# Machine Learning prediction of the structural/textural-mechanical properties relationship of pyrolitic carbons

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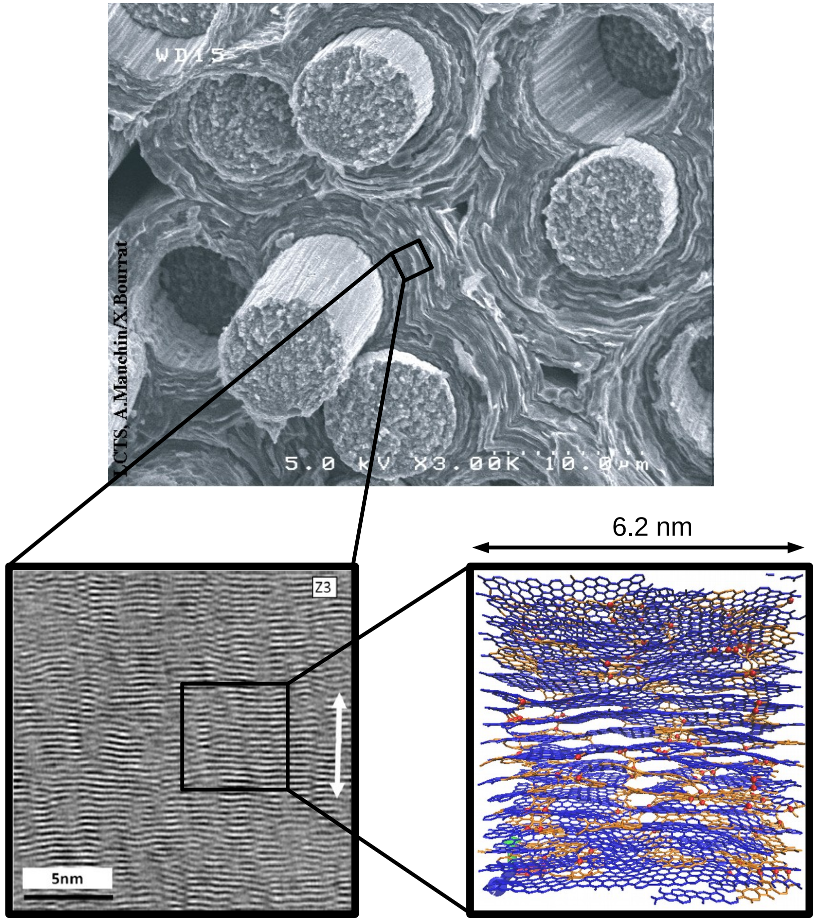
# Context / High textured pyrocarbons (pyCs)

- PyCs are carbon-based materials prepared by Chemical Vapor Infiltration (CVI)
- PyCs constitute interphases or matrices parts of elements




HRTEM image | Atomistic reconstruction

**[Weisbercker et al. Carbon 2012]** | **[Farbos et al. Carbon 2014]**

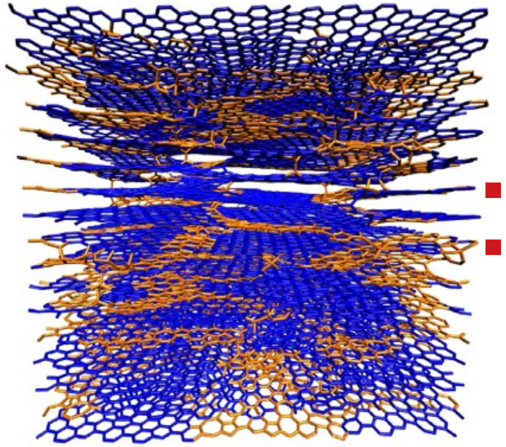
## High performance C/C composites applications

- Thermal protection systems
- Plasma-facing components
- ARIANE V boosters
- Conditions : very high temperature
- Difficulty for small-scale experiments

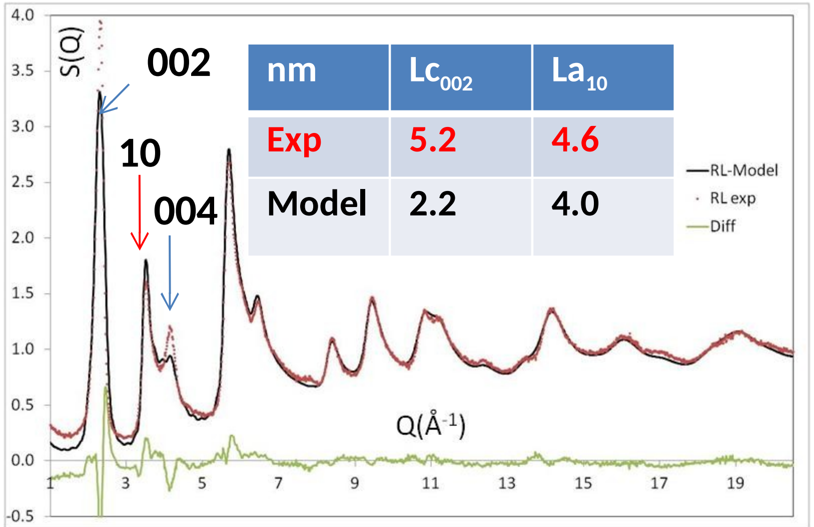



# Context / Results obtained with IGAR

**_Rough Laminar Pyc_**

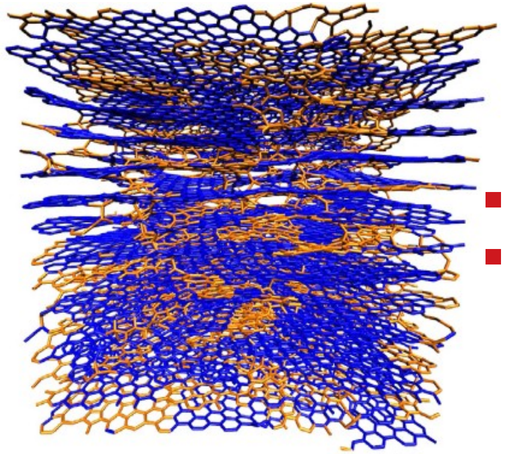


- Highly textured PyCs
- Low content of defects

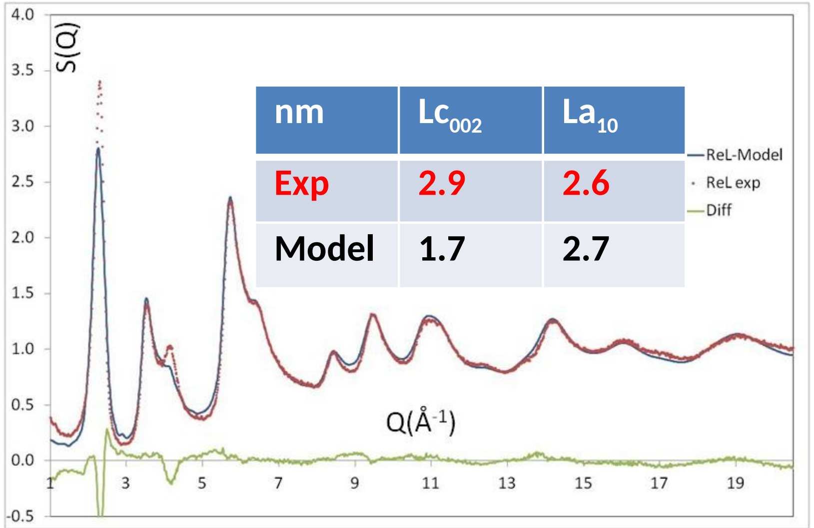



| nm | $Lc_{002}$ | $La_{10}$ |
|---|---|---|
| Exp | 5.2 | 4.6 |
| Model | 2.2 | 4.0 |

