Universitat d'Alacant
Universidad de Alicante

UNCUYO
UNIVERSIDAD
NACIONAL DE CUYO

The effect of strain and temperature on the mechanical properties of irradiated graphene

J. Martinez-Asencio[1], C. J. Ruestes[2], E. Bringa[2], M. J. Caturla[1]

[1] Dept. Física Aplicada, Universidad de Alicante, Spain
[2] CONICET and UNCuyo, Mendoza, Argentina

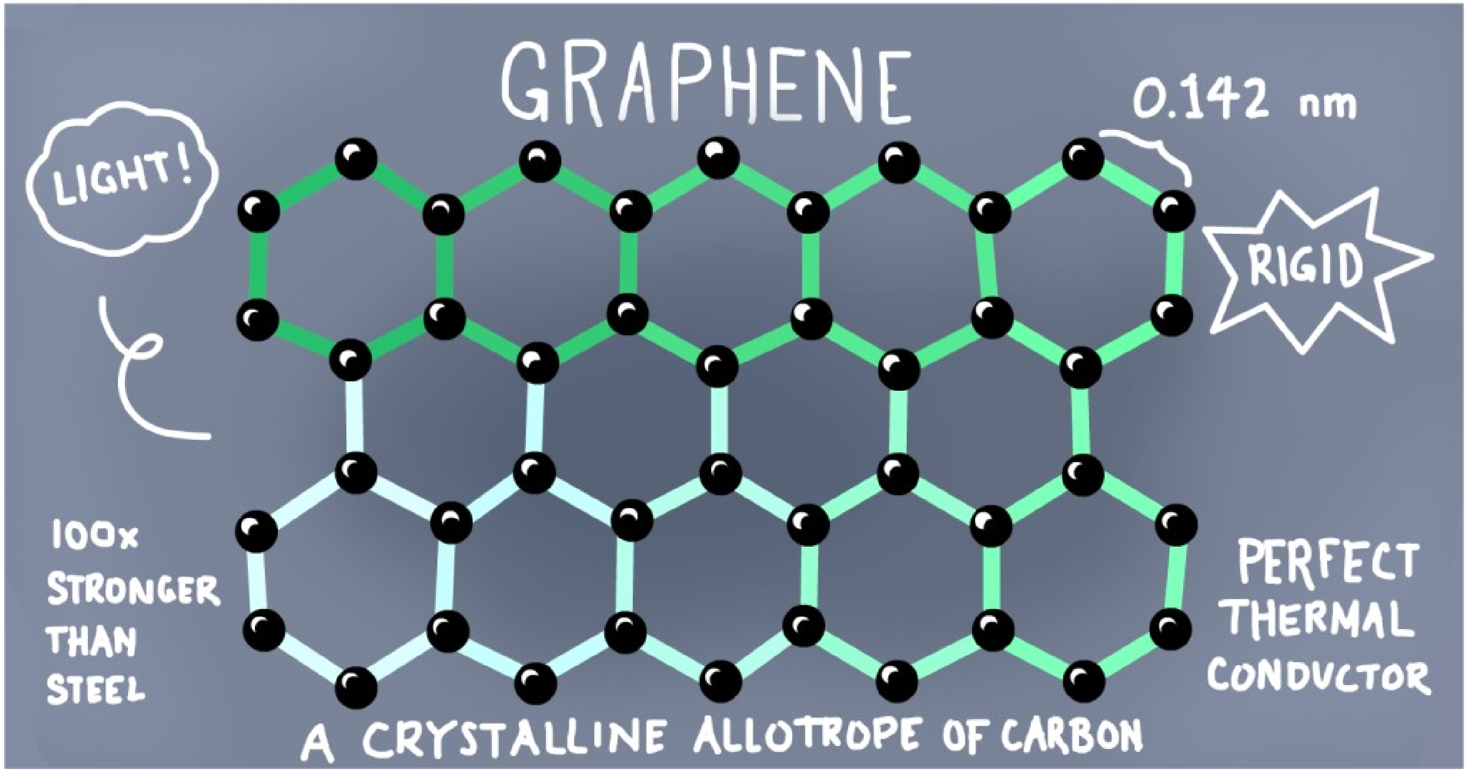
10th RES Users conference, León, September 2016

Outline

- Motivation
- Simulations set up and requirements
- Results
- Conclusions

Motivation

Graphene:
a one-atom thick material with special electrical and mechanical properties

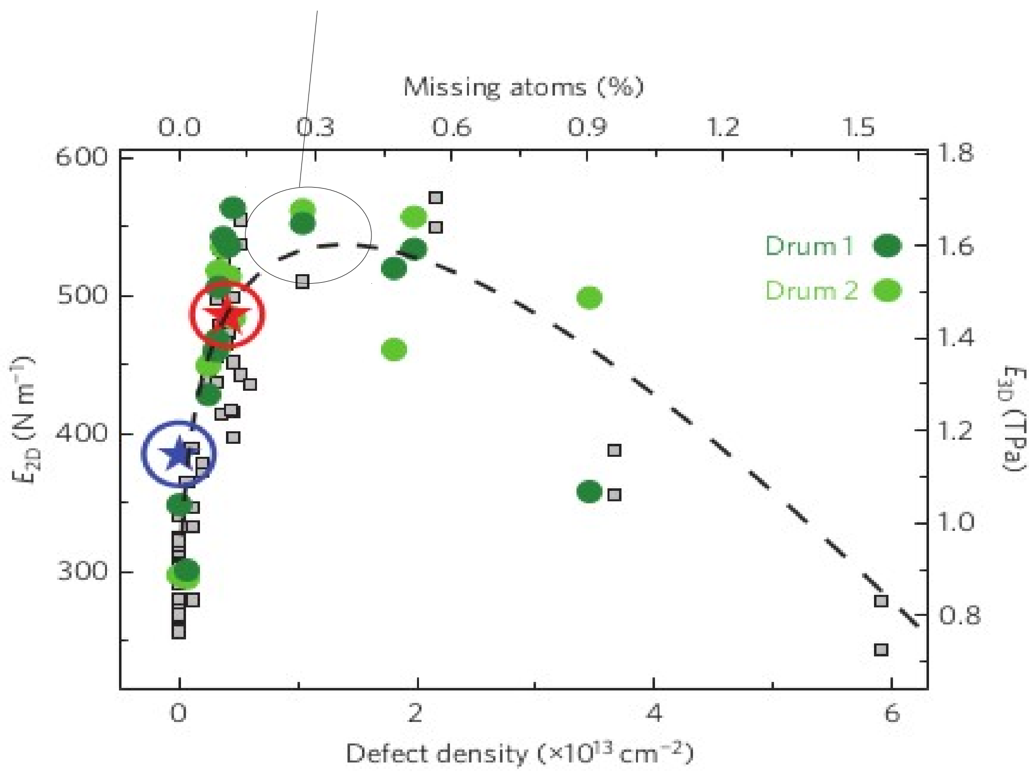


http://graphenewholesale.com/what-is-graphene/

Motivation

The mechanical properties of graphene can be modified by the introduction of defects → ion irradiation

~0.2% → E(2D)~550 Nm^{-1}



López-Polin et al. Nature Physics, 2015, 11, 26-31

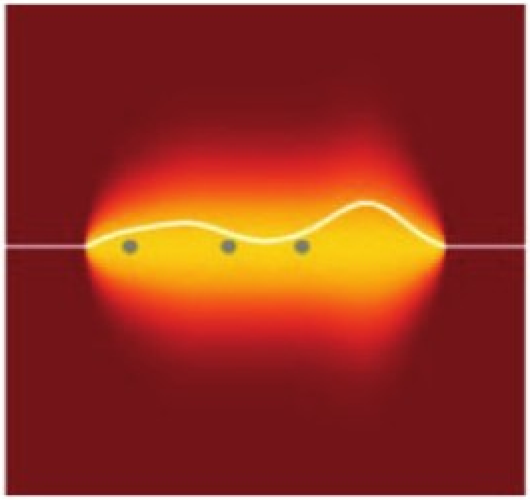
E2D increases for very low irradiation doses

