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Abstract — A multiscale approach to simulate graphene combining molecular mechanics and continuum mechanics is presented. A series of virtual expermiments is performed first, to develop computationally cheaper, substitute continuum model of the discrete molecular model of pristine graphene. Continuum and molecular models are then coupled providing the means to simulate a defected graphene while keeping the computational cost.
Key-words — graphene, virtual tests, elastic properties, molecular mechanics.

1 Introduction

Nanoscale systems and processes based on graphene [1] are becoming more viable for engineering applications, however, our ability to model their performance remains limited. The main challenge is that parts of graphene-based devices modelled with discreet models (such as molecular mechanics (MM) and molecular dynamics (MD)) typically contain extremely large number of particles, even though the actual physical dimension may be quite small. We focus in this work upon the MM neglecting both the dynamic effects and the thermal effects, used for quasi-static loading applications with the assumption of the zero Kelvin temperature. The equilibrium configuration of graphene corresponds to a state of minimum energy of the particle system. It is assumed here that the initial configuration is at equilibrium. Moreover, for the MM model we use classical interatomic potential depending on the position of atomic nucleus, namely modified Morse potential (see [2, 3]). Regardless of the above assumptions, following explicitly the trajectory for large number of degrees of freedom easily becomes intractable. Thus we reach for: 1. A substitute, continuum model which models the average mechanical behaviour of atomic system (in Sec. 2), and 2. A coupled multiscale (MS) model consisting of developed continuum and molecular model (in Sec. 3).

2 Continuum modeling of graphene

We tend here to give a combined MM and continuum approach for the study of the in plane mechanical behaviour of single layer graphene sheet under moderate deformations. We note that continuum model of graphene is extremely simplified, however, experimental investigation [4] show that graphene undergos large deformations, with highly nonlinear behavior and still remain *elastic*, with stable bonds and intact bond topology. Thus continuum modeling brings insight and quantitative information on the relevant physical phenomena, and seams optimal for the current and potential future applications. This further allows us to adapt and use a finite strain elastic model for the large range of phenomena and motivates us to benefit from somewhat simplified nonlinear membrane theory. We seek to adjust the nonlinear membrane theory which includes, as a special case, the hyperelastic model in terms of strain energy density (SED), W, as a function of principal stretches $W(\lambda_1, \lambda_2)$. This is an elegant alternative for the construction of the elastic constitutive response that satisfies the material indifference and isotropy restrictions (often used to characterise rubberlike materials [5]). In order to construct SED potential $W_{\text{fit}}(\lambda_1, \lambda_2)$, we determine the equilibrium potential energy of atomistic system for the series of biaxial loading cases performed on lattice sample, see Fig. 1 (left). The loading cases are designed to form the uniform grid in the space of λ_1, λ_2 resulting with the cloud of points shown as dots in Fig. 1 (right).

These results are further used to perform a surface fit given as $W_{\text{fit}}(\lambda_1, \lambda_2)$. With this result in hand, we

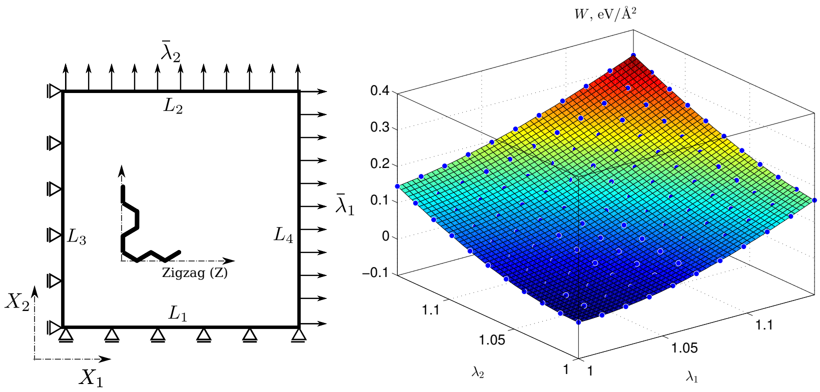

Figure 1: Scheme of the lattice sample with symmetry BCs used for biaxial tensile tests. The envelope of the sample is composed of lines L_i which coincides with boundary atoms (left). The surface fit W of SED obtained by series of biaxial tests performed by MM simulation.

