Des images aux calculs de propriétés dans les composites carbone/carbone à trois échelles

Gerard L. Vignoles, J.-M. Leyssale, B. Farbos, P. Weisbecker, O. Caty, G. Couégnat, M. Charron, P. Engerand, A. Gillard, J.-P. Da Costa, C. Germain

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33 410 Talence Cedex, France

From images to property computations in carbon/carbon composites at three scales

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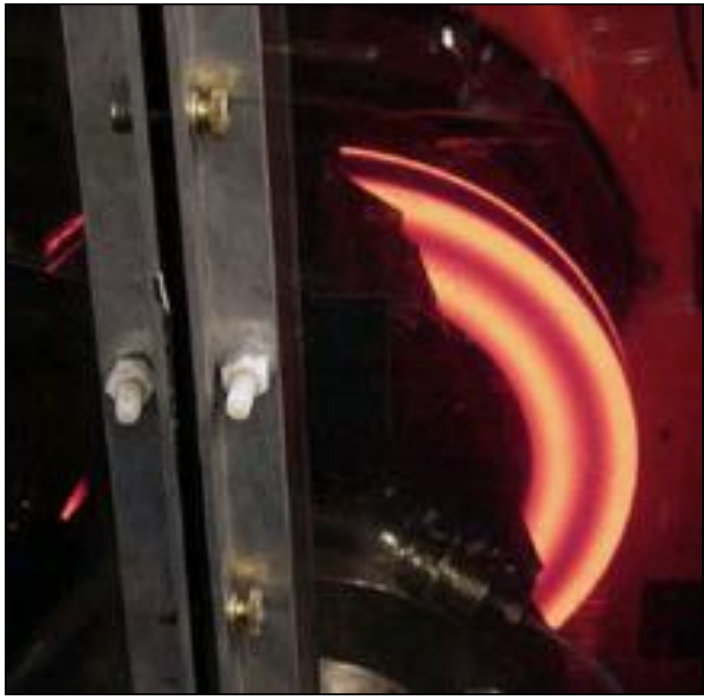
C/C Composites : applications

Rocket nozzles



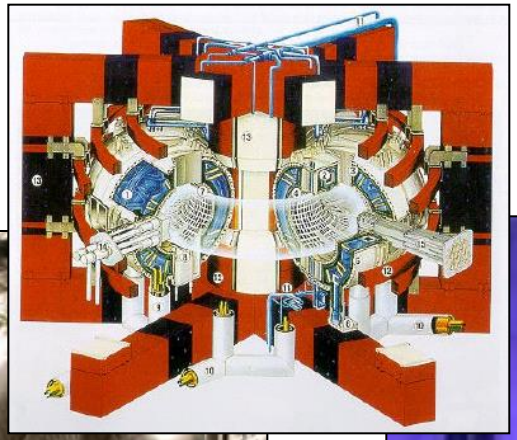
Aeroplane brake discs

C/C Composites : applications

Huygens mission

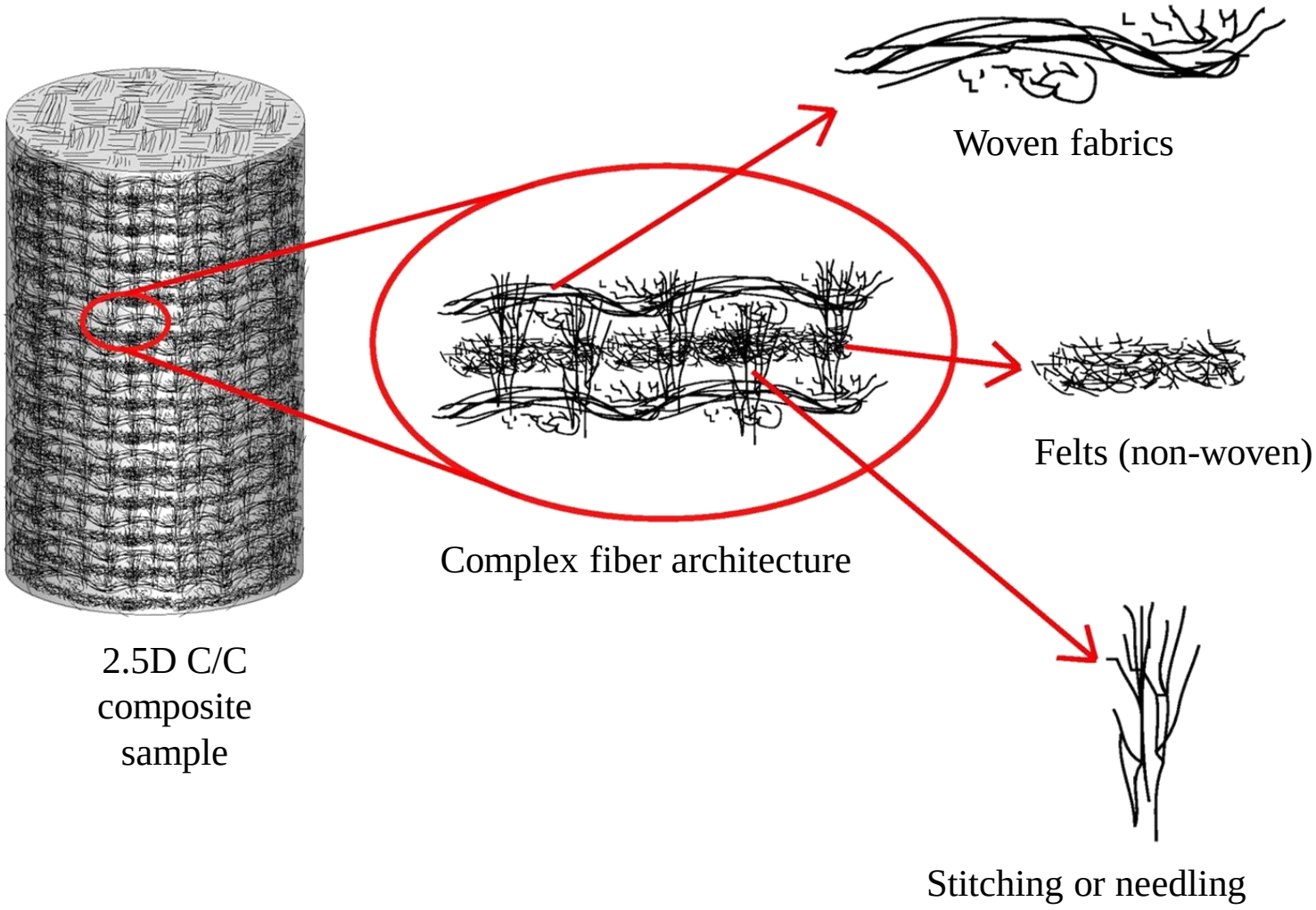
Atmospheric reentry
Thermal Protection
Systems (TPS)

C/C Composites : applications

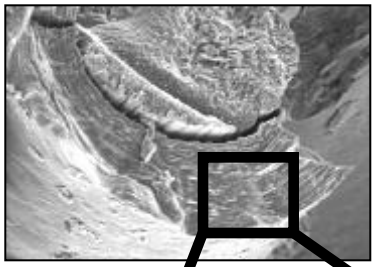


Tokamak plasma-facing components

3D stitched C/C (« 2.5D ») : a complex architecture

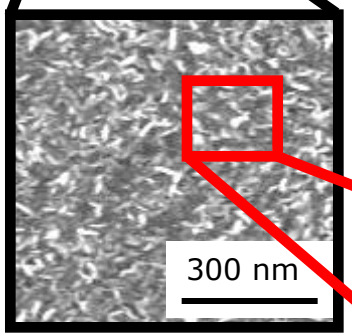


Multi-scale organization of dense carbons

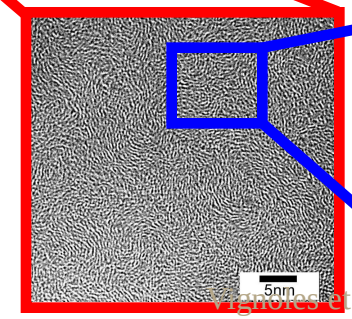


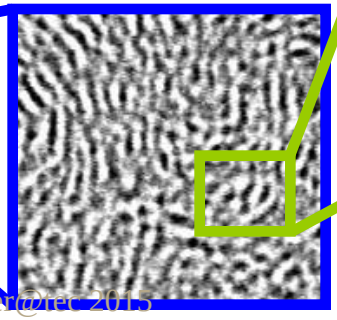
Fibers, matrices, interfaces

Macropores

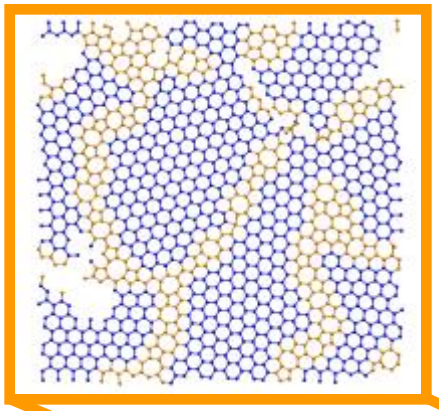


Organized domains

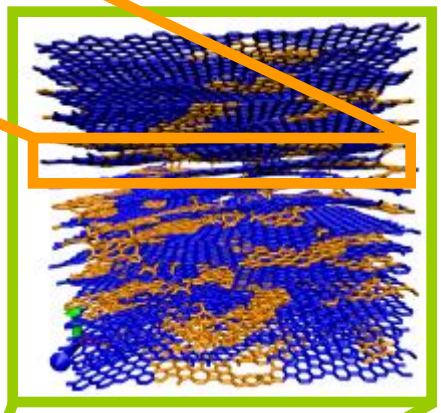




Sheet orientation distribution

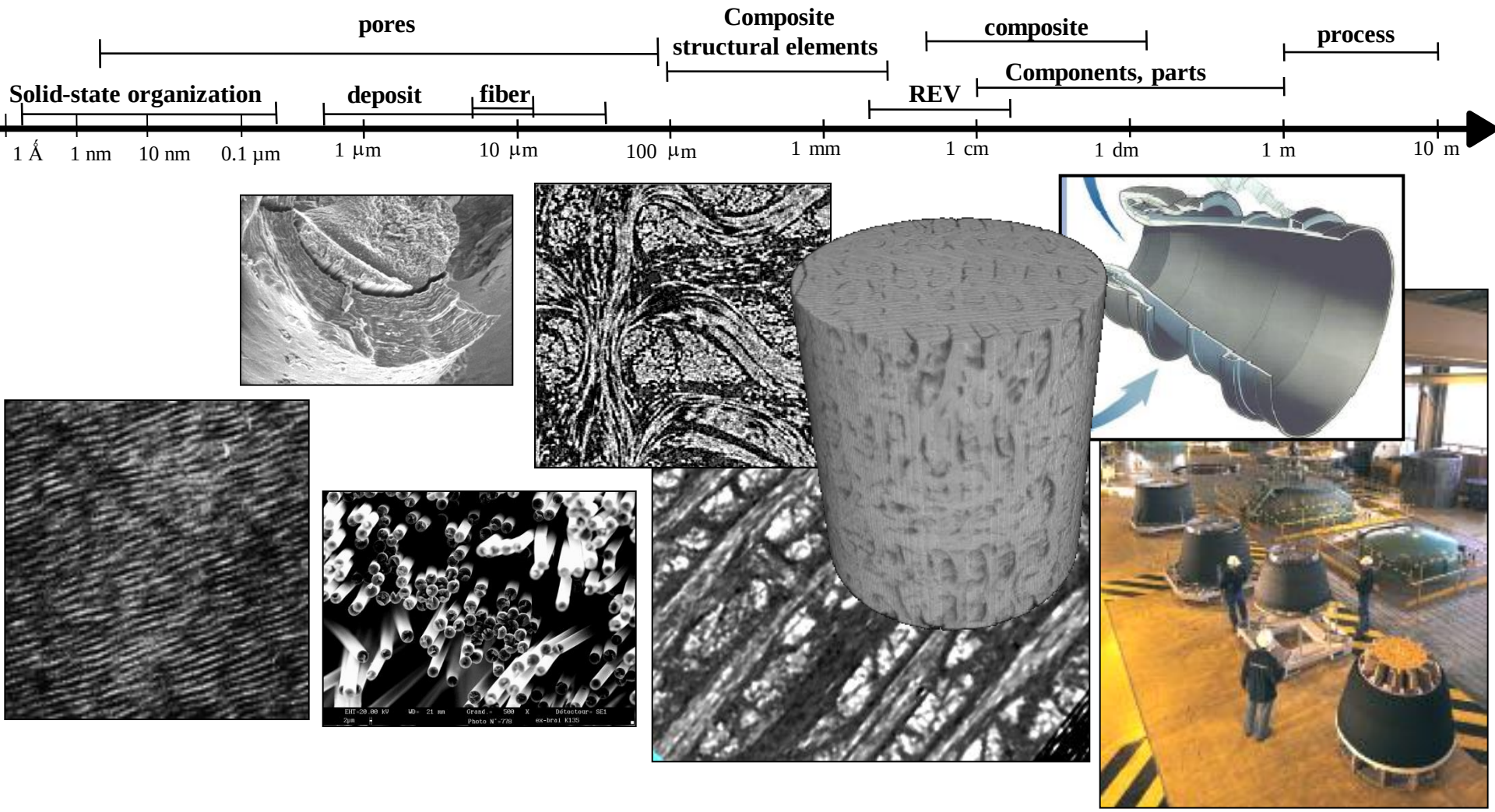


Inter- and intra-sheet defects



Texture is present at all scales !

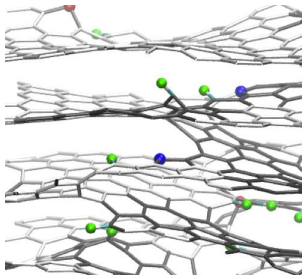
3D stitched (“2.5D”) C/C composites: multiscale materials

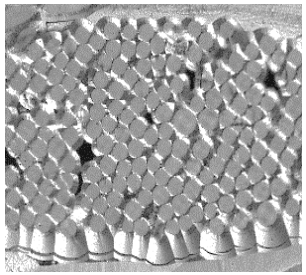


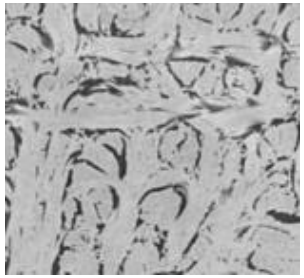
Motivations

- C/C : excellent structural & thermal properties at high T
- Very important criteria : stiffness, resistance, thermal expansion, heat conduction, …
- Thermomechanical behavior depends strongly on structure at all scales …
- A « difficult » architecture :
  - Pyrocarbon nanotexture : versatile and multi-scale organization
  - « 2.5D » stitched fabrics: very complex geometry
- High production costs ➔ motivates <u>numerical modeling</u>

Outline







Part I -

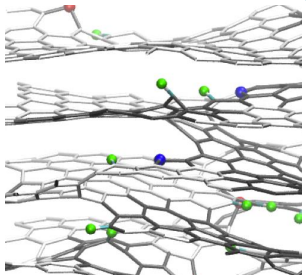
- Nanoscale : Pyrocarbon nanotexture modeling from HRTEM images

Part II -

- Micro-scale : C/C thermoelasticity modeling from micrographs
- Macro-scale : C/C thermoelasticity modeling from X-ray CT scans

Part I -

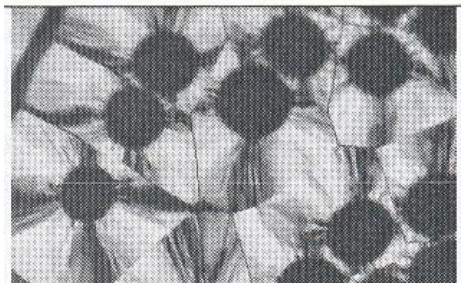
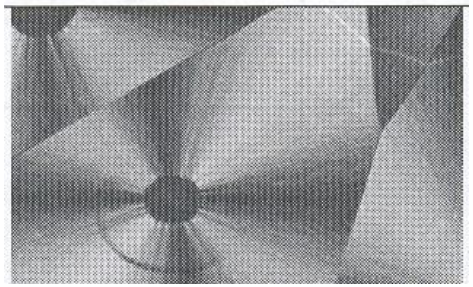
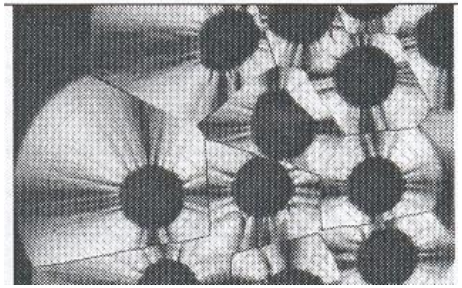
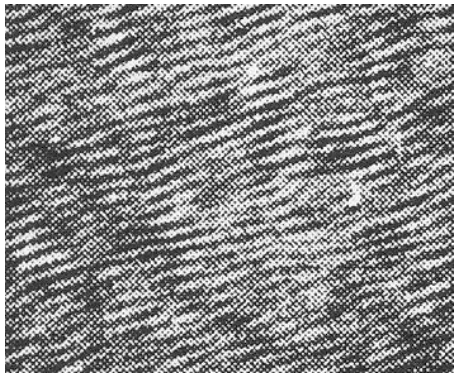
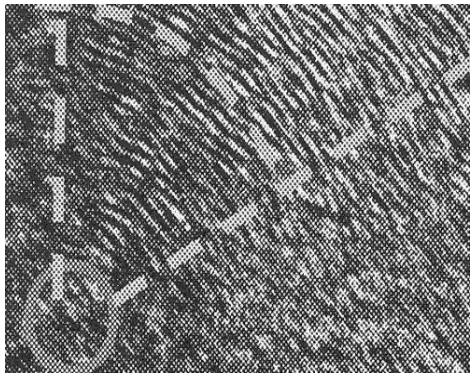
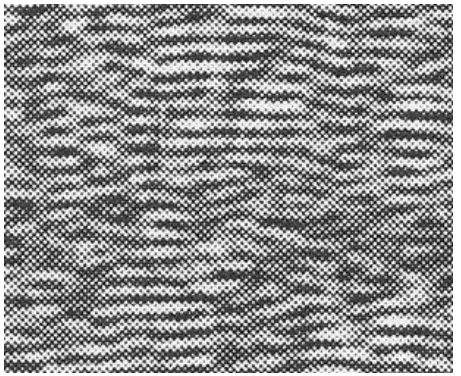
PYROCARBON MATRICES NANOTEXTURE



Atomistic modelling : structure-property relationships

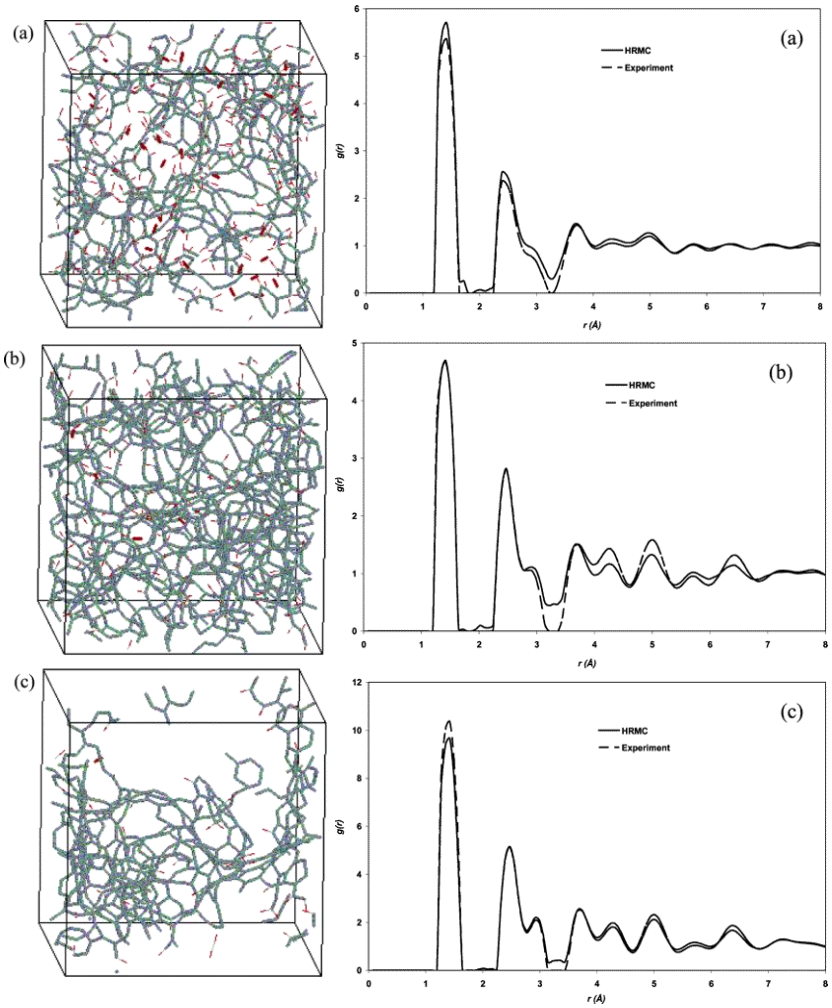
Pyrocarbons nanotexture & structure

At least 3 different PyCs, depending on processing conditions

Rough Laminar	Smooth Laminar	Regenerative Laminar
		
		
High anisotropy - long ranged straight fringes	Low anisotropy - long ranged curved fringes	High anisotropy - short ranged straight fringes

Can we build some atomistic models from experimental characterizations ?

