10 ANNIVERSARY
GRAPHENE AND 2DM VIRTUAL CONFERENCE & EXPO
Graphene online 2020
October 19-23

An atomistic model for flexoelectricity tensor in non-perfect graphene

Yida Yang, Gautier Lecoutre, Laurant Hirsinger, Michel Devel
Institut FEMTO-ST, Univ. Bourgogne Franche-Comté, CNRS, ENSMM, 15B avenue des Montboucons 25030 Besançon CEDEX, France

INTRODUCTION

When a strain gradient is applied to some possibly centrosymmetric dielectric, a strain gradient appears due to a phenomenon called flexoelectricity[1]. Being able to accurately compute flexoelectricity coefficients is a foundation for finding a combination of materials with a relatively large global flexoelectricity coefficient for future use. Recently, B. Javvaji et al published a paper[2] on the calculation of piezoelectricity and flexoelectricity coefficients for a patterned graphene, using molecular dynamics (MD) simulations coupled with a charge dipole model[3][4]. Reading this paper, however, it seemed to us that some terms were missing in the computation of the forces with respect to what we usually use, i.e. terms coming from gradient of effective charges and dipoles that change when the positions of atoms change. We thus decided to carry out the same simulations as Javvaji et al. to assess the importance of the missing terms and possibly provide improved numerical estimations. Hence, a comparison between our results and those reported by Javvaji et al. are presented in the poster, along with some preliminary results on 2D MoS_2.

Flexoelectricity

When a material is submitted to an external force, the resulting deformation gradient can break its spatial inversion symmetry and consequently induce a change of the polarization.

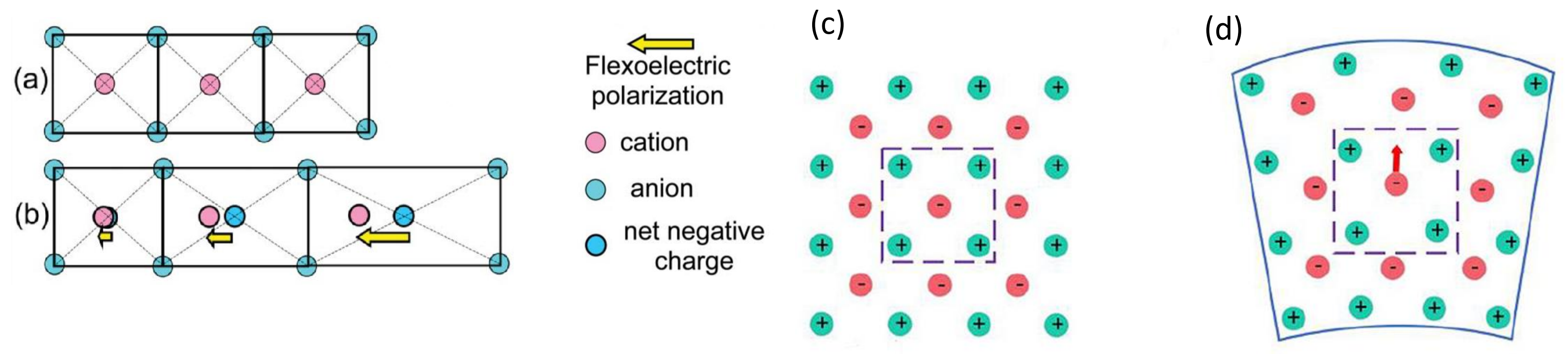

Figure 1. The origin of flexoelectric effects in solids.

(a)(c) non-deformed 2D structure of elementary charges. When the geometric centers of positive and negative charges coincide, the net dipole moment of the unit cell is zero. (b)(d) The material is deformed by an external force, the strain-gradient induces an uncompensated dipole moment via the flexocoupling mechanism.