can calculate the second Piola-Kirchhoff stress tensor ($\boldsymbol{S}$) and the elastic tangent modulus ($\mathbb{C}$)

$$\boldsymbol{S} := \sum_{i=1}^{2} \underbrace{\frac{1}{\lambda_i} \frac{\partial W(\lambda_1, \lambda_2)}{\partial \lambda_i}}_{s_i} \mathbf{n}_i \otimes \mathbf{n}_i, \qquad \mathbb{C} := 2\sum_{i=1}^{2} \frac{\partial s_i}{\partial \boldsymbol{C}} \mathbf{n}_i \otimes \mathbf{n}_i + 2\sum_{i=1}^{2} s_i \frac{\partial}{\partial \boldsymbol{C}} (\mathbf{n}_i \otimes \mathbf{n}_i). \tag{1}$$

Note that the matrix representations of these results can further be directly used for the calculation of the internal force vector and the element tangent stiffness matrix of the standard 2D large displacements elastic membrane finite element. Having the continuum description of the graphene behavior we turn to the coupling of molecular and continuum.

3 Multiscale atomistic-to-continuum coupling

In this paper coupling is based on the bridging domain (BD) method [6]. The basic idea is to divide the computational domain Ω in three subdomains, atomistic Ω^a, continuum Ω^c and their overlap $\Omega^b = \Omega^a \cap \Omega^c$, where the compatibility is enforced by Lagrange multiplier (LM) method. The total potential energy of the system can be written as

$$E_{tot,w} = E^a_{tot,w}(\mathbf{d}) + E^c_{tot,w}(\mathbf{u}), \tag{2}$$

where $\mathbf{d}$ and $\mathbf{u}$ are displacement vectors in the atomistic and continuum domains, respectively, and w in sequel denotes the energy weighting in Ω^b. The weighted atomistic and continuum energies are defined as

$$E^a_{tot,w} = \sum_i \left(\sum_{j \neq i} w^a_{ij} V_p + \sum_{j \neq k \neq i} w^a_i V_\theta \right) - \sum_{i \in \Omega^a} w^a_i \bar{\mathbf{f}}_i \cdot \mathbf{d}_i, \tag{3}$$

$$E^c_{tot,w} = \int_{\Omega^c} w^c(\mathbf{X}) W d\Omega^c - \int_{\Omega^c} w^c(\mathbf{X}) \mathbf{u} \cdot \mathbf{b} d\Omega^c - \int_{\Gamma^c_\sigma} w^c(\mathbf{X}) \mathbf{u} \cdot \bar{\mathbf{t}} d\Gamma^c, \tag{4}$$

where V_p and V_θ comes from interatomic potential (see [3]), and $\bar{\mathbf{t}}$, $\mathbf{b}$ is related to continuum tractions and volume forces, while $\bar{\mathbf{f}}_i$ denotes point forces on atom i. In order to enforce the compatibility between the atomistic and continuum domains, the coupling term, in terms of energy, is added to total energy forming

$$W_L := E_{tot,w} + C = E_{tot,w} + \int_{\Omega^b} \alpha_1 \boldsymbol{\lambda} \cdot (\mathbf{u} - \mathbf{d}^b) + \alpha_2 \nabla \boldsymbol{\lambda} (\nabla \mathbf{u} - \nabla \mathbf{d}^b) d\Omega. \tag{5}$$

where the choice of the weighting parameters α_1 and α_2 determines the coupling by mixing the displacement and strain coupling terms, $\mathbf{d}^b(\mathbf{X})$ is the interpolated atomistic displacement field in Ω^b, and $\boldsymbol{\lambda}$ denotes LM field.

