Bending admissible continuum approximation for mono-layered 2D materials

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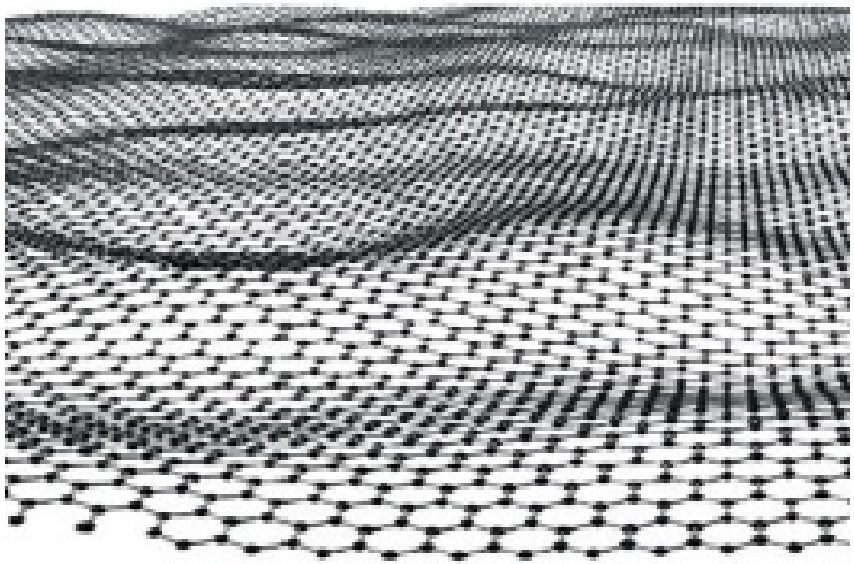
Outline

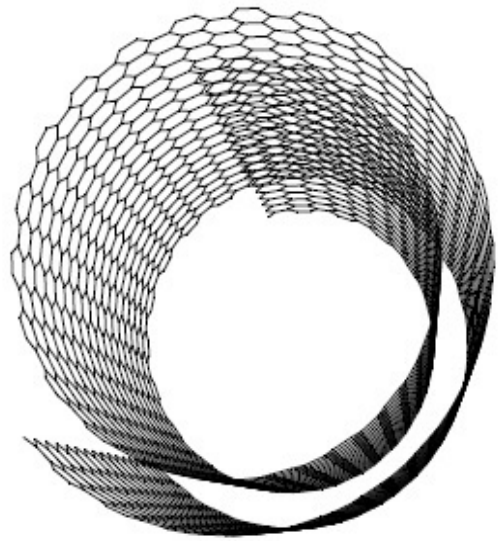
- Motivation: non-flat configuration
- Continuum model: von-Kármán for mono-layered system
  - assumptions on the empirical atomistic energy
  - scaling rules of the displacement fields
  - appropriate norm that reflects the observed stability
- Stability and error analysis
  - linear stability analysis
  - force consistency analysis
  - displacement fields error estimate
- Ongoing work
- Summary

Experimental observations

Graphene is locally two-dimensional but not flat. Nanoscale **ripples** appear in suspended samples and **rolling up** often occurs when boundaries are not fixed. [1] [2]



(a) Ripple[3]



(b) Rolling up [4]

Need an appropriate continuum model for multiscale modelings.

[1] Novoselov et. al. Nature, 446, 60-63, (2007)
[2] U. Stefanelli, et. al. U. Z. Angew. Math. Phys. 69:70, (2018)
[3] Deng and Berry, Materials Today, Volume 19 (4), (2016)
[4] P. Lambin et. al. Applied Sciences, 4, 282-304, (2014)

A few existing continuum models

- **Based on Cauchy–Born rule**
  - Exponential Cauchy–Born rule; [5]
  - High order Cauchy–Born rule. [6] [7]