The total polarization in an electromechanical system can be expressed as follows:

$$P_i = \varepsilon_0 \chi_{ij} E_j + e_{ijk} u_{jk} + \boxed{\mu_{klij} u_{kl,j}} \quad \textbf{flexoelectricity}$$

$$u_{jk} = \left(U_{jk} + U_{kj}\right)/2$$

where, e_{ijk} is a piezoelectricity tensor, χ_{ij} is the dielectric susceptibility, u_{jk} the symmetric strain tensor and $u_{jk,l}$ the strain gradient. μ_{klij} is the flexoelectricity tensor. The last term is the direct flexoelectric effect written using a symmetrized strain tensor.

Gaussian regularized Charge dipole model

In the charge dipole model, each atom is supposed to carry an effective charge q and dipole moment **p**. The total electrostatic interaction energy E is expressed as

$$E = E^{\text{q-q}} + E^{\text{q-p}} + E^{\text{p-p}} + E^{\text{ext}}$$

where,

$$E^{q-q} = \frac{1}{2}\sum_{i,j} q_i T_{ij}^{q-q} q_j \qquad T_{ij}^{q-q} = \frac{\operatorname{erf}\left(r_{ij}/\sqrt{R_i^2 + R_j^2}\right)}{4\pi\varepsilon_0 r_{ij}}$$

$$E^{q-p} = \frac{1}{2}\sum_{i,j} q_i \bar{T}_{ij}^{q-p} \bar{p}_j \qquad \bar{T}_{ij}^{q-p} = -\nabla T_{ij}^{q-q}$$

$$E^{p-p} = \frac{1}{2}\sum_{i,j} \bar{p}_i \bar{\bar{T}}_{ij}^{p-p} \bar{p}_j \qquad \bar{\bar{T}}_{ij}^{p-p} = \nabla\nabla T_{ij}^{q-q}$$

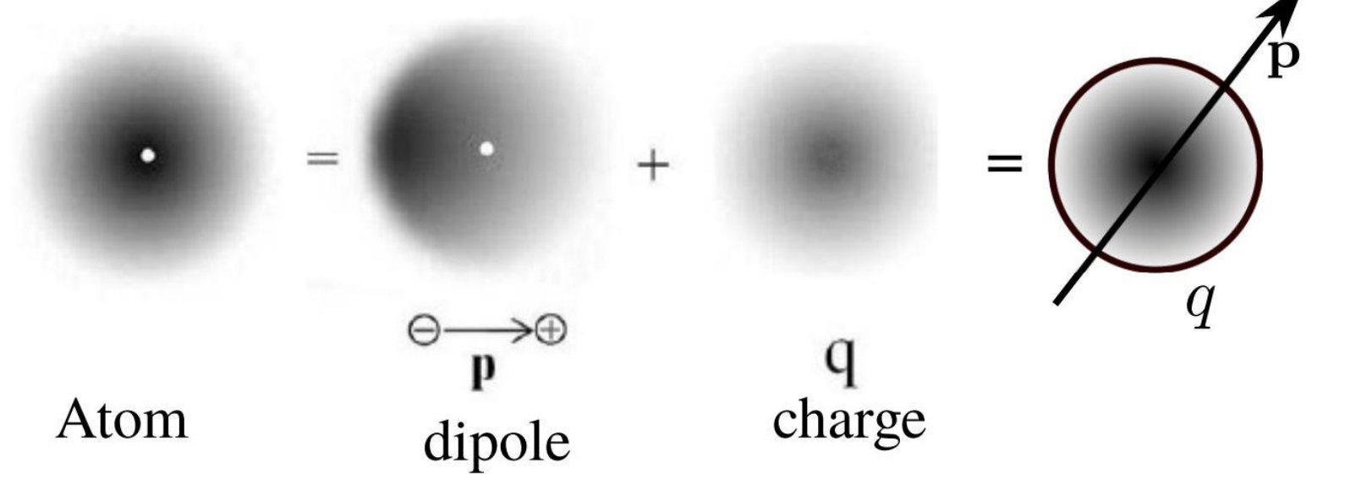

Fig. 2 Schematic representation of atom in charge-dipole scheme

T^{q-q}, T^{q-p} and T^{p-p} are interaction tensors, between point charges and dipoles, that diverge when 2 atoms are too close. In order to overcome this, the tensors are regularized by convolution with a radial Gaussian function[3][4], i.e