Atomistic reconstruction from experiments



Jain, Langmuir 2006

(Hybrid) Reverse Monte Carlo (RMC)

Reconstruction by fitting the Pair Distribution Function (PDF)

$$g(r) = \frac{1}{r_0 N}\left\langle \sum_{i=1}^{N} \sum_{j \neq i} \delta(r - r_{ij}) \right\rangle$$

or

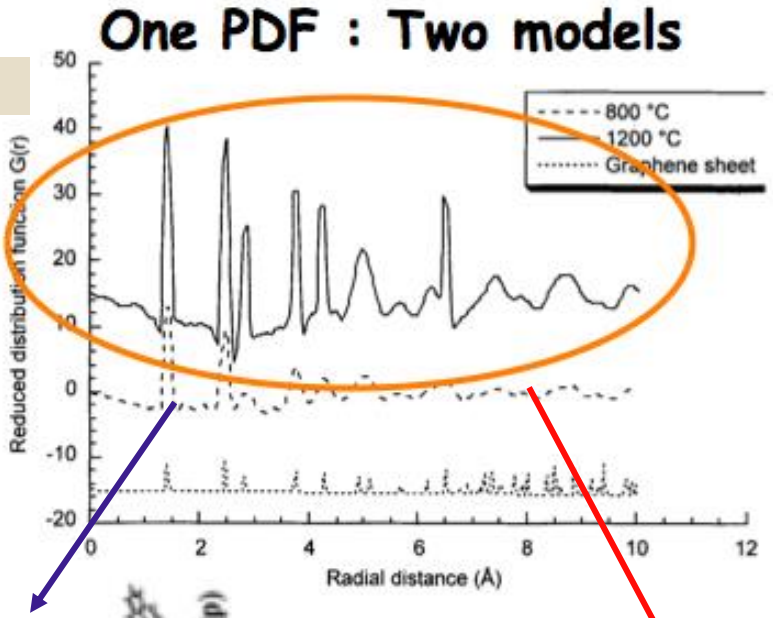
$$G(r) = 4\pi\rho_0 r\left[g(r) - 1\right]$$

obtained from neutron or X-ray diffraction

Works for isotropic & disordered systems

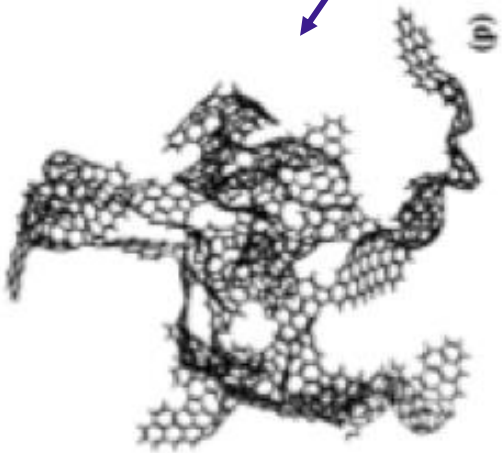
Atomistic reconstruction from experiments

Petkov Phil. Mag. B 1999



H(R)MC is irrelevant when systems contain ***local order*** and ***mesoscopic scale disorder***

Relevant info lies in HRTEM images



Acharya et al., Phil. Mag. B 79 (1999) 1499.

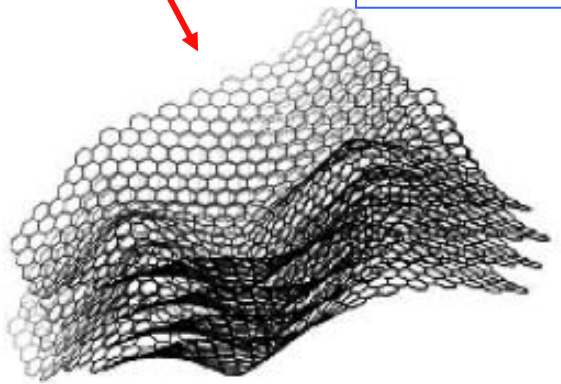
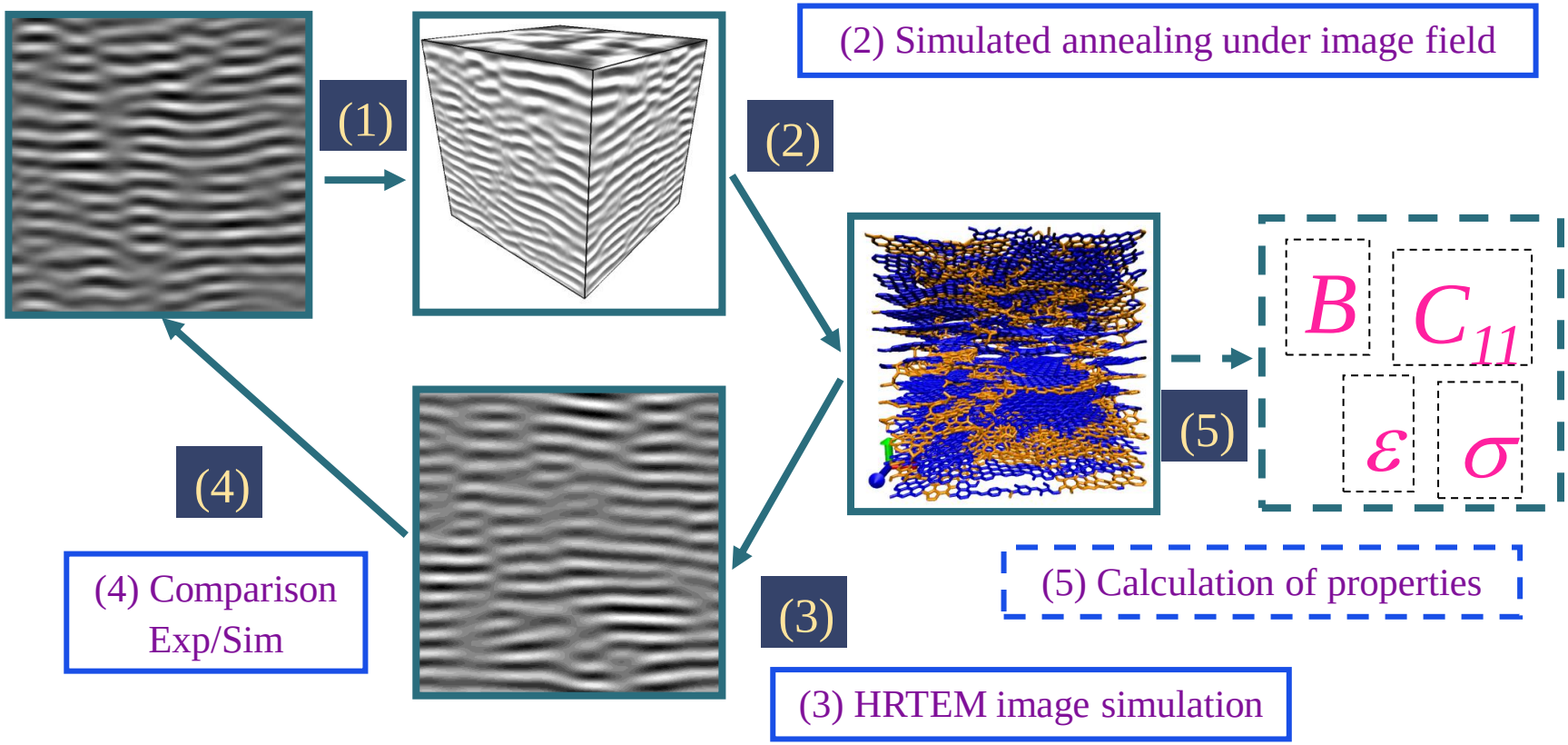


Fig. 3. A model of structure consisting of four turbostratically layered sheets. Models containing typically 10 sheets were used to calculate the PDF.

Smith et al, Carbon 42 (2004) 2041.

Reconstruction from HRTEM : The IGAR method

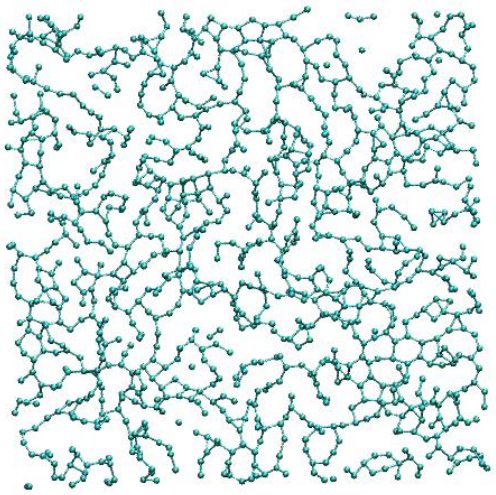
(1) Statistical analysis of HRTEM images & Synthesis of 3D HRTEM-like images

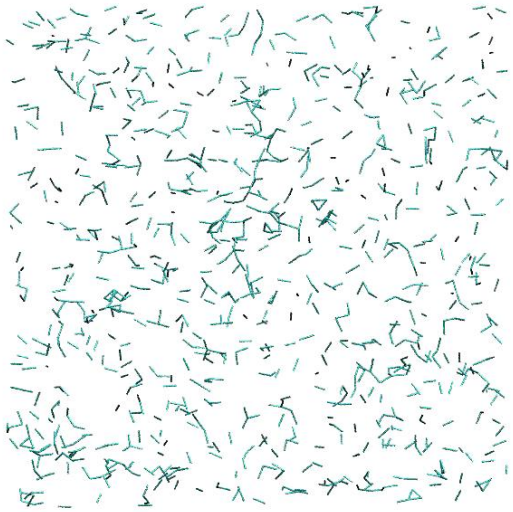


Leyssale *et al.* , App. Phys. Lett. 95 (2009) 231912

J.M. LEYSSALE, J.-P. DA COSTA, C. GERMAIN, P. WEISBECKER and G. L. VIGNOLES, Carbon (2012), **50**, 4388–4400.

IGAR : Image Guided Atomistic Reconstruction





Images are filled with carbon atoms with

$d = 2.1\ g/cm^2$
$d_{002} = 3.5\ Å$

Molecular Dynamics quenching

$$\frac{d^2\mathbf{r}_i}{dt^2} = \frac{d\mathbf{v}_i}{dt} = -\frac{1}{m_i}\frac{d\left(U_{REBO} + U_{IMAGE}\right)}{d\mathbf{r}_i}$$

with empirical reactive potential

$$U_{REBO} = \sum_{i=1}^{N}\sum_{j=i+1}^{N}\left[V^R(r_{ij}) - b_{ij}V^A(r_{ij})\right]$$

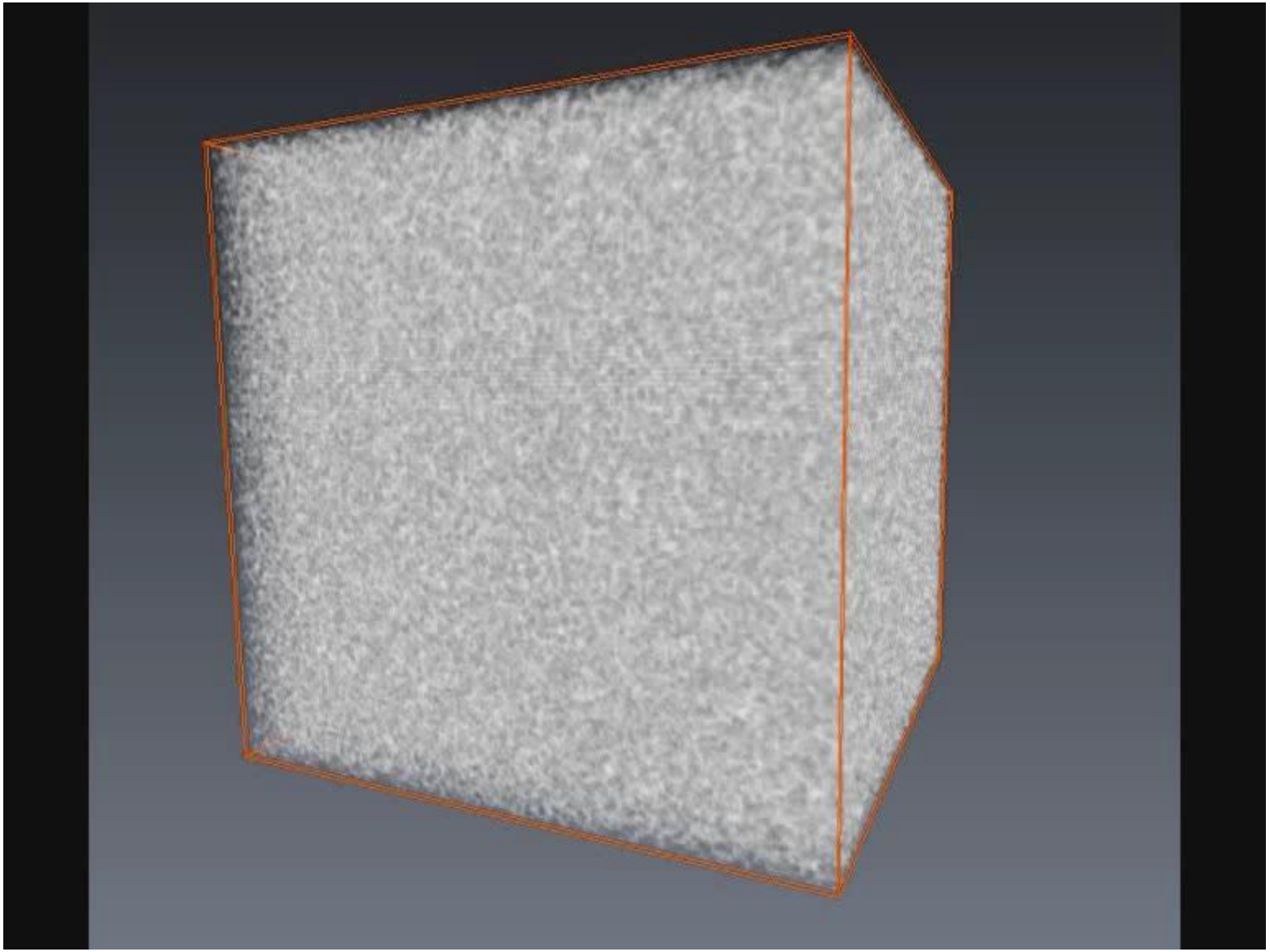
$$\begin{pmatrix} V^R(r) = f^c(r)\left[1 + Q/r\right]A\exp(-\alpha r); \\ V^A(r) = f^c(r)\sum_{n=1,3} B_n \exp(-\beta_n r) \end{pmatrix}$$

and an external force field imposed by the 3D image :

$$U_{IMAGE}(eV) = 2\sum_{i=1}^{N} I(\mathbf{r}_i)$$

J.M. LEYSSALE, J.-P. DA COSTA, C. GERMAIN, P. WEISBECKER and G. L. VIGNOLES, Carbon (2012), **50**, 4388–4400.

IGAR in a single movie …



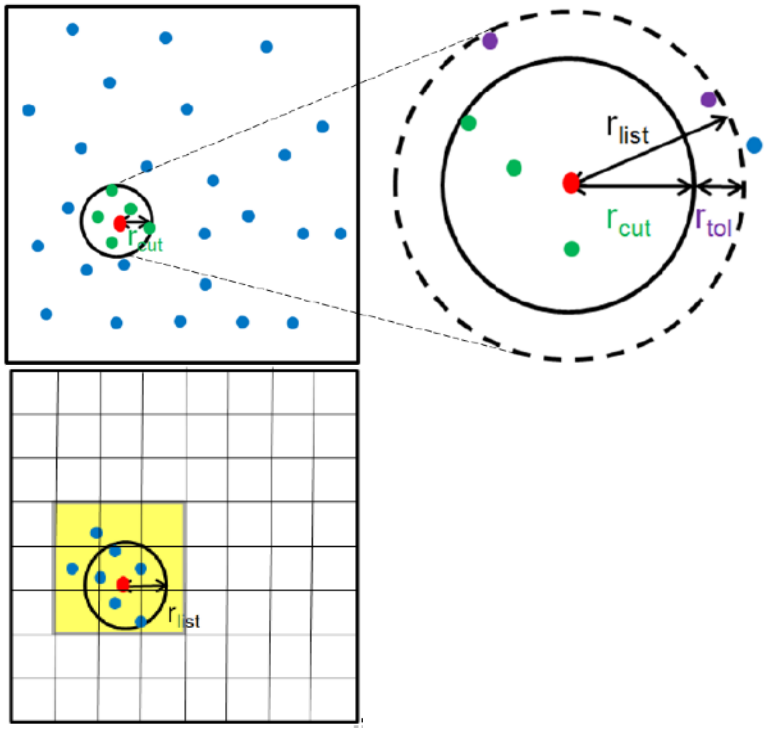
HPC effort for IGAR simulations

Code optimization

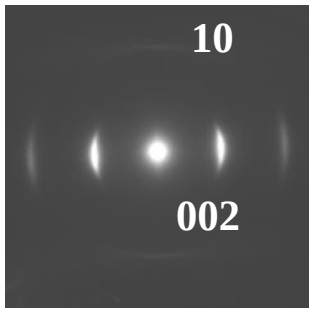
- Implementation of Verlet neighbours lists for the computation of potentials
- Space partitioning for lists updates

Parallelization

- Using OpenMP
- Avoiding « reductions »
- Speedup almost linear until 70 CPUs

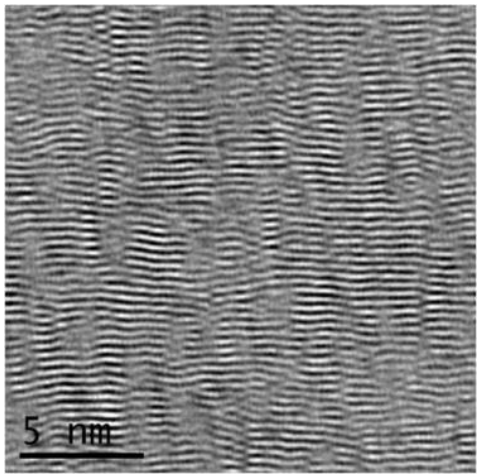


Two distinct Highly Textured pyrocarbons



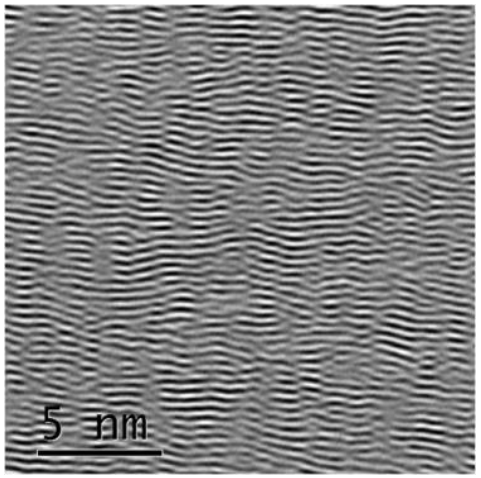
OA=22-25°

a) RL-PyC



OA=25-30°

b) ReL-PyC

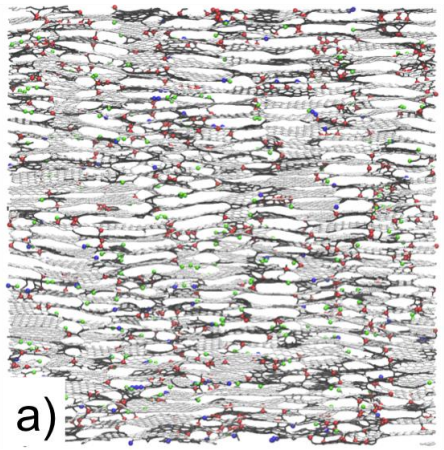


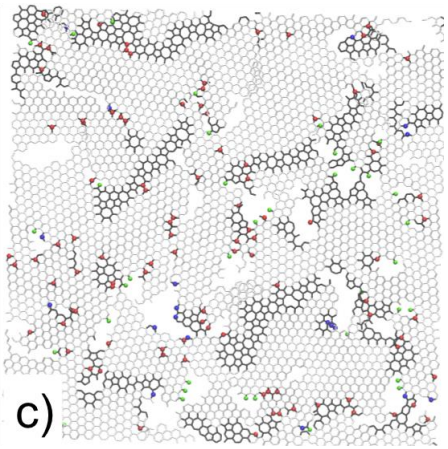
Sample	PLOM Ext. angle Ae (°)	*Raman Aniso. ratio* R_A	*Raman FWHM of D band* (cm^{-1})	SAED Angular Opening AO (°)	XRD d_{002} (A)	XRD L_a, L_c (nm)	*Density d* (g/cm^3)	*SIMS-ERDA H at.%*
RL	25°	8.2	83	22-25°	3.45	4.6 ; 5.2	2.12	0.7
ReL	20°	7.2	204	25-30°	3.49	2.6; 2.9	2.11	2.5

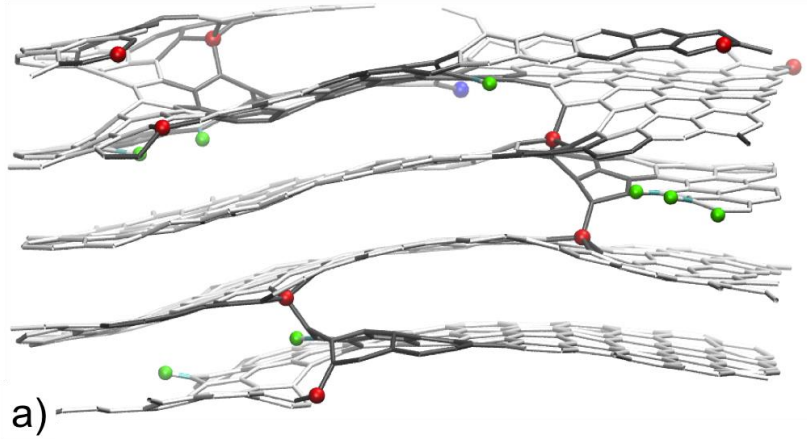
P. WEISBECKER, J.-M. LEYSSALE, H. E. FISCHER, V. HONKIMAKI, M. LALANNE, G. L. VIGNOLES, Carbon (2012), **50**, 1563–1573

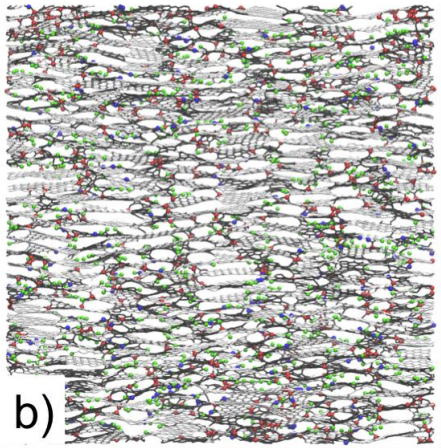
Large models (>200 000 at.)

