Multiscale mechanical analysis of cellulose nanofibril suspensions

Crystalline cellulose plastifies by localized shear

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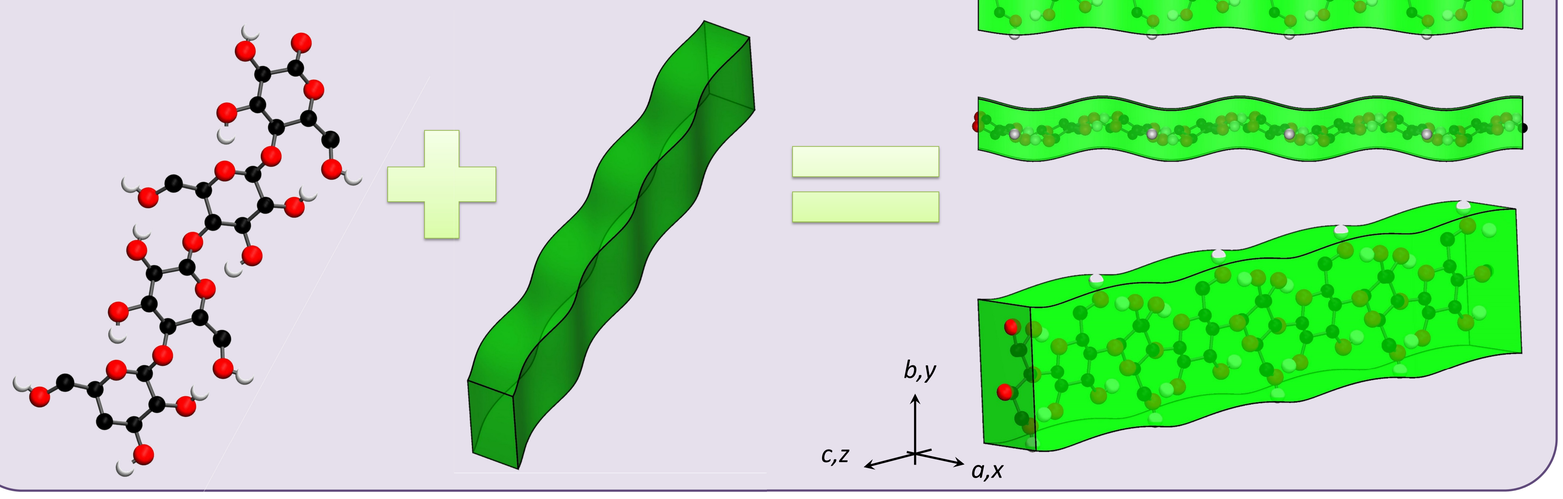
Introduction

Objective

- **Anisotropic** response of cellulose
- **Elasto-plastic** deformation

Main idea

- Analogy with **corrugated** structures



Methods

Initial structure

- GROMACS package version 5.1.
- Gromos 56A$_{carbo}$ force field
- no LINCS algorithm
- **periodic** boundaries
- **cellulose Iβ** with 30-150 chains (3360-50000 atoms)

Deformation of crystals

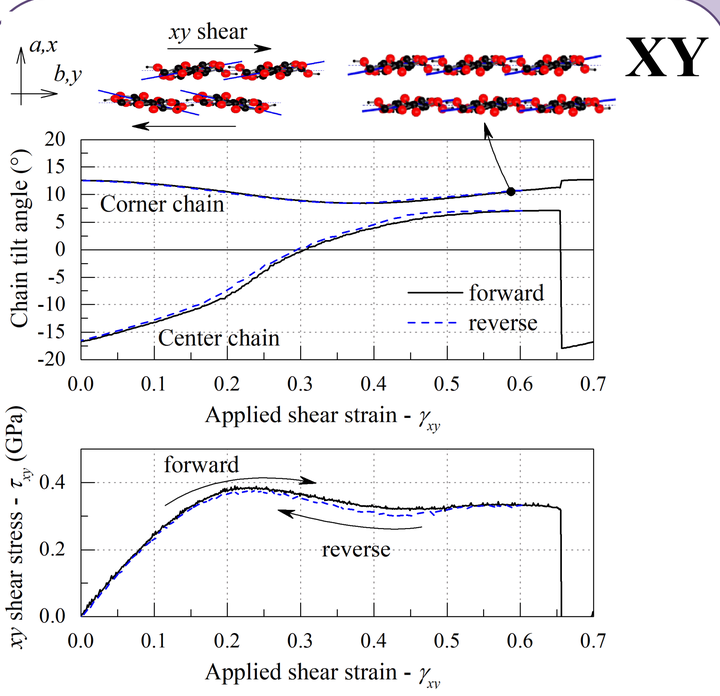
- The cellulose crystals were deformed **quasi-statically** using molecular mechanics
- The shear deformation was applied at constant **zero pressure**