**_Regenerative Laminar Pyc_**



- Highly textured PyCs
- High content of defects




| nm | $Lc_{002}$ | $La_{10}$ |
|---|---|---|
| Exp | 2.9 | 2.6 |
| Model | 1.7 | 2.7 |

**[Leyssale et *al*. Carbon (2012)]**

✔ Very good in-plane agreement for $L_a$
✖ Graphene layers stacking underestimated

## Aim of the PhD thesis

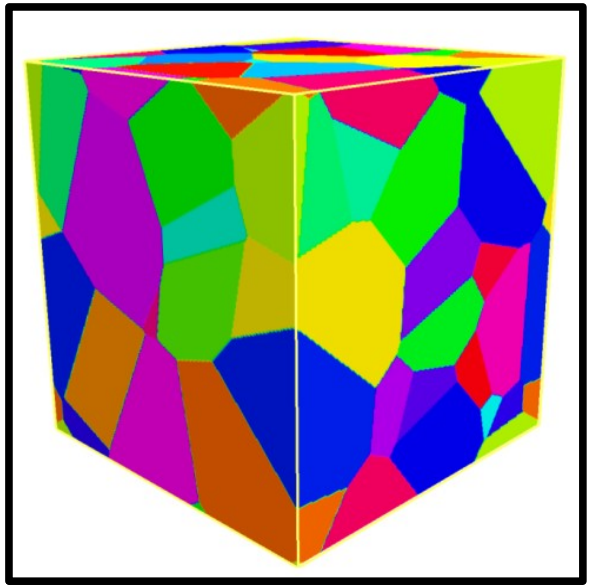
- Improve the numerical synthesis methodology and propose evolutions
- Build a large database of microstructures
- Compute structural and textural properties of reconstructed PyCs
- **Compute elastic constants of reconstructed PyCs**
- **Structure-properties relationship (ML)**
- Start thinking about the setup of a constitutive law at the mesoscopic scale

## Numerical aspects

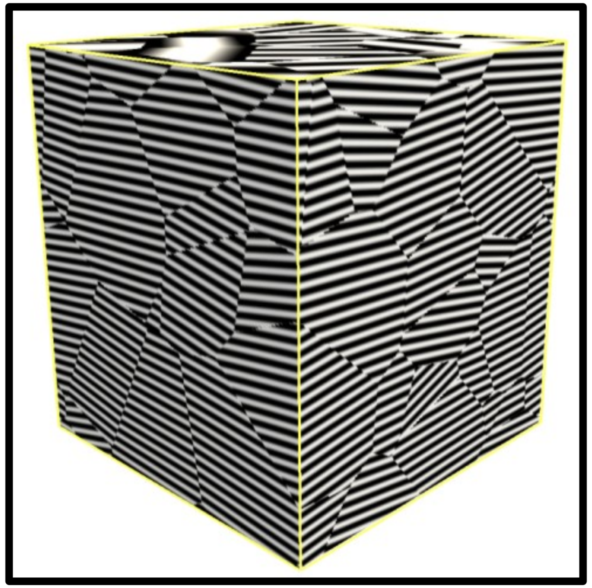
- Implement the reconstruction methods in CEA MD codes
- Setup the full analysis chain of structural, textural and elastic properties at very high temperature
- Identify structural/textural/elastic properties relationships
- Bridge atomic and continuum scales

# This work : Polygranular image guided reconstruction

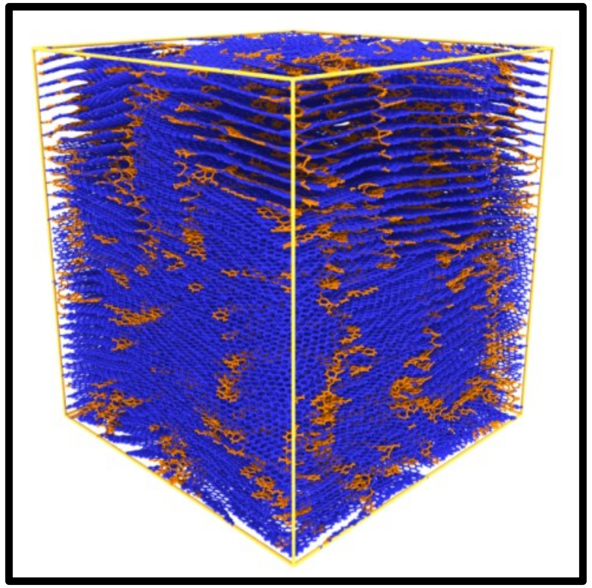
**Anisotropic Voronoï tesselation**



**Decorated with straight fringes**



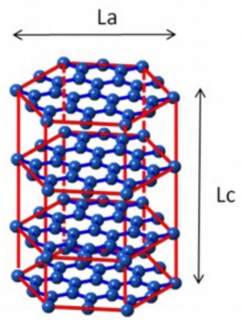
**Atomistic model after IGAR quench**



Quench (REBO[1]), Relaxation (LCBOPII[2])

[[1]Brenner et al. J. Phys. Condens. Matter 2002; [2]Los et al. Phys. Rev. B. 2005]

