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Orientation and assembly of cellulose nanofibrils in flow focusing

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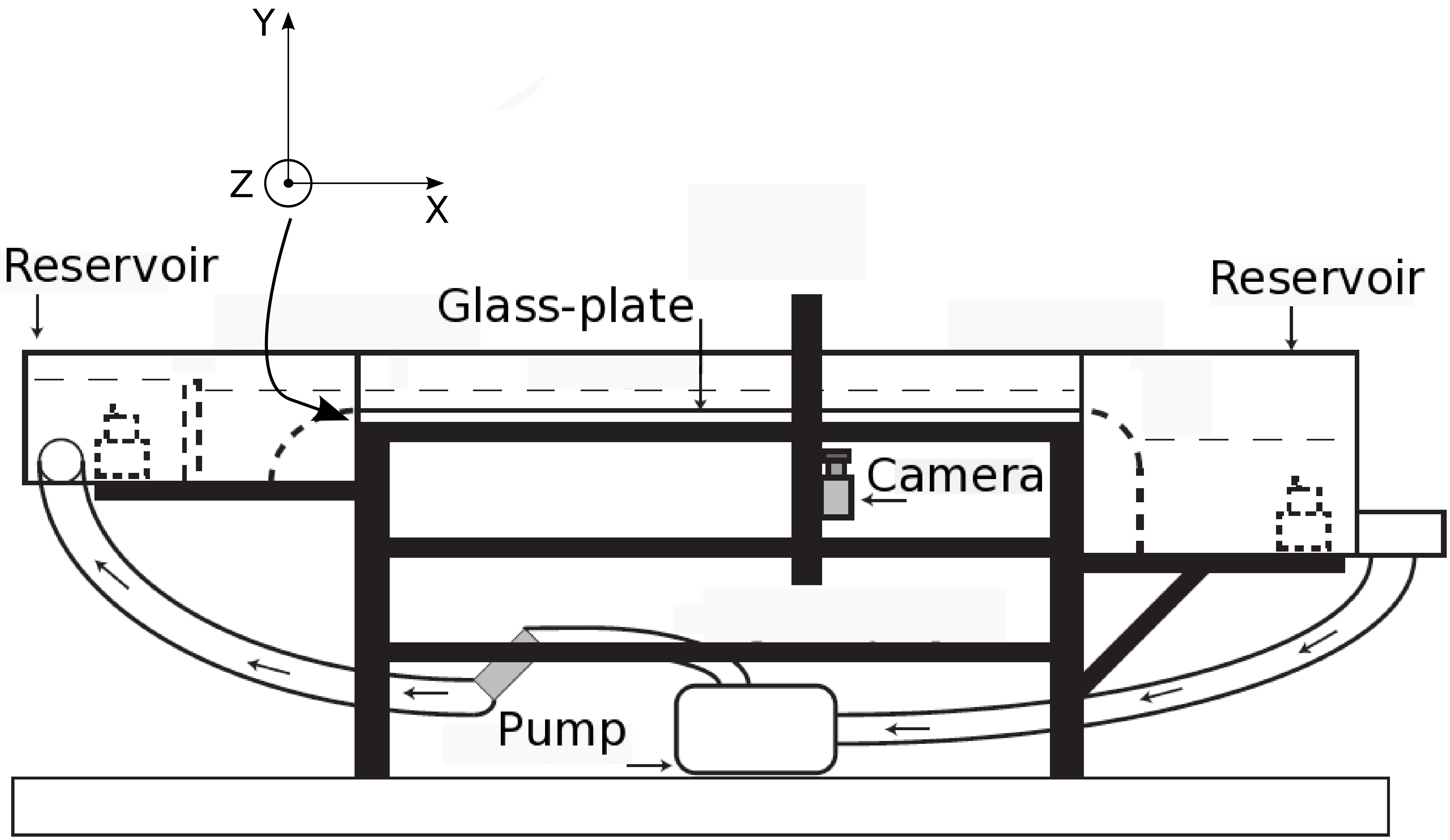
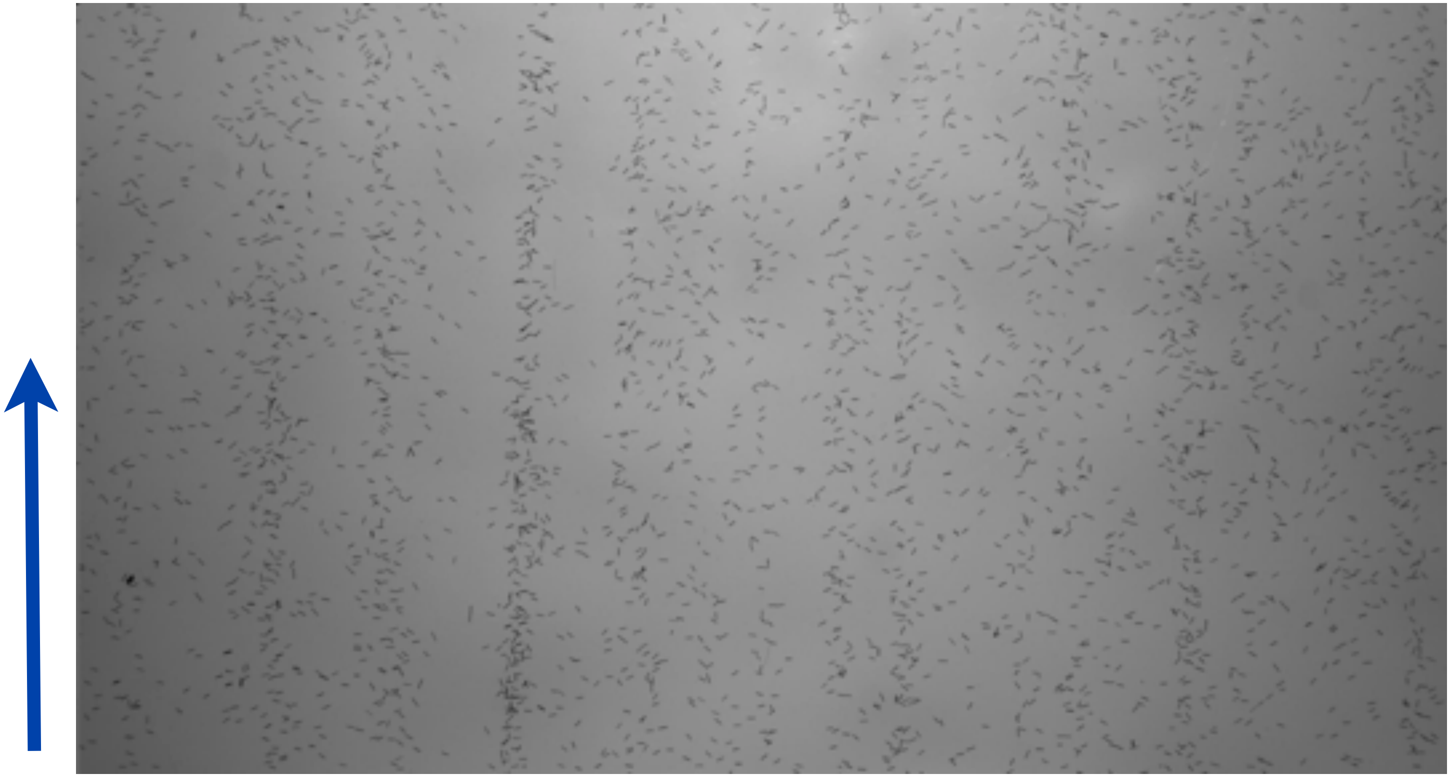
NORDITA, June 11th, 2014