Motivation

Possible explanations

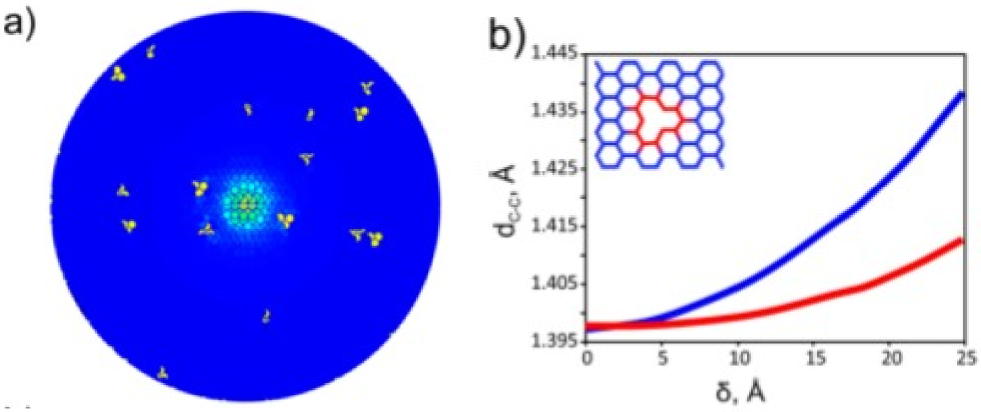
López-Polin et al.
Nature Physics 2015, 11, 26-31

Effect of defects on thermal fluctuations



Kvashnin & Sorokin,
J. Phys. Chem. Lett. 2015, 6, 2384

MD simulations: monovacancies

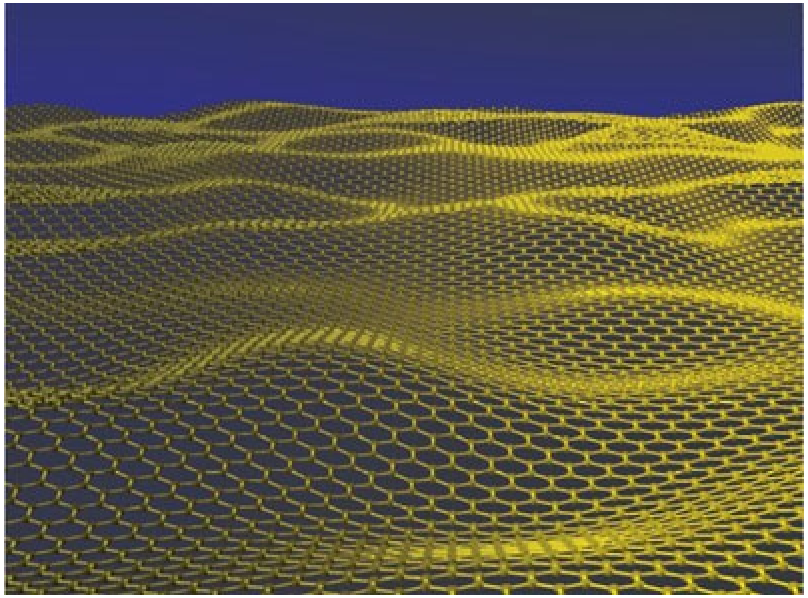


Limitations: 0 K / no irradiation

Motivation

General question:

What is the role of ripples on the mechanical response of graphene?



IOP Physics World,
1 February 2011

P. San-Jose, J. González, and F. Guinea
Phys. Rev. Lett. 106, 045502, 2011

IRRADIATION + TEMPERATURE + STRAIN

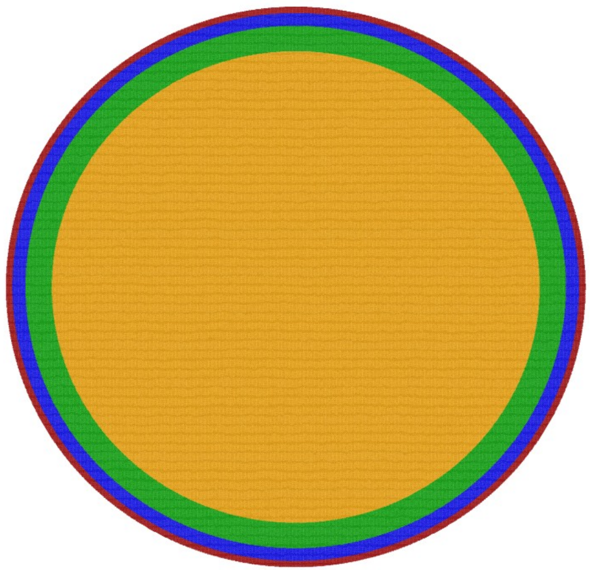
Simulation set up and requirements
Irradiation

Two types of simulations → (1) Irradiation

- LAMMPS code.
- 3-body Hybrid Tersoff/ZBL (C-C) and ZBL potential (Ar-C) (Ar-Ar)
- T=300 K (as in experiments) and 10 K (to check T effects)
- 75 nm radius graphene drumhead (674644 atoms)
- Fixed boundaries by a thermal langevin bath (300 K or 10 K)

- Relaxation:
  - Minimization + box relax (cg).*
  - NPH ensemble 3 ps

- Bombardment with Ar+ ions:
  - NVE ensemble
  - Low energy: 140 eV
  - 1000 ions. 1 every 5000 steps (dt/reset)
  - Randomly (perpend.) within a R=65 nm circle.
  - Equilibration 35000 timesteps (1 fs each)



- *Also for different initial strains: -0.25% (compressive) to 0.25% (tensile)

Simulation set up and requirements
Nanoindentation

Two types of simulations → (2) Nanoindentation

- Spherical indenter tip with repulsive potential

Kelchner et al. (Phys.Rev. B, 1998, 58, 11085)

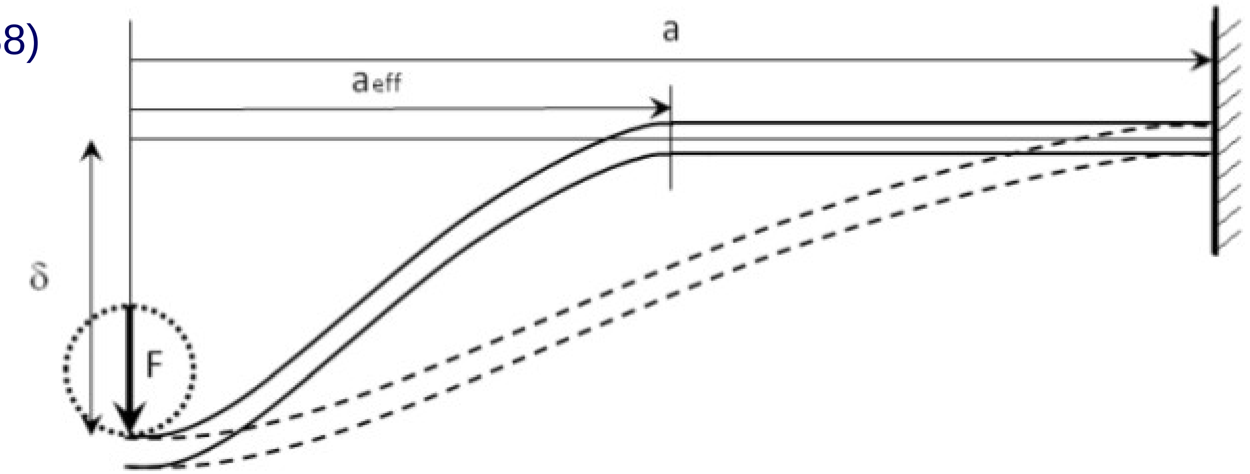
- Indenter radius R= 10 nm.
- Penetration rate of 5 m/s
- Upon contact: force computed every 0.1 ps
- Membrane model (following the non-linear Föppl theory)

$$F(\delta) = \pi\, \sigma_0^{2D}\, \delta + q^3/a^2\, E_{2D}\, \delta^3$$

Lee et al. (Science 2008, 321, 385-388)

q=1.02 value from

Komaragiri et al.
J. Appl. Mech. 2005, 72, 203)