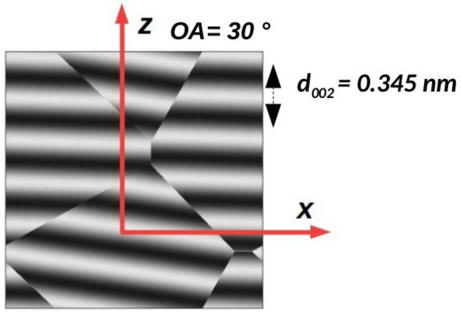
Based on crystallographic parameters

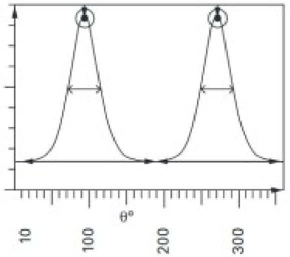



$$N_G = V / V_G$$

$$V_G = \frac{\pi}{4} L_a^2 L_c$$

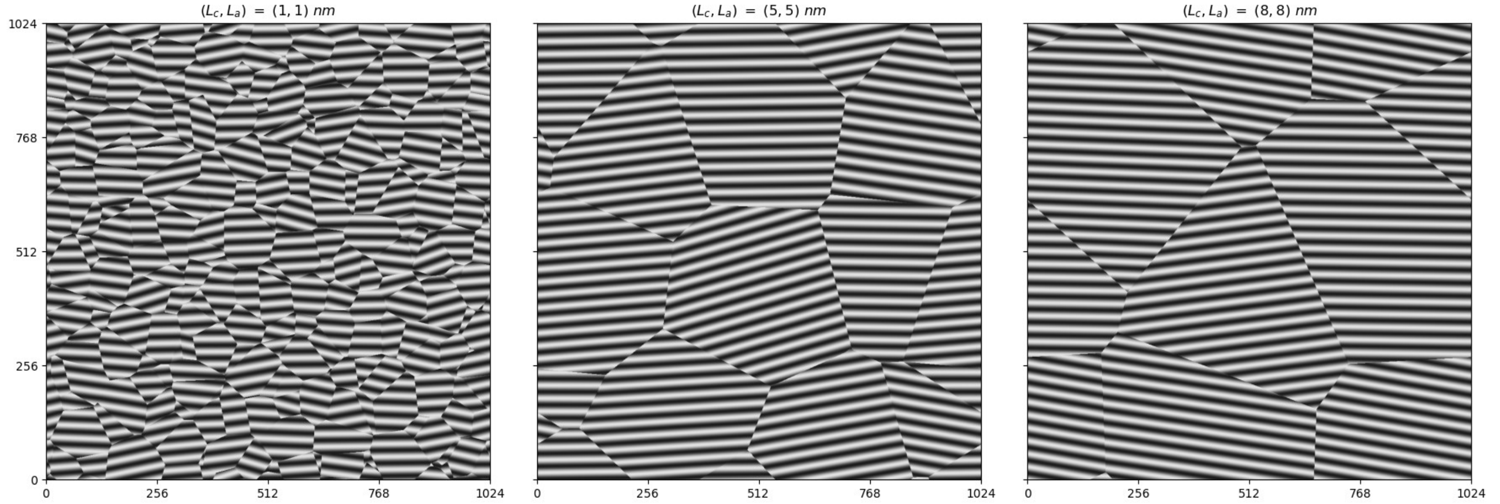
$$r_a = L_c / L_a$$



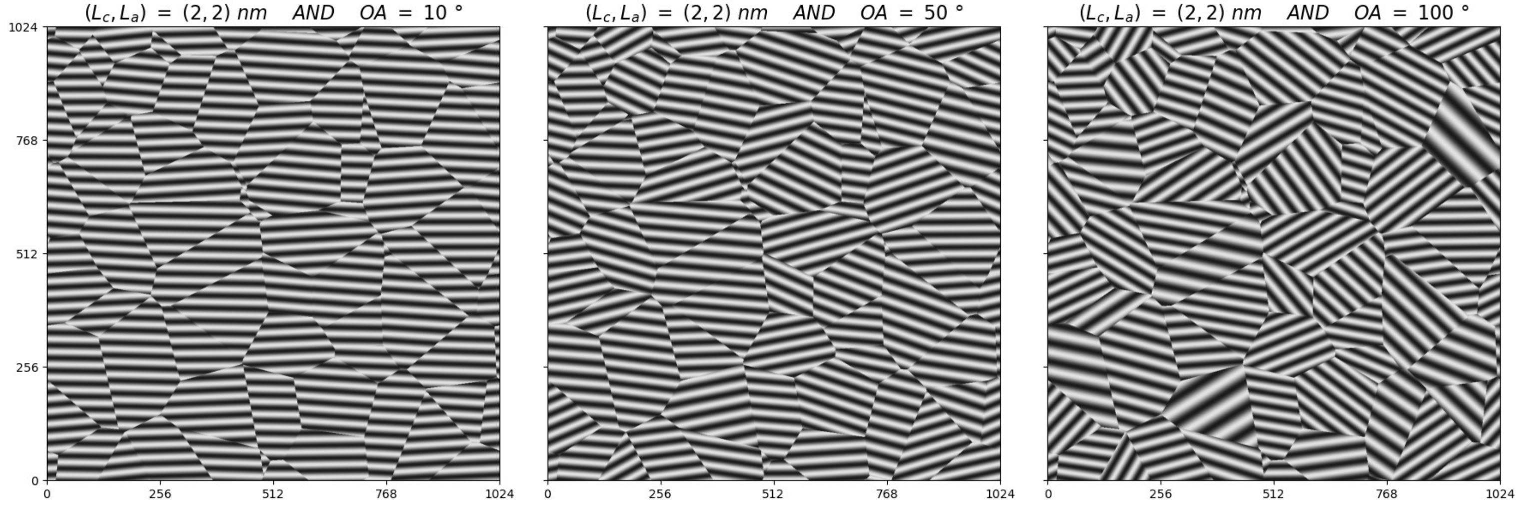



# Parametric approach - setup

Variation of $L_a$ ( = $L_c$ ) from 1 nm to 8 nm ; OA = 30°



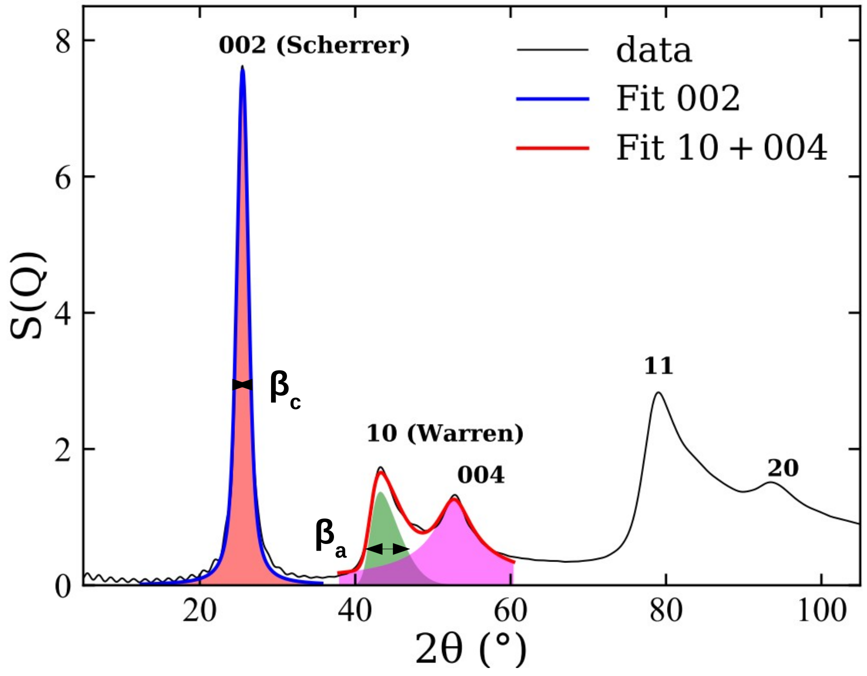

Variation of OA from 10° to 100° ; $L_a$ ( = $L_c$ ) = 2, 4 & 6 nm




- Cell length = 12.4 nm
- ρ = 2.16 g.cm$^{-3}$
- 5 different quench times (0.7, 1.5, 4.0, 4.4 & 8.4 ns)