WWSC is a joint research center at KTH and Chalmers

Fibres in/under a turbulent shear flow



Fibers in the flow



Flow direction

Dimensional analysis

- 9 physical parameters, 3 dimensions ->
  6 (!) dimensionless groups

Friction Reynolds number: $Re_\tau = \frac{hu^+}{\nu}$

Rotational particle Reynolds number: $Re_p = \frac{\dot{\gamma} l_f^2}{\nu}$

Rotational particle Stokes number: $St = \frac{\rho_p}{\rho_f} Re_p$

Concentration: $c = n l_f^2$

Particle aspect ratio: $r_p = \frac{l_f}{d_f}$

Sedimentation parameter: $S = \frac{T_{Jeff}}{T_{sed}}$

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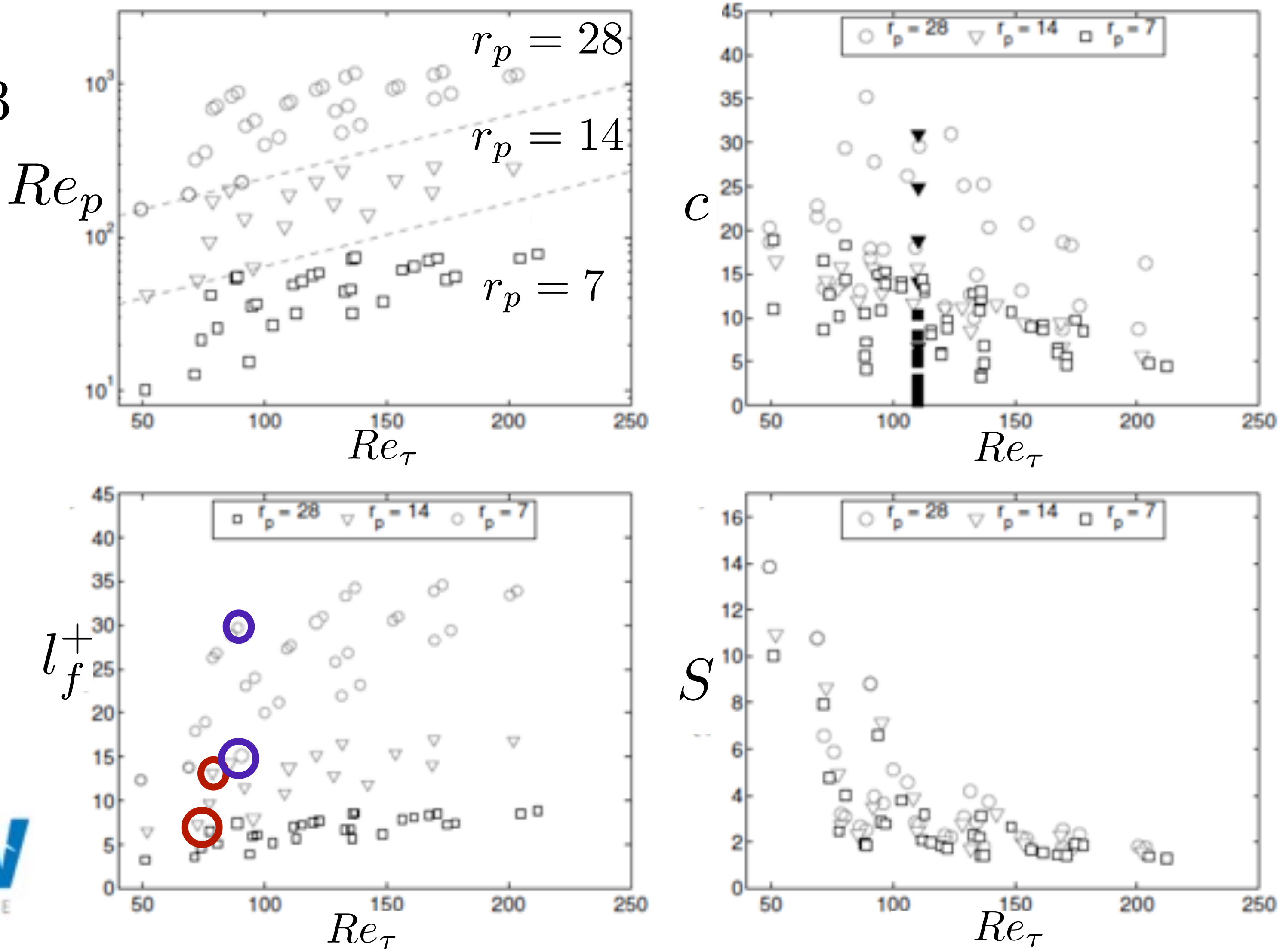
Parameters covered

