Structural deformations of two-dimensional planar structures under uniaxial strain: The case of graphene - (Supplementary File)

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Abstract. (of the main manuscript): In the present work, a method for the study of the structural deformations of two dimensional planar structures under uniaxial strain is presented. The method is based on molecular mechanics using the original stick and spiral model and a modified one which includes second nearest neighbor interactions for bond stretching. As we show, the method allows an accurate prediction of the structural deformations of any two dimensional planar structure as a function of strain, along any strain direction in the elastic regime, if structural deformations are known along specific strain directions, which are used to calculate the stick and spiral model parameters. Our method can be generalized including other strain conditions and not only uniaxial strain. We apply this method to graphene and we test its validity, using results obtained from *ab initio* Density Functional Theory calculations. What we find is that the original stick and spiral model is not appropriate to describe accurately the structural deformations of graphene in the elastic regime. However, the introduction of second nearest neighbor interactions provides a very accurate description.

Keywords: structural deformations, planar structures, uniaxial strain, graphene, molecular mechanics

Fitting functions for the bond length and bond angle deformations

The bond length and bond angle deformations (δl_i and $\delta\phi_{ij}$, respectively) for uniaxial strain ε in the range [-0.05, 0.05] are fitted to quadratic functions of ε. These fitting functions are presented bellow. The intercepts of the fitting functions are not forced to be zero. However, their values are of the order of 10^{-4} – 10^{-5} in Å and rad units for δl_i and $\delta\phi_{ij}$, respectively. Therefore, they are negligible. The units of δl_i and $\delta\phi_{ij}$ in those fitting functions are Å and rad, respectively. For the expressions of the form $\delta l_i = 3a_0 f_i(\varepsilon)$, a_0 is the bond length of graphene at equilibrium for $\varepsilon = 0$, ($a_0 = 1.4216$ Å according to our DFT calculations), the quadratic function $f_i(\varepsilon)$ is dimensionless and its intercept is set to zero.

- For strain along $\boldsymbol{L} = \boldsymbol{a} + \boldsymbol{b}$ ($\theta_0 = 0$, i.e. along arm chair direction).
  - $\delta l_1 = -0.188811\varepsilon^2 + 0.283636\varepsilon - 3.85 \times 10^{-5} = 3a_0(-0.0442719\varepsilon^2 + 0.066506\varepsilon)$
  - $\delta l_2 = \delta l_1$
  - $\delta l_3 = 2.289044\varepsilon^2 + 1.184000\varepsilon + 1.10 \times 10^{-5} = 3a_0(0.536730\varepsilon^2 + 0.277621\varepsilon)$
  - $\delta\phi_{12} = 3.563471\varepsilon^2 - 1.309408\varepsilon - 2.52 \times 10^{-5}$
  - $\delta\phi_{23} = -1.781735\varepsilon^2 + 0.654704\varepsilon + 1.26 \times 10^{-5}$
  - $\delta\phi_{31} = \delta\phi_{23}$
- For strain along $\boldsymbol{L}_\perp = \boldsymbol{a} - \boldsymbol{b}$ ($\theta_0 = 90^o$, i.e. along zig-zag direction).
  - $\delta l_1 = 1.065268\varepsilon^2 + 0.880364\varepsilon - 11.1 \times 10^{-5} = 3a_0(0.249781\varepsilon^2 + 0.206426\varepsilon)$
  - $\delta l_2 = \delta l_1$
  - $\delta l_3 = -0.744755\varepsilon^2 - 0.00663636\varepsilon + 2.66 \times 10^{-5} = 3a_0(-0.174628\varepsilon^2 - 0.001556\varepsilon)$
  - $\delta\phi_{12} = -2.605373\varepsilon^2 + 1.315279\varepsilon - 10.15 \times 10^{-5}$
  - $\delta\phi_{23} = 1.302690\varepsilon^2 - 0.657640\varepsilon^2 + 5.07 \times 10^{-5}$
  - $\delta\phi_{31} = \delta\phi_{23}$
- For strain along $\boldsymbol{L} = 2\boldsymbol{a} + \boldsymbol{b}$, ($\theta_0 = 10.893395^o$).
  - $\delta l_1 = -0.442890\varepsilon^2 + 0.118545\varepsilon + 4.29 \times 10^{-5} = 3a_0(-0.103848\varepsilon^2 + 0.027796\varepsilon)$
  - $\delta l_2 = 0.142191\varepsilon^2 + 0.495818\varepsilon - 5.13 \times 10^{-5} = 3a_0(0.0333406\varepsilon^2 + 0.116258\varepsilon)$
  - $\delta l_3 = 2.047786\varepsilon^2 + 1.139182\varepsilon - 0.23 \times 10^{-5} = 3a_0(0.480160\varepsilon^2 + 0.267113\varepsilon)$
  - $\delta\phi_{12} = 3.166204\varepsilon^2 - 1.218509\varepsilon - 1.03 \times 10^{-5}$
  - $\delta\phi_{23} = -2.409895\varepsilon^2 + 1.032964\varepsilon - 1.29 \times 10^{-5}$
  - $\delta\phi_{31} = -0.740239\varepsilon^2 + 0.185116\varepsilon + 1.04 \times 10^{-5}$
- For strain along $\boldsymbol{L}_\perp = 4\boldsymbol{a} - 5\boldsymbol{b}$, ($\theta_0 = 79.106605^o$).
  - $\delta l_1 = 1.728438\varepsilon^2 + 1.053000\varepsilon + 3.52 \times 10^{-5} = 3a_0(0.405280\varepsilon^2 + 0.246905\varepsilon)$
  - $\delta l_2 = 0.453380\varepsilon^2 + 0.665909\varepsilon - 5.34 \times 10^{-5} = 3a_0(0.106307\varepsilon^2 + 0.156141\varepsilon)$
  - $\delta l_3 = -0.700204\varepsilon^2 + 0.0368182\varepsilon - 4.10 \times 10^{-5} = 3a_0(-0.164182\varepsilon^2 + 0.008633\varepsilon)$
  - $\delta\phi_{12} = -2.619619\varepsilon^2 + 1.221761\varepsilon + 0.48 \times 10^{-5}$
  - $\delta\phi_{23} = 2.475599\varepsilon^2 - 1.031171\varepsilon + 2.34 \times 10^{-5}$
  - $\delta\phi_{31} = 0.142596\varepsilon^2 - 0.190542\varepsilon - 2.68 \times 10^{-5}$