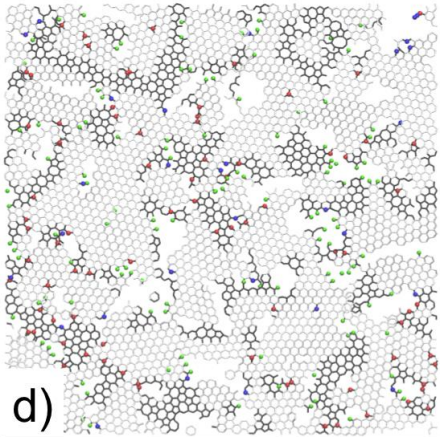
RL

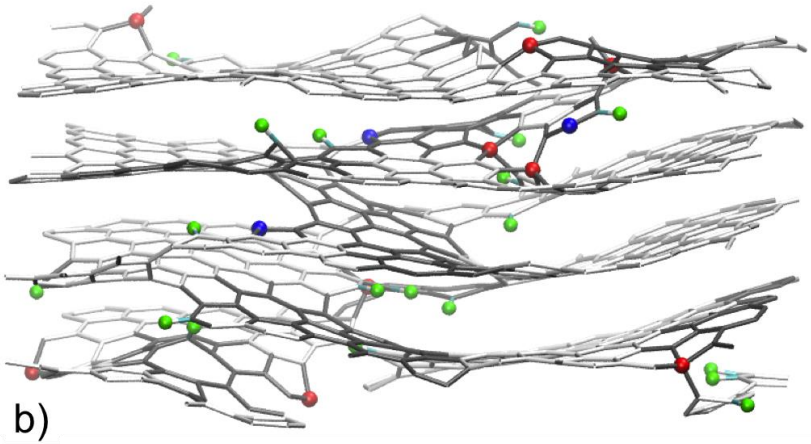






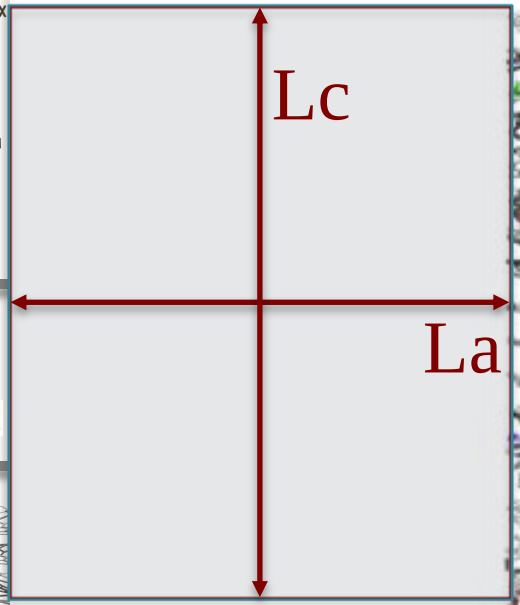


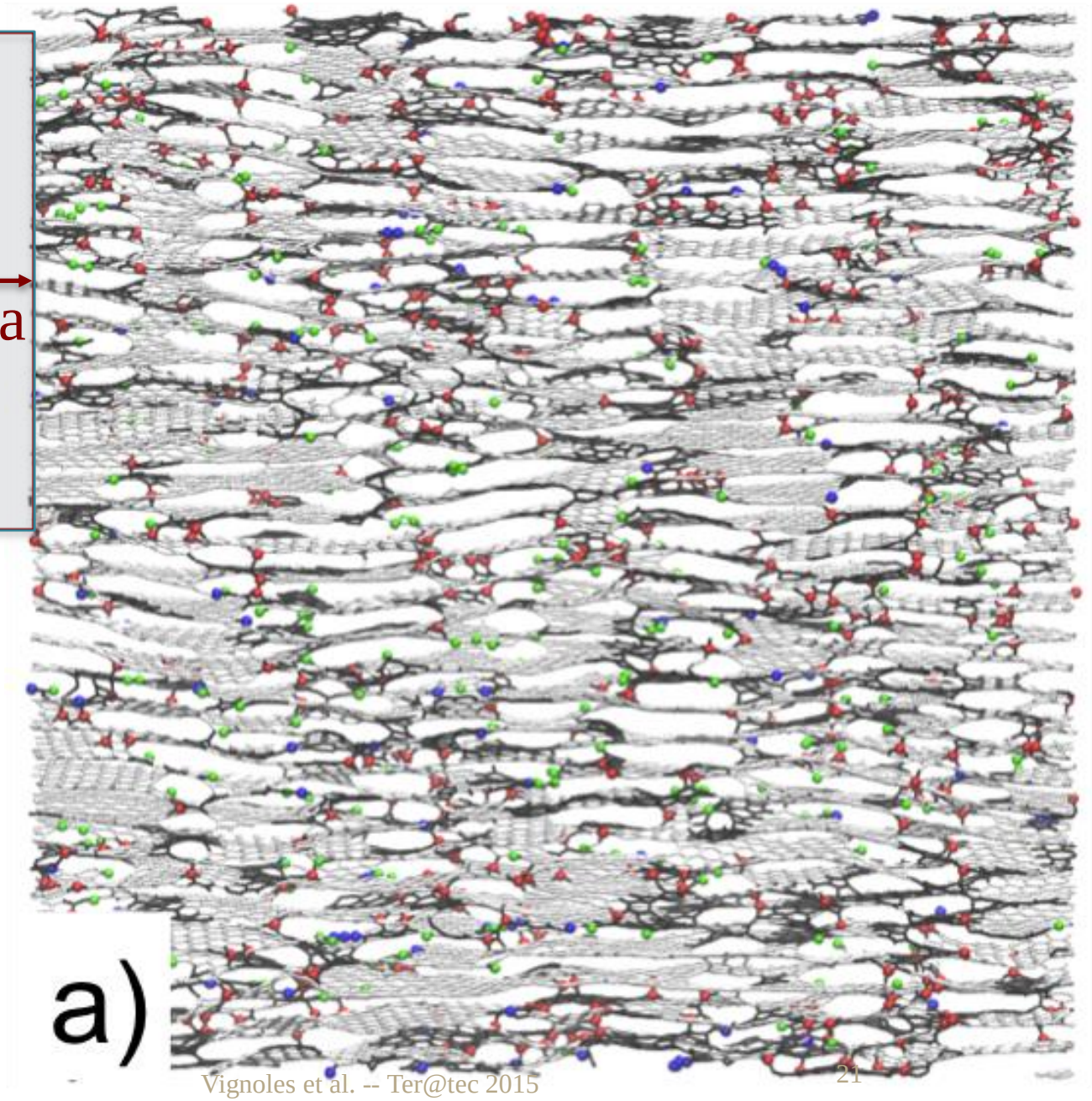




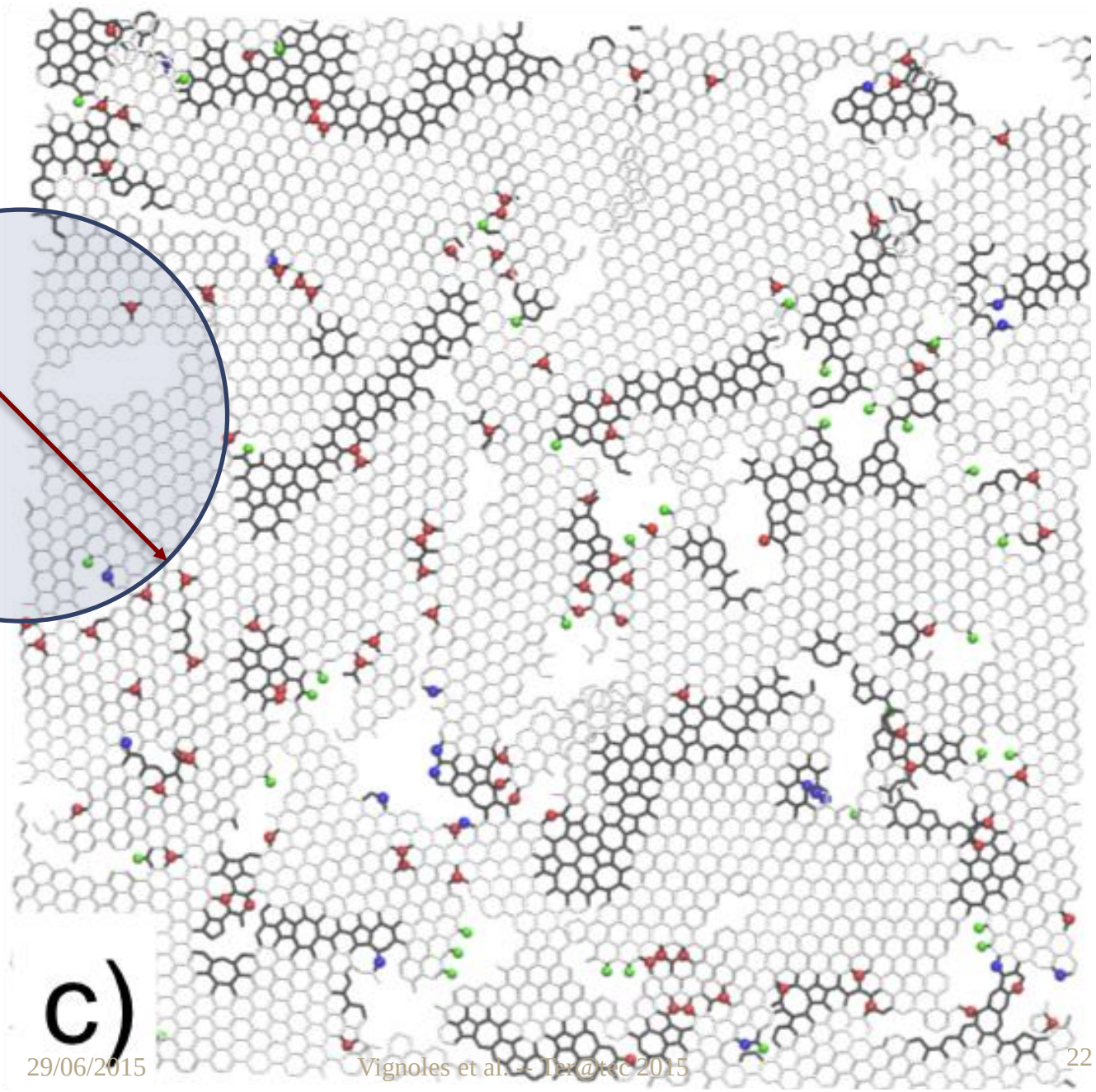
ReL

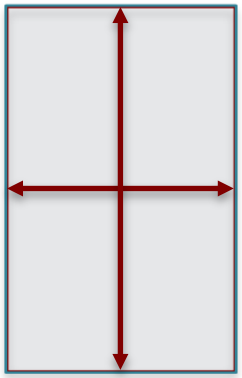
B. Farbos, et al., *Carbon* (2015) **84**, 160-173

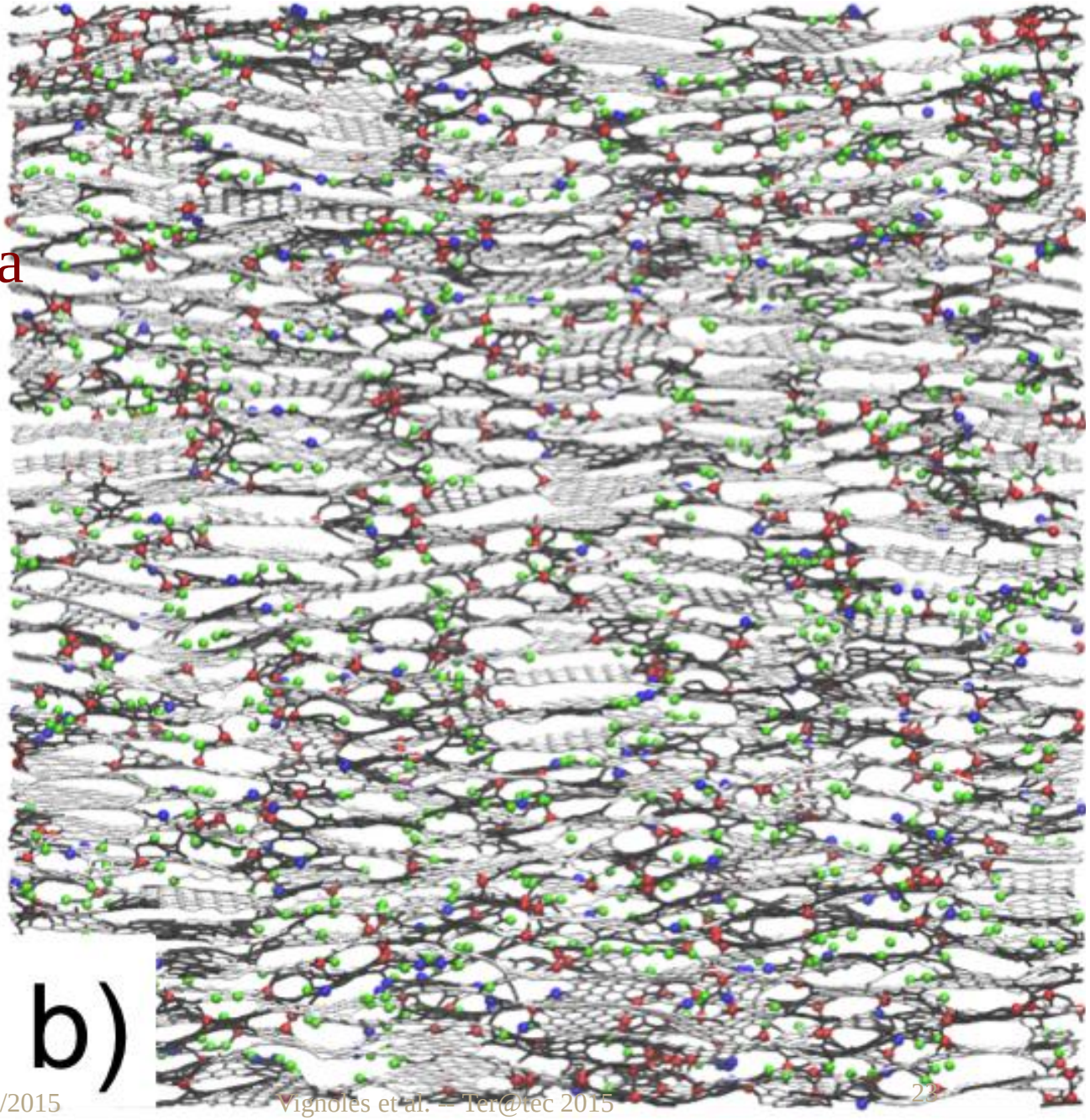

Lc
La

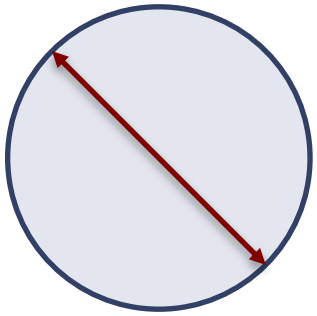

a)

La

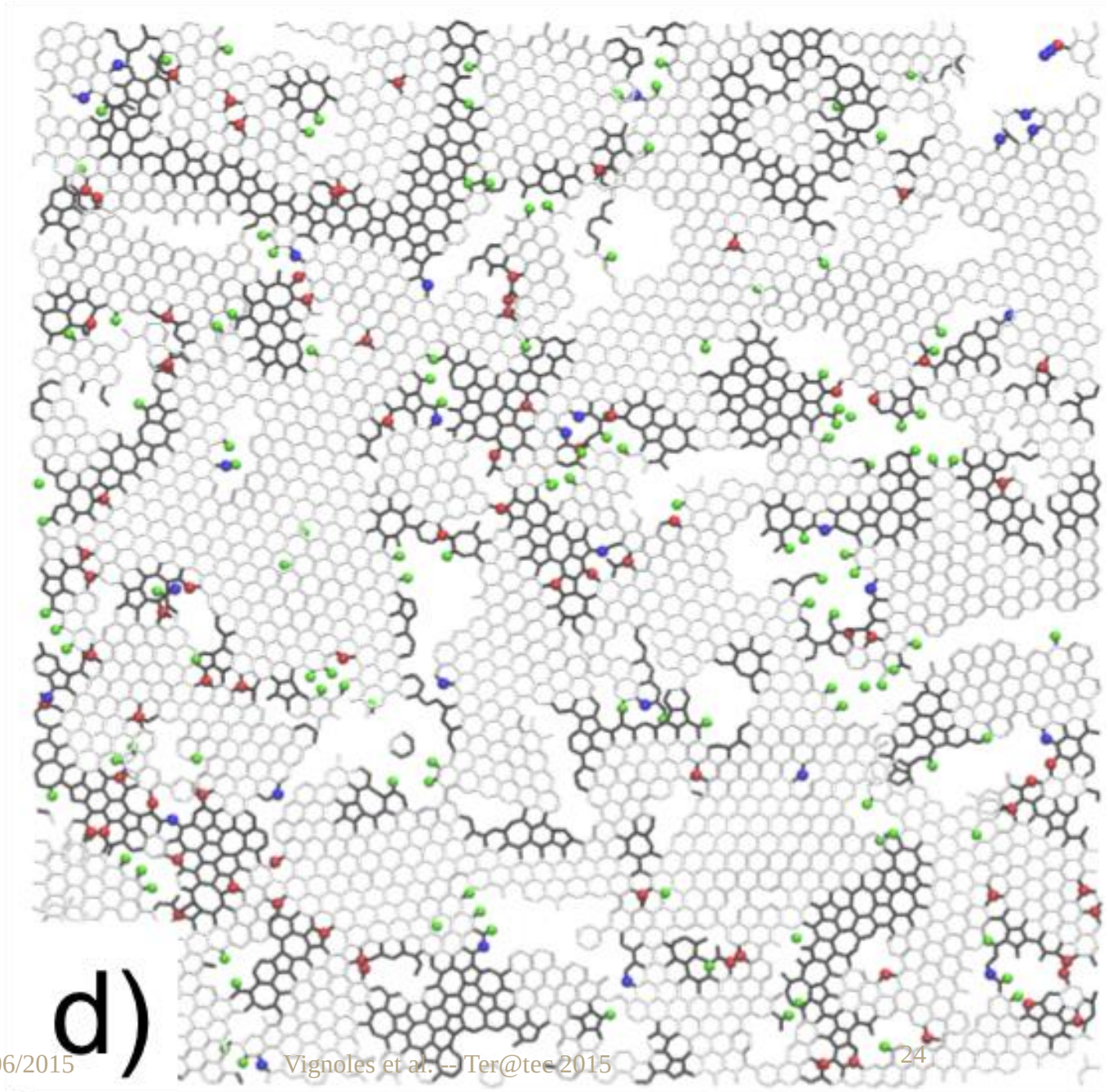



Lc
La

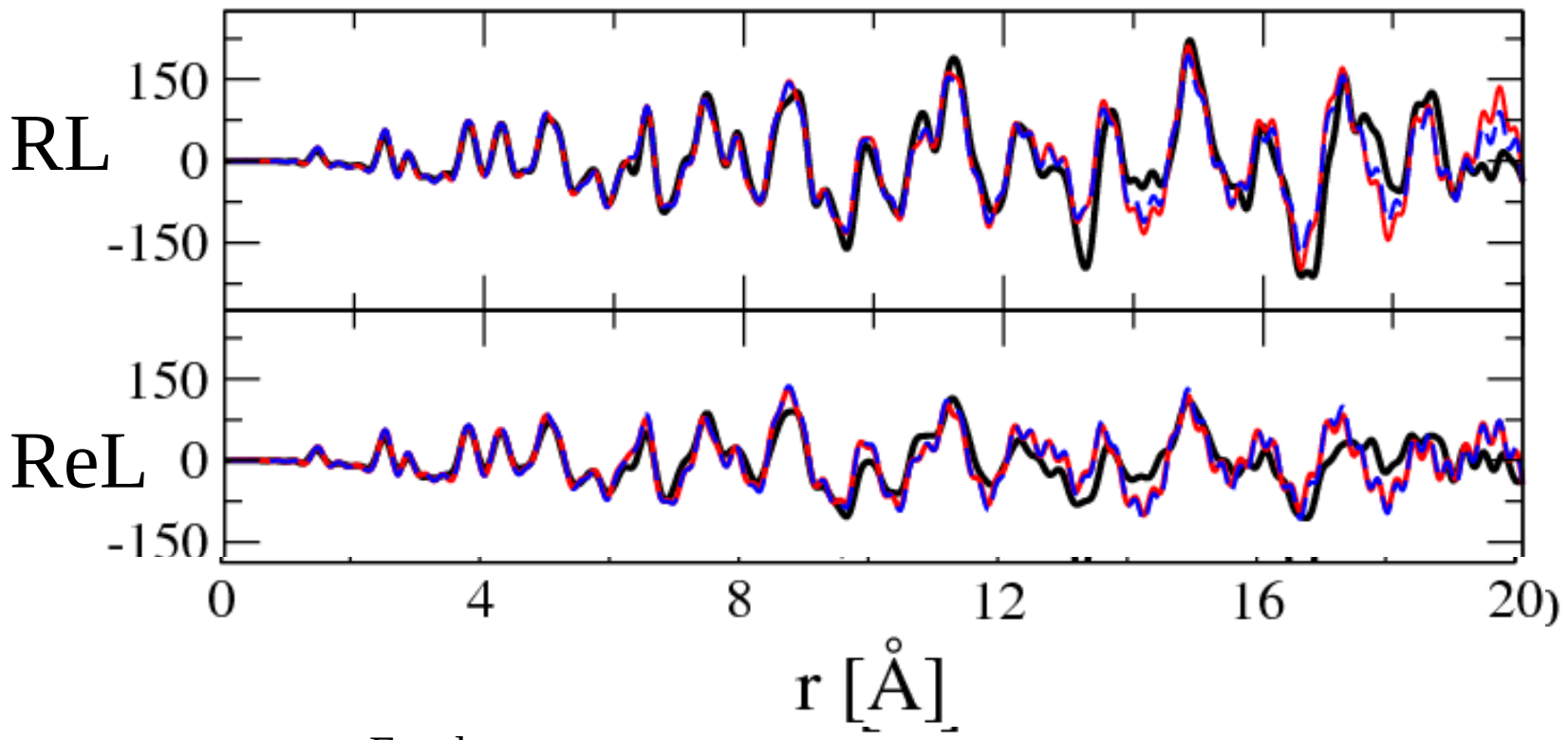

b)



La



Structural properties: PDFs $r^2G(r)$



Exptl.