$$\frac{\rho_p}{\rho_f} = 1.3$$



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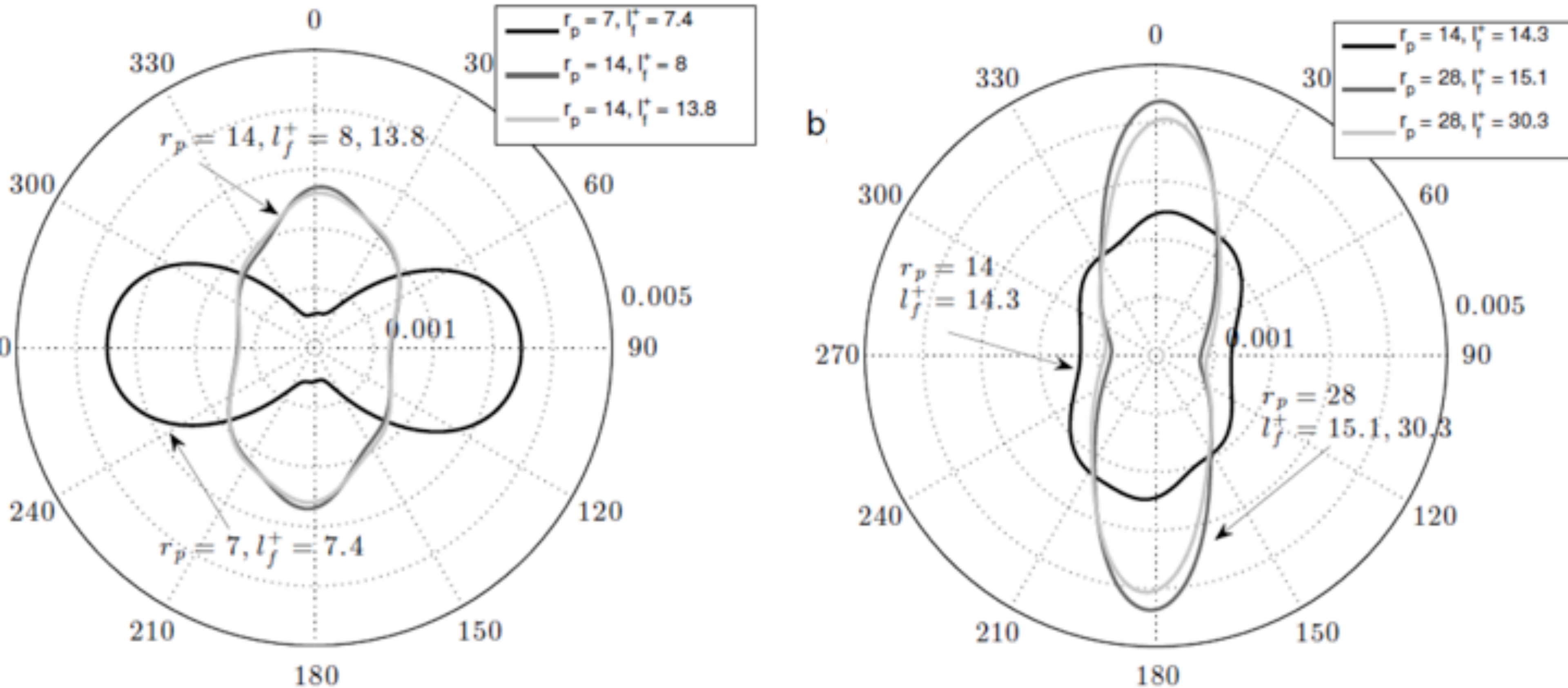
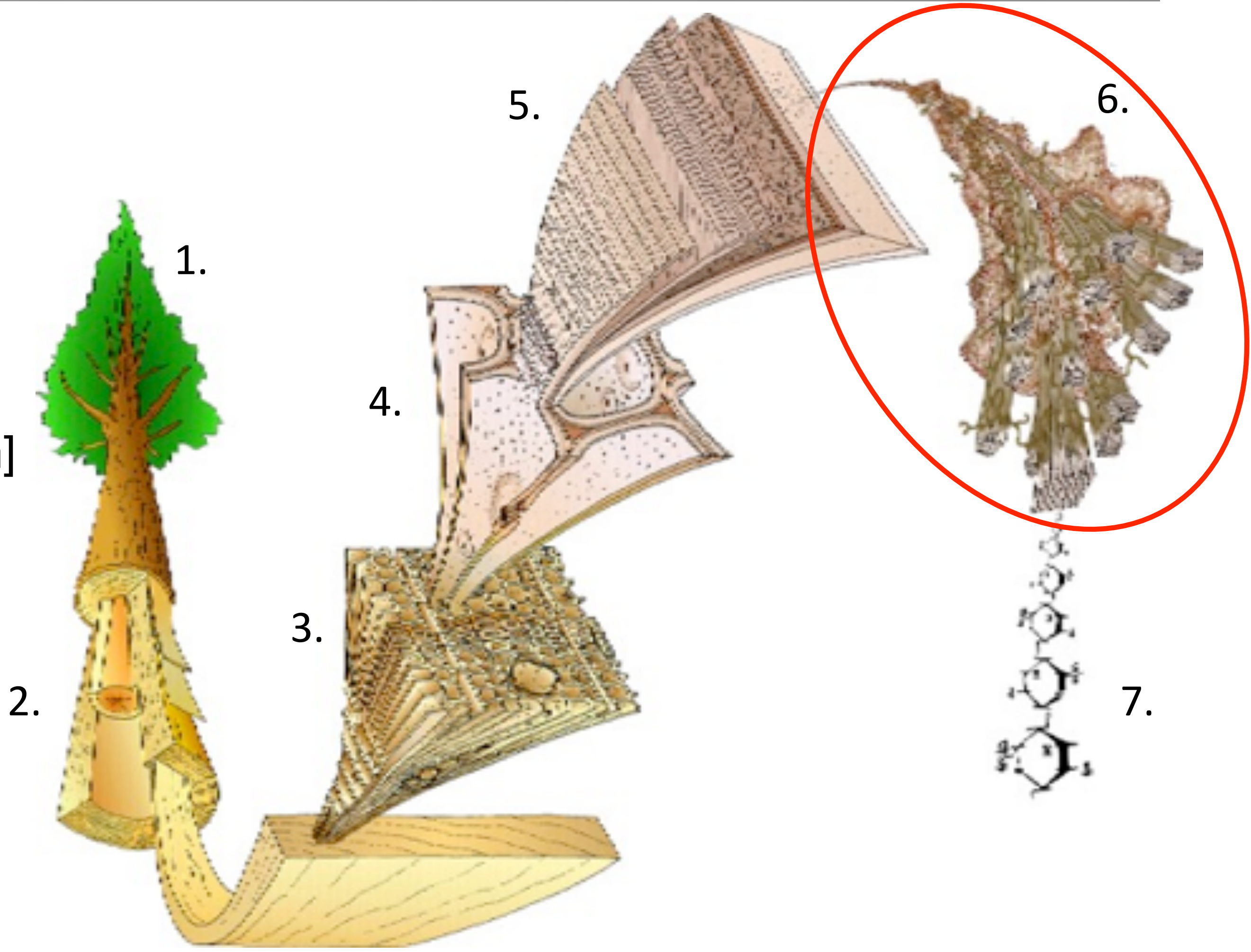
Orientation *not* determined by l_f^+