Simulation set up and requirements
Some numbers

Irradiation

512 cores (32 nodes) in MareNostrum
Time ~ 14 h / calculation = 12kCPUh

At least 26 calculations performed:
different temperatures & strains

Large files for output: ~ 5Gb/simulation

Indentation

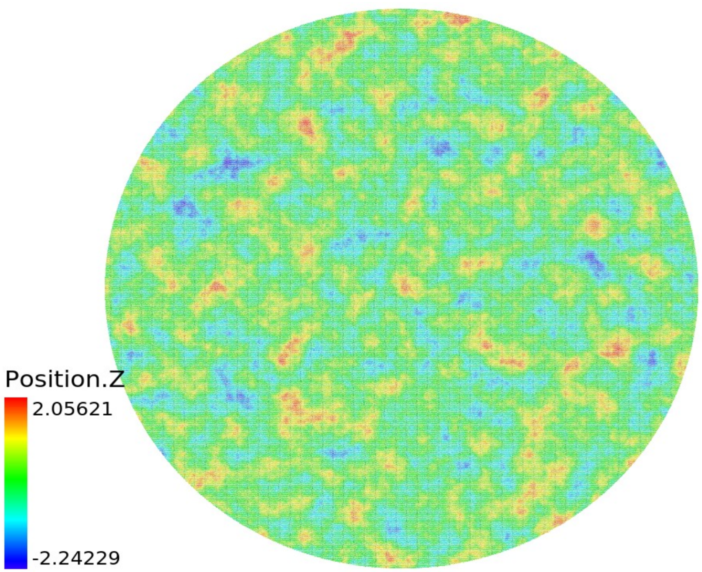
256 cores (16 nodes) in MareNostrum
Time ~ 6 h / calculation = 1.5kCPUh

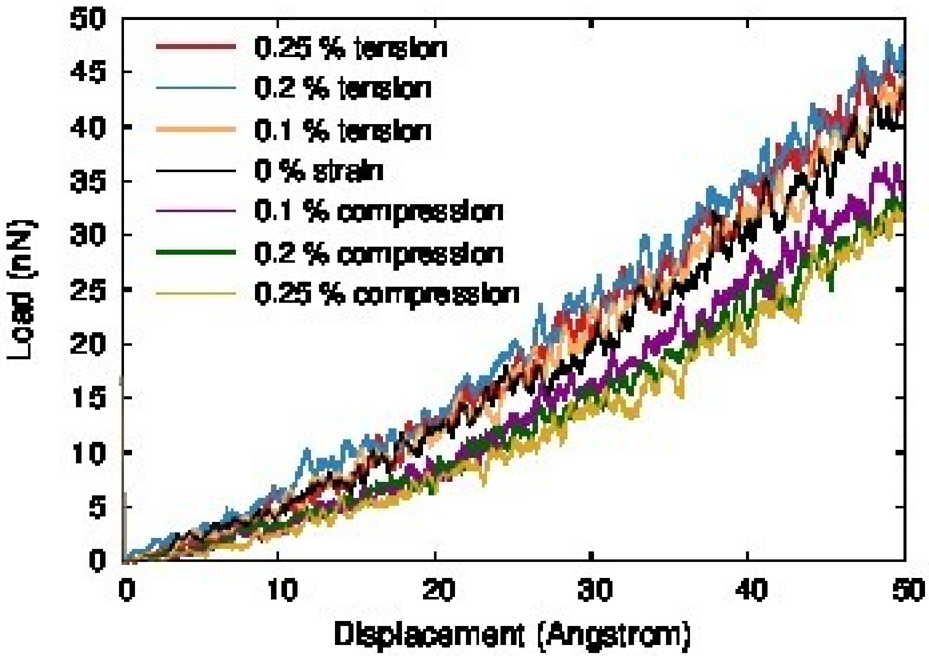
At least 140 calculations performed:
different temperatures & strains & doses

Large files for output: ~ 2Gb/simulation

Results:
Irradiation and nano-indentation

T = 300K



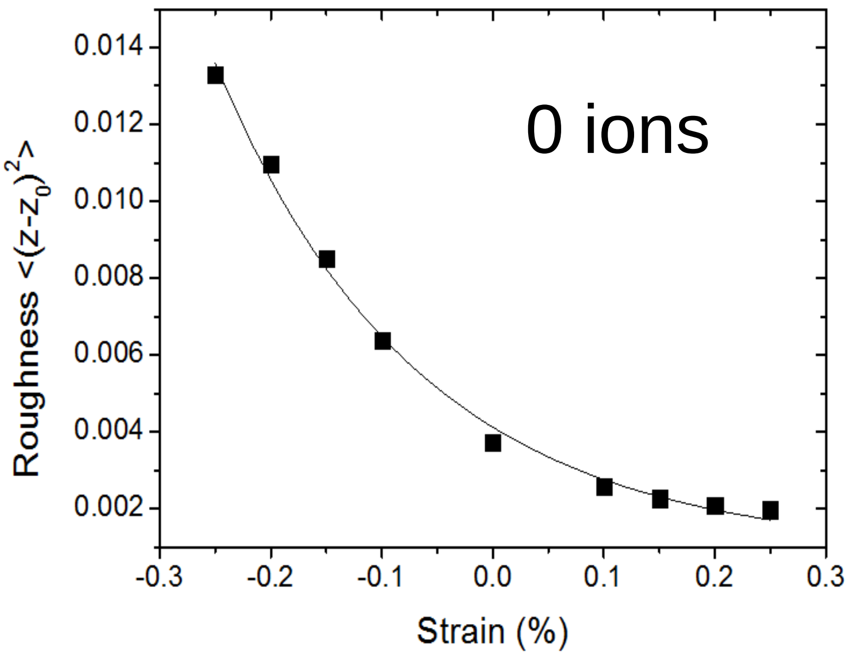


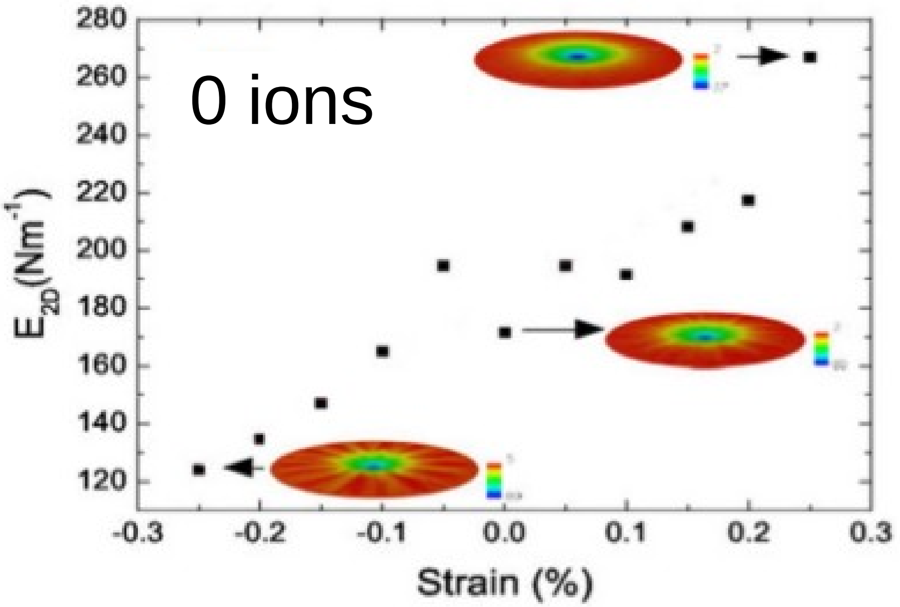
Initial distribution of ripples in the graphene membrane (no initially applied strain)