We give next the weak form of the coupling problem. Let $\mathcal{V}^a$ and $\mathcal{V}_0^a$ be the sets of trial ($\mathbf{d}$) and test ($\mathbf{w}$) functions in Ω^a, respectively. Analogously, for the continuum domain let the standard space of admissible solutions be $\mathcal{V}$ and the space of test functions $\mathcal{V}_0$. Furthermore, the space of LM is denoted as $\mathcal{M} = \{\boldsymbol{\lambda}, \boldsymbol{\mu} \in H^1(\Omega)\}$. We further proceed to the minimising of the functional in (5) which leads to the saddle point problem, which can be written in terms of its weak form:
Find $(\mathbf{u}, \mathbf{d}, \boldsymbol{\lambda}) \in \mathcal{V} \times \mathcal{V}^a \times \mathcal{M}$ such that

$$\begin{aligned} G_w^c(\mathbf{u};\mathbf{v}) + G_w^a(\mathbf{d};\mathbf{w}) + (\boldsymbol{\lambda}, \mathbf{v} - \mathbf{w}^b_{i|_{i\in\Omega^b}})_C = 0 \qquad & \forall(\mathbf{v},\mathbf{w}) \in \mathcal{V}_0 \times \mathcal{V}_0^a, \\ (\boldsymbol{\mu}, \mathbf{u} - \mathbf{d}^b)_C = 0 \qquad & \forall \boldsymbol{\mu} \in \mathcal{M}, \end{aligned} \tag{6}$$

where $\mathbf{w}^b$ denotes interpolated test atom displacement field, and the terms defining the weak form of equilibrium with the scaling in the overlap are as follows

$$G_w^c(\mathbf{u};\mathbf{v}) := \int_{\Omega^c} w^c \nabla^s \mathbf{v} \cdot \boldsymbol{\sigma}(\nabla^s \mathbf{u}) d\Omega - \int_{\Omega^c} w^c \mathbf{v} \cdot \mathbf{b} d\Omega - \int_{\Gamma_\sigma^c} w^c \mathbf{v} \cdot \bar{\mathbf{t}} d\Gamma, \tag{7}$$

$$G_w^a(\mathbf{d};\mathbf{w}) := \sum_{i\in\Omega^a} \left(\sum_{j\neq i} w_{ij}^a \frac{\partial V_p}{\partial \mathbf{d}_i} \cdot \mathbf{w}_i + \sum_{j\neq k\neq i} w_i^a \frac{\partial V_\theta}{\partial \mathbf{d}_i} \cdot \mathbf{w}_i \right) - \sum_{i\in\Omega^a} w_i^a \bar{\mathbf{f}}_i \cdot \mathbf{w}_i. \tag{8}$$

The numerical implementation of the given coupling formulation pertains to the choice of the standard finite element and LM field approximation with corresponding shape functions (for the details see [7]).

We give next one numerical example of graphene sheet with initial crack-like defect, where the defect is modeled simply by removing a line of bonds parallel with the vertical direction. The coupled model will be compared with the fully molecular, see both models in Fig. 2. Moreover, we present the energy