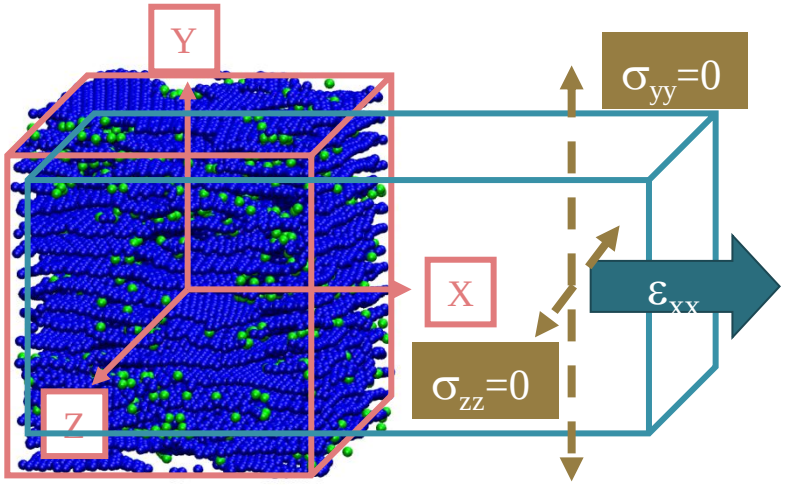
Small models (6.2^3 nm^3)

Large models (12.4^3 nm^3)

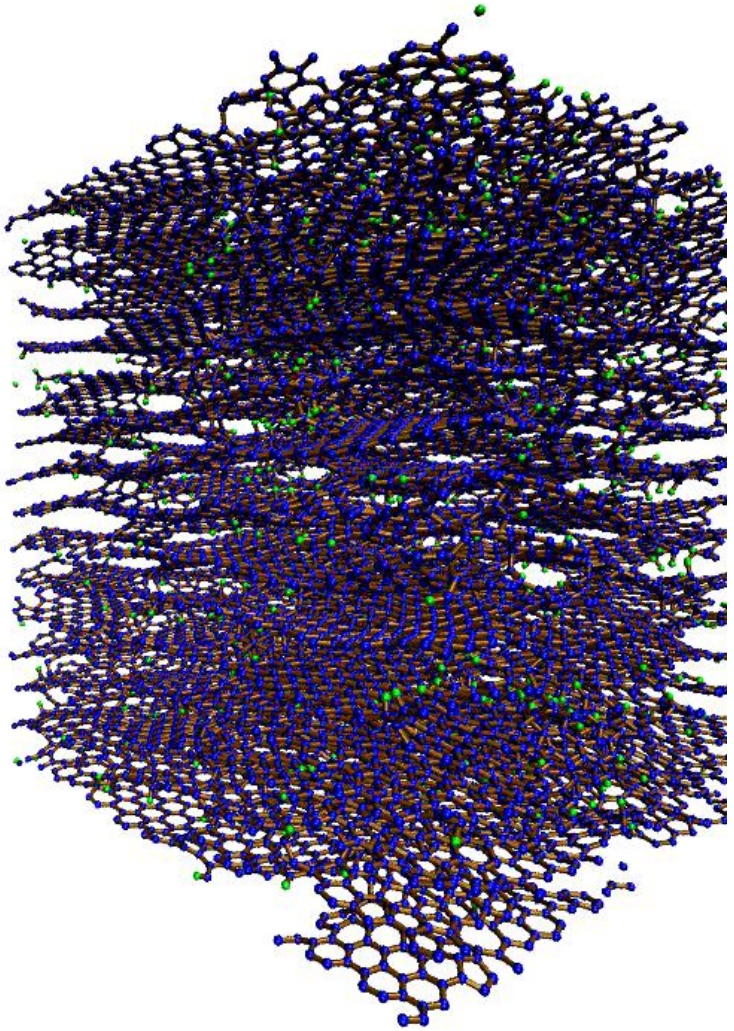
P. WEISBECKER, J.-M. LEYSSALE, H. E. FISCHER, V. HONKIMAKI, M. LALANNE, G. L. VIGNOLES, Carbon (2012), **50**, 1563–1573

Uniaxial tensile test of a ReL PyC

Uniaxial tensile test simulation



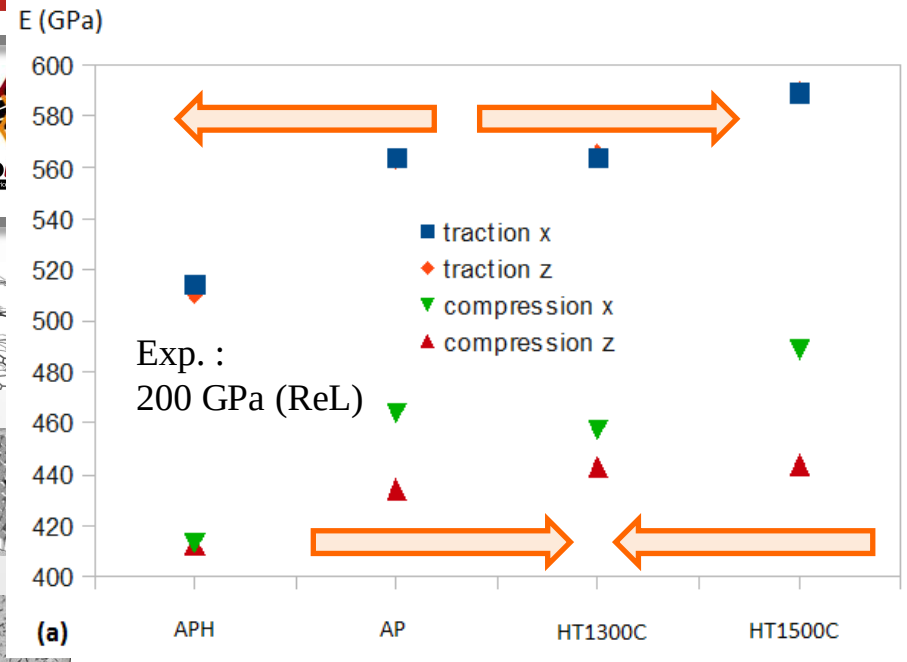
HMC **N(ε_{xx})(σ_{yy}=0)(σ_{zz}=0)T**



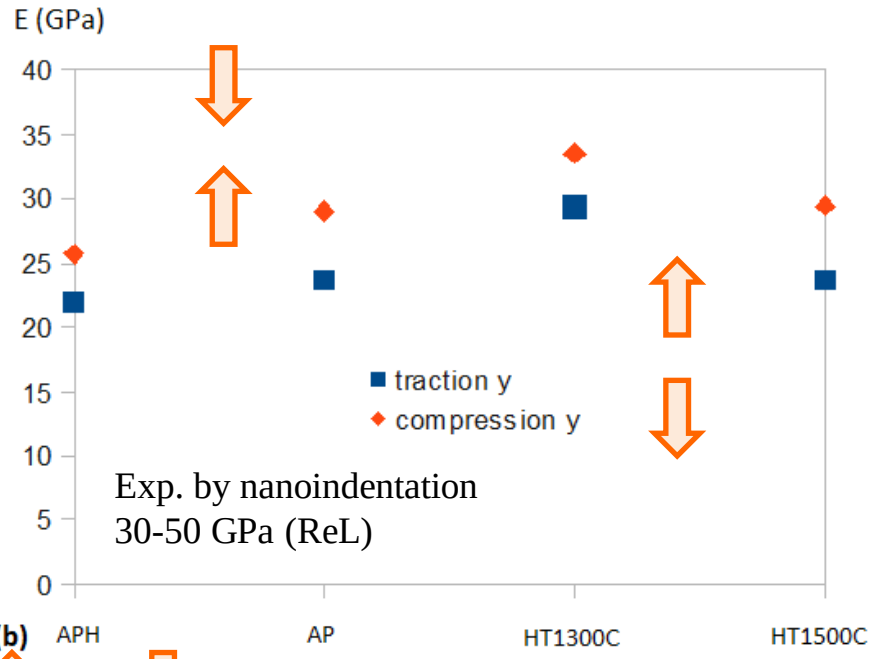
J.-M. LEYSSALE, B. FARBOS, J.-P. DA COSTA, P. WEISBECKER, G. CHOLLON, G. L. VIGNOLES, Ceram. Trans. **248**, 42-50 (2014)

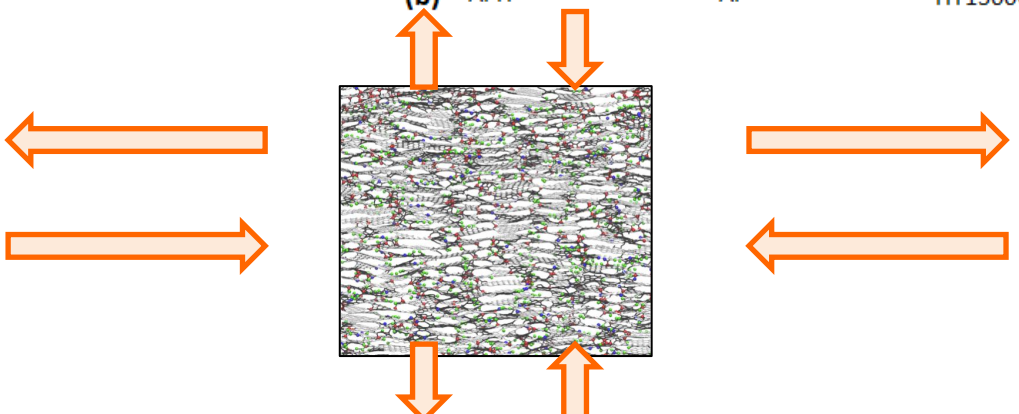
ReL : Young modulus computation

002 in-plane modulus



002 normal modulus



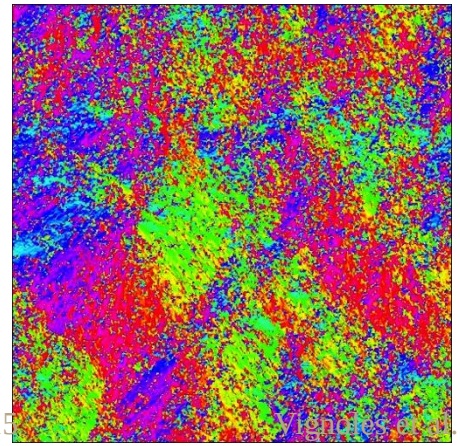


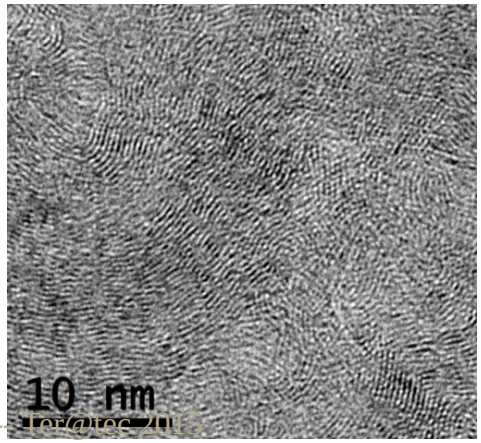
Conclusion – Part I

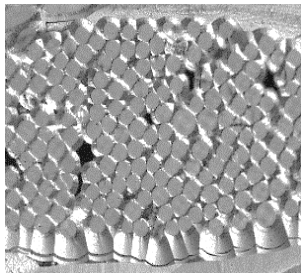
- IGAR method : OK for HT pyrocarbons
- Models are > 200 000 atoms: realistic but …

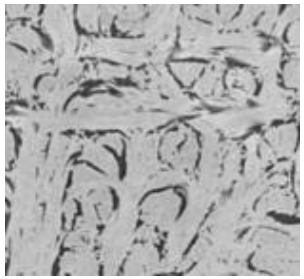
  should be larger (for tensile tests)
- Stiffness : interplane compression related to nano-indentation values
- Also compute heat transfer and heat expansion ?
- How to move to larger, less anisotropic models ?

SL pyC







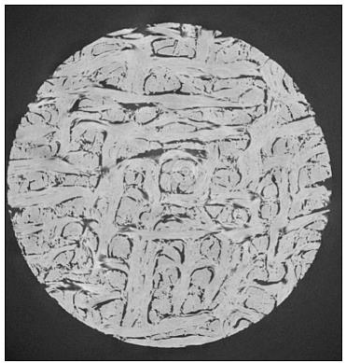


Part II -

IMAGE-BASED THERMAL EXPANSION MODELING OF C/C COMPOSITES

Aim & method

- From the detailed knowledge of the spatial arrangement of the constituents and of their elementary properties, obtain the effective thermomechanical behavior
- Utilization of X-ray µCT for structural description
- Image processing + FE computation
- Checking wrt. TMA tests



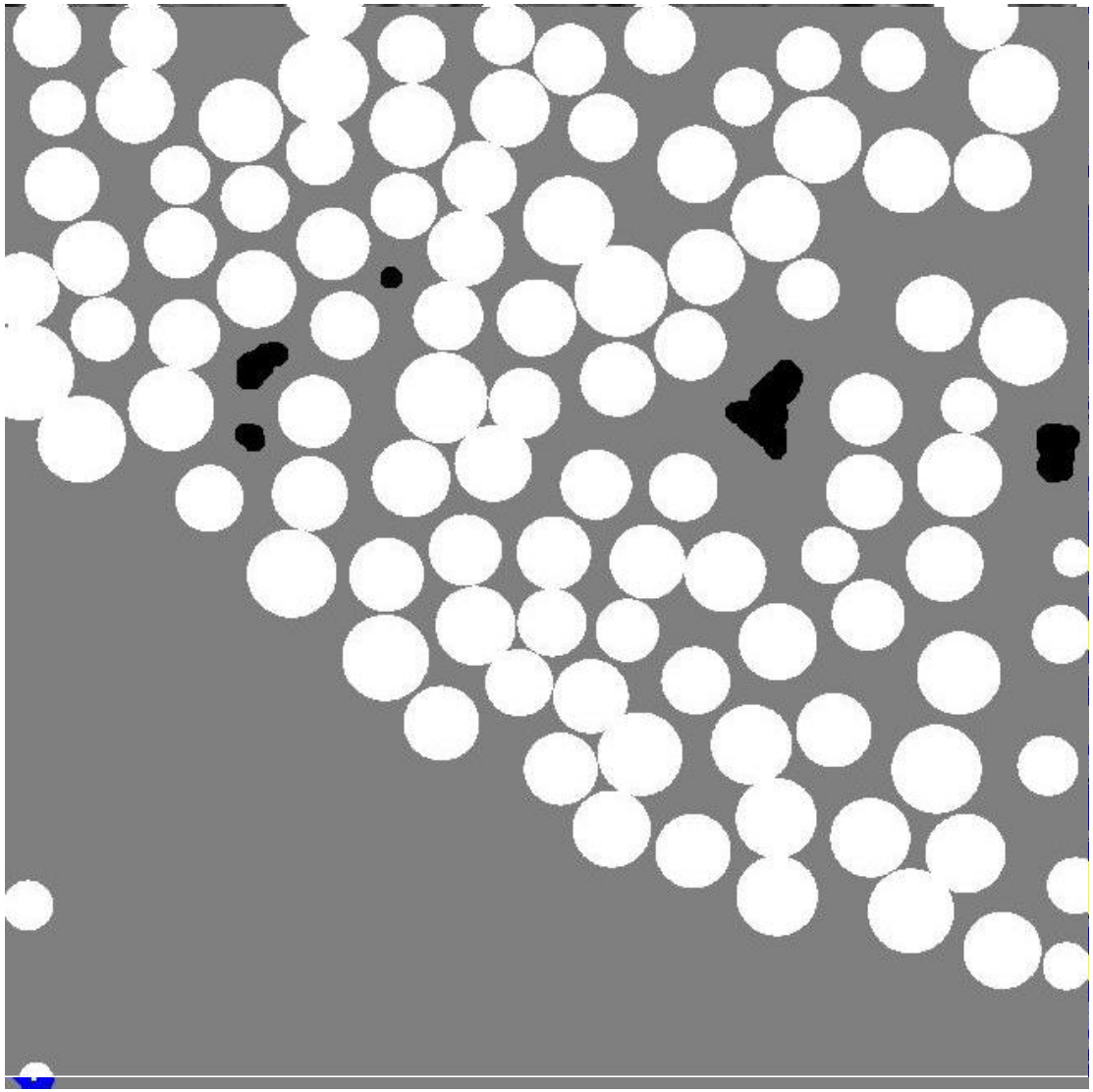
PLOM image analysis

Segmentation
of pores

Identification
of fiber centers

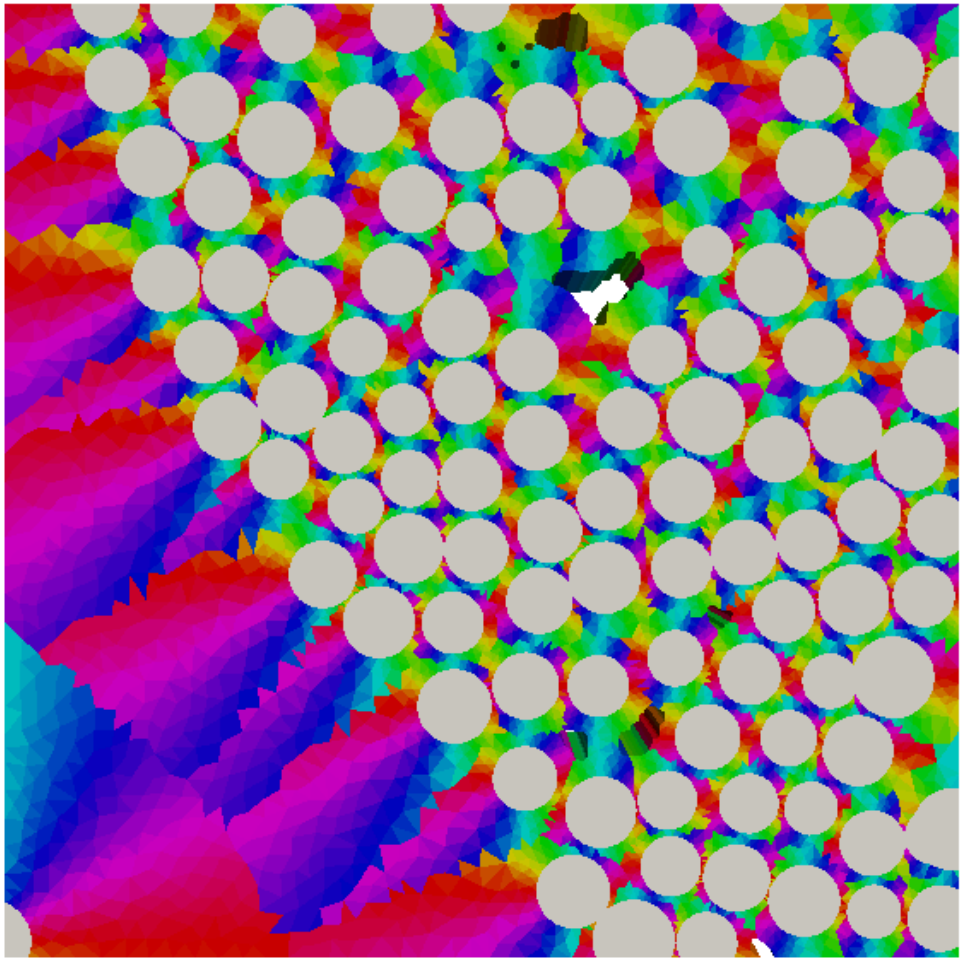
Fiber segmentation

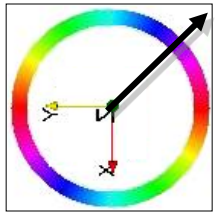
⇒ Full F/M/P
segmentation



PLOM image analysis

Matrix orientation field determination in GenCell* software



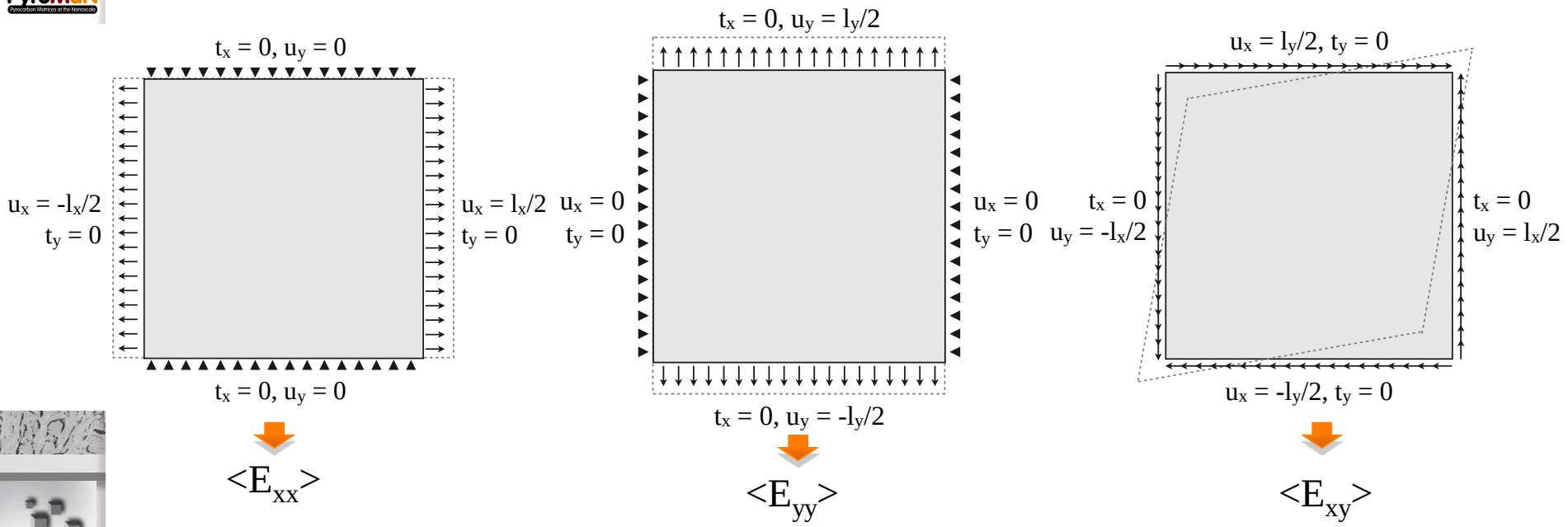


* G. COUEGNAT (2008). *Approche multiéchelle du comportement mécanique de matériaux composites à renforts tissé.* PhD Université Bordeaux I.