[5] Arroyo and Belytschko, 40, 455-469, Meccanica, (2005)
[6] Yang and E, PRB 74, 184110, (2006)
[7] Cazeaux and Luskin, M2AN, 52, 729-749, (2018)
[8] Ariza and Ortiz, J. Mech. Phys. Solids 58, (2010)
[9] Yang and Tewary, PRB 77, 245442, (2008)
[10] Xiaojie Wu and Xiantao Li, manuscript
[11] Nelson et al. Int J Numer Meth Eng 30, 517-539, (1990)
[12] Zhang et. al. J. Mech. Phys. Solids, 67, 2-13, (2014)

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  - Iterative method by using force corrections;[8]
  - Discrete lattice Green function and obtain the linear Kirchhoff plate theory for the far-field considering a point defect/ point force loading. [9] [10]

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- **Based on nonlinear plate model**
  For example, using the von-Kármán plate theory. [11] [12]

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Some of them **do not target for modeling ripples**.

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Literatures related to the von-Kármán theory

- **Applicational point of view:** The von-Kármán has been used for modeling and computational simulations of ripples and wrinkles, see for example, Kumar et. al. (2015) [13] and Gao et. al. (2019). [14]

[13] Kumar et. al. Scientific Reports, 5:10872, (2015)
[14] Gao et. al. Handbook of Graphene: 2, 1-44, (2019)
[15] Braun et. al. arXiv:1907.00197, (2019)
[16] Friesecke et. al. Arch. Ration. Mech. Anal., 180, 183-236, (2006)

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- **Multilayered graphene system:** Braun et. al. rigorously derived the von-Kármán model via analyzing scaling of energy in terms of the lattice constant ε and the layer distance h. [15]
- **Continuum plate theories**: Friesecke et. al. rigorously studied different plate models by scaling of the elastic energy per unit volume in terms of the thickness h of plates [16]
  - f^{ext} and total energy $\sim h^2$: Kirchhoff plate
  - f^{ext} and total energy $\sim h^4$: von-Kármán plate
  - general case with external force $\sim h^\alpha$ with $\alpha > 0$, summarized in Table 1 of Friesecke et. al. (2006)

[13] Kumar et. al. Scientific Reports, 5:10872, (2015)
[14] Gao et. al. Handbook of Graphene: 2, 1-44, (2019)
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Goal

1. Rigorously derive the continuum von-Kármán model which captures the governing out-of-plane rippling type deformations in a mono-layered system.

2. Prove rigorous stability and error analysis.

Problem setup and notations

- Consider a simple lattice with reference configuration

$$\mathcal{L} := \varepsilon \mathsf{F}\mathbb{Z}^d, \quad \text{for some } \mathsf{F} \in \mathbb{R}^{d\times d}, \quad \det(\mathsf{F}) = 1, \quad d \in \{1, 2\},$$

  where ε is the lattice constant. It is deformed into $\mathbf{y} : \mathcal{L} \to \mathbb{R}^{d+1}$.

- For simplicity, consider a 1D periodic chain of $N \in \mathbb{N}$ atoms. Assume $\varepsilon N = 1$, thus the reference atomic chain is $\mathcal{L} = (0,\ 1]$. Denote the physical displacement field by $\mathbf{u} : \mathcal{L} \to \mathbb{R}^2$ where

$$\mathbf{u}(\varepsilon i) := \mathbf{y}(\varepsilon i) - \begin{pmatrix} \varepsilon i \\ 0 \end{pmatrix} = \begin{pmatrix} u_1(\varepsilon i) \\ u_2(\varepsilon i) \end{pmatrix} = \begin{pmatrix} u_{i,1} \\ u_{i,2} \end{pmatrix}.$$

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  For bulk materials, u_1 and u_2 are scaled in the **same** way with respect to ε.

  Challenge: For 2D materials, need to identify **different** scaling rules of u_1 and u_2 wrt ε.