Coarse-graining for post treatment

The coarse-graining technique is a method which uses a Gaussian convolution to calculate continuum quantities (e.g. strain) based on discrete results (e.g. the displacement).

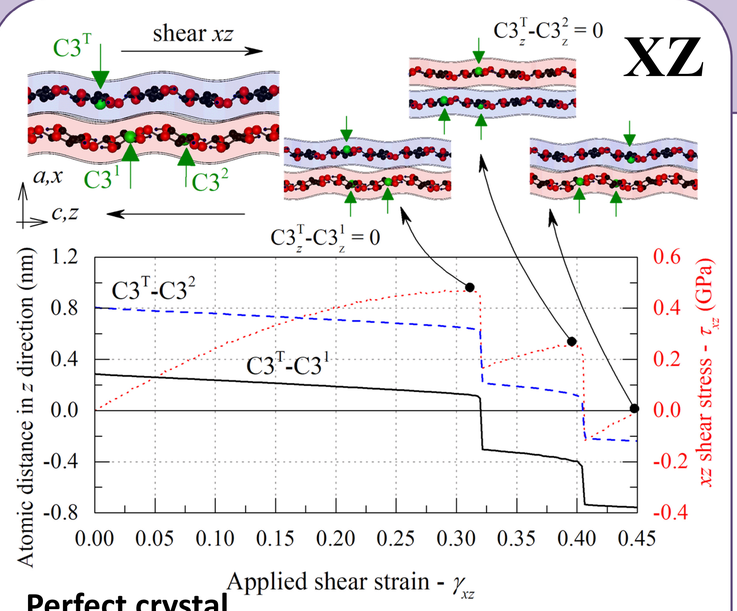
Shear deformation

XY



Internal rotation around chain axis (*c,z*)

XZ

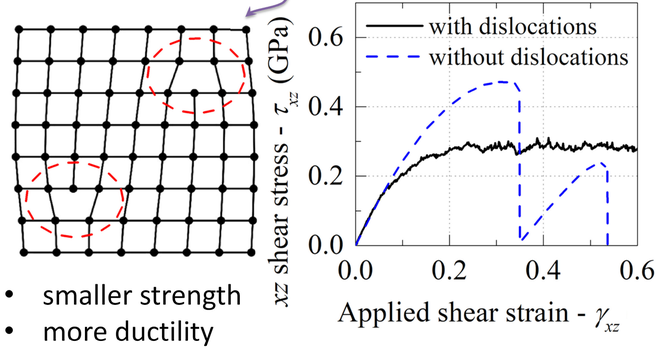


Perfect crystal

- **Corrugated** ribbon like response
- Two equilibrium positions
- Expansion in *a,x* direction