E2D obtained from Force vs. displacement calculations

Results:
unirradiated graphene – effect of applied strain

Before irradiation, T = 300K





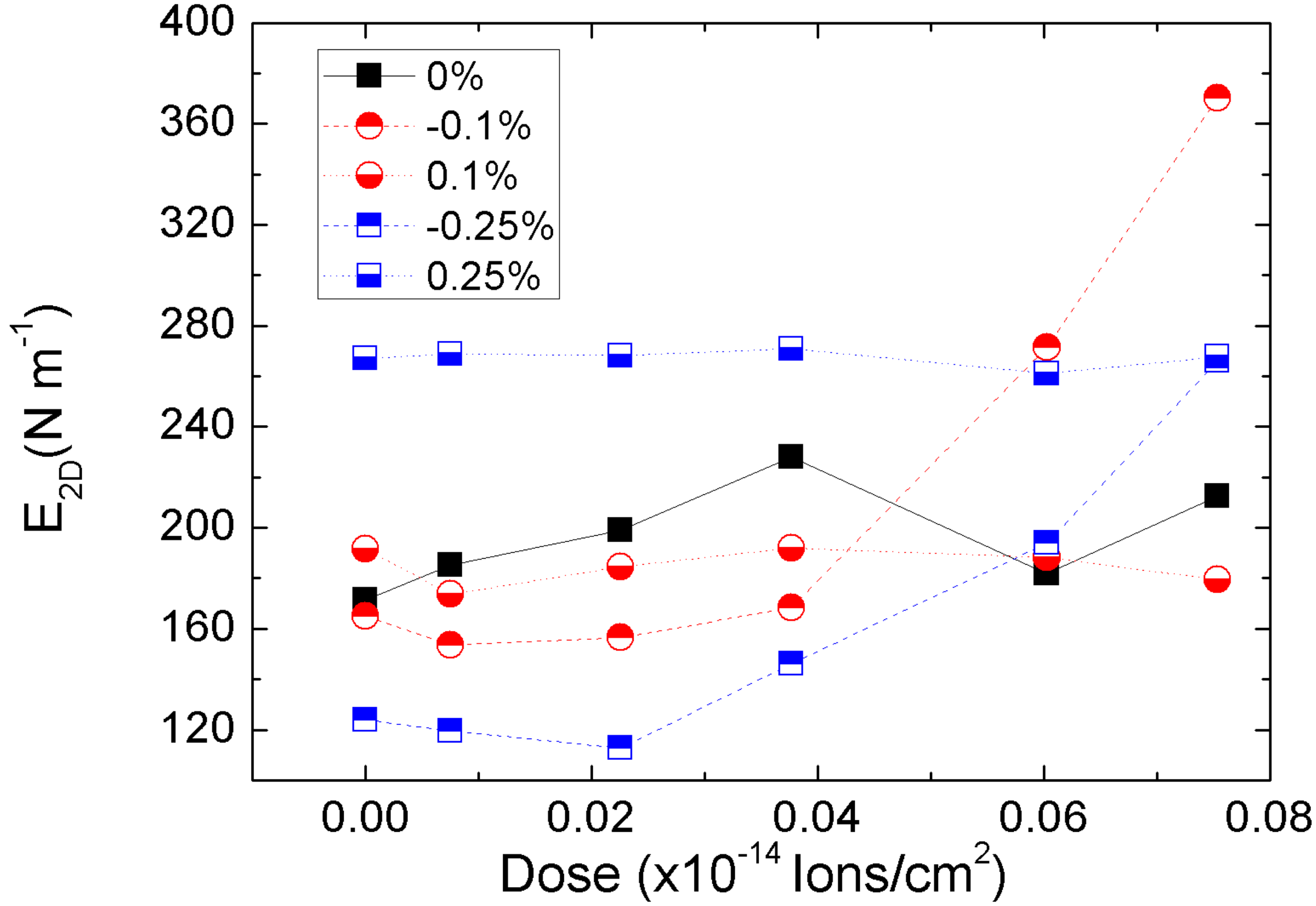
Small changes in roughness under tensile strain but increases under compression

Softening of the membrane under compression and stiffening under tension. In agreement with Lee, Nanoscale Res. Lett., 2015, 10, 422

J. Martinez-Asencio, et. al, Phys. Chem. Chem. Phys, 2016

Results:
irradiated graphene – E2D as a function of dose

Changes due to the irradiation, T = 300K

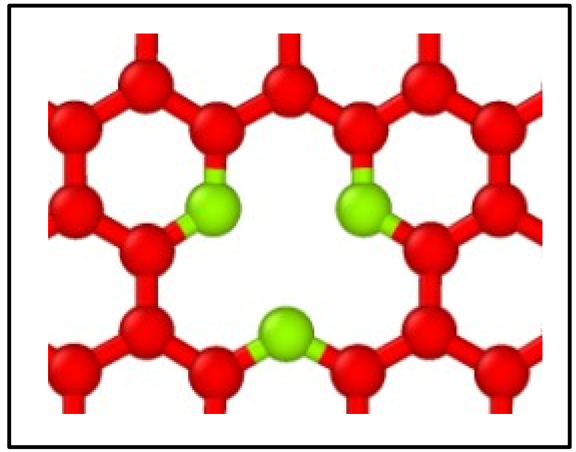


J. Martinez-Asencio, et. al, Phys. Chem. Chem. Phys, 2016

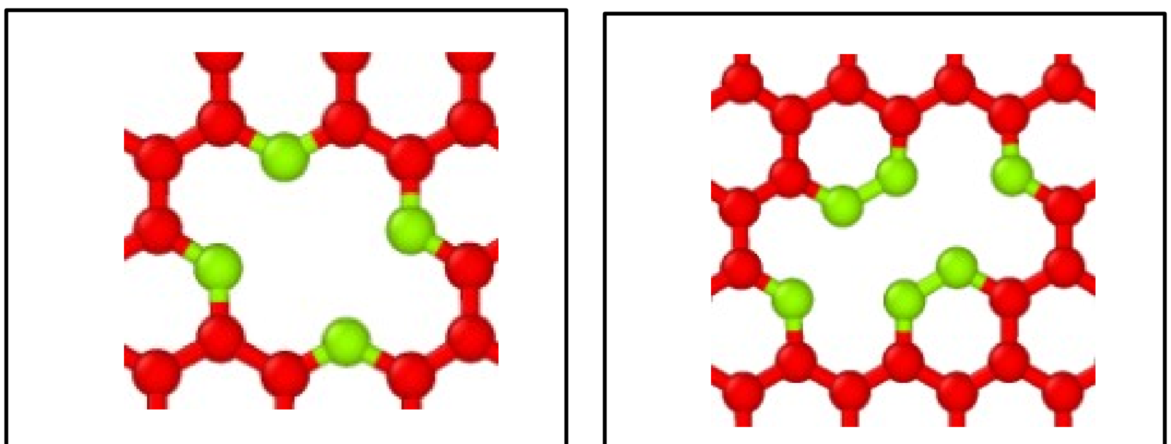
E2D constant for tensile strain and increases non-linearly with dose for compressive strains

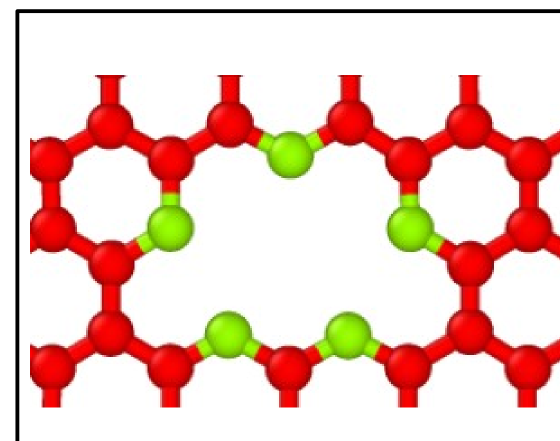
Results: irradiation → Types of defects produced