point charges ⇒ Gaussian charges

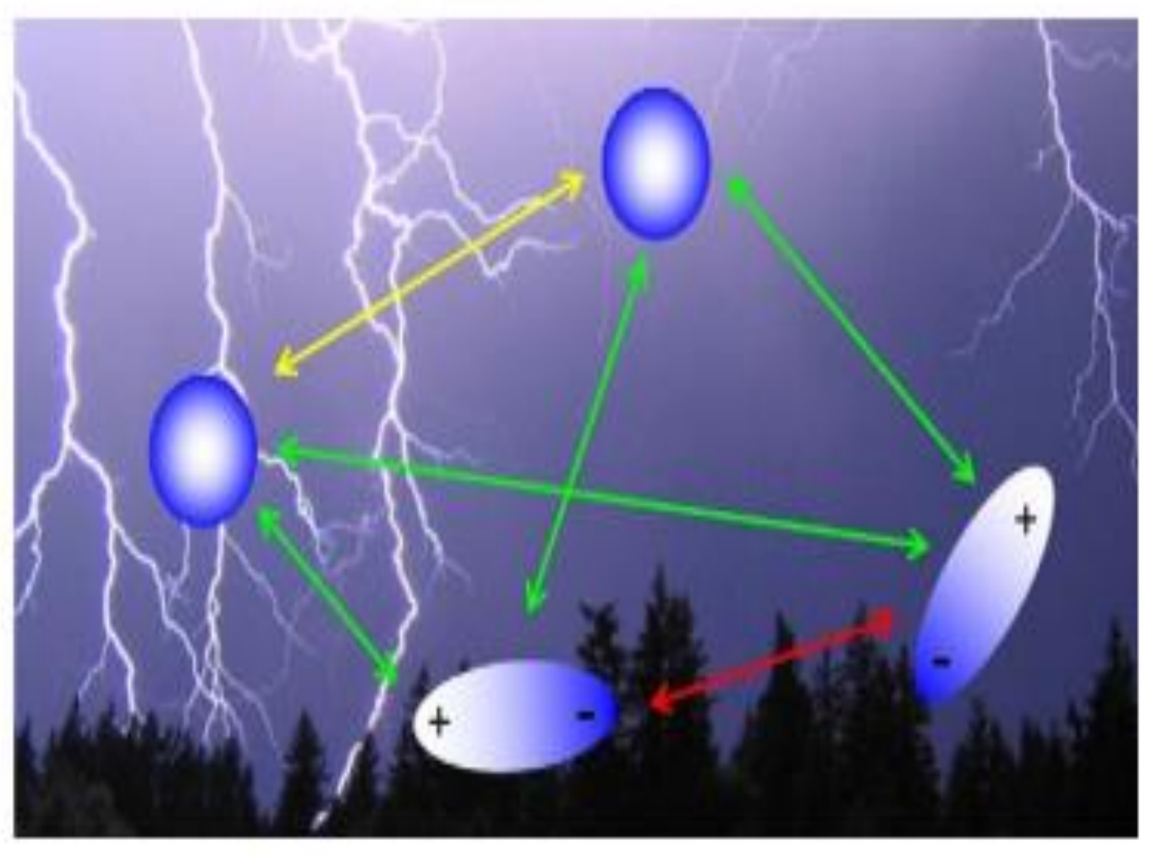

Fig. 3 The five types of energy contribution in charge dipole scheme.