Hierarchical structure of a tree

1. Tree, [m]
2. Wood, [dm]
3. Annual rings, [cm]
4. Wood fibres, [mm]
5. Fibre wall, [µm]
6. Fibrill aggregates, [nm]
7. Polymer chains, [Å]



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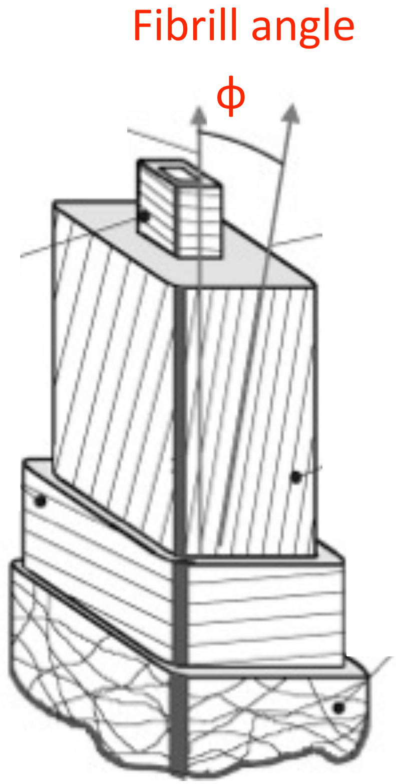
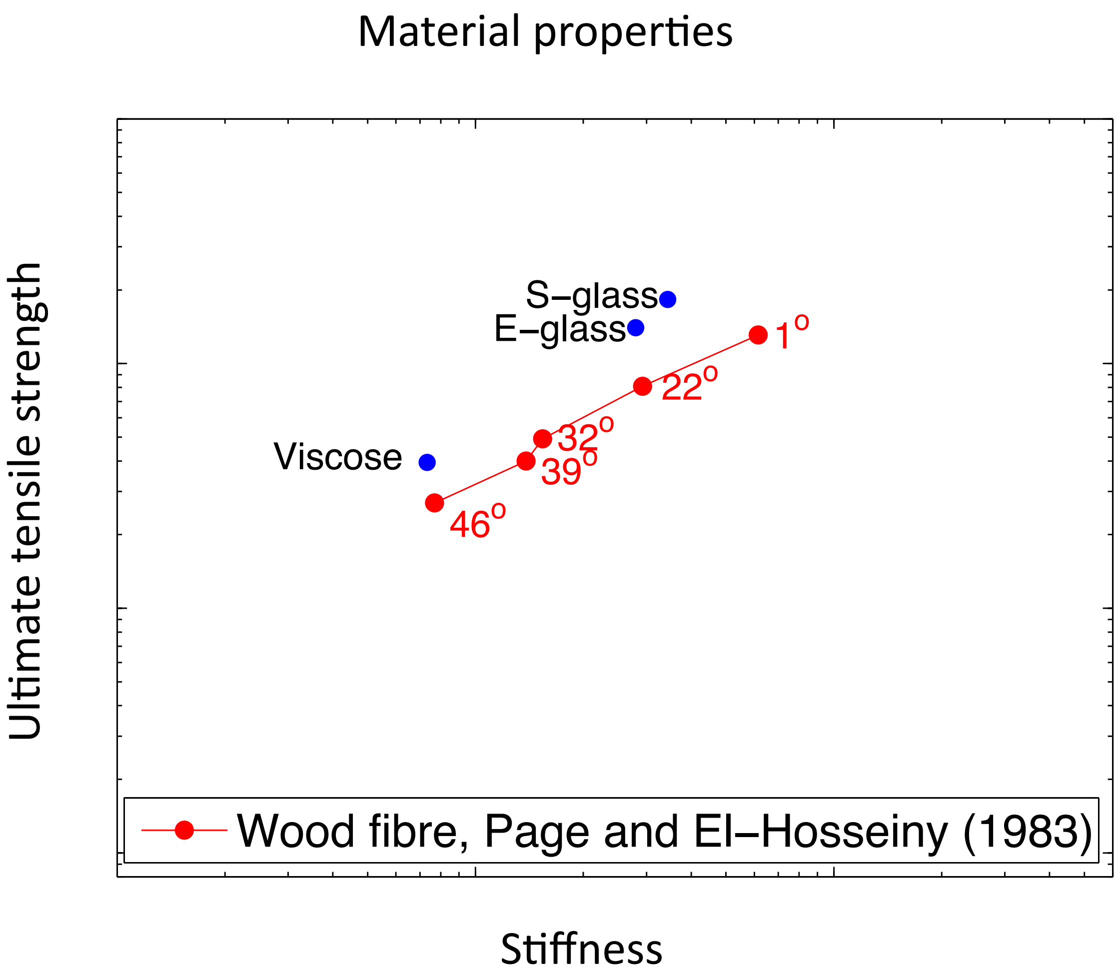
Vision