## Calculation of $L_a$, $L_c$

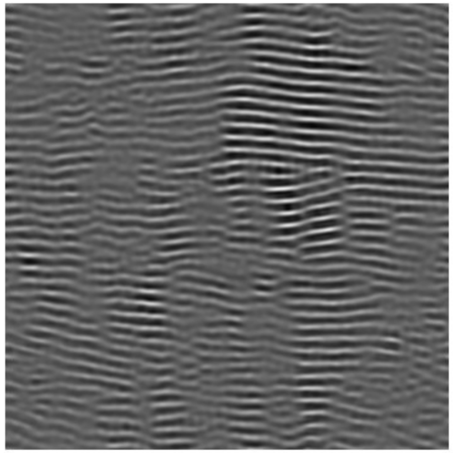
$$S(Q) = 1 + \frac{1}{Q}\int_{O}^{r_{max}} G(r)\sin(Qr)dr$$




$$L_{a,c} = K_{a,c}\,\lambda / \beta_{a,c}\cos(\theta)$$

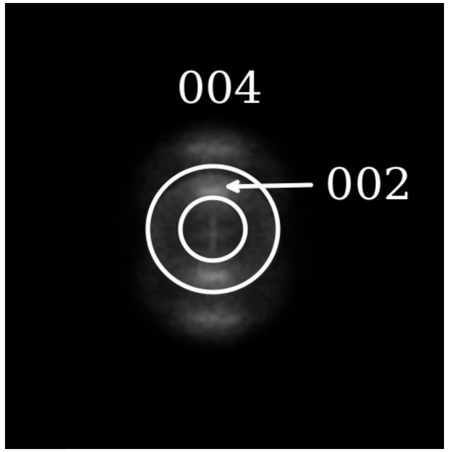
$K_a$ = 1.7 and $K_c$ = 1

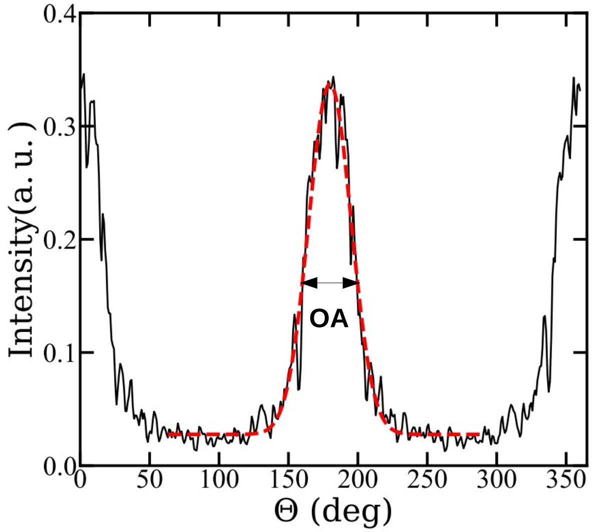
## Calculation of OA



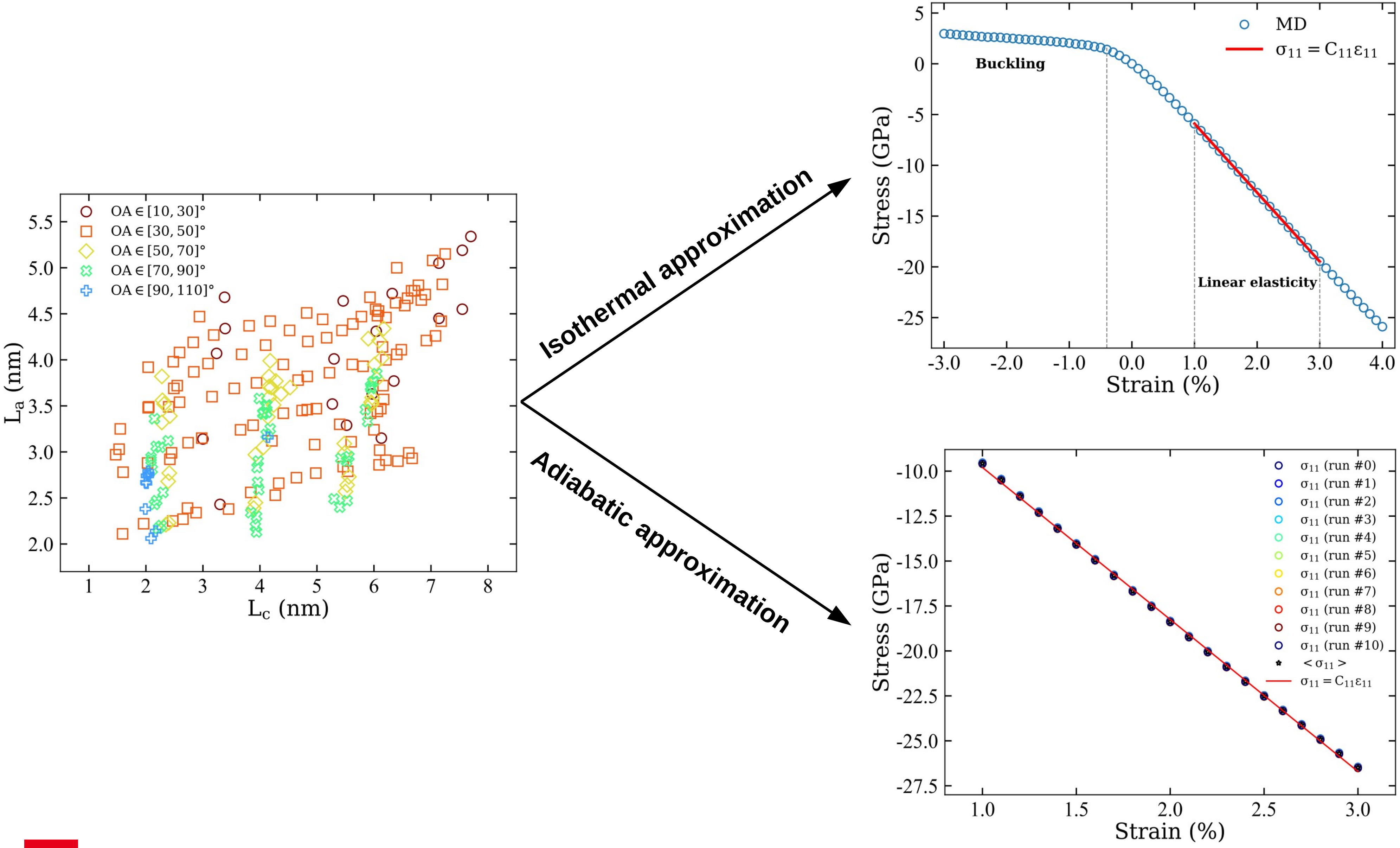
[Dr Probe, J. Barthel. Ultramicroscp. 2018, 193, 1-11]