Atomistic energy

The total atomistic energy for 1D chain is

$$E^{\mathrm{a}}(\mathbf{y}) = E^{\mathrm{a}}_{pp}(\mathbf{y}) + E^{\mathrm{a}}_{ap}(\mathbf{y}) = \varepsilon \sum_{i=1}^{N} \left[V\left(\frac{|\mathbf{y}_{i+1} - \mathbf{y}_i|}{\varepsilon} \right) + f(\theta_i) \right].$$

E^{a}_{pp} denotes the pairwise potential and E^{a}_{ap} denotes the angular potential.

We assume the following properties:

- $V(s)$ behaves like a quadratic function, it reaches local minimum at $s = 1$ with $V(1) = V'(1) = 0$ and $V''(1) > 0$;
- $\theta_i := \theta(i)$ measures the angle between vector $\{\mathbf{y}_i - \mathbf{y}_{i-1}\}$ and vector $\{\mathbf{y}_{i+1} - \mathbf{y}_i\}$ and $0 < \theta_i \ll 1$;
- $f(\theta)$ denotes the angular potential that also behaves like a quadratic function with local minimum at $\theta = 0$, and $f(0) = f'(0) = 0$, $f''(0) > 0$, $f(\theta) \approx \theta^2$ when $0 < \theta \ll 1$.

Scaling of displacement field

- Total energy is scaled as $\sim \varepsilon^4$ for the von-Kármán plate:

$$E^{\mathrm{a}}(\mathbf{y}) \leq C\varepsilon^4.$$

We then proved that the deformed configuration $\mathbf{y}$ is close to a rigid motion: $\mathbf{x} \to \mathrm{R}(\mathbf{x} + \mathbf{c})$, where $\mathbf{c}$ denotes a translation and R denotes a rotation.

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- Hence, the displacement field $\mathbf{u}$ must be scaled as

$$\mathbf{u} = \begin{pmatrix} u_1 \\ u_2 \end{pmatrix} = \begin{pmatrix} \varepsilon^2 \tilde{u}_1 \\ \varepsilon \tilde{u}_2 \end{pmatrix},$$

  where $\tilde{u}_1$, $\tilde{u}_2$ are scalar functions and are independent of ε.

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- Hence $\frac{1}{\varepsilon^4} E^{\mathrm{a}}$ is $\Gamma-$convergent to the von-Kármán energy as $\varepsilon \to 0$

$$E^{\mathrm{c}}(\hat{\mathbf{u}}) = E^{\mathrm{c}}(\hat{u}_1(x), \hat{u}_2(x)) = \int_0^1 \Big[\frac{1}{2} V''(1) \left(\hat{u}_1'(x) + \frac{1}{2}(\hat{u}_2'(x))^2\right)^2 + \frac{1}{2} f''(0)(\hat{u}_2''(x))^2\Big] dx.$$

  $\hat{u}_1(x)$ and $\hat{u}_2(x)$ are smooth interpolation of the scaled displacements $\tilde{u}_1$ and $\tilde{u}_2$ from $\mathcal{L} \to \mathbb{R}$.

Appropriate norm for the observed stability

Define the first order differentiation operator

$$D_1^\epsilon v_\ell = \frac{v_{\ell+1} - v_\ell}{\varepsilon},$$

and the second order differentiation operator

$$D_1^{(2),\epsilon} v_\ell = \frac{D_1^\epsilon v_\ell - D_1^\epsilon v_{\ell-1}}{\varepsilon} = \frac{v_{\ell+1} - 2v_\ell + v_{\ell-1}}{\varepsilon^2}$$

For a physical displacement field $\mathbf{u}$, the semi-norm is defined as

$$\|\mathbf{u}\|_*^2 := \|D_1^\varepsilon u_1\|_{\ell_\varepsilon^2}^2 + \varepsilon^2 \|D_1^{(2),\epsilon} u_2\|_{\ell_\varepsilon^2}^2,$$

where for any scalar function v

$$\|v\|_{\ell_\varepsilon^2}^2 = \varepsilon \sum_{i=1}^{N} |v_i|^2.$$

For a continuum scaled displacement field $\hat{\mathbf{u}}$, the semi-norm is defined as

$$\|\hat{\mathbf{u}}\|_{*^{\mathrm{c}}}^2 := \int_0^1 \left[(\hat{u}_1')^2 + (\hat{u}_2'')^2\right] dx.$$