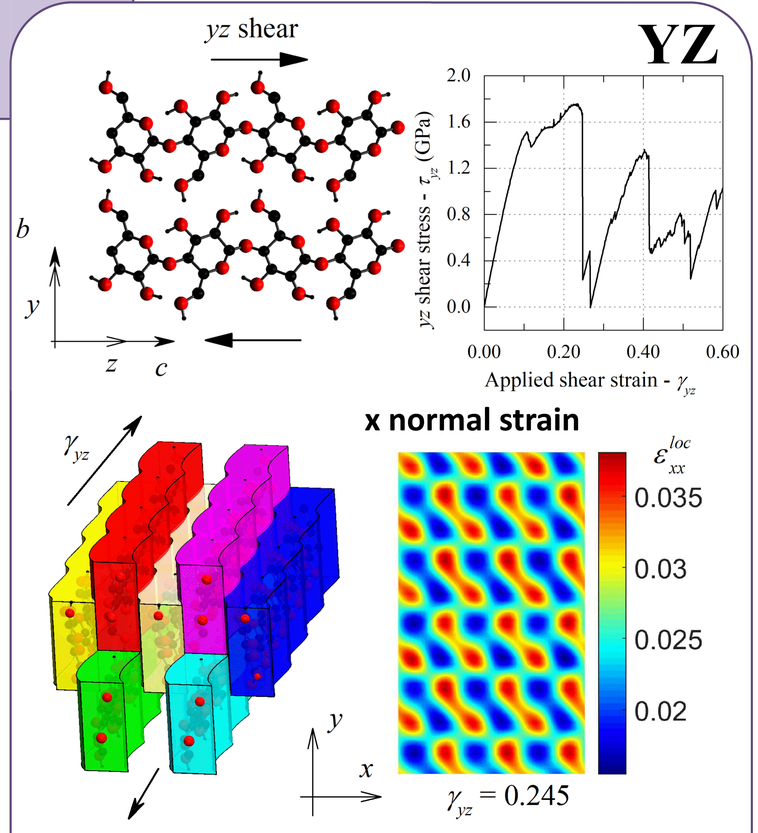
With 2 dislocations

schematic representation of crystalline unit cells (xz)



- smaller strength
- more ductility

YZ



Corrugated ribbons

- **Shear band** is larger and localized between chains.
- **Extension** in the two perpendicular (*a,x* and *b,y*) directions → noticeable dilatancy.
- Center and corners chains overlap → **interlocking**.
- Chains can only slide on top each other if they overcome the **resistance due to their corrugated shape.**

Summary

Global

- shear **stiffness** and **strength** is 5x higher in *yz* direction
- the actual **shape** of the **atomic structure** has a leading role on the macroscopic stress/strain response
- non isovolumic plasticity with dilatancy

XY plane

- **rotation** around axis *z* of each individual chain has a significant but reversible effect on the quasi-**hyperelastic** response of the material
- plasticity induced by **shear bands**

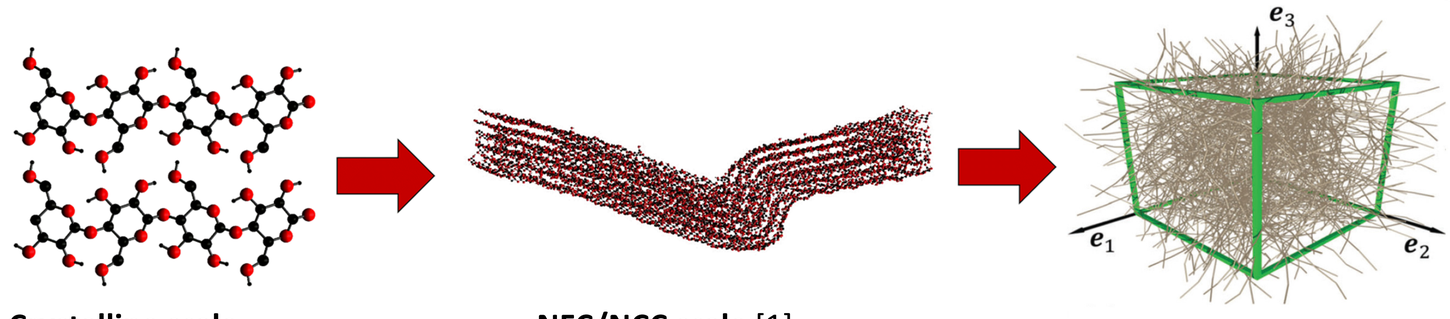
XZ plane

- **two equilibrium positions** based on local composition
- significant effect of possible **defects** such as dislocations
- plasticity induced by **shear bands**

YZ plane

- due to the more pronounced corrugated shape in this direction the structure is **interconnected** and thus stronger
- more **diffused** plasticity

Outlook



Crystalline scale

- Elementary mechanisms
- Shear strength
- Structural defects

NFC/NCC scale [1]

- Finite size effects
- Free surfaces
- Imperfect structure

NCF/NCC gels [2,3]

- Discrete element method
- Large deformations
- Continuum yield criterion

Contacts

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References

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[2] D Rodney, B Gadot, O. Riu Martinez, S Rolland du Roscoat, L Orgéas. Nature Materials, 15, 72-77 (2016).

[3] F. Martoia, P. J. J. Dumont, L. Orgéas, M. N. Belgacem, J.-L. Putaux , Micro-mechanics of electrostatically stabilized suspensions of cellulose nanofibrils under steady state shear flow, Soft Matter, 12, 1721 (2016).