Boundary conditions for FEM analysis

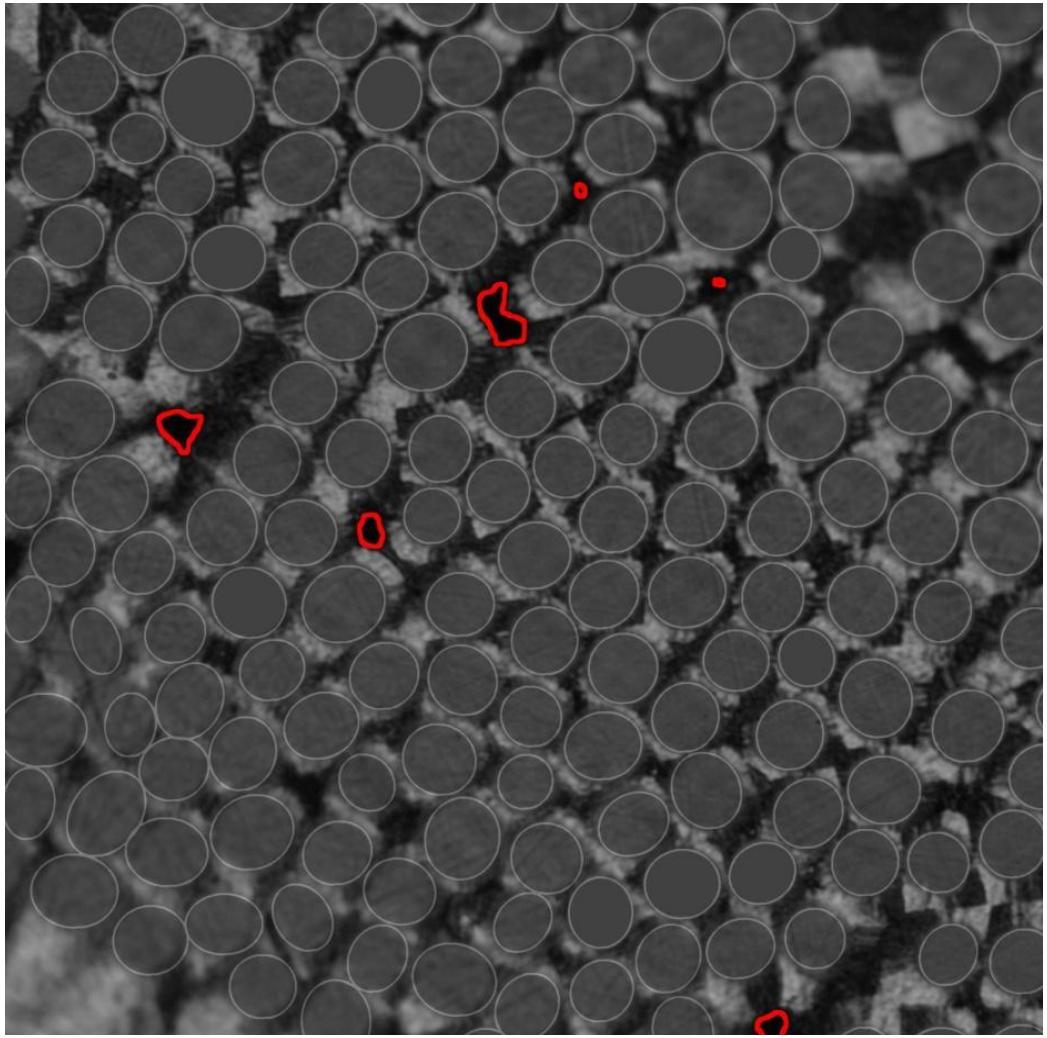
Periodic Mixed Uniform Boundary Conditions (Pahr & Zysset*):

- Satisfy Hill's lemma
- Respect material anisotropy
- Give less « boundary mismatch artifacts » than Periodic BCs



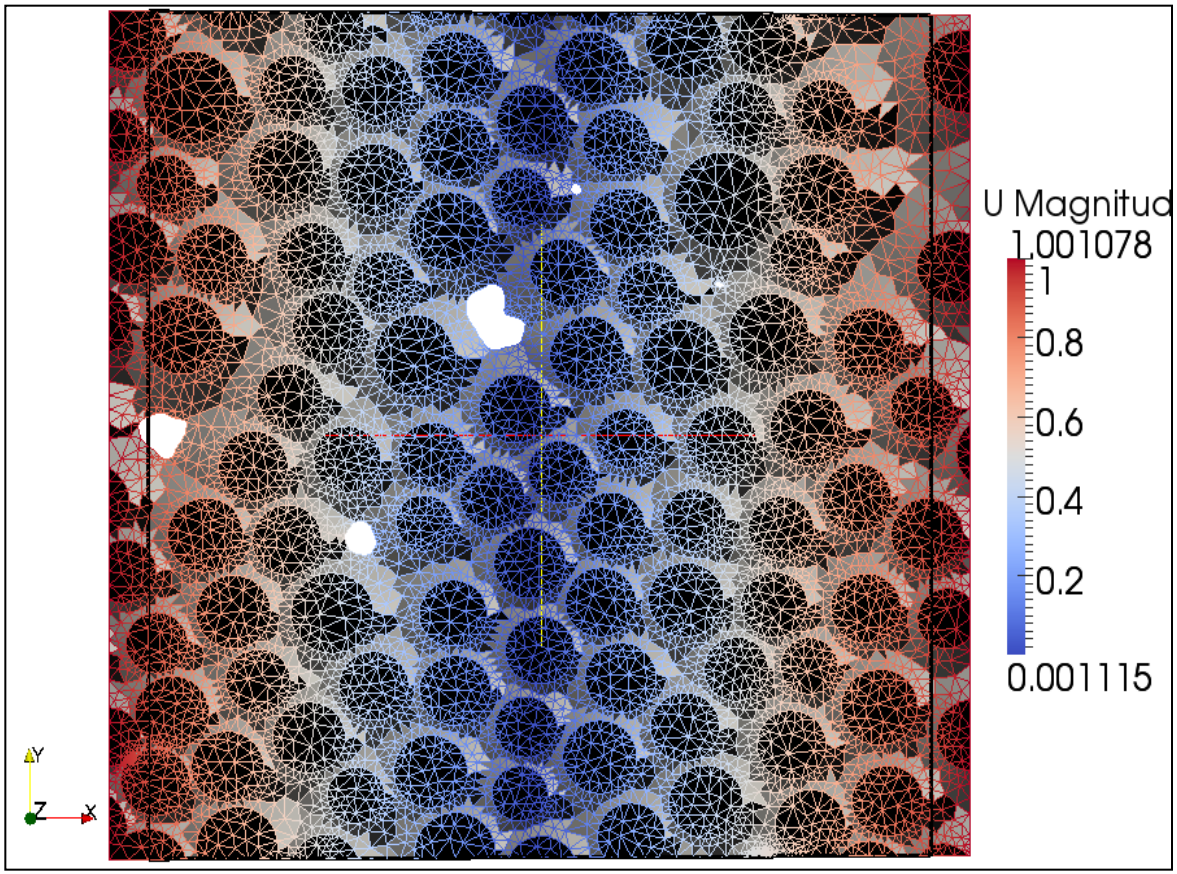
* DH Pahr & PK Zysset (2008). Influence of boundary conditions on computed apparent elastic properties of cancellous bone. *Biomech Model Mechanobiol* , 7:463-476.