MD simulation-AIREBO many-body potential

As Javvaji et al. we use the AIREBO many-body potential to describe the covalent and van der Waals bonds between carbon atoms. This potential has been used by many other authors to predict some physical and chemical properties of various carbon materials. The AIREBO potential consists of three terms:

$$E = \frac{1}{2}\sum_{i}\sum_{j\neq i}\left[E_{ij}^{\text{REBO}} + E_{ij}^{\text{Lj}} + \sum_{k\neq i,j}\sum_{l\neq i,j,k} E_{kijL}^{\text{TORSiON}}\right]$$

$$E_{ij}^{REBO} = \varphi^R(r_{ij}) - b_{ij}\varphi^A(r_{ij})$$

$$b_{ij} = \frac{1}{2}\left(b_{ij}^{\sigma-\pi} + b_{ji}^{\sigma-\pi} + b_{ji}^{RC} + b_{ji}^{DH}\right)$$

E^{LJ} is a Lennard-Jones 12-6 potential term, E^{tor} is a single-bond torsion term and E^{REBO} is composed of interatomic repulsion φ^R and attraction terms φ^A. The bond order function b_{ij} includes the many-body effects, where $b_{ij}^{\sigma-\pi}$ depends on the atomic distance and bond angle, b_{ji}^{RC} represents the influence of bond conjugation and b_{ji}^{DH} is a dihedral-angle term for double bonds.

Calculation of piezoelectricity and flexoelectricity coefficient for trapezium graphene

The local atomic strain $\bar{\bar{\varepsilon}}^{at}$ for every atom is given as $\bar{\bar{\varepsilon}}_i^{at} = \frac{1}{2}[(\bar{\bar{F}}_i)^T \bar{\bar{F}}_i - \bar{\bar{I}}]$

where $\bar{\bar{F}}_i$ is the deformation gradient of atom i and $\bar{\bar{I}}$ stands for identity matrix. The $\bar{\bar{F}}_i$ tensors were calculated according to a method reported in paper[5].