Linear stability

- The atomistic model is linear stable wrt to the $\|\cdot\|_*$:

$$\langle \delta^2 E^{\mathrm{a}}(\mathbf{0})\mathbf{v}, \mathbf{v}\rangle \geq \gamma^{\mathrm{a}} \|\mathbf{v}\|_*^2.$$

- The continuum model is linear stable wrt to the $\|\cdot\|_{*^{\mathrm{c}}}$:

$$\langle \delta^2 E^{\mathrm{c}}(\mathbf{0})\hat{\mathbf{v}}, \hat{\mathbf{v}}\rangle \geq (1+\varepsilon^2)\gamma^{\mathrm{a}} \|\hat{\mathbf{v}}\|_{*^{\mathrm{c}}}^2.$$

- Consequently, both atomistic and continuum models are linear stable and with similar stability constants.

Approximation error

Approximation error in terms of energy difference.

- Applying Taylor expansion to the atomic site energy of site i:
  Pair interaction:

$$V(\mathbf{u}_i) = V\left(\sqrt{(1+D_1^{\varepsilon}u_{i,1})^2+(D_1^{\varepsilon}u_{i,2})^2}\right)$$
$$\approx \frac{1}{2}V''(1)\left(D_1^{\varepsilon}u_{i,1}+\frac{1}{2}(D_1^{\varepsilon}u_{i,2})^2\right)^2+O(\varepsilon^6)$$
$$= \varepsilon^4\frac{1}{2}V''(1)\left(D_1^{\varepsilon}\tilde{u}_{i,1}+\frac{1}{2}(D_1^{\varepsilon}\tilde{u}_{i,2})^2\right)^2+O(\varepsilon^6).$$

  Angular interaction:

$$f(\theta_i) \approx f''(0)\theta_i^2 \approx \varepsilon^2 f''(0)\left(D_1^{(2),\epsilon}u_{i,2}\right)^2+O(\varepsilon^6)$$
$$= \varepsilon^4 f''(0)\left(D_1^{(2),\epsilon}\tilde{u}_{i,2}\right)^2+O(\varepsilon^6).$$

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$$= \varepsilon^4 f''(0)\left(D_1^{(2),\epsilon}\tilde{u}_{i,2}\right)^2+O(\varepsilon^6).$$

- Hence, **the continuum total energy is of second order consistent comparing to the scaled atomic total energy.**

Approximation error

Approximation error in terms of force difference.

- Compute $\mathbf{F}_i^{\mathrm{a}}(\mathbf{u})$ and get

$$
\begin{aligned}
\mathbf{F}_i^{\mathrm{a}}(\mathbf{u}) &= -\frac{1}{\varepsilon}\frac{\partial E^{\mathrm{a}}}{\partial \mathbf{u}_i} \\
&= V''(1)\begin{pmatrix} \varepsilon^2\left(\tilde{u}''_{i,1} + \tilde{u}'_{i,2}\tilde{u}''_{i,2} + O(\varepsilon)\right) \\ \varepsilon^3\left(\tilde{u}''_{i,2}\tilde{u}'_{i,1} + \tilde{u}'_{i,2}\tilde{u}''_{i,1} + \frac{3}{2}\tilde{u}''_{i,2}(\tilde{u}'_{i,2})^2 + O(\varepsilon)\right) \end{pmatrix} \\
&\quad + f''(0)\begin{pmatrix} 0 + O(\varepsilon^3) \\ \varepsilon^3\left(\tilde{u}^{(4)}_{i,2} + O(\varepsilon)\right) \end{pmatrix} \\
&= \begin{pmatrix} \varepsilon^2\left(\mathbf{F}^{\mathrm{c}}_{i,1}(\tilde{\mathbf{u}}) + O(\varepsilon)\right) \\ \varepsilon^3\left(\mathbf{F}^{\mathrm{c}}_{i,2}(\tilde{\mathbf{u}}) + O(\varepsilon)\right) \end{pmatrix}.
\end{aligned}
$$

