Supplementary Material

Unveiling the mechanism of structure-dependent thermal transport behavior in self-folded graphene film: A multiscale study

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Here we provide more details about the simulation model for the calculation of k_g and the values of input parameters for the analytical model as well as discussions on structural stability analysis.

1. Simulation model for the calculation of k_g

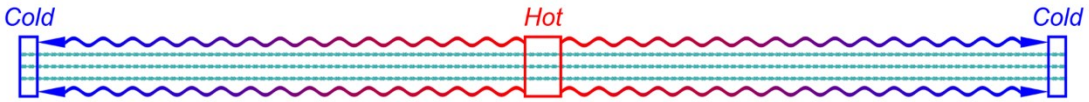

Fig. S1 Schematics of MD simulation models for the calculations of in-plane thermal conductivity k_g in the analytical model.

2. Values of input parameters for the analytical model

l_{f} (nm)	k_{g} ($\mathrm{Wm^{-1}K^{-1}}$)	λl_{f}
14.7	82	0.86
24.9	93	1.37
48.8	107	2.51
98.6	169	4.03

Table S1 MD simulation results of k_g and corresponding values of λl_f that are adopted in the analytical model for predictions of temperature profiles and thermal conductivities of SF-GF.

3. Structural stability analysis of SF-GF

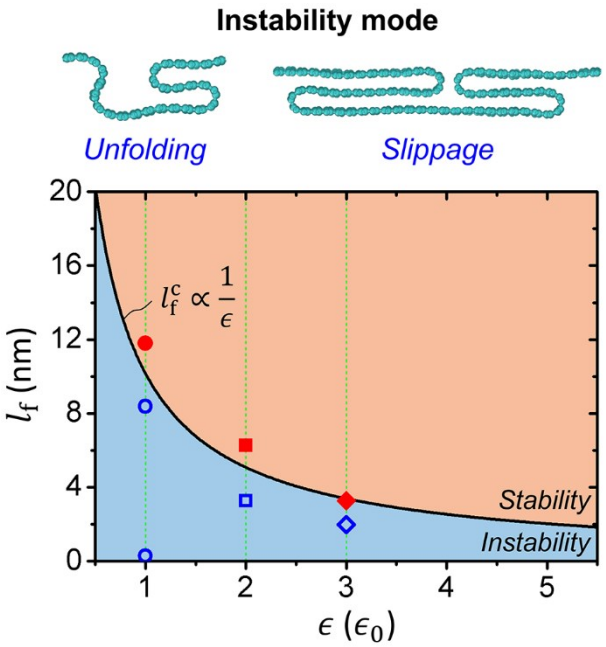

Fig. S2 Structural stabilities of SF-GF models under 300 K with different fold lengths l_f and interlayer interaction parameters ϵ. The solid and hollow data points denote stable and instable structures of SF-GF, respectively. The critical fold length for stability l_f^c is represented by the black boundary between orange and blue domains. Two instability modes, unfolding and slippage, are observed in MD simulations of SF-GF.

The stability of such folded graphene structure is investigated by MD simulation, which is a major concern in folded nano-materials and nano-origamis[1,2]. Fig. S2 shows the stabilities of different SF-GF models with varying fold lengths l_f and potential parameters ϵ that reflect the strength of interlayer interaction in MD simulations based on the 12-6 Lennard-Jones potential. Here, ϵ_0 stands for the parameter setting for pristine interface between graphene layers, which is adopted by MD simulations for the calculation of thermal conductivity in this paper. A reciprocity law is observed between the critical fold length for stability l_f^c and ϵ, namely, $l_f^c \propto 1/\epsilon$. For $\epsilon = \epsilon_0$, the folded configuration of SF-GF fails when its fold length is less than l_f^c around 10 nm.

Substituting $l_f^c = 10$ nm and other known parameters into the expression of λl_f, we can obtain the lower bound of λl_f at around 0.2 that is used in Section 4.2 of this paper. Two instability modes are observed in MD simulations, one is unfolding mode, and the other is slippage mode, as shown in Fig. S2. For the first mode, the grafold is unfolded and released. For the second mode, the folded structure is maintained, but the graphene layers slip like a wiggling snake, which breaks the symmetry of simulation model about the z-aix. In addition, the unfolding mode only occurs for l_f less than 1 nm in MD simulations.

It is known that a stable folded structure of graphene results from the balance between the interlayer adhesion over the flat folded domains and the out-of-plane bending over the curved joints between folded layers. The adhesion caused by van der Waals interaction tends to fold the graphene while the bending resists this folding tendency and makes the fold length smaller. Defining the flat graphene as the ground state, the total energy for the grafold in the RVE model is expressed as

$$U_{\text{tot}} = U_{\text{adhesion}} + U_{\text{bending}} \tag{S1}$$

with adhesion energy $U_{adhesion}$ (negative) and bending energy $U_{bending}$ (positive). If $U_{tot} < 0$, the grafold is energetically favorable and it can be kept in a stable configuration. On the contrary, the grafold is instable if $U_{tot} > 0$ since the adhesion energy fails to compete against the bending energy.

The adhesion energy $U_{adhesion}$ between multilayer graphene shows a relationship as[2]

$$U_{\text{adhesion}} = -\gamma l_{\text{f}} \tag{S2}$$

where γ is the binding energy per unit length (treated as a 2D model). For MD simulations, previous theoretical study[3] proposed that γ is proportional to the interlayer interaction parameter ϵ. It indicates the scaling law between $U_{adhesion}$ and ϵ as $U_{adhesion} \propto -\epsilon l_f$. From the equilibrium atomic structure by MD simulations, the shape of bending joints between layers can be approximated as a semicircle with fixed length $l_{bending} = \pi h/2$ and the curvature $\kappa = 2/h$. With the bending stiffness D, the bending energy can be approximately calculated as[4]

$$U_{\text{bending}} = \frac{D\kappa^2 l_{\text{bending}}}{2} = \frac{\pi D}{h} \tag{S3}$$

which can be regarded as a constant here in our model. Substituting the descriptions of $U_{adhesion}$ and $U_{bending}$ into Eq. (S1), there must exist a critical fold length l_f^c, above which U_{tot} is negative and the stable configuration of grafold is achieved. Obviously, the scaling law between l_f^c and ϵ should follow the form as $l_f^c \propto 1/\epsilon$, which is consistent with the result from foregoing MD simulations.

It should be mentioned that above theoretical stability analysis is more like a qualitative discussion due to the simplification of the shape of the bending joints. It has been reported that the bending shape of this curved domain varies under different fold lengths[1]. Although the difference of bending shape is relatively small between models with different l_f in MD simulations, it is necessary to take into account this issue for the more precise analytical models. Moreover, the effect of thermal fluctuation on the stability is ignored here, which causes significant influence on the interlayer adhesion energy[5]. The in-depth understanding on the stability of SF-GF or the similar graphene-origami will be explored in our future work.

References

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