FFT



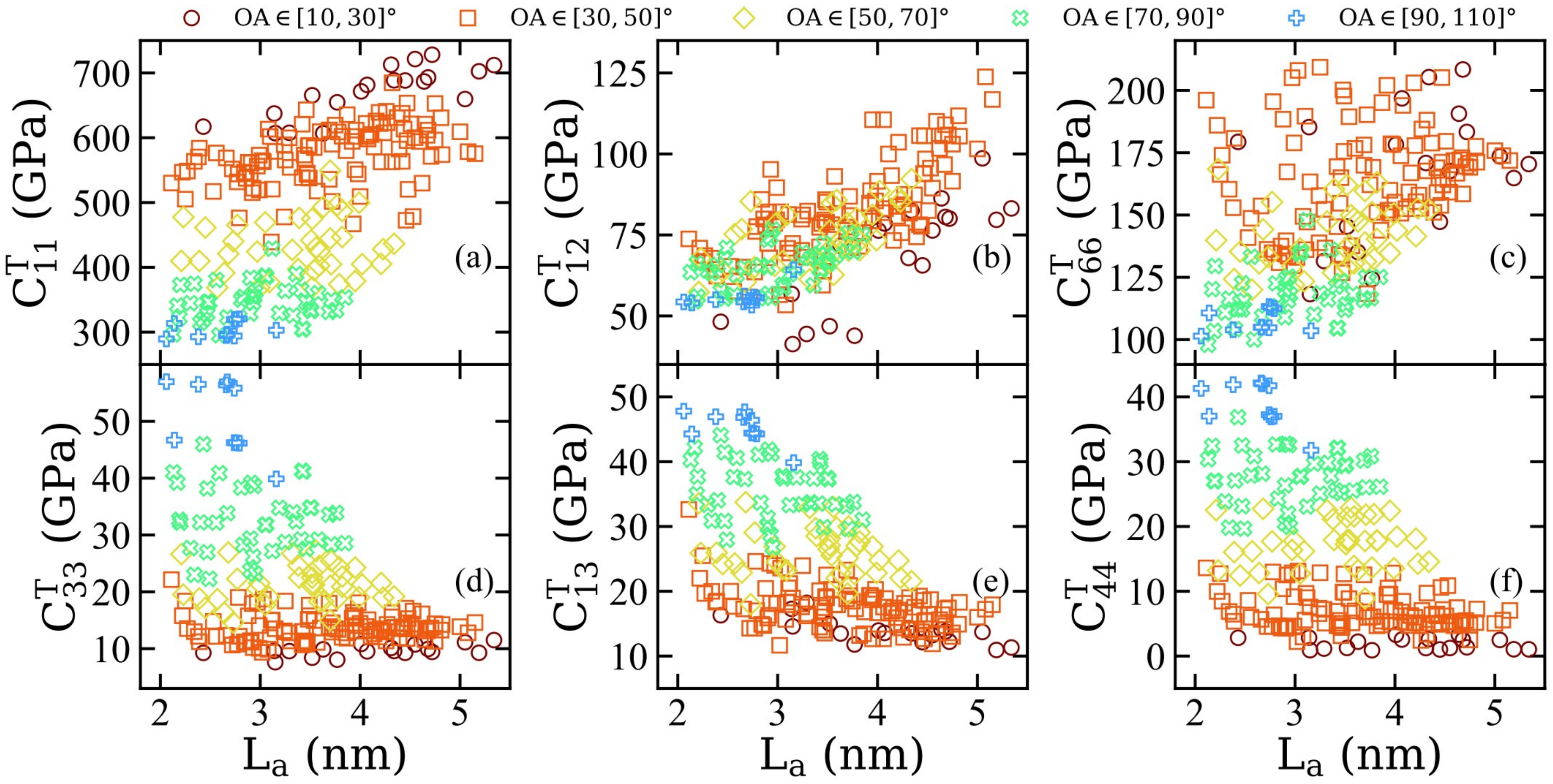
# Presentation of the database & elastic properties setup




- NVT trajectories during 100 ps for each $\varepsilon_{ij}$
- Averaging stress $\sigma_{ij}$ over the last 20 ps
- Linear fit of the stress vs strain curve

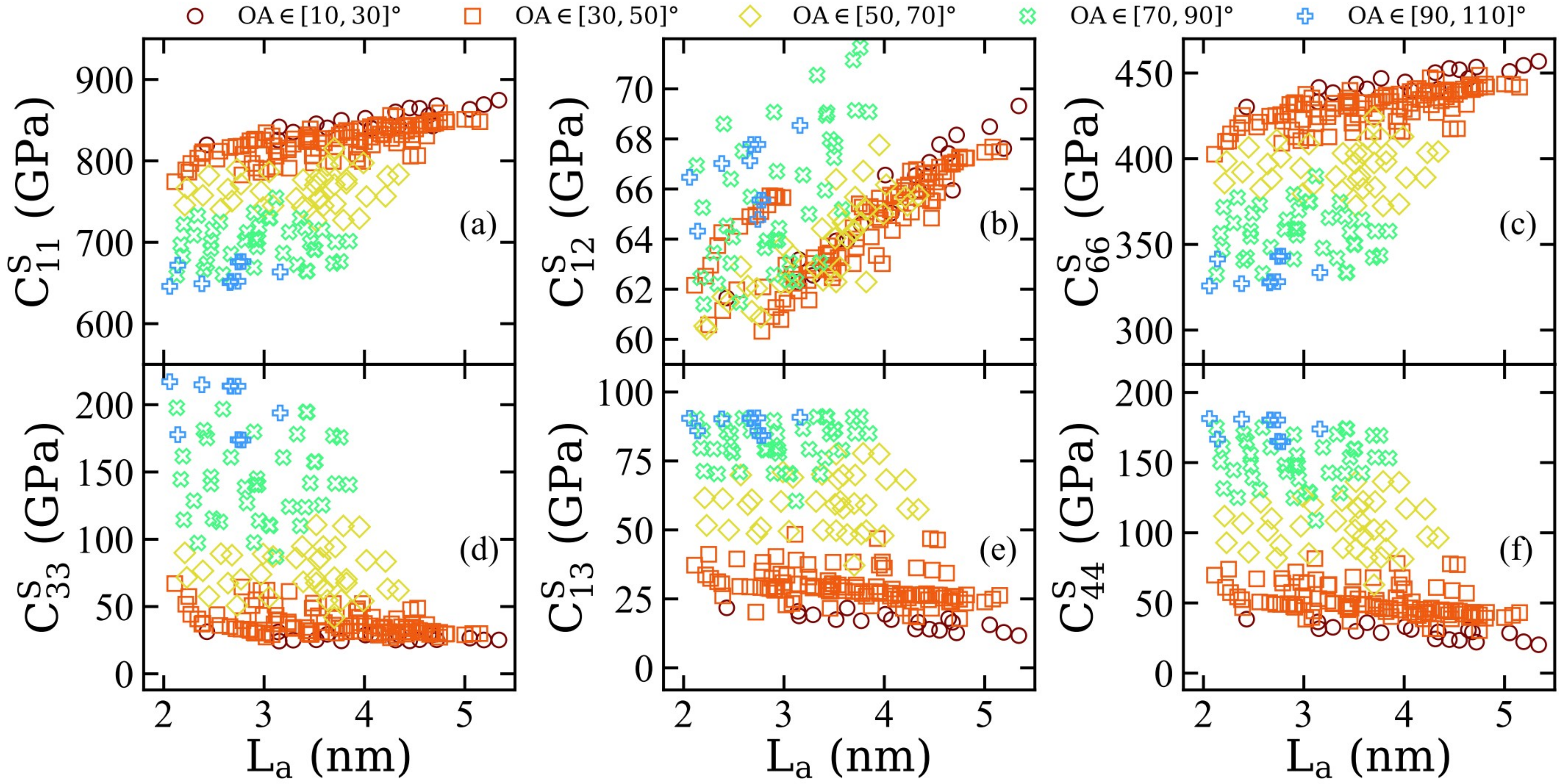
- NVT trajectory during 100 ps at $\varepsilon_{ij}$ = 0 %
- 10 independent configurations selected from the last 20 ps
- Single step calculation for each $\varepsilon_{ij}$
- Linear fit of the average stress vs strain curve