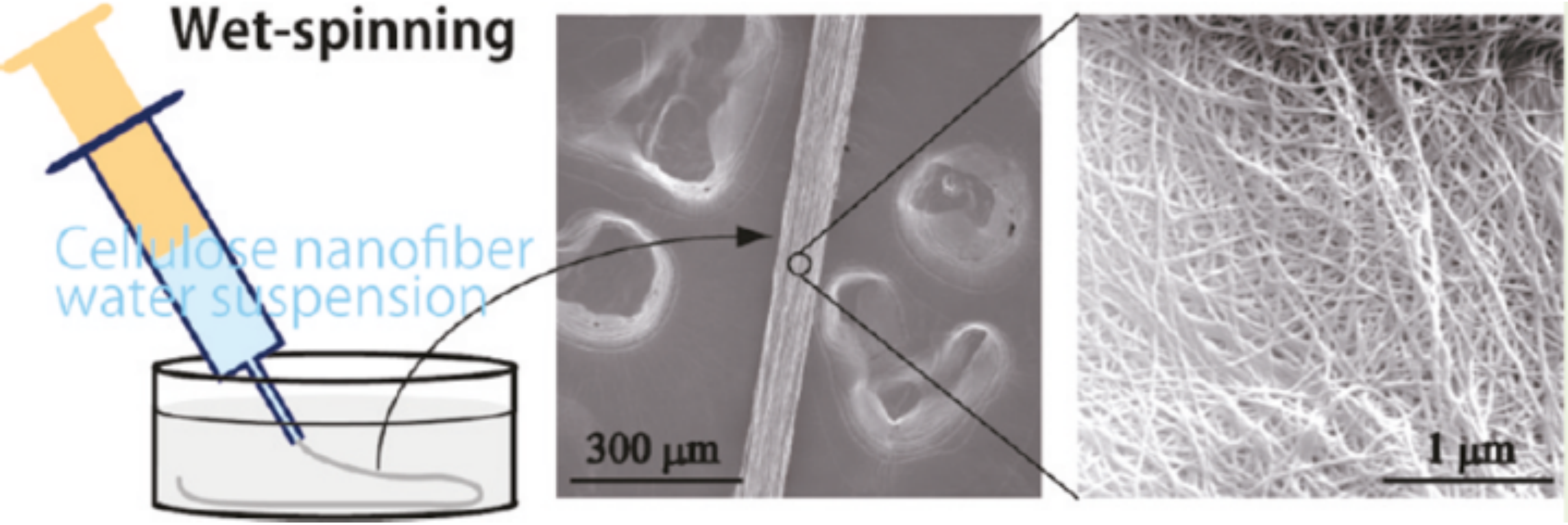
Return of cellulose in high-tech applications!

Material properties of cellulose fibres





Filament preparation by ejection from syringe

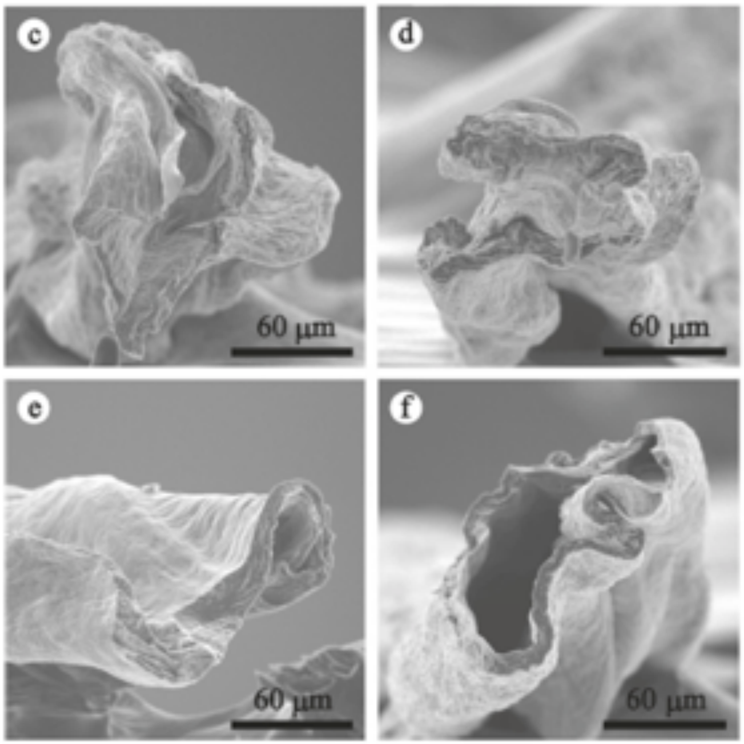


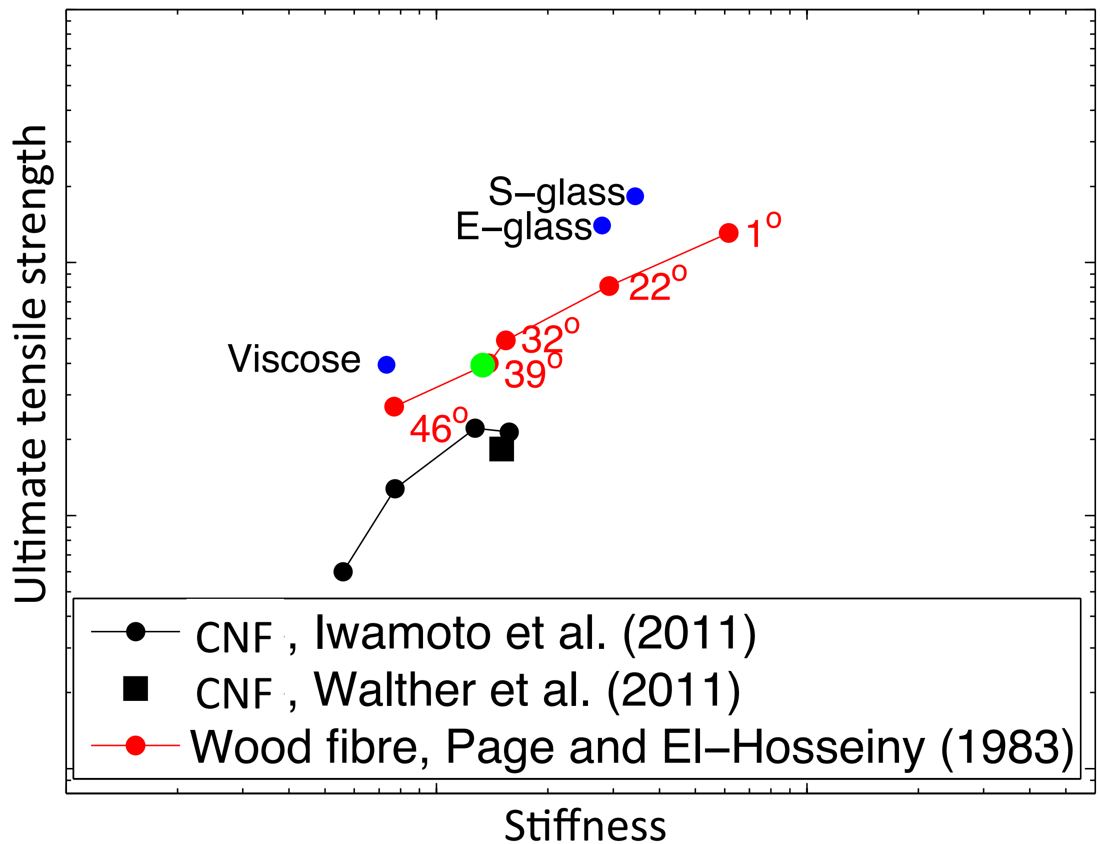
Iwamoto et al. (2012)