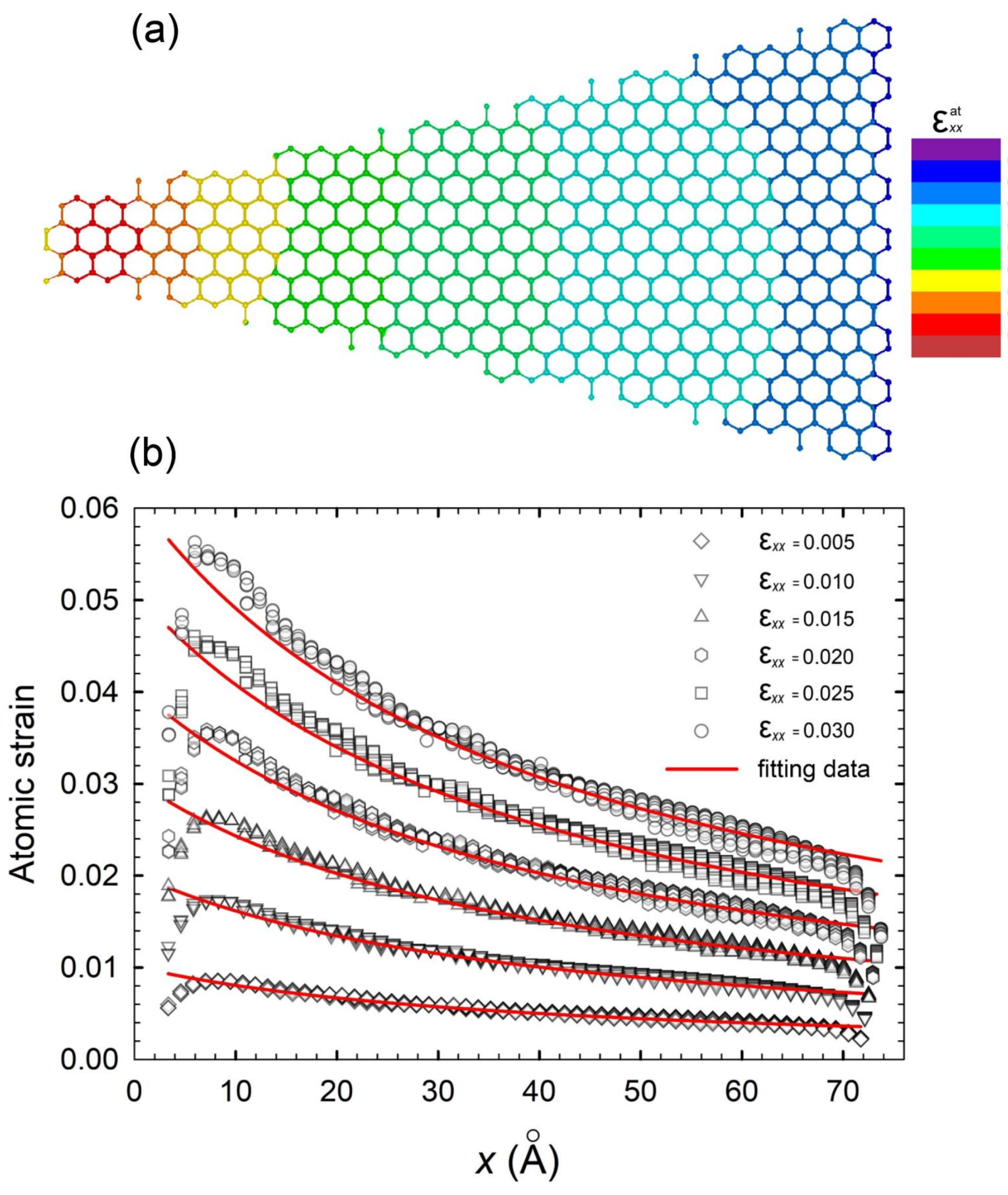

Fig. 4 (a) Atomic strain distribution for trapezium-shaped graphene. (b) Atomic strain along x axis for every atoms. x denotes the atomic position along x axis.