Mostly mono-vacancies



Single vacancy





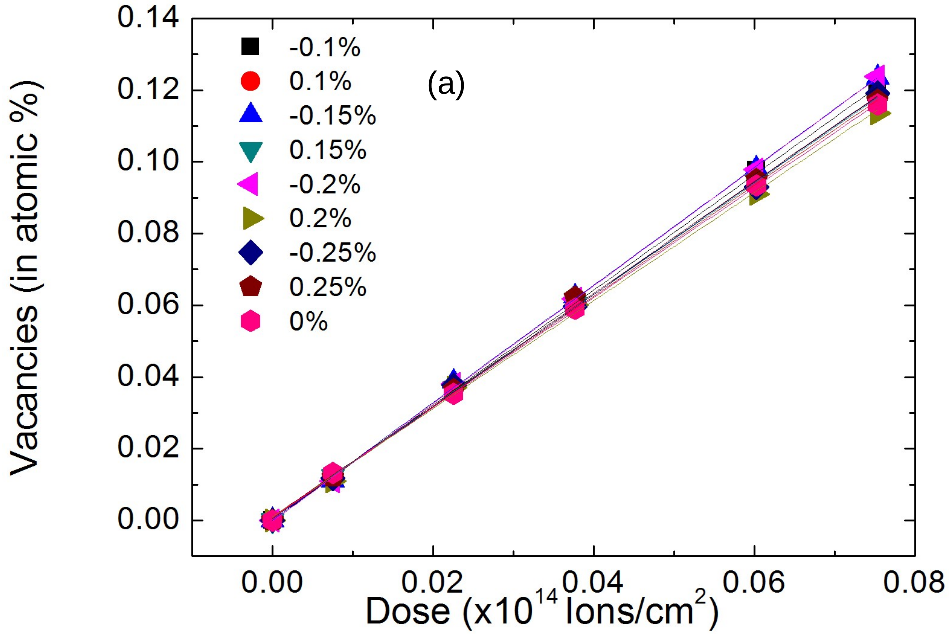
Di-vacancies

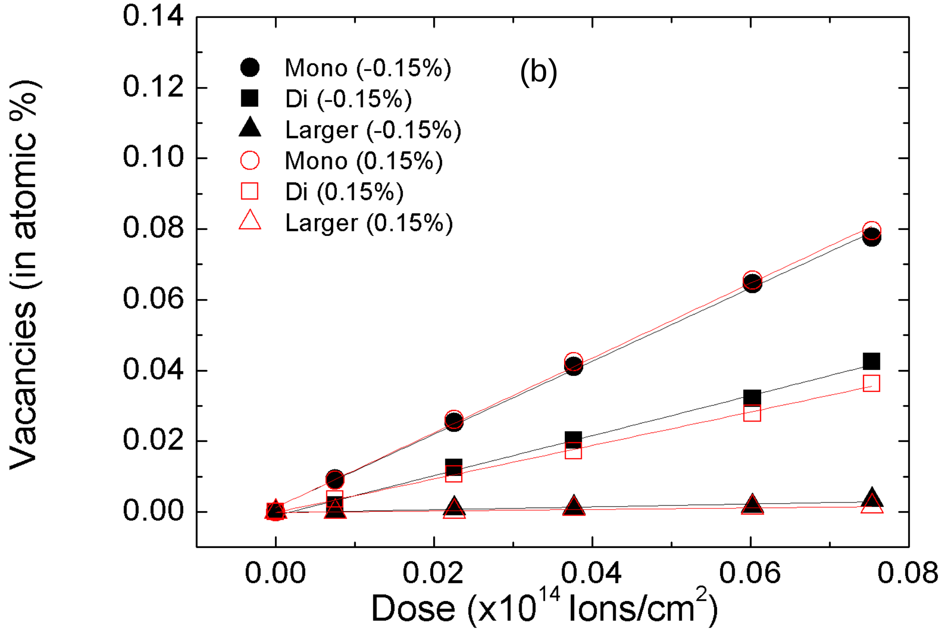
Triple-vacancy

J. Martinez-Asencio & M. J. Caturla, NIMB 2015, 352, 225
J. Martinez-Asencio, et. al, NIM B, under review

Results: defect production

Defects as a function of dose for different initial strains





Total number of defects increases linearly with dose and it is very similar for all initial strains: higher defect production for compressed samples

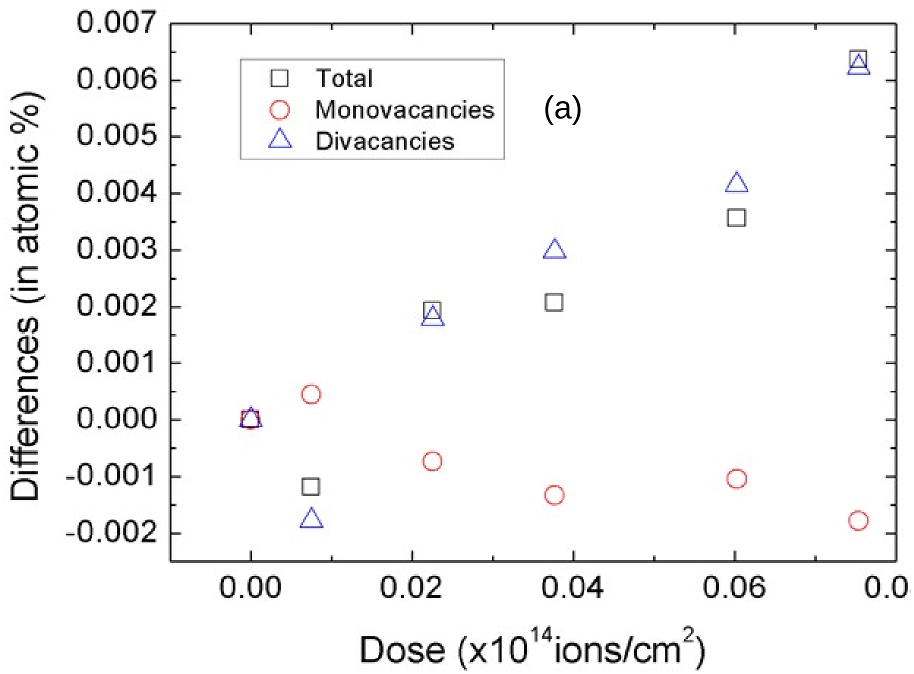
Higher concentration of di-vacancies for compressed samples

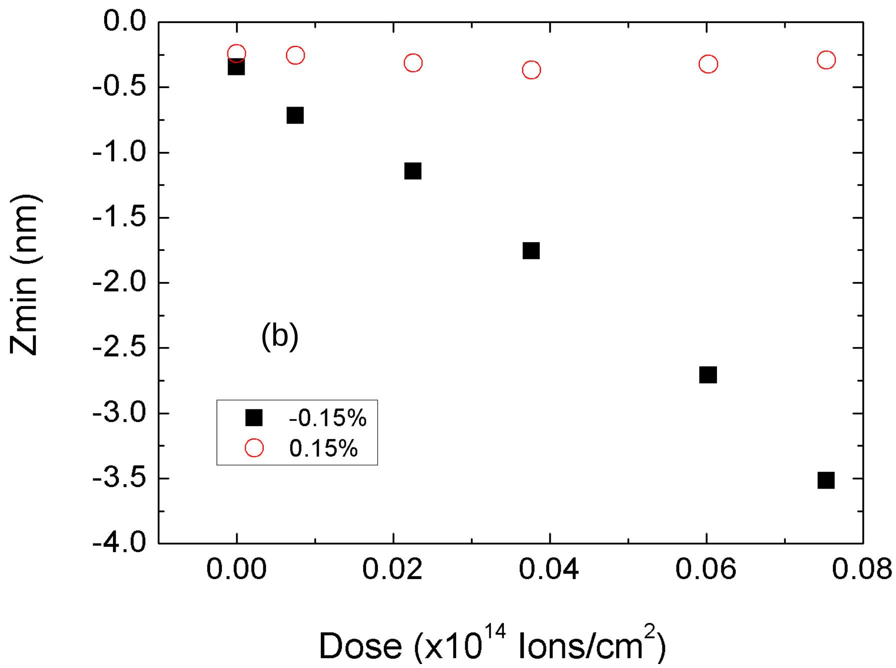
No correlation to E2D dependence with dose!!