Resulting filament and its properties

Iwamoto et al. (2012)

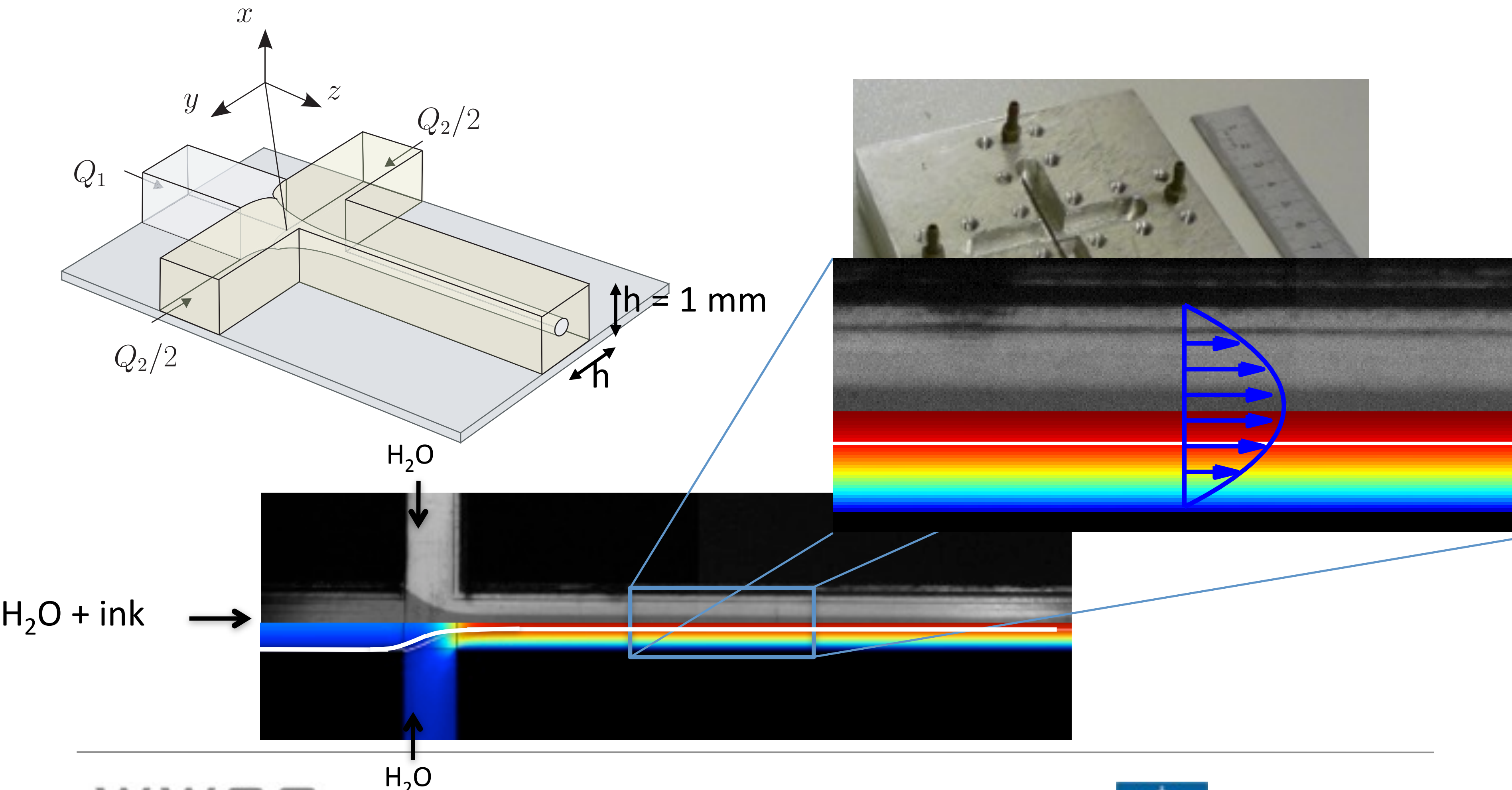
Material properties



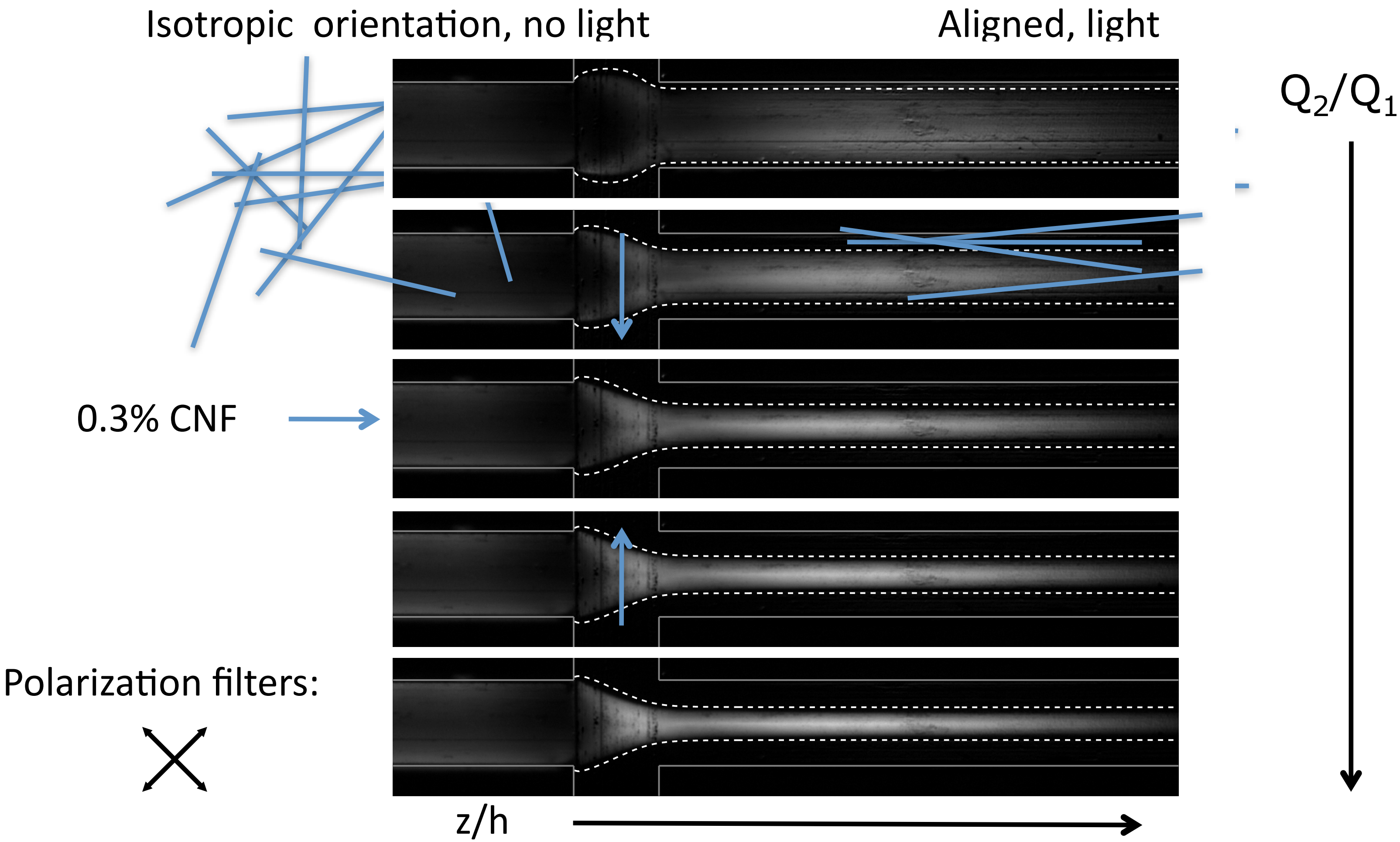