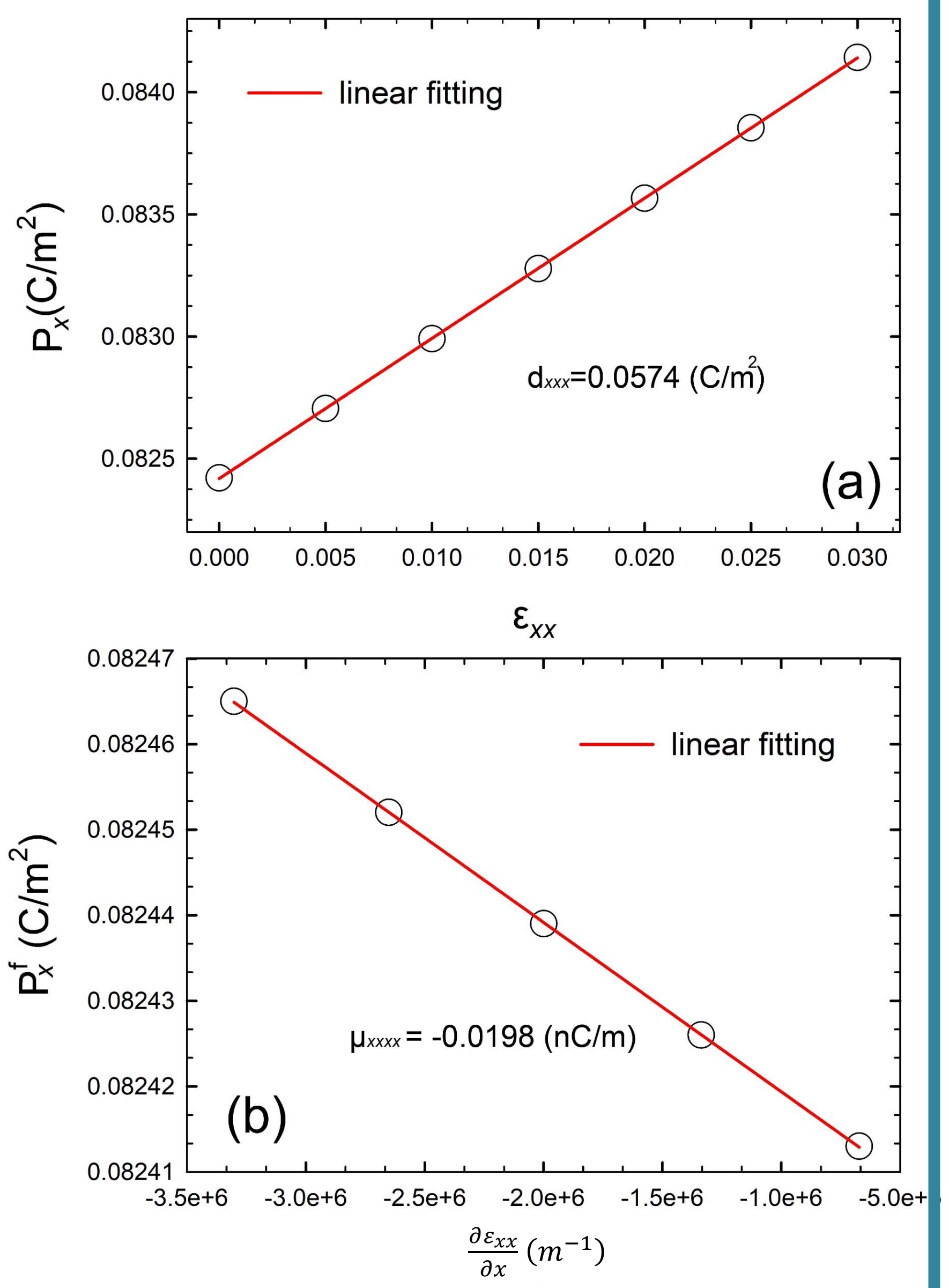

Fig. 5 Polarization as a function of strain (a) and strain gradient (b), respectively. Slopes stand for piezoelectricity(a) and flexoelectricity(b) coefficient.

	B. Javvaji	present result
piezoelectric coefficient (C/m^2)	0.08013	0.0574
flexoelectric coefficient (nC/m)	-0.789886	-0.0198

Table 1 : Comparison of piezoelectric and flexoelectricity coefficient between results reported by B. Javvaji and present calculated result.

Perspectives: Calculation of a flexoelectricity coefficient for MoS_2

The Gaussian regularized charge dipole model is adopted to calculate the polarization distribution for an MoS_2 nanoribbon with 732 atoms under an electric field[6].

