classical mechanics to describe the propagation of model-III cracks in graphene as a function of applied stress, lateral dimension and loading geometry. We also perform MD simulations to investigate fracture dynamics. For different geometries (along the zigzag and armchair directions), cracks propagate in the same (zigzag) direction but yield different fracture surface roughnesses. The propagation velocity of the tip increases with increasing applied stress and lateral dimension, and the portion of debonding power decreases. Direct MD simulations and the analytical model show excellent agreement in crack propagation velocities, critical stresses and critical sizes for initiating steady crack propagation, which provides a fundamental understanding of crack propagation in a graphene monolayer.

Conflicts of interest

There are no conflicts to declare.

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