# Isothermal elastic properties



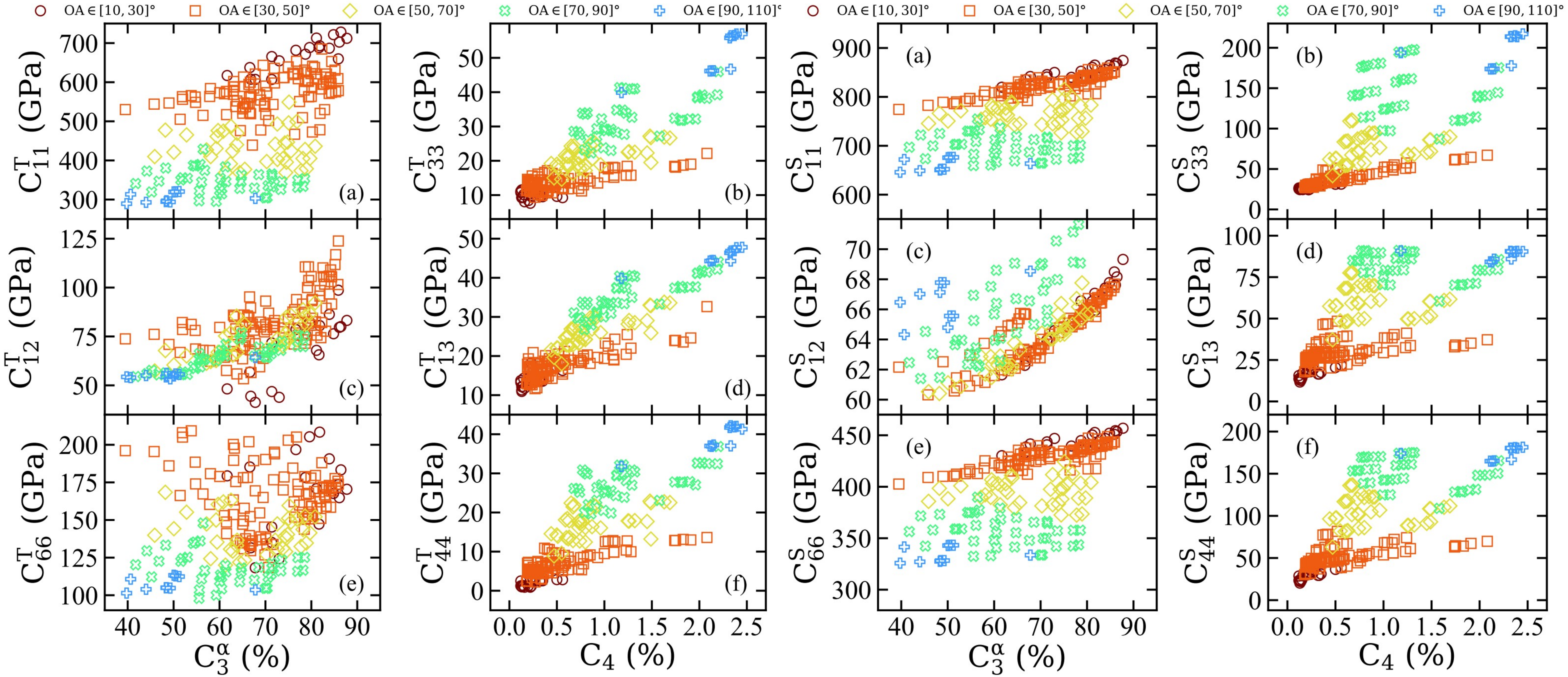

- Low OA :
  High in-plane stiffness & Low transverse properties
- Medium OA :
  Small evolution with $L_a$
- High OA :
  Low in-plane stiffness & High transverse properties