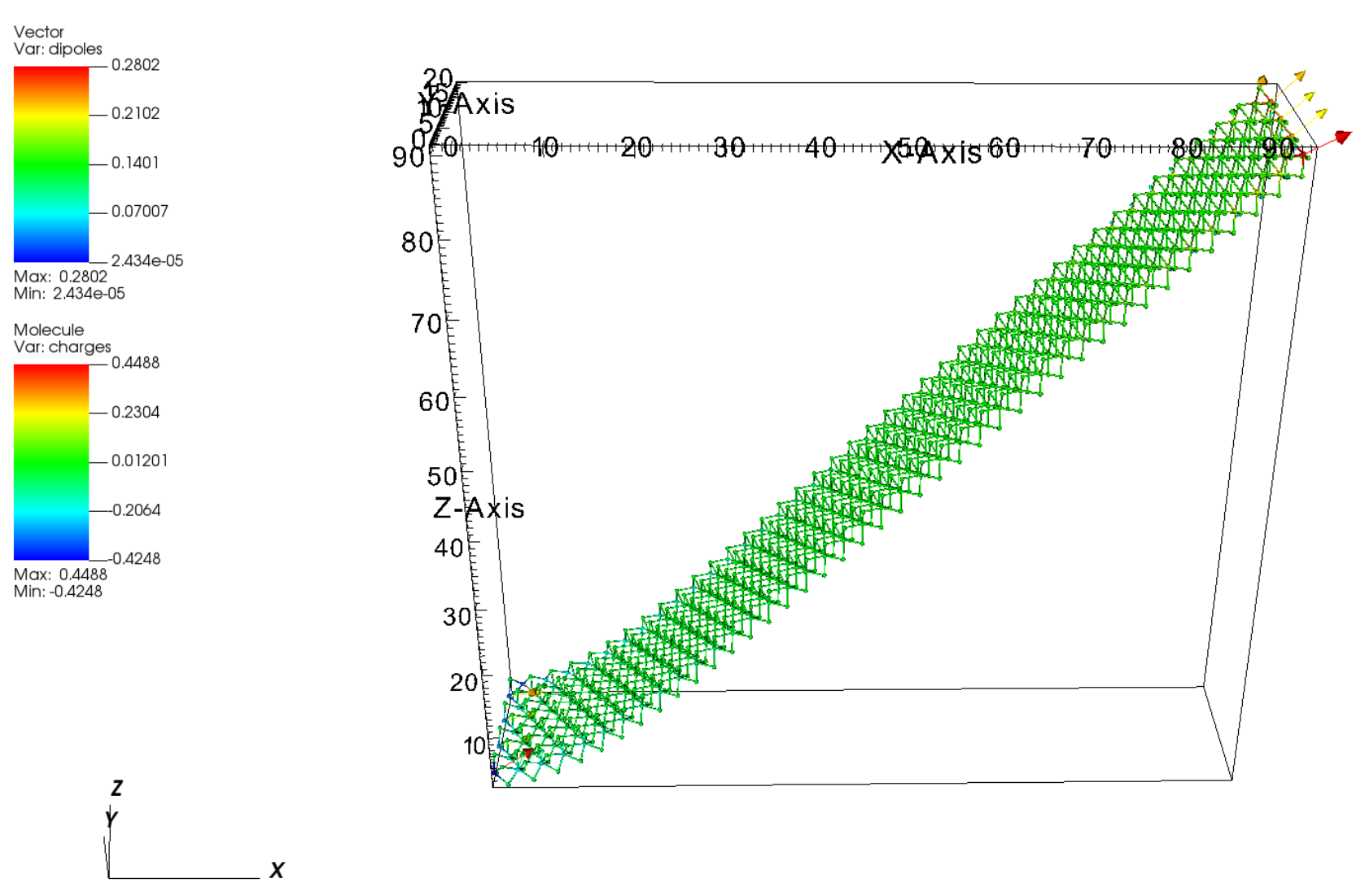

Fig. 7 Induced-field deflection in MoS_2 nanoribbon, subjected to a 2.83 V/nm external electric field, with the nanoribbon fixed at its left edge.