An alternative:
flow focusing

Extensional flow with *minimum shear*

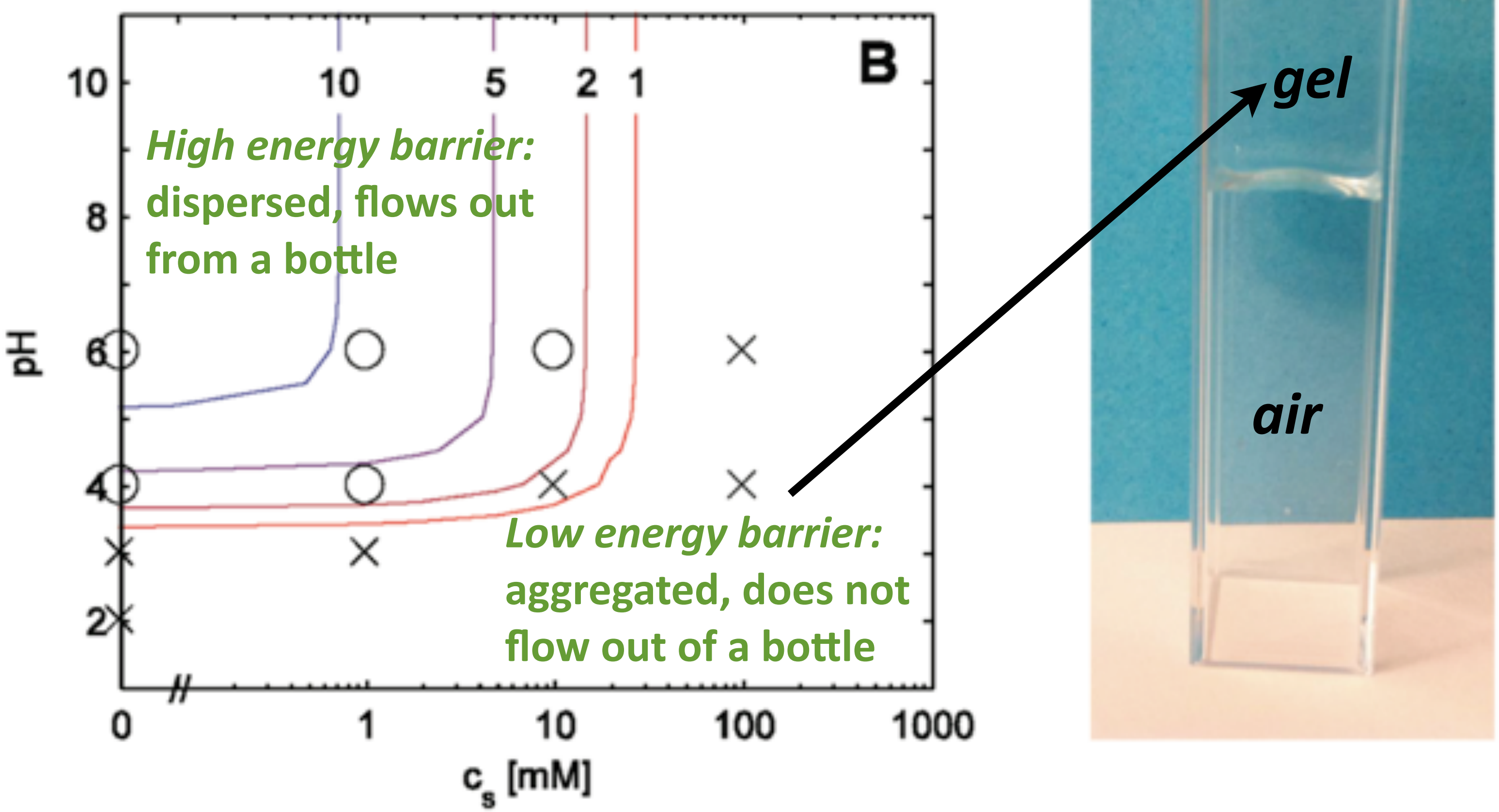


Polarized light visualizes alignment!