Results: defect production

Effect of strain: -0.15% vs. 0.15% at T = 300K

$\Delta N = N(-0.15\%) - N(0.15\%)$





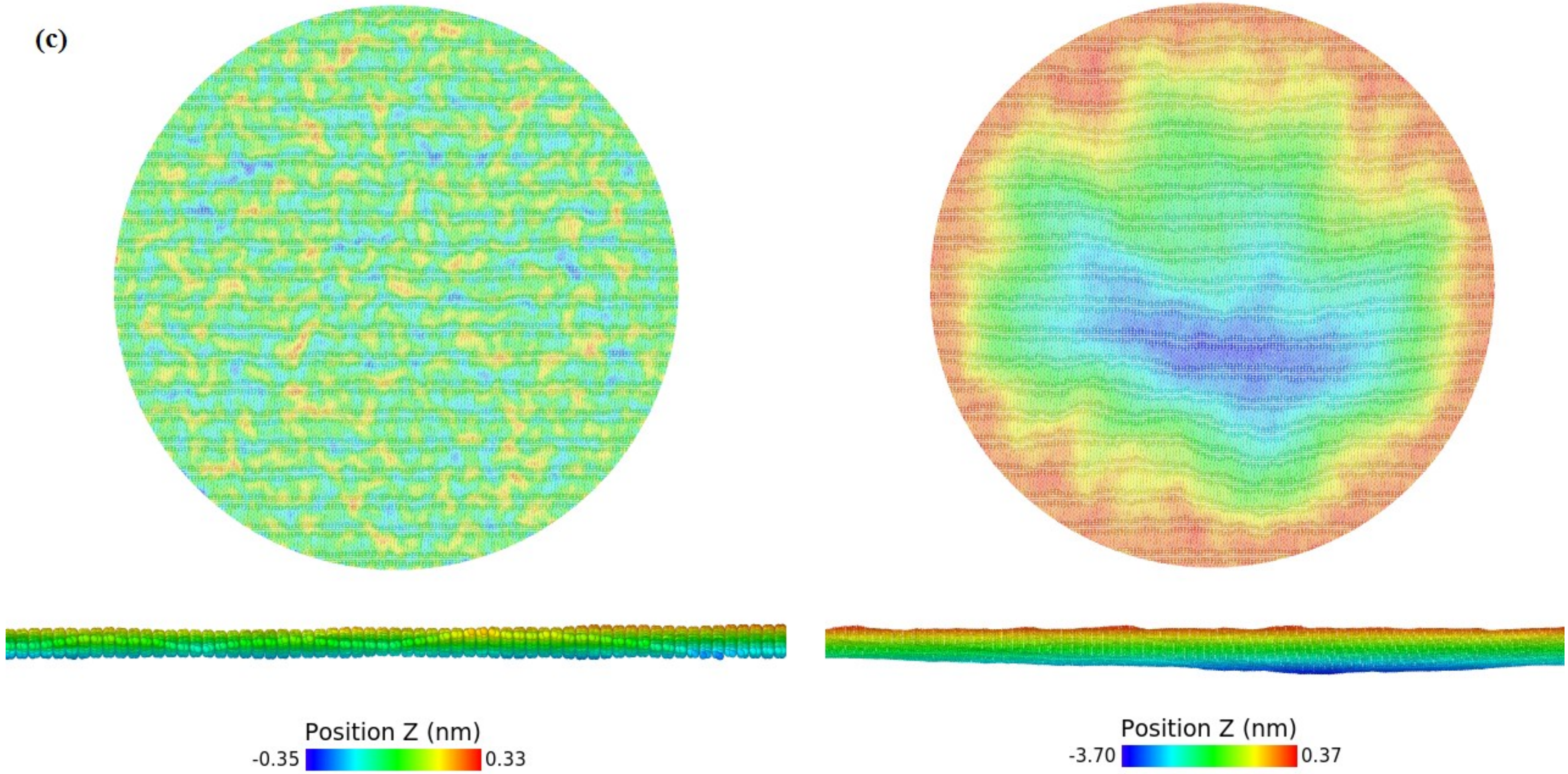
For compressed samples: number of di-vacancies increases with dose and mono-vacancies decreases

Large out-of-plane displacements for compressed samples

J. Martinez-Asencio, et. al, NIM B, under review

Results: out-of-plane displacements before and after irradiation

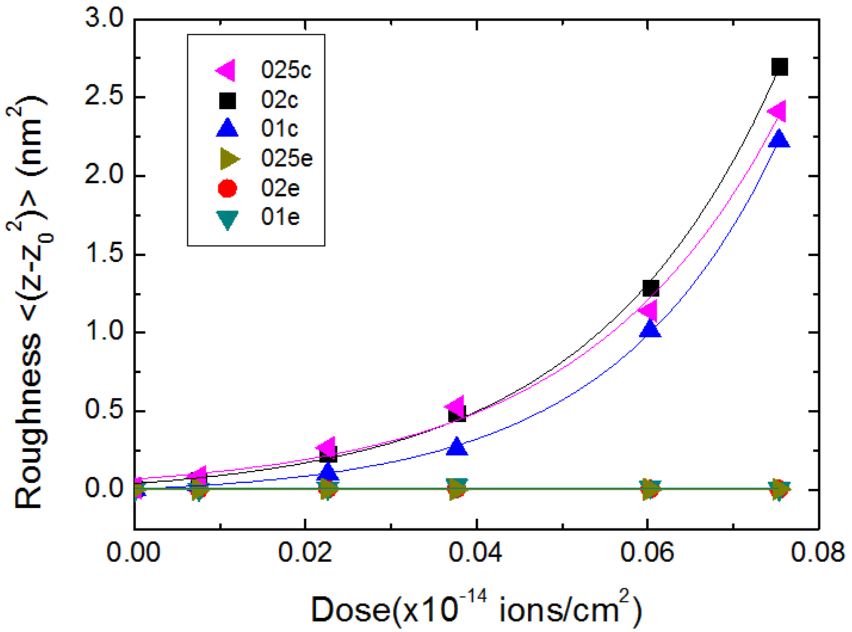
T = 300K   Initial strain: -0.15% (compression)