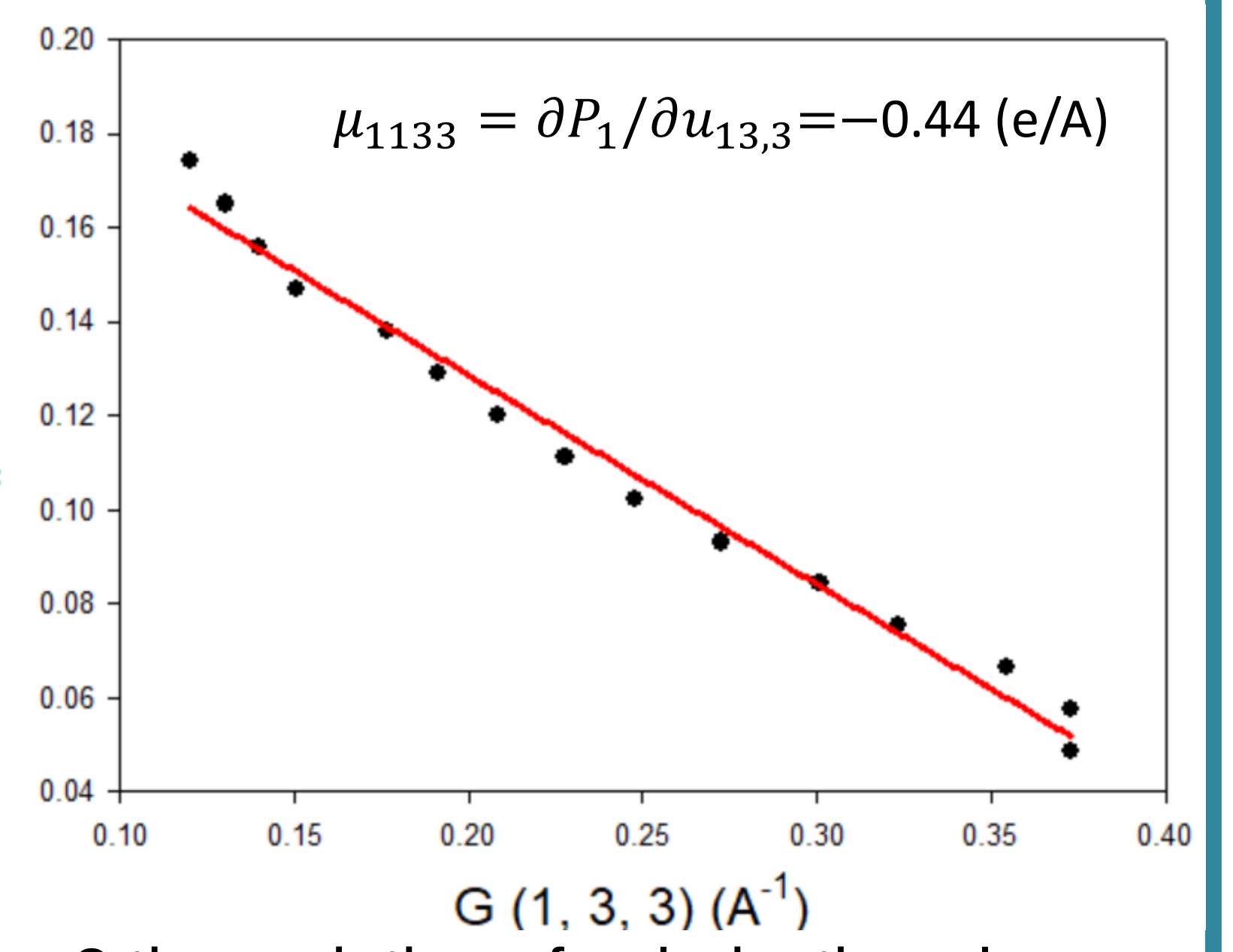

Fig. 8 the variation of polarization along x axis as a function of second order gradient of transformation $G(1,3,3) = u_{13,3}$

We expect to extend our current method to **combinations of monolayers of various 2D materials** for the computation of their flexoelectricity tensor components.

CONTACT PERSON

Yida YANG

yida.yang@femto-st.fr
PhD student

REFERENCES

[1] Qian Deng, Mejdi Kammoun, Alper Erturk, Pradeep Sharma, Nanoscale flexoelectric energy harvesting, International Journal of Solids and Structures, (2014) **51**, 3218-3225
[2] Brahmanandam Javvaji, Bo He and Xiaoying Zhuang, The generation of piezoelectricity and flexoelectricity in graphene by breaking the materials symmetries, Nanotechnology (2018) **29**, 225702
[3] Alexandre Mayer, Philippe Lambin, Rachel Langlet, Charge-dipole model to compute the polarization of fullerenes, Applied Physics Letters (2006) **89**, 063117
[4] Zhao Wang, Michel Devel, Electrostatic deflections of cantilevered metallic carbon nanotubes via charge-dipole model, Physical Review B (2007) **76**, 195434
[5] Javvaji B, Shenoy B M, Mahapatra D R, Ravikumar A, Hegde G M and Rizwan M R, Stable configurations of graphene on silicon, Applied Surface Science (2017) **414**, 25–33
[6] Yida Yang, Michel Devel and Zhao Wang, An atomistic model for the charge distribution in layered MoS_2, The Journal of Chemical Physics, (2018) **149**, 124102

Graphene online 2020