# Adiabatic elastic properties




- Same trend than isothermal
- $C_{ij}$ more higher

# Correlation with atomic-scale environment



# Structure/properties relationship using Supervised Learning Model

**Dataset :** 210 models

**Target :** Elastic constants $C_{ij}$

**Features :** $L_c$, $L_a$ & OA

**Train set & Test set :** 80 % & 20 %

**Model :** Random Forest Regressor[1] (RFR)
[[1]L. Breiman, Random forests, Machine Learning 45 (2001)]

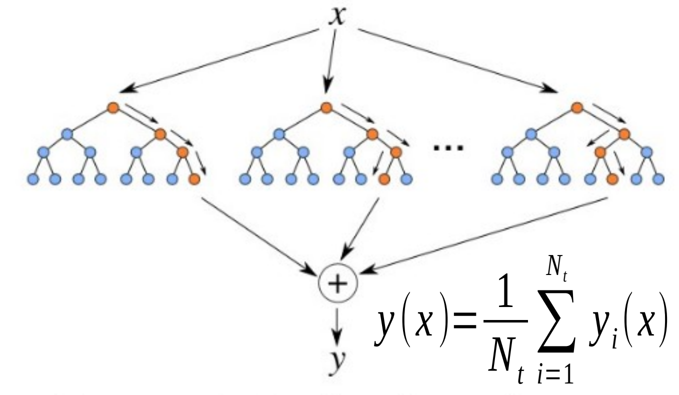
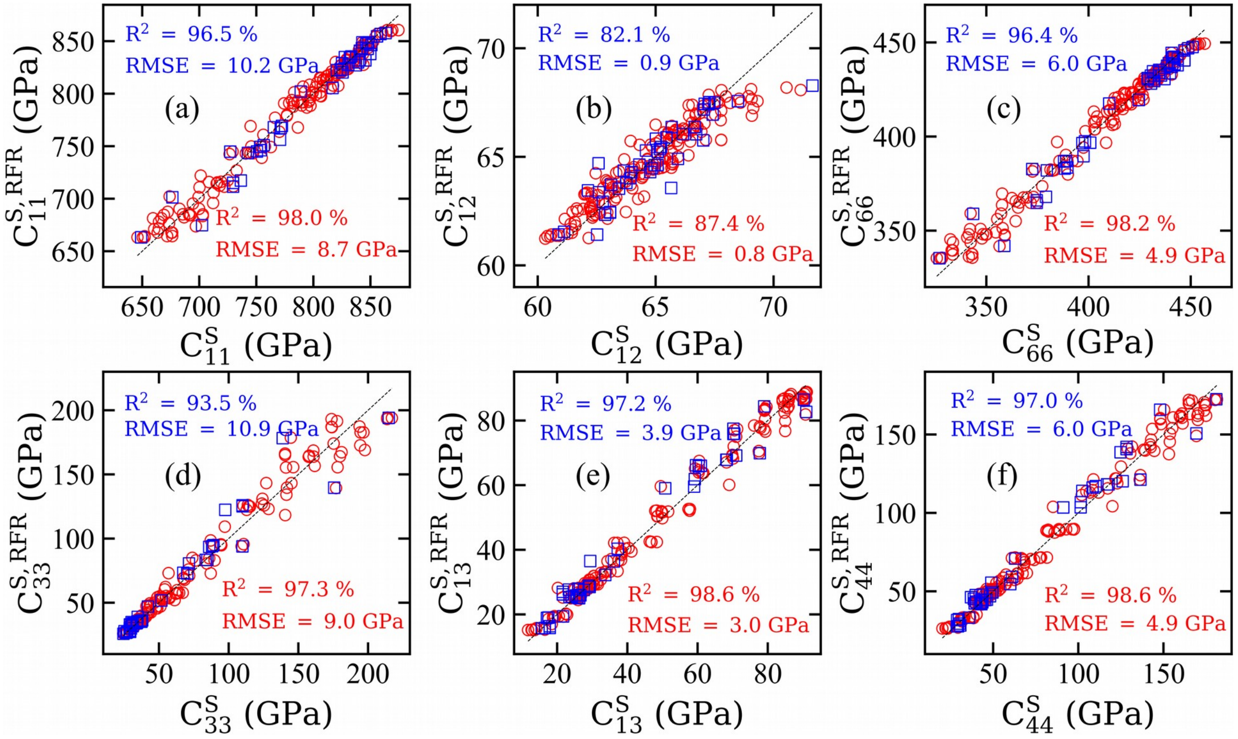
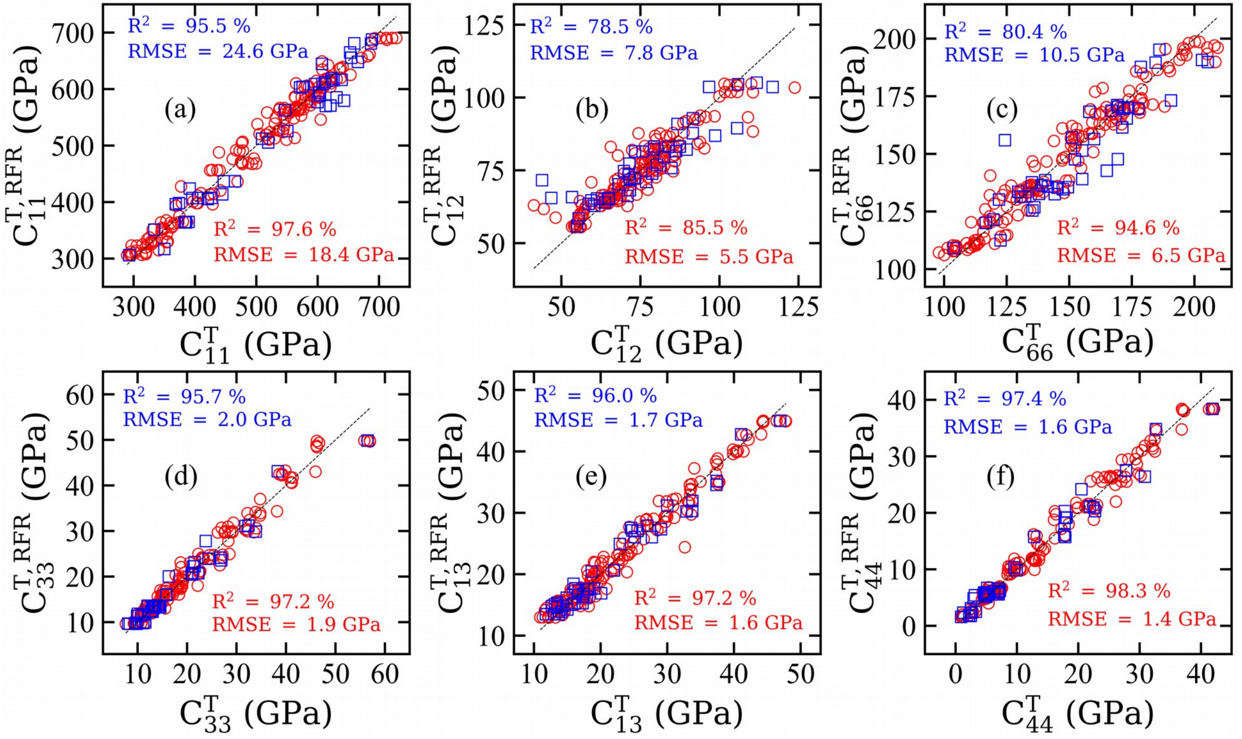



*Diagram of a Random Forest Regressor*

- $N_t = 100$
- $D_t = 15$
- $N^{samples} = 5$

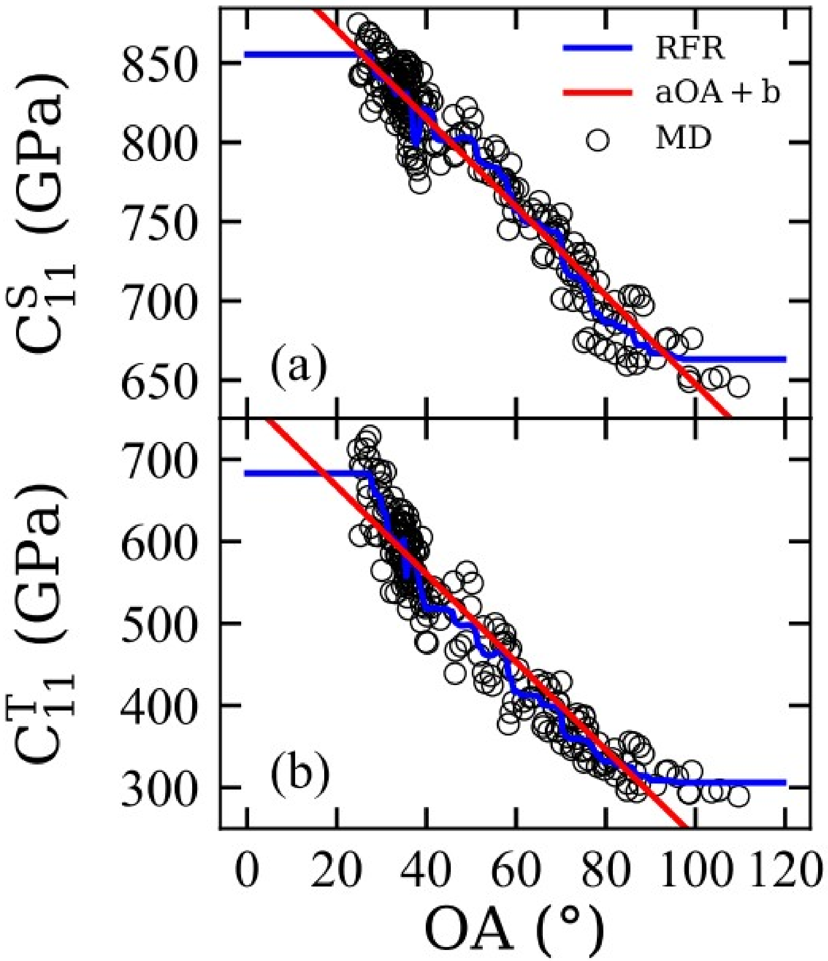


# Features importance of the RFR model

- Inputs : fitted predictive model ***m*** & tabular dataset (train or test set) **D**
- Compute the reference score ***s*** of the model on data **D** ($R^2$)
- For each feature ***j*** (column of **D**) :
  - For each repetition ***k*** in 1, …, K :
    - Randomly shuffle column of dataset ***D*** to generate a corrupted version of the data named $D_{k,j}'$
    - Compute the score $s_{k,j}$ of model ***m*** on corrupted data $D_{k,j}'$
  - Compute importance $i_j$ for feature $f_j$ defined as :