FEM analysis



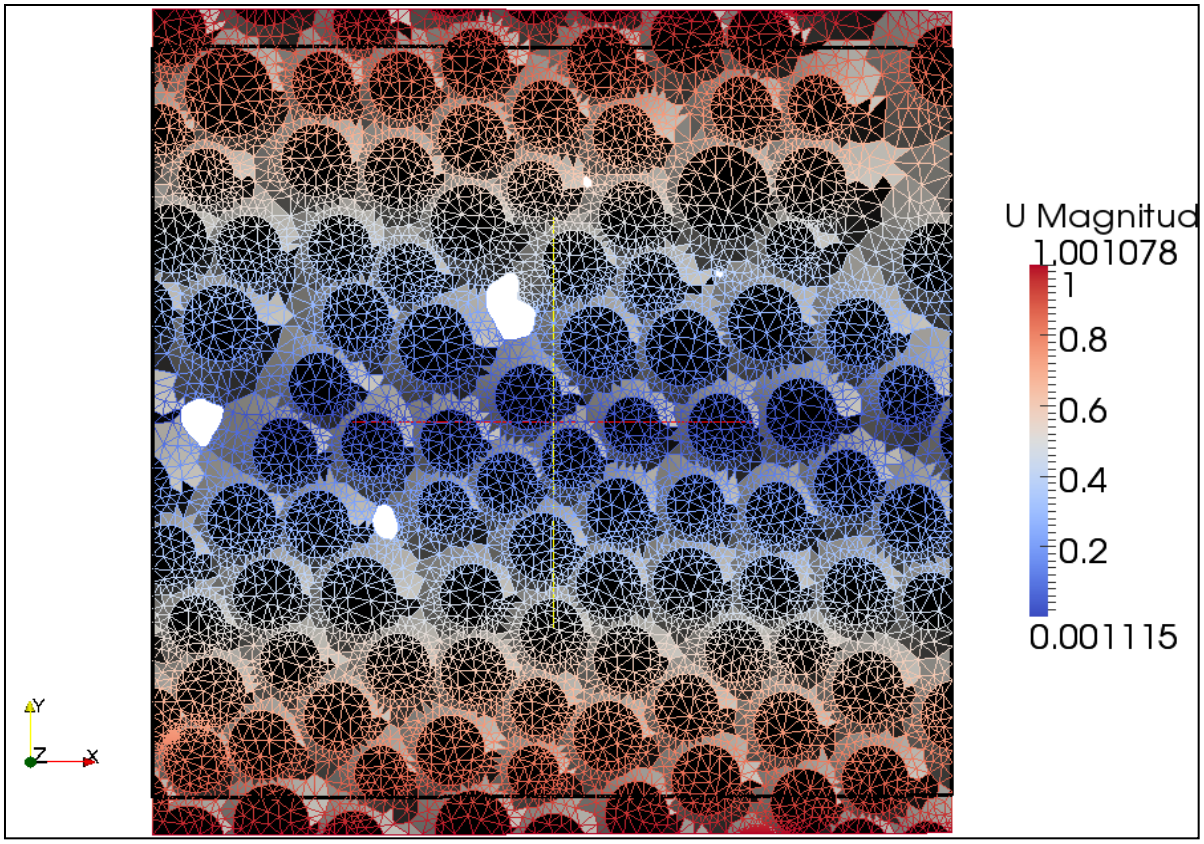
FEM analysis

Tensile along X



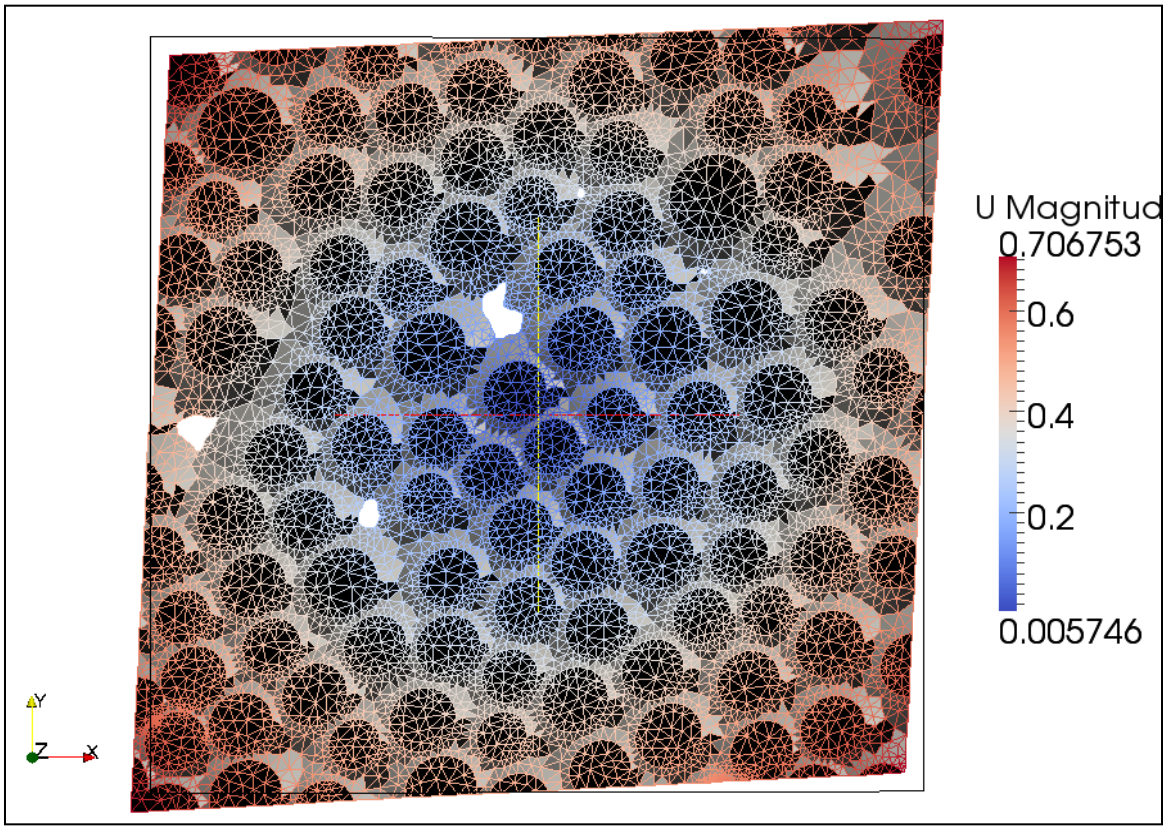
FEM analysis

Tensile along Y



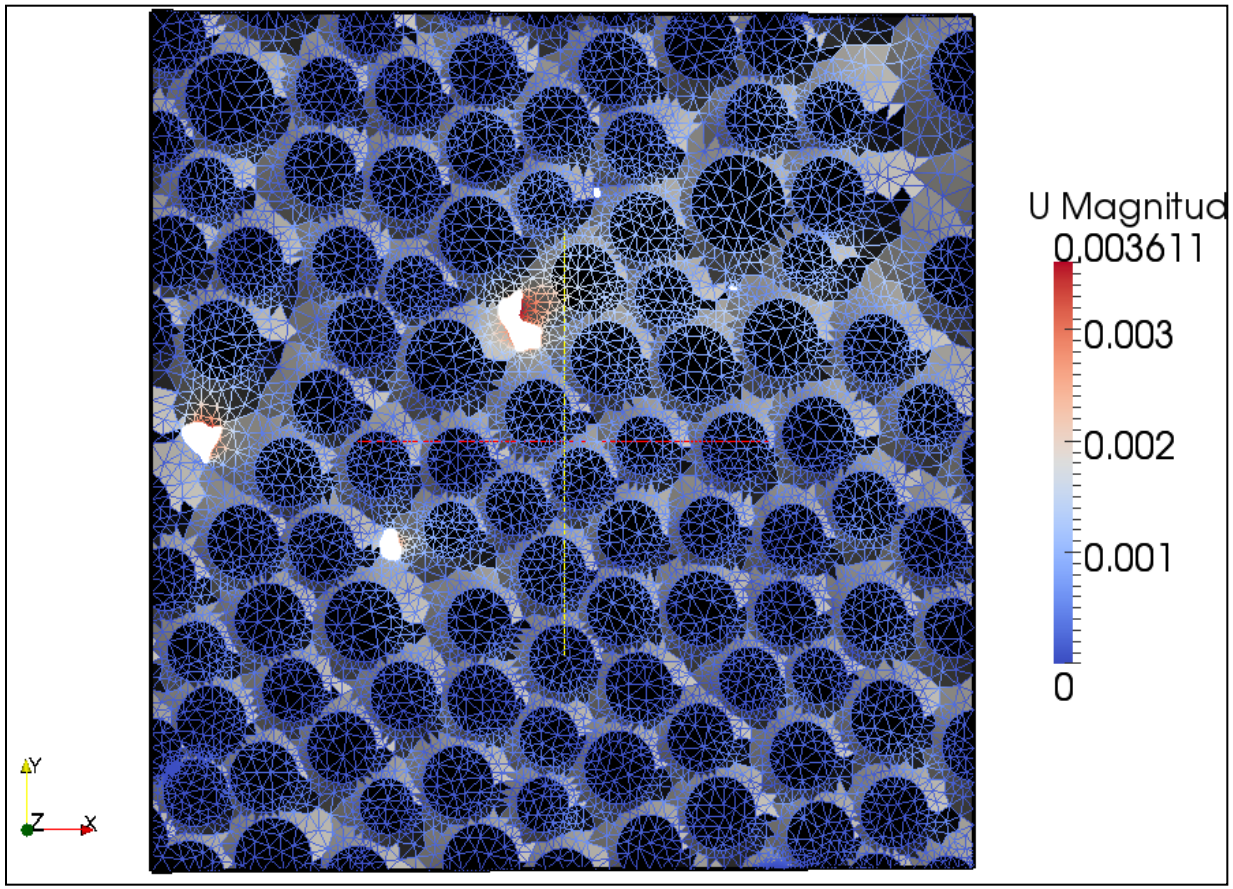
FEM analysis

XY shear

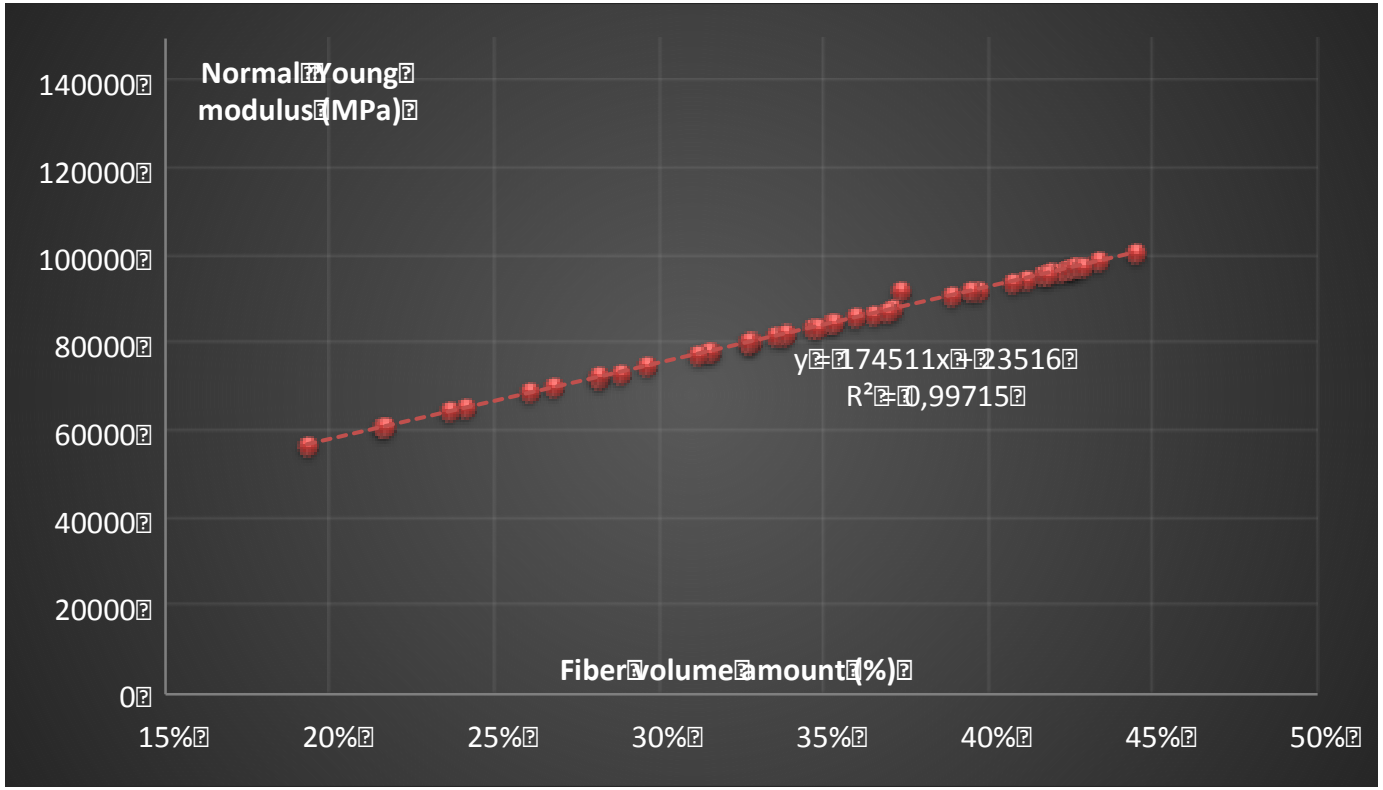


FEM analysis

Heat expansion

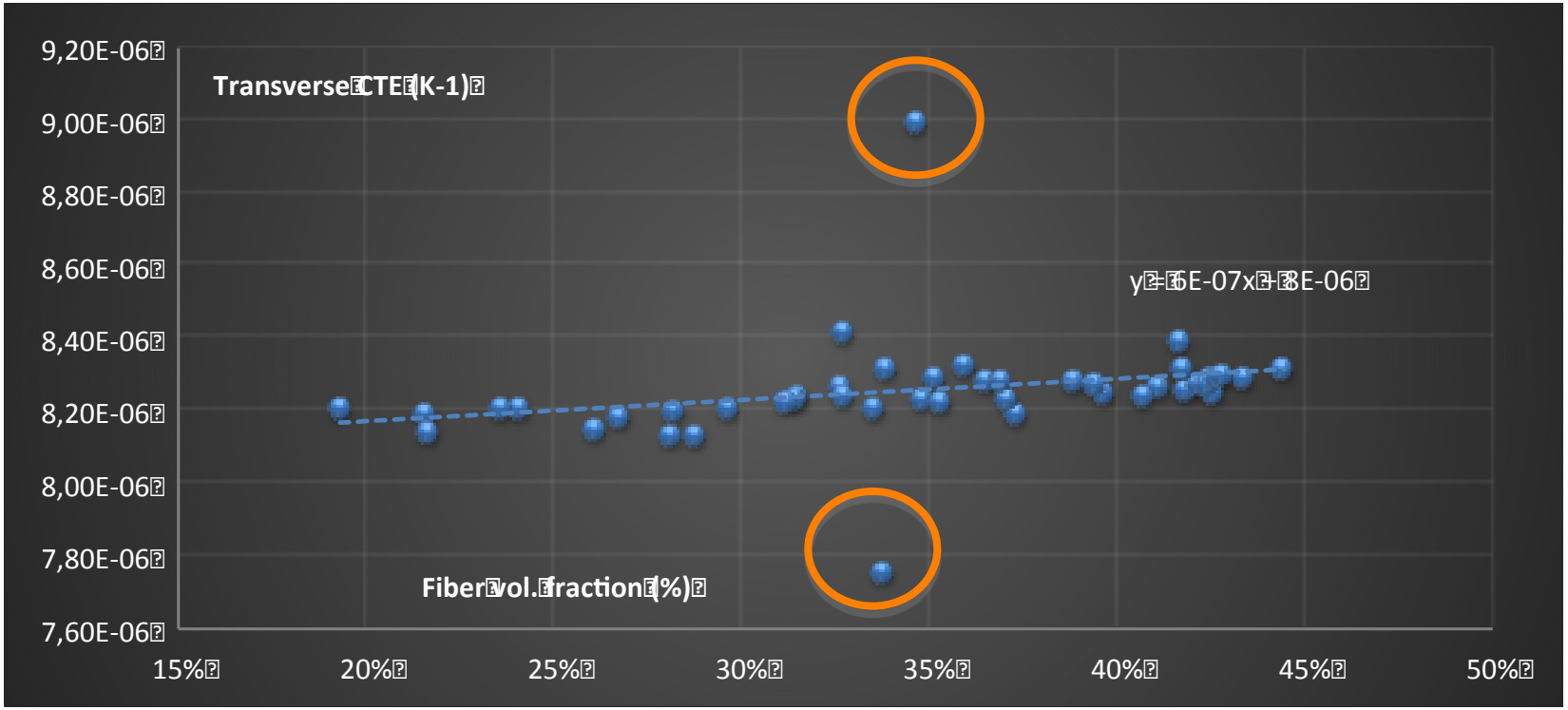


Micro-scale results

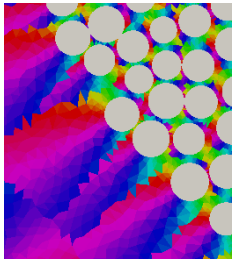


Elastic constants scale with the fiber volume amount

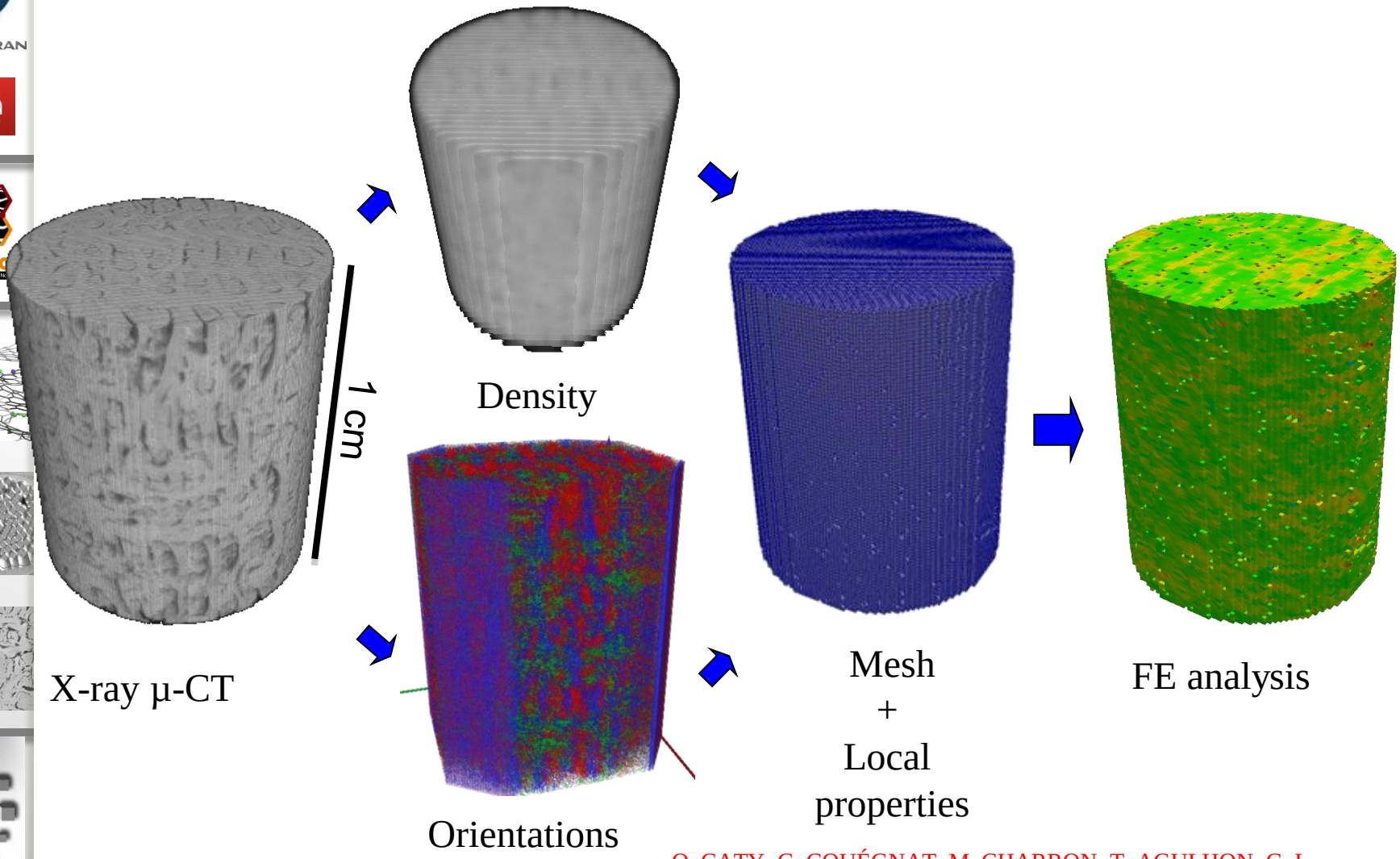
Micro-scale results



CTE scales with the fiber volume amount
… except close to bundle edges !



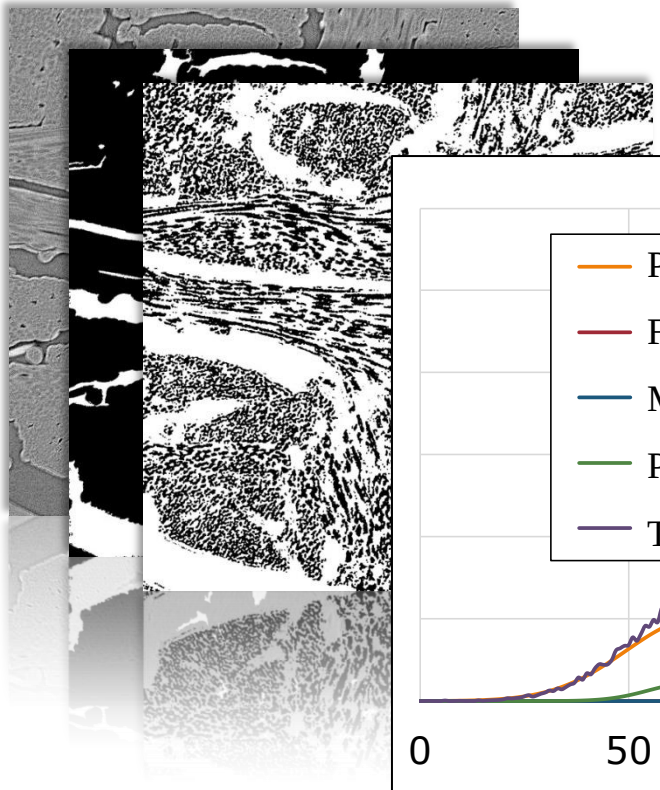
Macro model



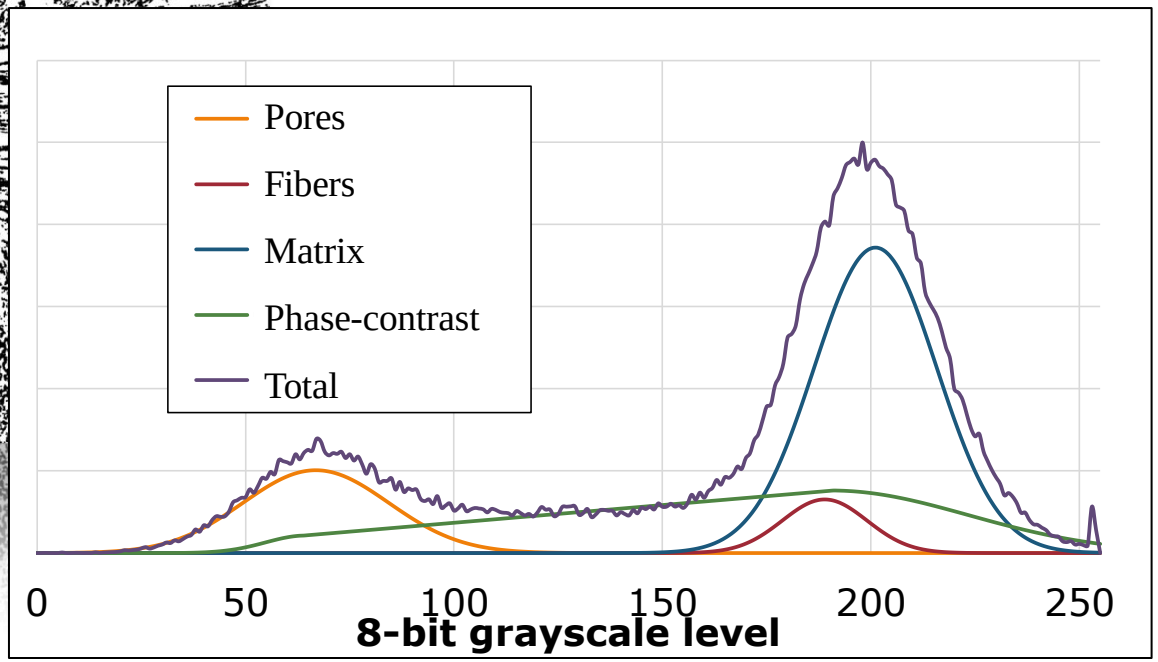
O. CATY, G. COUÉGNAT, M. CHARRON, T. AGULHON, G. L. VIGNOLES, *Ceram. Trans.*. 248, *36-41 (2014)*

Grayscale/composition relationship

Study of the composition on detailed images

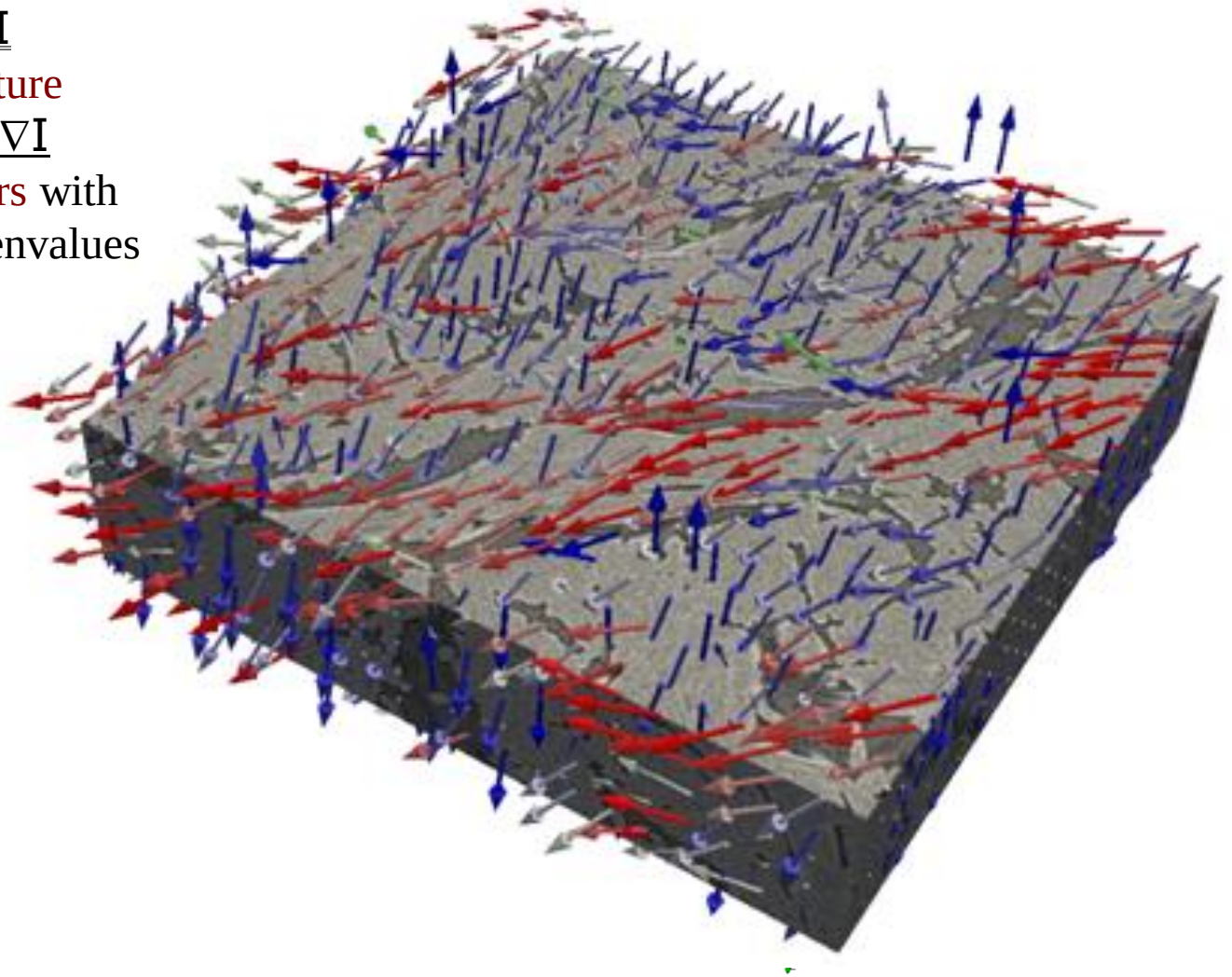


Histogram modeling

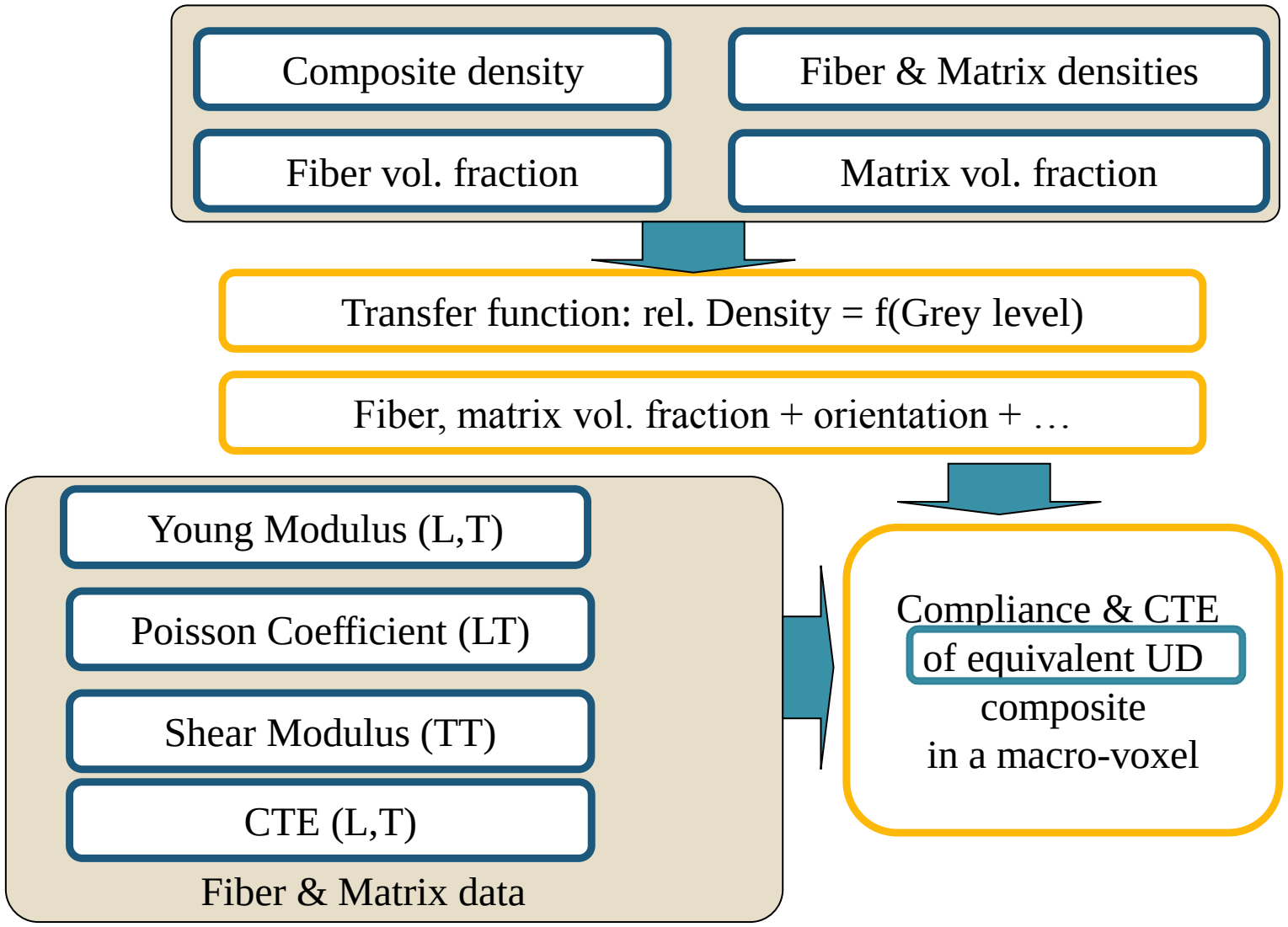


Fiber direction detection

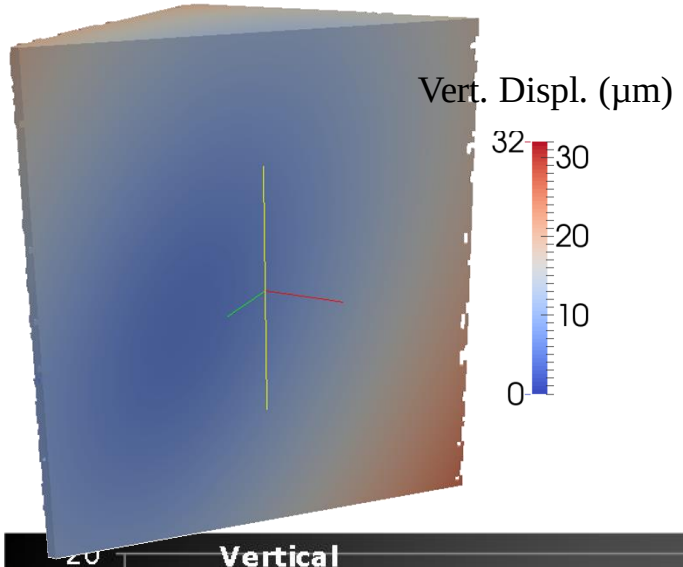
Based on Hessian matrix $\underline{\underline{\nabla^2 I}}$
or on structure tensor ${}^t\nabla I . \nabla I$
eigenvectors with lowest eigenvalues