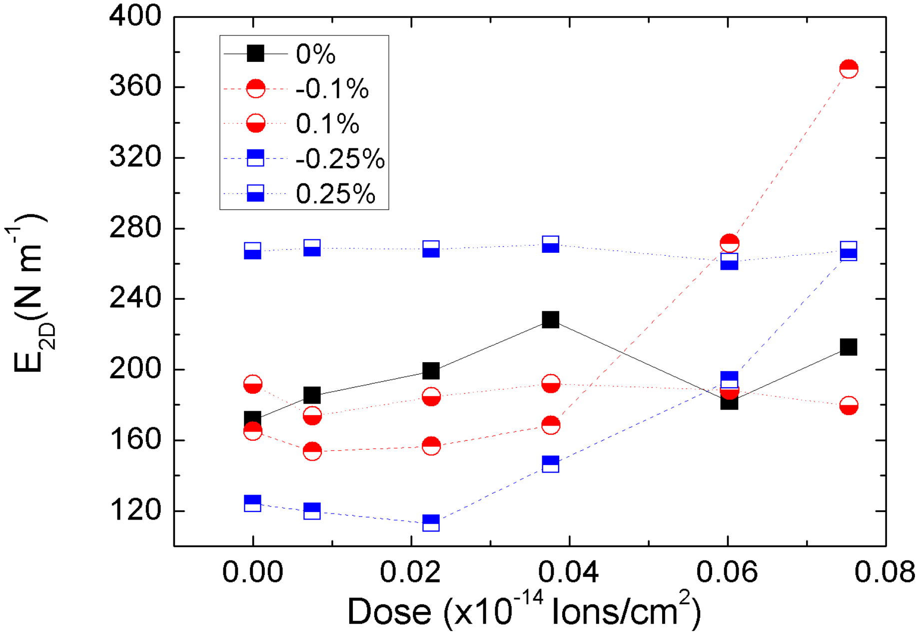


Ripples initially transform into a deep well after irradiation

Results:
unirradiated graphene – effect of applied strain

Changes due to the irradiation, T = 300K





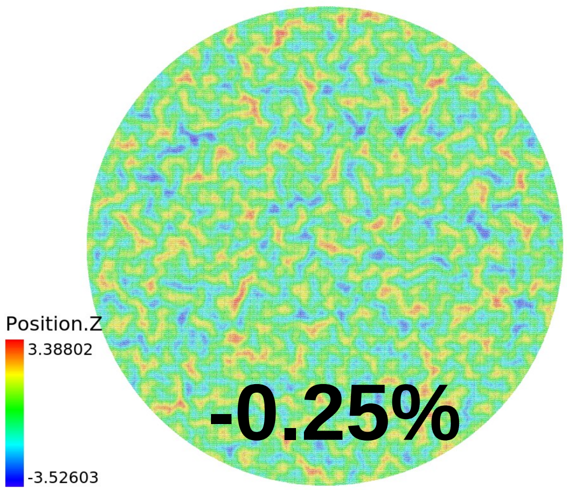
Under tensile strain no changes in roughness, but large changes under compression that increase non-linearly with dose

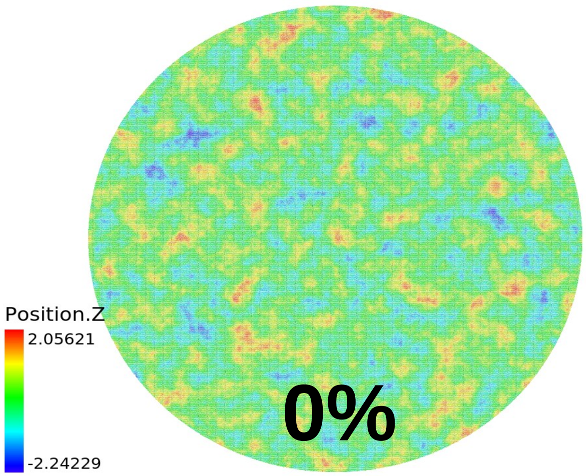
Similar trends as those observed in the E2D dependence with dose

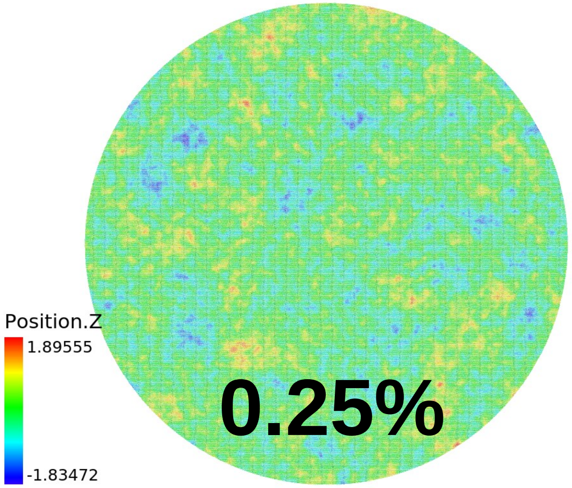
J. Martinez-Asencio, et. al, Phys. Chem. Chem. Phys, 2016