$$i_j = s - \frac{1}{K}\sum_{k=1}^{K} s_{k,j}$$

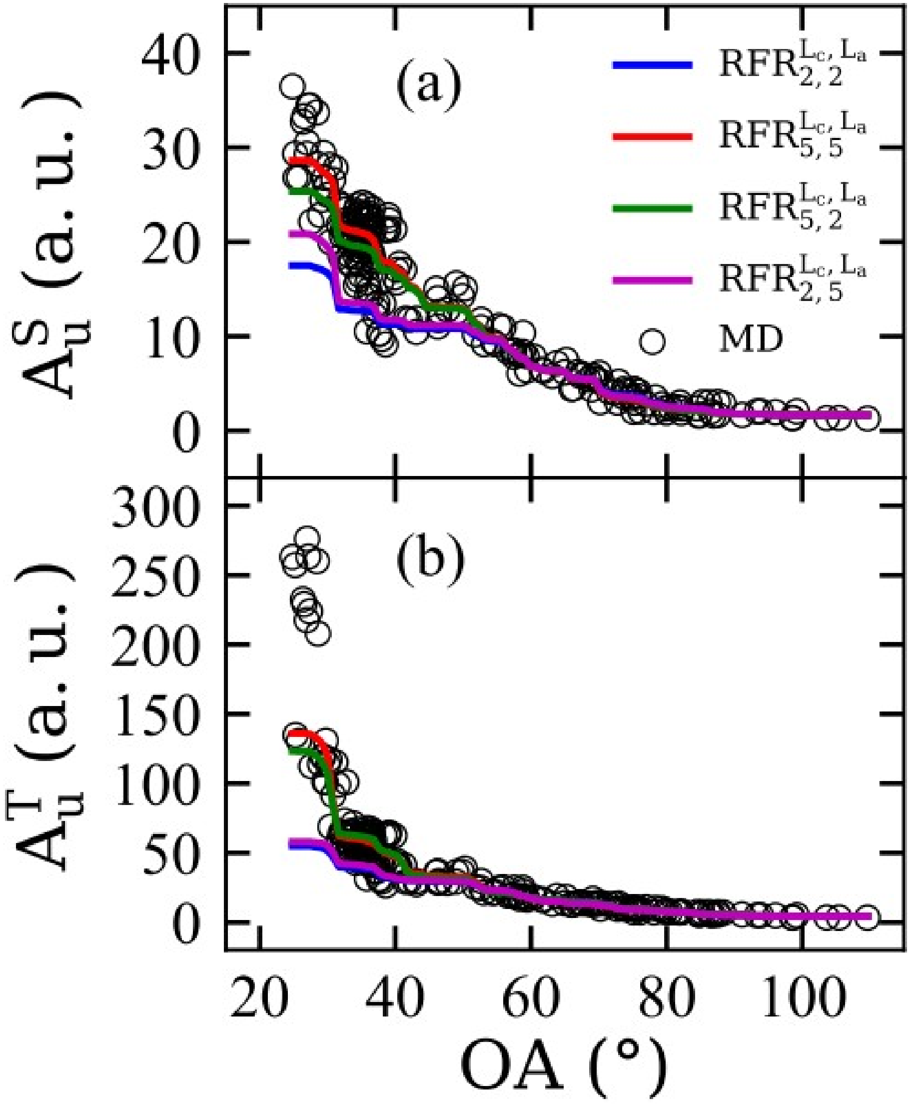



→ **Orientation Angle (OA) is the most important feature**

# Elastic anisotropy sensitivity to textural parameters

$$A^U = \boldsymbol{c} : \boldsymbol{s} = 5\frac{G^V}{G^R} + \frac{K^V}{K^R} - 6$$

- $A^U$ : Universal Anisotropy
- $G^V$ : Shear modulus using Voigt approximation
- $G^R$ : Shear modulus using Reuss approximation
- $K^V$ : Shear modulus using Voigt approximation
- $K^R$ : Bulk modulus using Reuss approximation



# Conclusion

- We have built a large dataset of pyC models using a new, polygranular, image guided atomistic reconstruction method
- We have characterized elastic properties under quasi-static and high-strain rate loading
- Elastic properties are strongly dependent on the atomic environment ($sp^2$ and $sp^3$ carbon atoms)
- Elastic properties were successfully connected to structural/textural parameters using ML
- ML reveals that textural anisotropy governs the elastic properties of pyCs
- Universal anisotropy index $A^U$ was also calculated in order to relate the elastic properties to the texture of the pyCs and shows that the orientation of the graphitic domains plays an important role on the elastic properties

| pyC | $E_1^T$ | $E_3^T$ |
|---|---|---|
| LL | 385 | 22 |
| LR | 663 (644) | 11 (22) |
| LRe | 530 (612) | 12 (29) |
| $LRe_{1300}$ | 575 (599) | 12 (29) |
| $LRe_{1500}$ | 554 (652) | 12 (24) |
| $LRe_{1700}$ | 683 (658) | 9 (24) |

# Thank you for your attention

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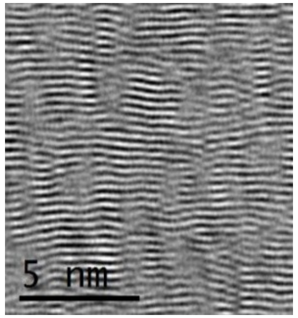
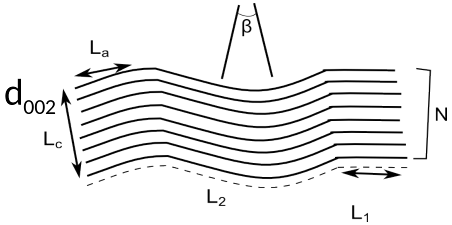
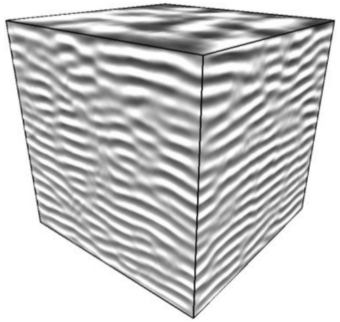
# Context / Image guided atomistic reconstruction (IGAR)

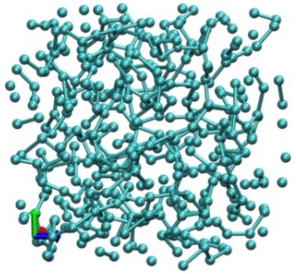



*002 lattice fringes image*




3D HRTEM-like synthesis





***Atoms***
***C / H***
***d=2.1g/cm³***

Image constrained
liquid quench simulation with REBO2 + HRTEM

Atomistic model (>200 000 atoms)
*Leyssale et al., App. Phys. Lett. 95 (2009), 231912.*

**An entire procedure to go from experimental imaging (HRTEM) to numerical synthesis of PyCs at the atomic level**

**Both length and time scales are compatible with classical MD simulations : ideal for a multiscale « *bottom-up* » approach**