Macro model input data

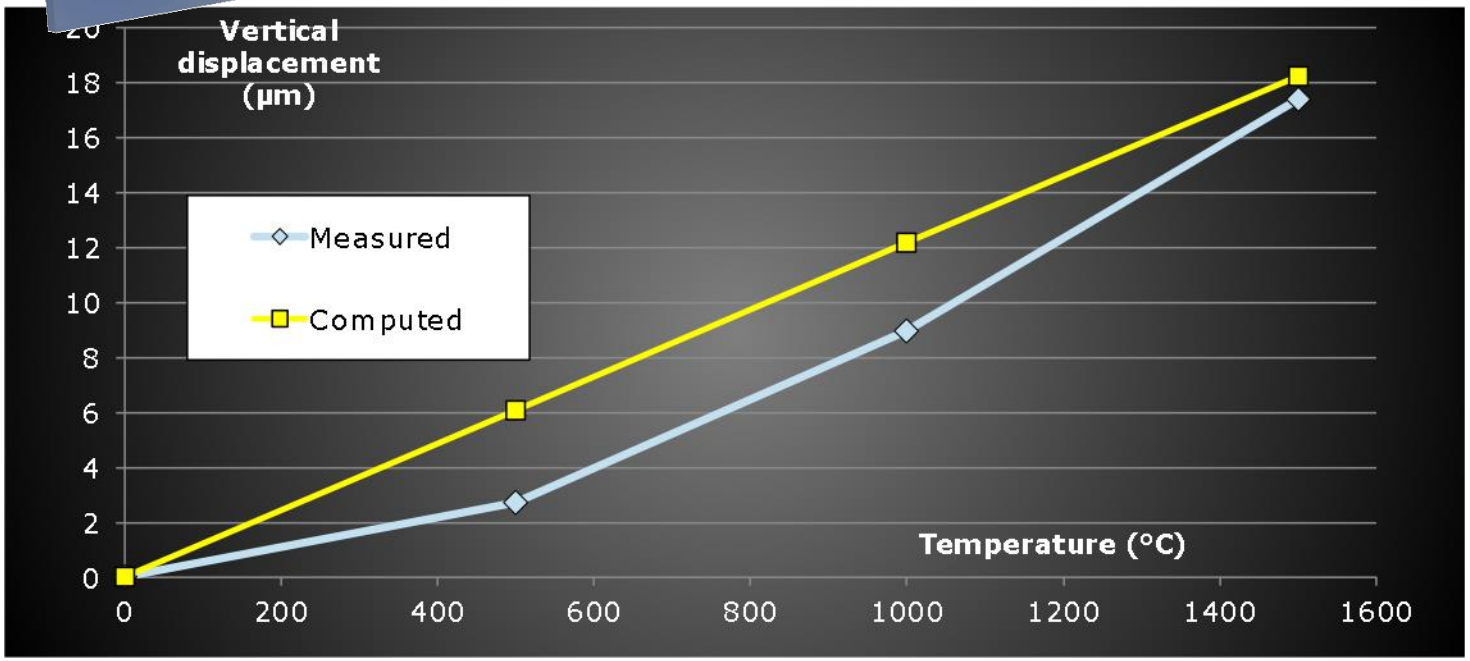


Large-scale results



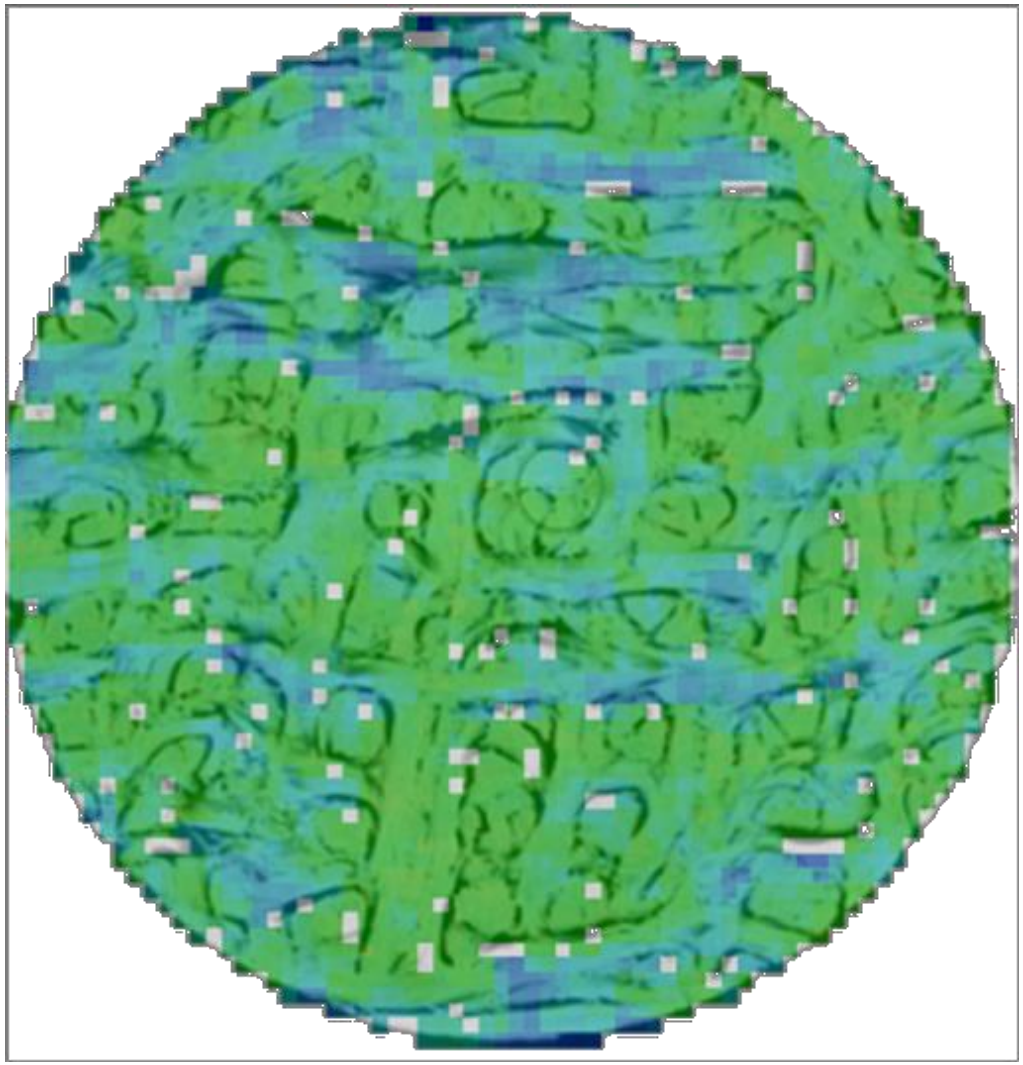
Properties set from:
Tsukrov I., Drach B., Gross T., 2012.
Int. J. Eng. Sci **58**, 129–143.
→ Do not depend on temperature !

~ 10^7 dof
CPU times: Assembly : 1 hr…,
Matrix solution: 10 min.



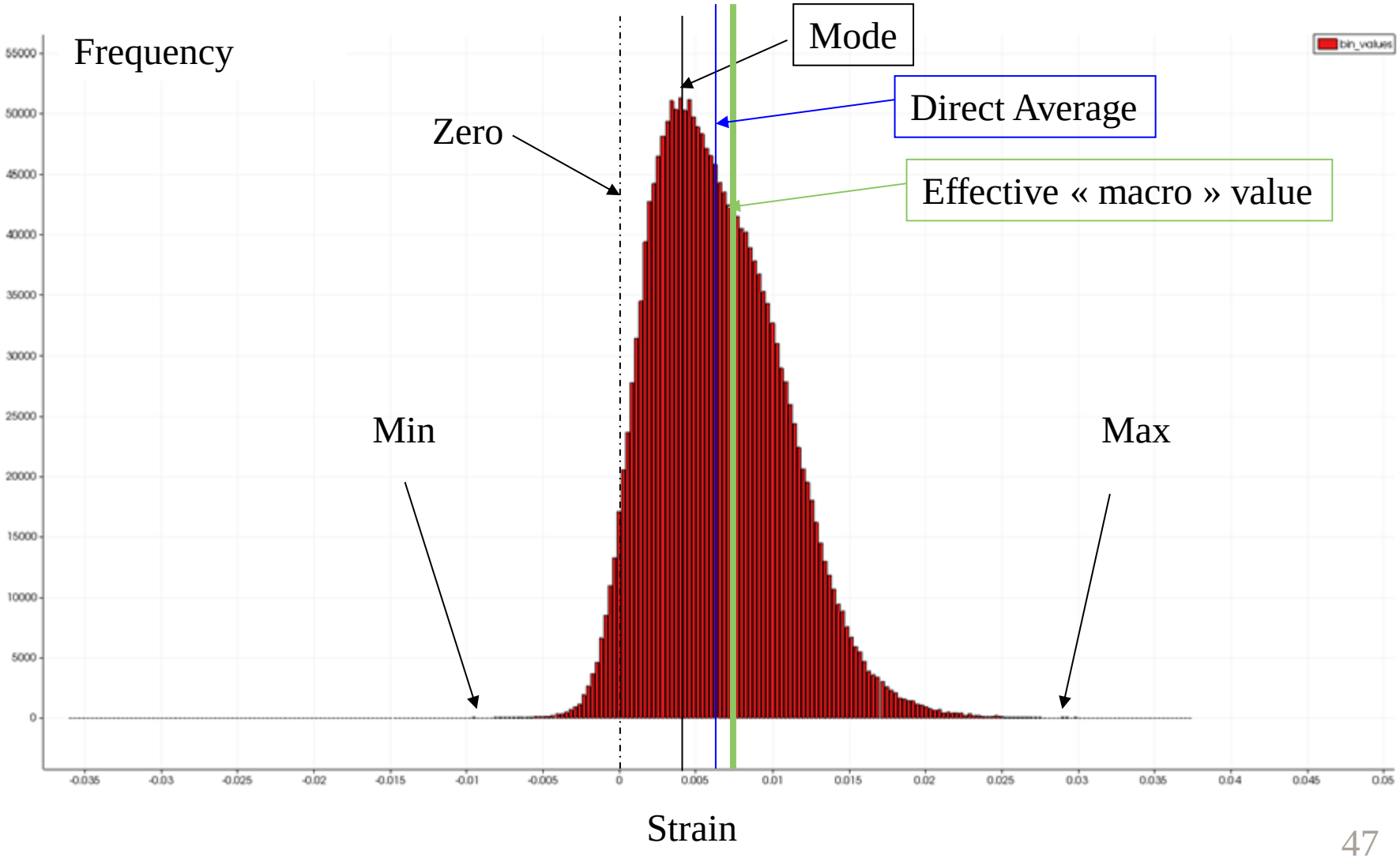
Macro model results

YY Strain & comparison with GL



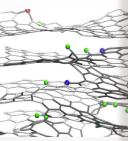
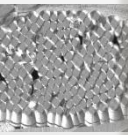
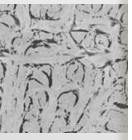
O. CATY, G. COUÉGNAT, M. CHARRON, T. AGULHON, G. L. VIGNOLES, *Ceram. Trans.*. 248, *36-41 (2014)*

Strain histogram



Part II - Discussion

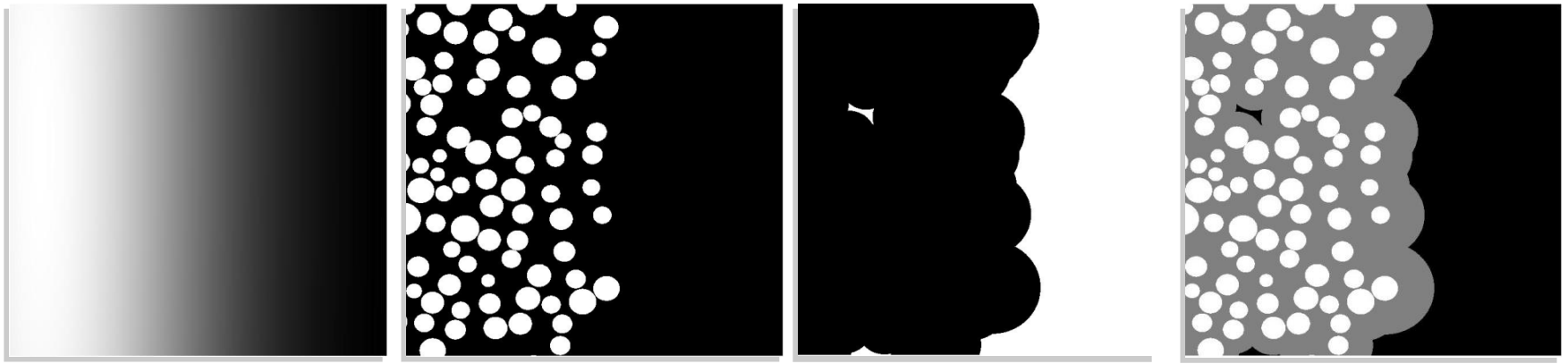
- Local study on PLOM images
  - Computation of effective properties
  - Watch out ! Gradient-containing images …
- Properties correlate well with fiber vol. fraction
  - … except when gradients are present (bundle edges)
- Macroscale study on X-ray CT 3D blocks
  - Captures well the effect of material structure on CTE
  - Depends on the input values for the constituents
- Comparison w.r.t. TMA : satisfactory
  - But input properties still only come from literature …

Part II - Outlook

- Input data values : still an open question
  - Local characterization is ongoing
  - Get input values from atomistic modelling …

- Gradient-containing subimages : how to deal ?

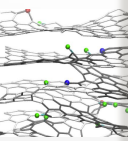
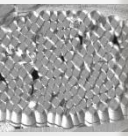
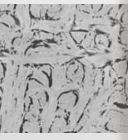


- Insert damage mechanics

- Another step toward « realistic virtual » materials …

Conclusion

- Image-based modeling : a versatile tool
  - Image-Guided Atomistic Reconstruction
  - Image-to-FE mesh procedures
- Able to deal with « realistic » media
  - Involves constrained image generation
  - Anisotropy has to be handled
- Gives insight into structure-properties relationships
- Developed tools may apply to other materials …
  either **real** or **virtual**.

Acknowledgements

PyroMaN Pyrocarbon Matrices at the Nanoscale

http://pyroman.dr15.cnrs.fr

PARTNERS

CEMES

LCTS

ims

FUNDING

INSTITUT CARNOT MIB

ANR

aerospace valley

Work on thermal expansion modelling:

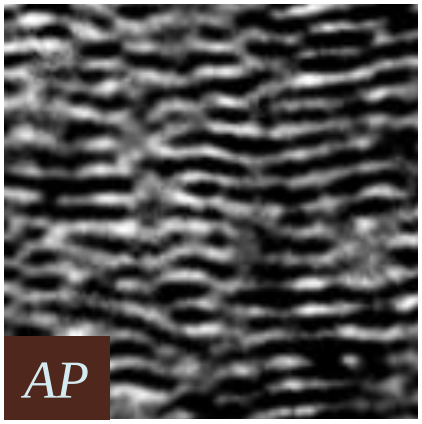
cea

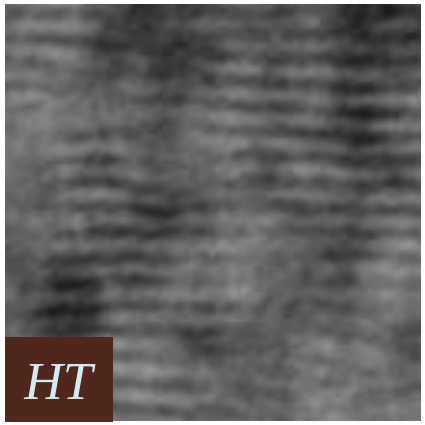
SAFRAN Herakles

Thank you for your attention

Any question ?

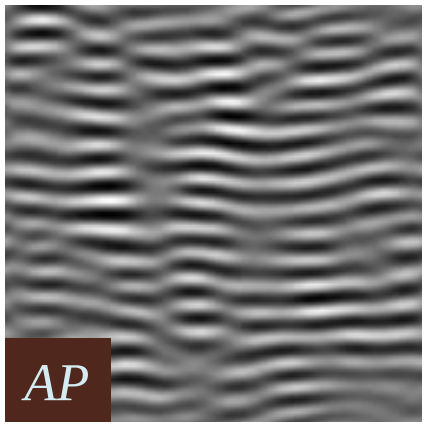
Generation of 3D HRTEM-like images - 1

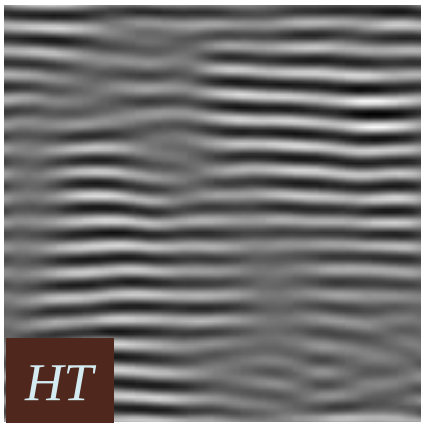




1) Image Filtering

Application of radial and directional band-pass filter
→ removes low frequency (background gradients) and high frequency (noise) artifacts





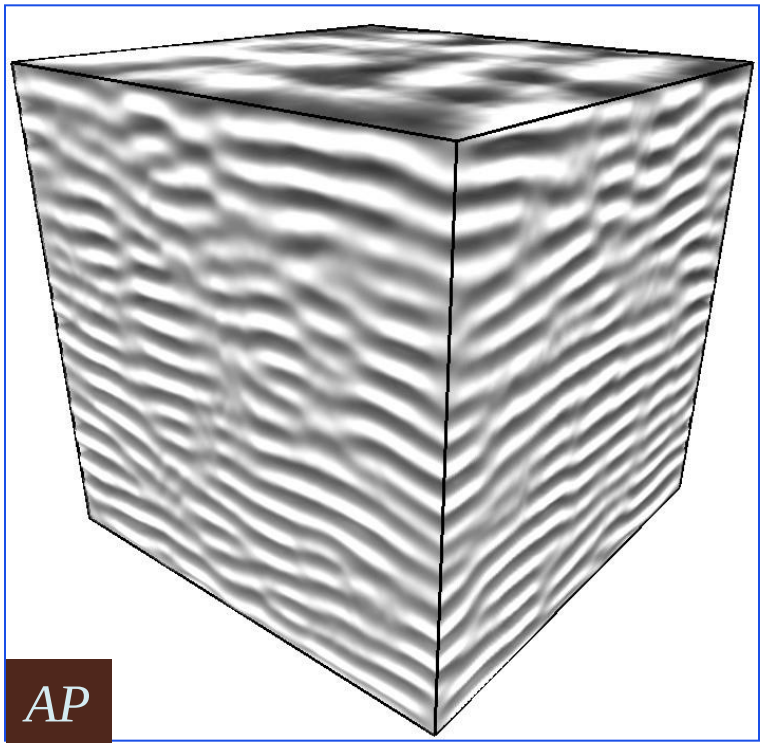
2) Image Analysis

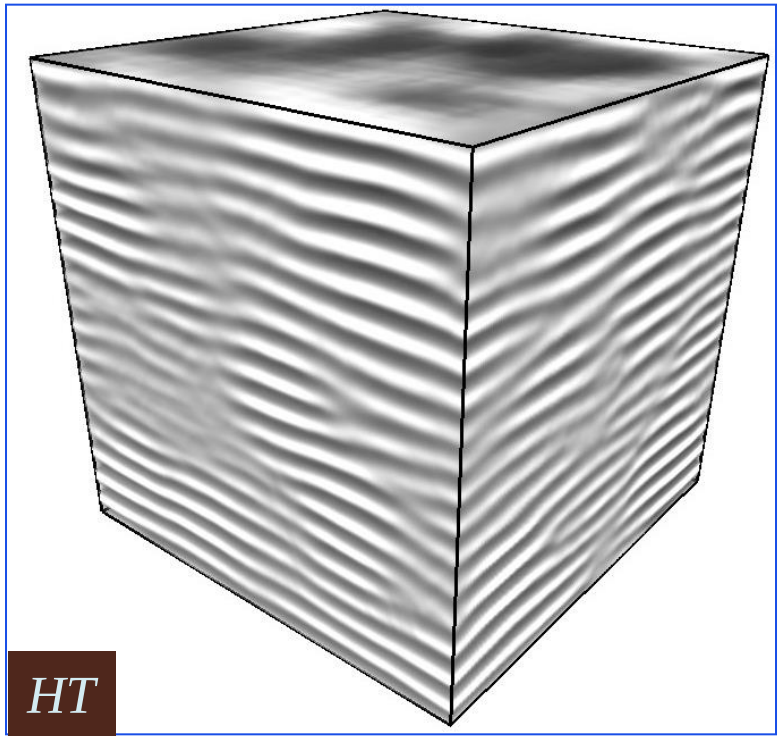
Pyramidal decomposition + calculation of 1st order (mean, variance, skewness, kurtosis) and 2nd order (autocorrelation) statistics

Generation of 3D HRTEM-like images - 2

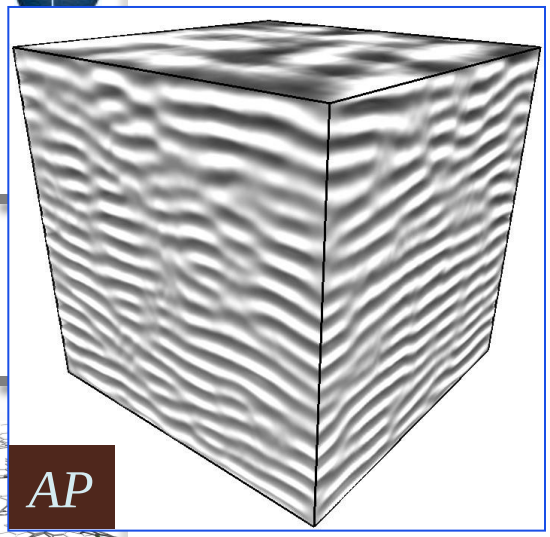
3) 3D Image synthesis

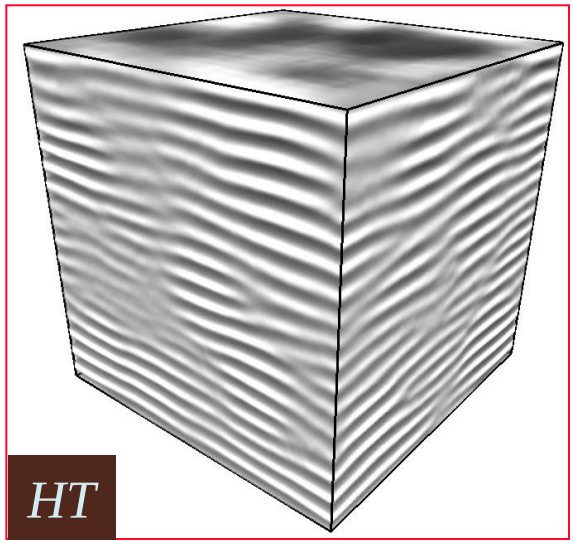
1) 3D extension of the 2D pyramidal statistics under an orthotropy hypothesis
2) Iterative refinement of an initially random 3D image to satisfy the 3D statistics

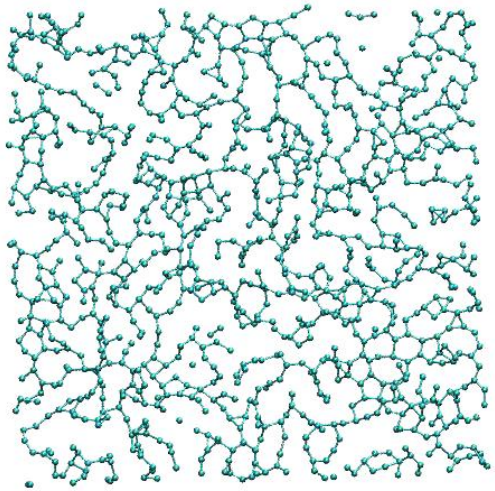


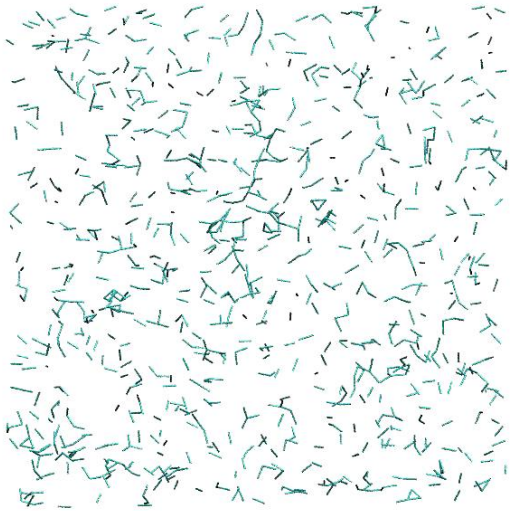


IGAR : Image Guided Atomistic Reconstruction









Images are filled with carbon atoms with

$d = 2.1\ g/cm^2$

$d_{002} = 3.5\ Å$

Molecular Dynamics quenching

$$\frac{d^2\mathbf{r}_i}{dt^2} = \frac{d\mathbf{v}_i}{dt} = -\frac{1}{m_i}\frac{d\left(U_{REBO} + U_{IMAGE}\right)}{d\mathbf{r}_i}$$

with empirical reactive potential

$$U_{REBO} = \sum_{i=1}^{N}\sum_{j=i+1}^{N}\left[V^R(r_{ij}) - b_{ij}V^A(r_{ij})\right]$$

$$\begin{pmatrix} V^R(r) = f^c(r)\left[1 + Q/r\right]A\exp(-\alpha r); \\ V^A(r) = f^c(r)\sum_{n=1,3} B_n \exp(-\beta_n r) \end{pmatrix}$$

and an external force field imposed by the 3D image :