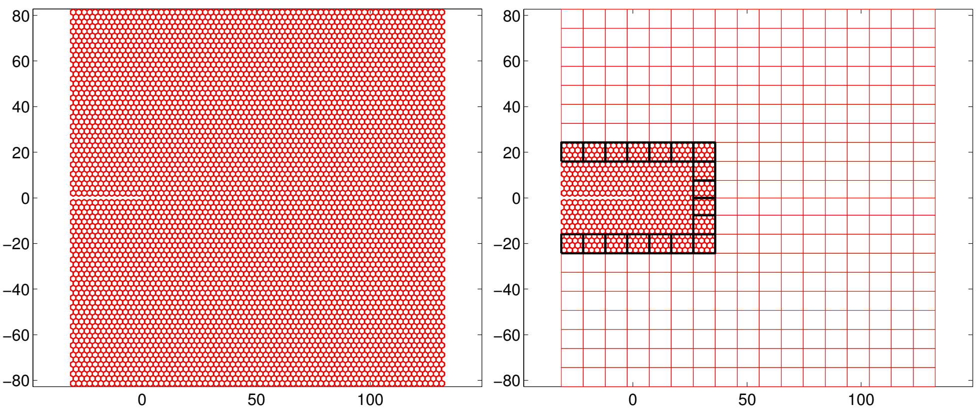

Figure 2: Graphene sheet with a hypothetical initial crack modeled using the fully molecular model (left) consisting of 10960 atoms and coupled model (right) with the size of atomistic domain 67.4×48.7 Å.

error convergence, as a basis for adaptivity procedure, with the increase of the Ω^a, for the mode-I type loading of the model from Fig. 2. Thus we define the global the global measure of the energy error as

$$e_E = \frac{\|\boldsymbol{\epsilon} - \boldsymbol{\epsilon}^{\text{ref}}\|_{\Omega^a \setminus \Omega^b}}{\|\boldsymbol{\epsilon}^{\text{ref}}\|_\Omega}, \qquad \|\boldsymbol{\epsilon}\| = \frac{1}{n_a} \sum_i^{n_a} \boldsymbol{\epsilon}_i : \boldsymbol{C} : \boldsymbol{\epsilon}_i, \tag{9}$$

where $\boldsymbol{\epsilon}^{\text{ref}}$ is the strain from the fully molecular model. The description of the models presented in the following results is given in Table 1 for the three different sizes of the Ω^a, and referential model. Model parameters are: number of atoms n_a, number of FE nodes n_n, number of LM nodes n_λ, number of degrees of freedom $n_{dof} = 2(n_a + n_n + n_\lambda)$.

Table 1: The data for the models 1, 2 and 3 used in convergence study. The size of the atomistic domain is defined by $L_1 \times L_3$ and given in Å.

id	ref	1	2	3
n_a	10960	1368	2080	2920
n_n	0	360	343	322
n_λ	0	38	46	54
n_{dof}	21920	3532	4938	6592
L_1	163.7134	67.4100	77.0400	86.6700
L_3	165.4100	48.6500	65.3300	82.0100

Not surprisingly the convergence is achieved as the size of the molecular domain is increased see Figure 3, where the type of coupling and weighting influences the error (for details see [7]).

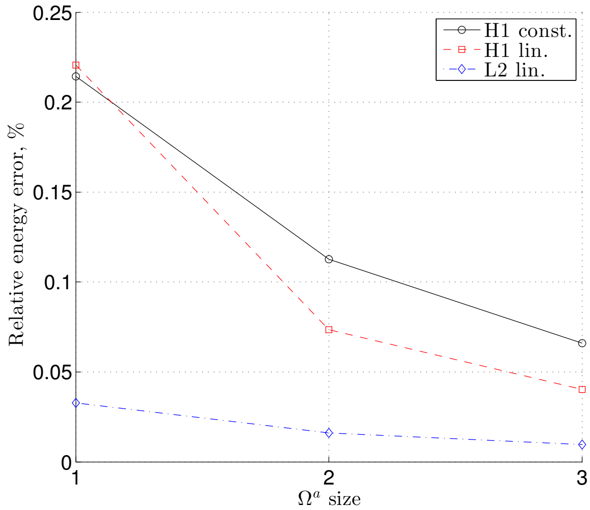

Figure 3: Convergence of the energy error e_E given for different molecular domain dimensions and the different couplings from (5) (L2 $(\alpha_1 = 1, \alpha_2 = 0)$, H1 $(\alpha_1 = 1, \alpha_2 = 1)$).

To conclude, we note that for the model denoted as 1 in the Table 1 n_{dof} is reduced by 84%, while the corresponding solution yields negligible error (less then 0.25%) with respect to the fully atomistic model showing really good performance of the modeling strategy.

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