- Hence, the pointwise error between **the scaled $\mathbf{F}_i^{\mathrm{a}}$** and the $\mathbf{F}_i^{\mathrm{c}}$ is of first order.
- As a result, **the scaled continuum displacement field has first order modeling error wrt atomistic $\|\cdot\|_*$.**

Extension to 2D

- Suppose $\mathbf{y} : \varepsilon\mathbb{Z}^2 \to \mathbb{R}^3$ and $E^{\mathrm{a}} \leq C\varepsilon^4$ with

$$E^{\mathrm{a}}(\mathbf{y}) = \varepsilon^2 \sum_i \left(V(D_1^\varepsilon \mathbf{y}_i) + f(\theta_i) + g(\omega_i) \right),$$

  where V denotes the pair potential, f denotes the bond angle θ potential and g denotes torsion angle ω (angle between two triangles) potential.

- Assume a similar scaling rule of displacement field

$$\mathbf{u} = \begin{pmatrix} u_1 \\ u_2 \\ u_3 \end{pmatrix} = \begin{pmatrix} \varepsilon^2 \tilde{u}_1 \\ \varepsilon^2 u_2 \\ \varepsilon u_3 \end{pmatrix}.$$

- So $\frac{1}{\varepsilon^4} E^{\mathrm{a}}$ is Γ-convergent to the von-Kármán model

$$E^{\mathrm{c}}(\tilde{\mathbf{u}}) = \int \mathbb{C} \left(\mathrm{sym}(\nabla \tilde{\mathbf{u}}_{1,2}) + \frac{1}{2} \nabla \tilde{u}_3 \otimes \nabla u_3 \right) : \left(\mathrm{sym}(\nabla \tilde{\mathbf{u}}_{1,2}) + \frac{1}{2} \nabla \tilde{u}_3 \otimes \nabla \tilde{u}_3 \right) dx_1 dx_2 + \int \frac{\kappa}{2} |\nabla^2 \tilde{u}_3|^2 dx_1 dx_2.$$

Extension to 2D

Four-body interactions must be included; and Tersoff and Brenner type interatomic potentials satisfy the assumptions.

Analysis is similar to the 1D case.

- The continuum total energy is **second order consistent** comparing to the **scaled** atomic total energy.
- The continuum force is pointwisely **first order consistent** comparing to the **scaled** atomic total energy.
- The **scaled** continuum displacement field has **first order** modeling error wrt the atomistic $\|\cdot\|_*$.

Physical continuum boundary conditions

- In a 1D atomistic model, Stefanelli proved that the boundary needs to specify the length of the chain in direction e_1 [17]

$$y_{N,1} - y_{1,1} = \mu(N-1)\varepsilon, \quad \mu \in (2/3, 1).$$

  In direction e_2, simply apply the clamped condition

$$y_{1,2} = D_1^\varepsilon y_{1,2} = y_{N,2} = D_1^\varepsilon y_{N,2} = 0.$$

  This corresponds to suspending the atomistic sample. [18]

- In the 2D continuum simulations , Zhang et al. 2014 obtained ripples with clamped boundary conditions applied to direction e_3. [19]

[17] U. Stefanelli, et. al. arxiv.1802.05053, (2018)

[18] Novoselov et. al. Nature, 446, 60-63, (2007).

[19] Zhang et. al. J. Mech. Phys. Solids, 67, 2-13, (2014)

Summary

- A continuum von-Kármán model is derived from atomistic descriptions by carefully specifying the scaling rules and homogenization.
- The continuum model is proved to be of second order in terms of energy, is of first order in terms of forces and displacement fields.
- A rigorous study of physical continuum boundary conditions is needed for both 1D and 2D atomistic and continuum models.
- Development of multiscale model is an ongoing project.

Thank you for your attention!

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