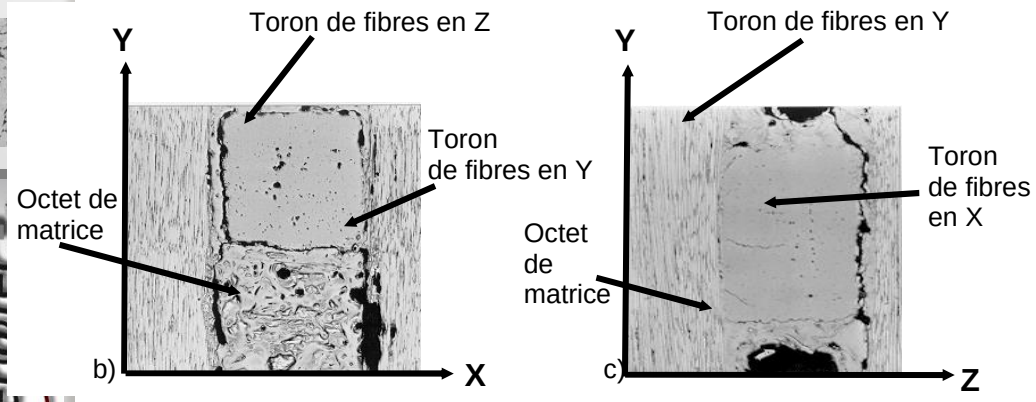
$$U_{IMAGE}(eV) = 2\sum_{i=1}^{N} I(\mathbf{r}_i)$$

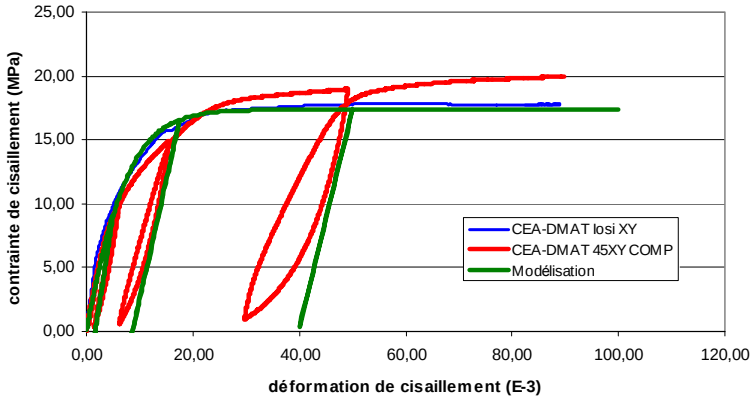
J.M. LEYSSALE, J.-P. DA COSTA, C. GERMAIN, P. WEISBECKER and G. L. VIGNOLES, Carbon (2012), **50**, 4388–4400.

Some past studies:3D

- U. Sydney
  - Micromechanics models for mechanical and thermomechanical properties of 3D through-the-thickness angle interlock woven composites
    - P. Tan, L.Tong, G.P. Steven, *Composites Part A* **30**(5) 637–648 (1999)

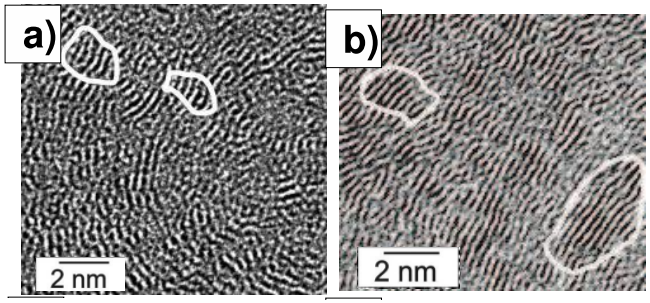
- CEA+INSA Lyon+ENSMA Poitiers
  - Macroscale model for 3D C/C mechanical behavior
    - C. Tallaron, S. Barre. *Adv. Comp. Lett.* **8**(5) 239-247 (1999)
    - L. Flacelière, C. Tallaron, *Proc. Matériaux 2006* (France)

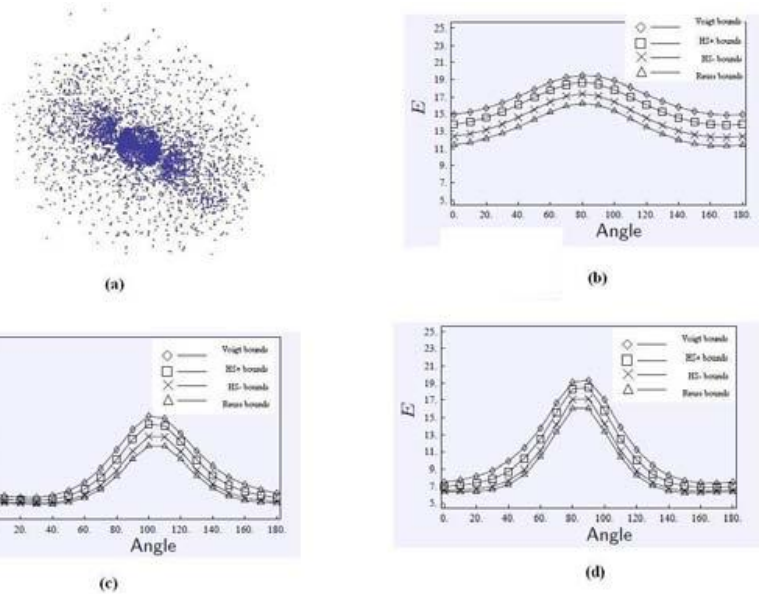




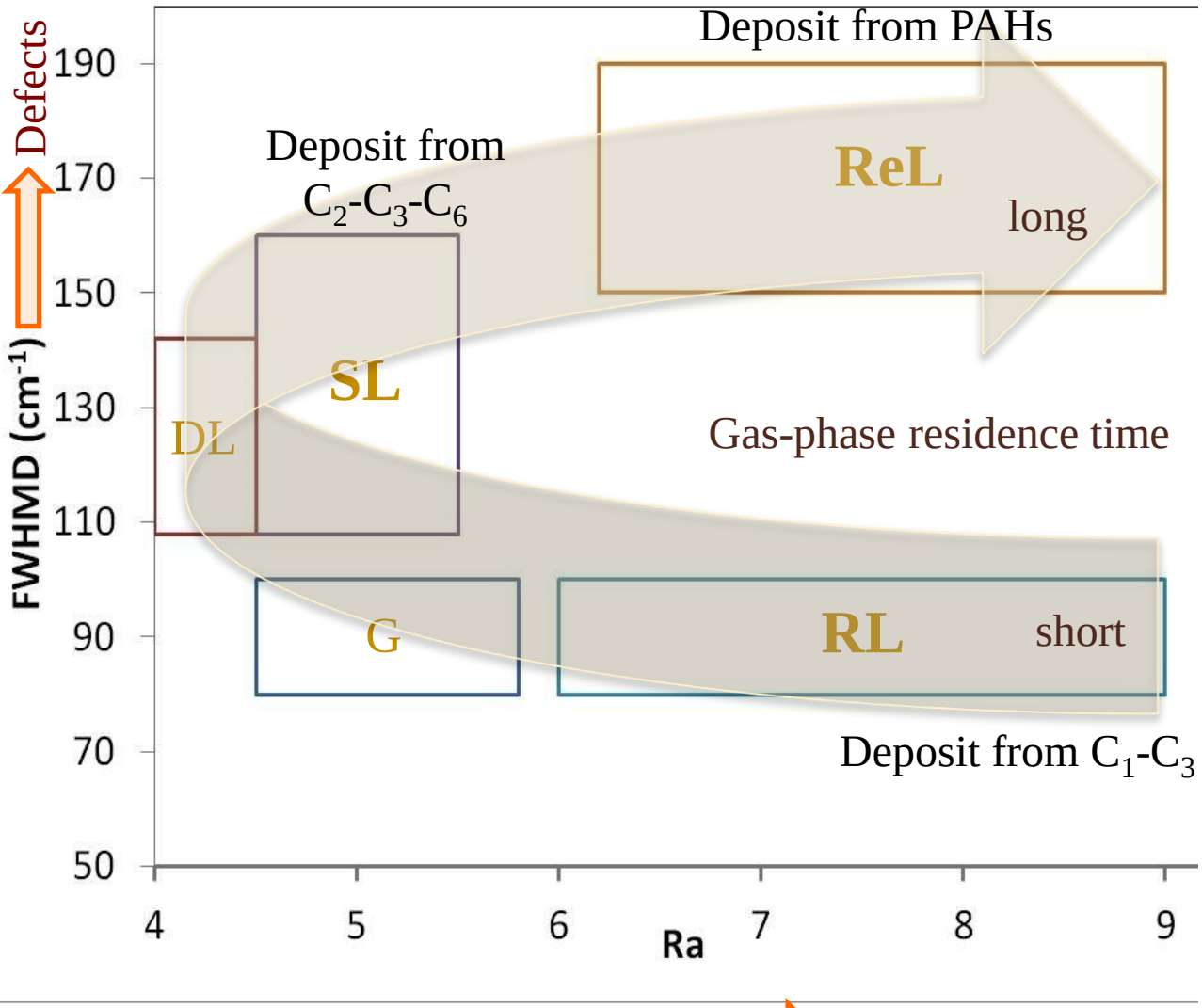
Structure-property relationship: former work

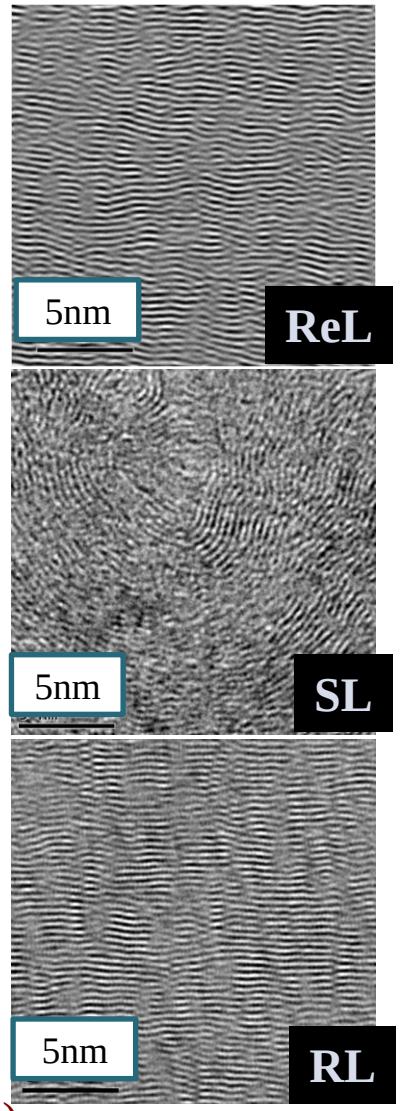
- KIT
  - Pyrocarbons at nanoscale
  - TEM LF imaging → 2D image processing → statistics → upper & lower bounds estimates of **CTE** & elasticity
    - Böhlke, T., Langhoff, T.-A., Lin, S. and Gross, T. (2013), ZAMM **93**, 313–328.
    - Böhlke,T., Lin, S., Piat, R., Heizmann, M., Tsukrov, I. (2010) PAMM **10**, 281-282.





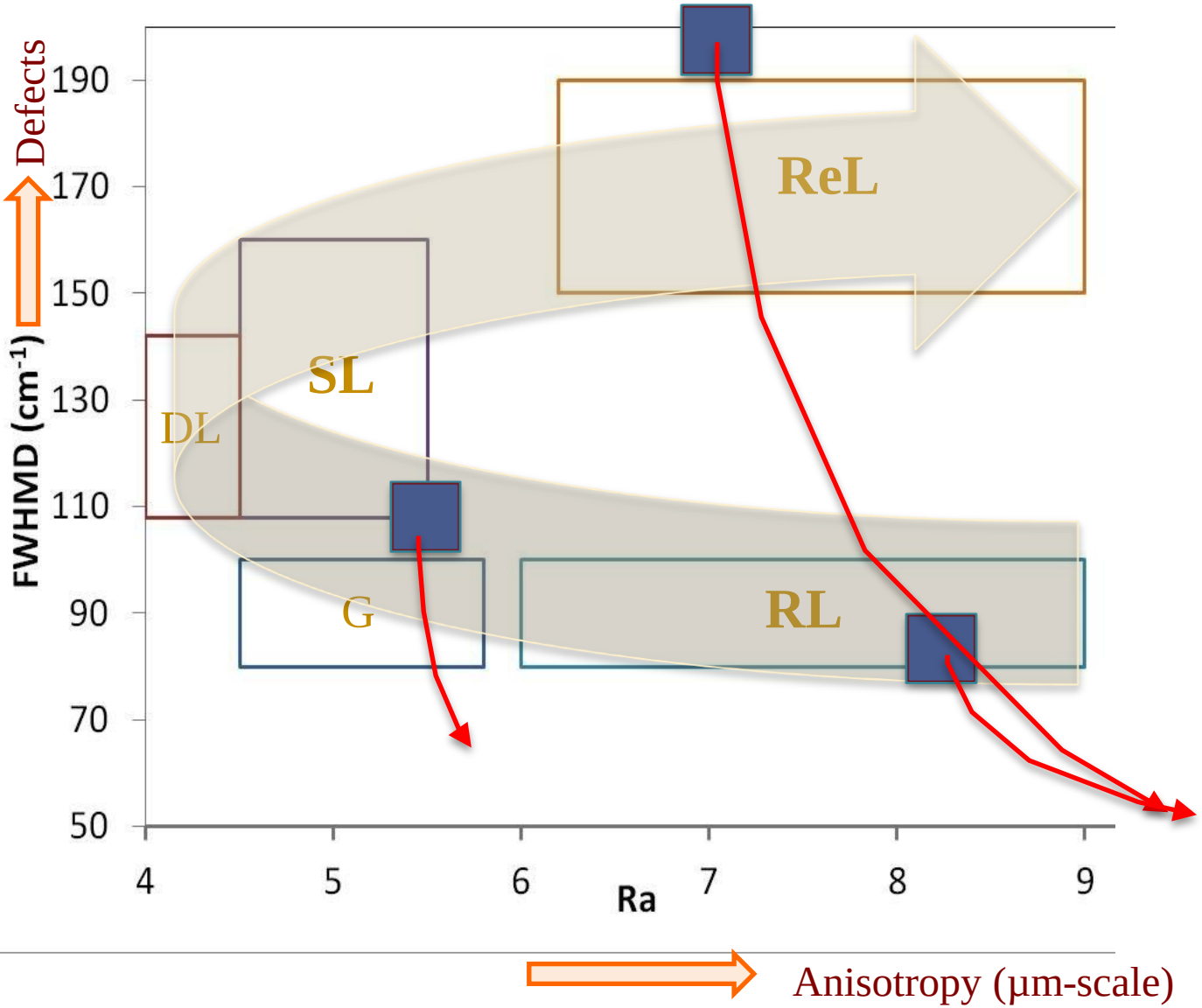
Raman-based Classification & relationship with processing parameters





(X. Bourrat et al., *J. Braz. Chem. Soc.* **17**(6) (2006),1090-1095.

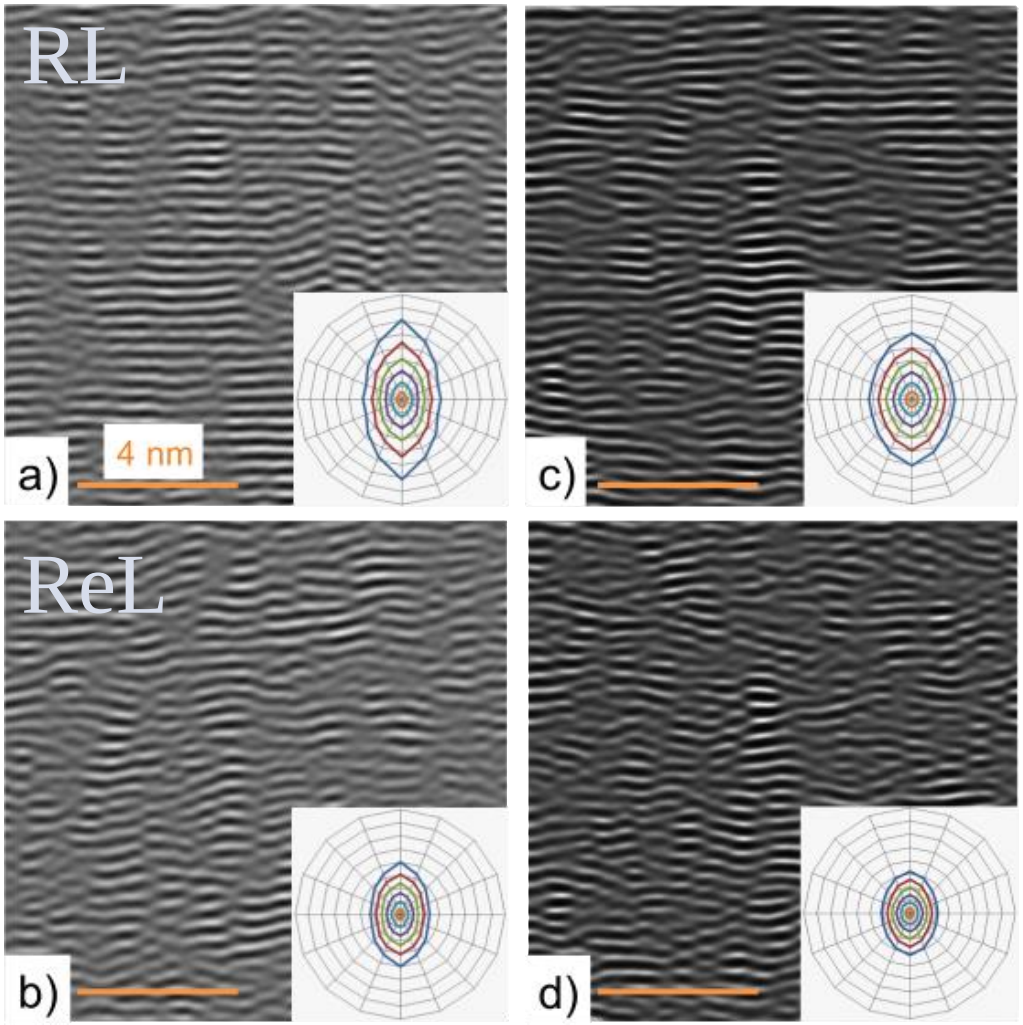
Our samples within the classification

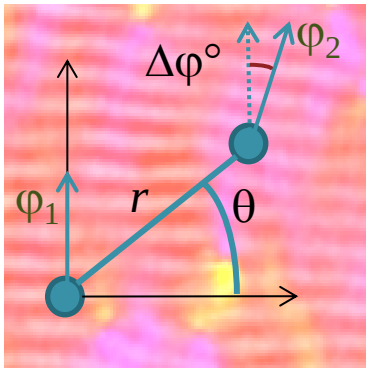


As prepared

Evolution by HTT

Fringe Orientation Correlation : model *vs.* experiment





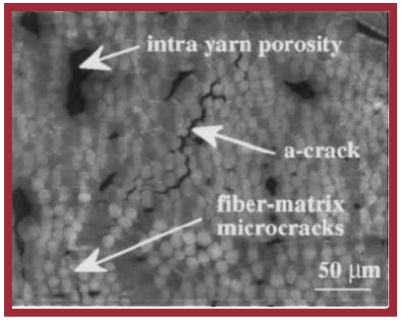
Maps of $\Delta\varphi^\circ = f(r,\theta)$

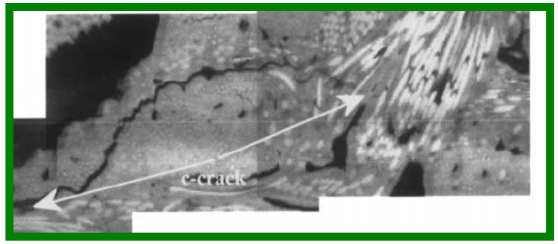
Excellent agreement in the horizontal direction

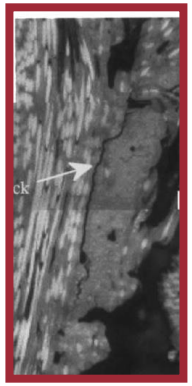
Order along 002 underestimated by models

Some past studies: stitched 3D

- LCTS:
  - 3D stitched C/C mechanical behavior : elasticity+ damage
    - O.Siron, J.Pailhès, J.Lamon, *Comp. Sci. & Tech.* **59**(1), 1–12 (1999)
    - J. Pailhès, G. Camus, J. Lamon, *Mech. Mater.* **34**(3), 161-177 (2002)

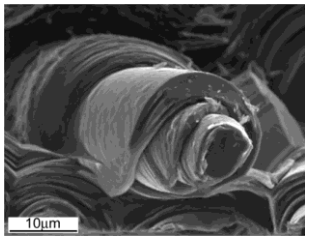






Some recent advances

- KIT & NHU:
  - « 3D » : felts & woven plies: elastic properties
  - CT-scans → image processing → FEM on parts of material + analytical mixing rules
    - Stasiuk, G., Piat, R., Deshpande, V. V., Mahajan, P., *Ceram. Eng. Sci. Procs* **34**(10), 213-220 (2013)
    - Drach, B., Tsukrov, I., Gross, T., Dietrich, S., Weidenmann, K., Piat, R., Böhlke, T., *IJSS* **48**(18), 2447-2457 (2011)
    - S. Dietrich, J.-M. Gebert, G. Stasiuk, A. Wanner, KA Weidenmann, O Deutschmann, I Tsukrov, R. Piat, *CSTE* **72**, 1892-1900 (2012)



Reznik & Gerthsen, Carbon 2001