Results:
changes in roughness with irradiation and strain

- **0 ions**

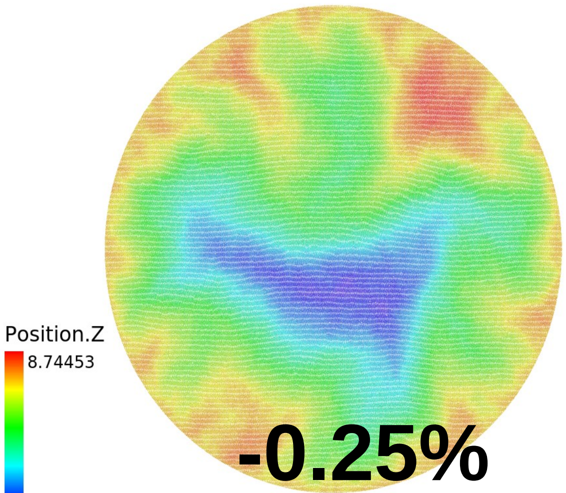


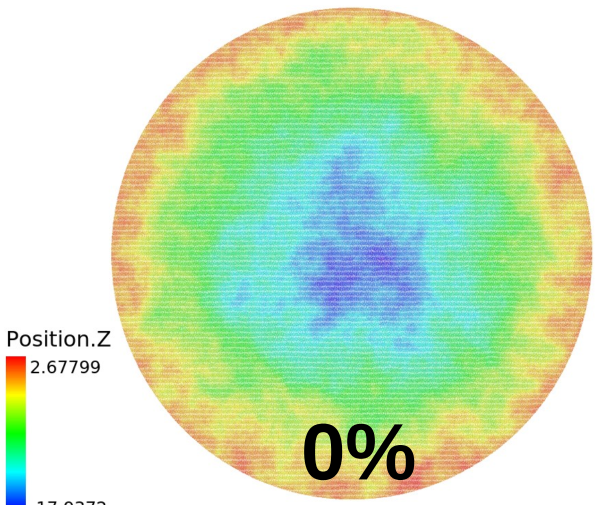


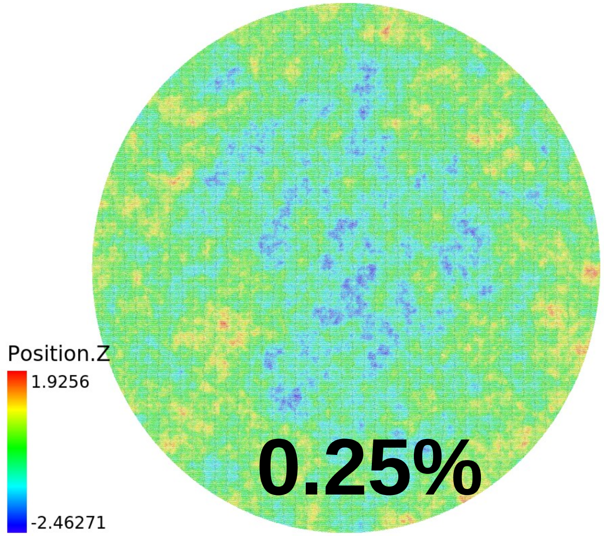


- **1000 ions**

Images obtained with OVITO

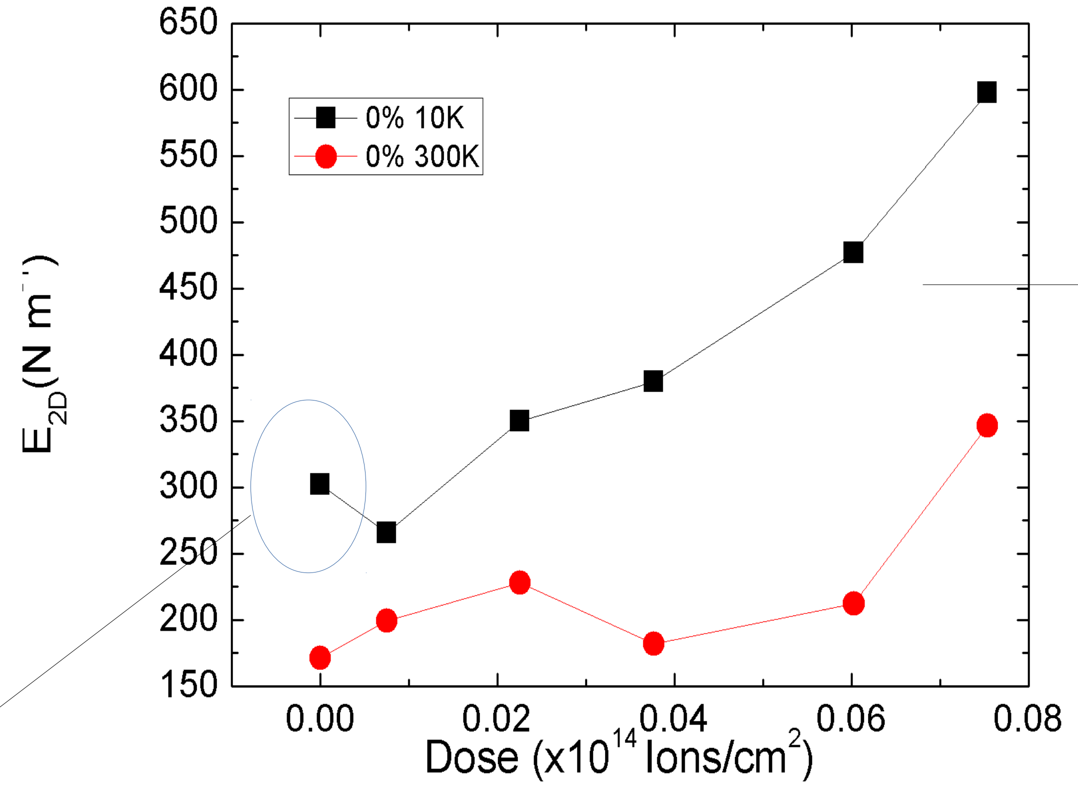






Simulation set up and requirements
Temperature effects

T = 300K vs. T = 10K without initially applied strain



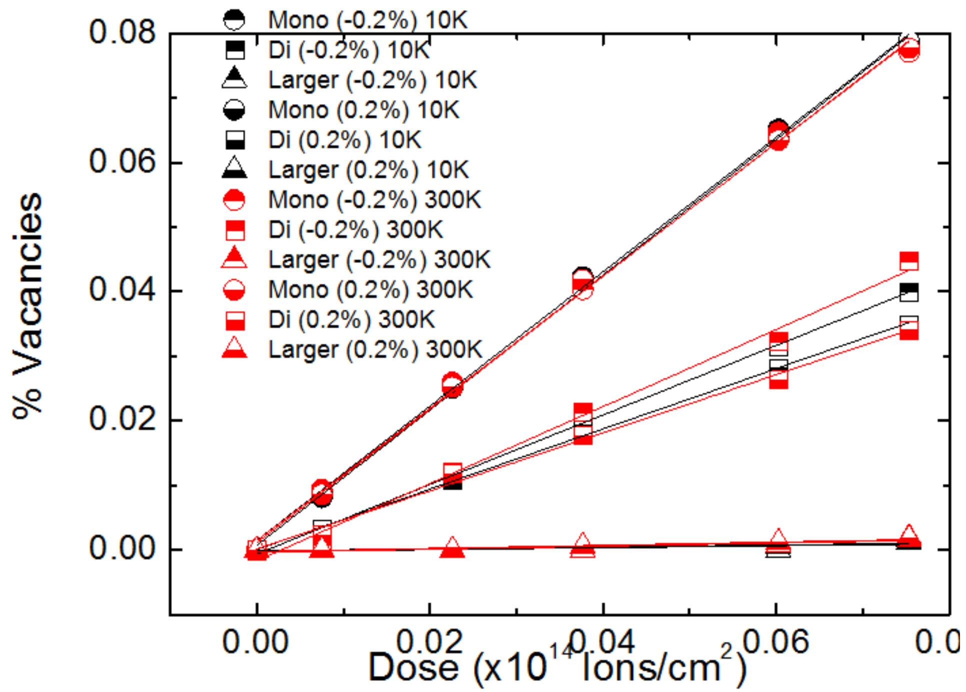
E2D increases with dose to values up to 600N/m, similar to experiments

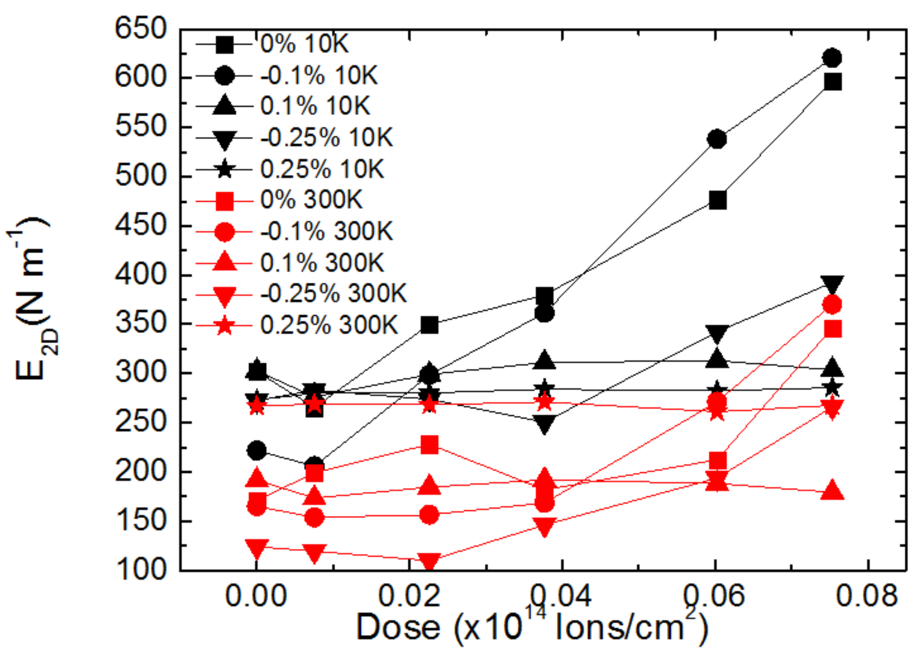
Initial value, before irradiation, closer to experimental values

Are corrugations due to temperature overestimated?

Results: effect of temperature

Two temperatures: 300K and 10 K





No significant changes in the number of defects produced at lower temperature

Similar trends for the E2D but starting values are higher, as expected, at lower temperature (quenching of ripples) and closer to experimental values at RT

Conclusions

- Irradiation with 140eV Ar ions produces mono-vacancies and a very small fraction of di-vacancies and triple-vacancies.
- Defect production increases linearly with dose (up to the doses studied here) independently of temperature or applied strain.
- E2D remains almost constant for samples under tension but increases non-linearly with dose with samples under compression. This effect dose not correlate with defect production.
- Roughness of the sample changes for tensile and compressive membranes and follow the trends observed for E2D.

Defect concentration alone can not explain the changes in mechanical properties of irradiated graphene

Future Work

- What happens at higher doses (defect overlap)?
- Why are the E2D values calculated at 300K too low compared to experimental and DFT calculations?
- Can we combine strain, irradiation and temperature to 'engineer' ripples in graphene?
- Other 2D materials

Acknowledgements

- Generalitat Valenciana PROMETEO2012/011
- Spanish Government FIS2010-21883
- Barcelona Supercomputing Center