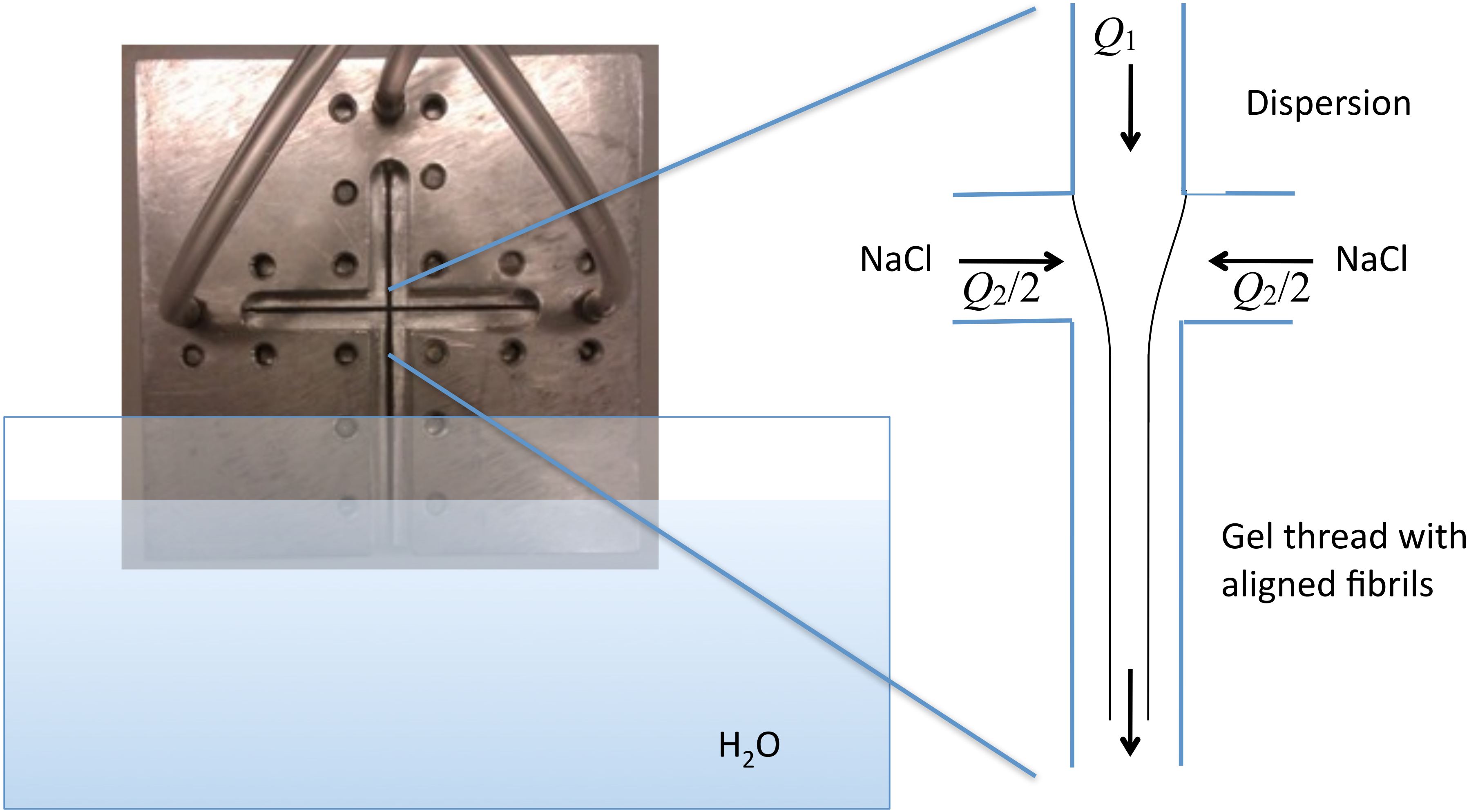
From aligned dispersed fibrills to a filament:
The disp-gel transition

Normalized energy barrier vs. pH and NaCl:



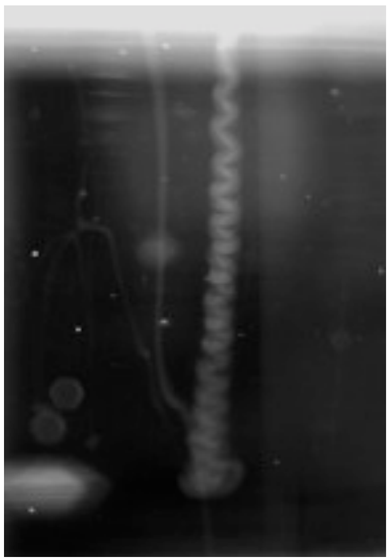
Fall et al. (2011)

Manufacturing process

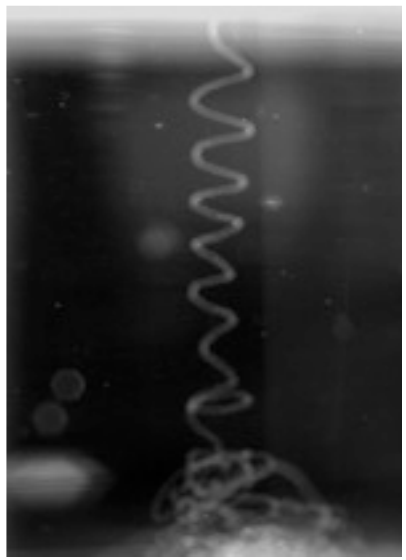


Ejection for further processing and drying

H_2O NaCl



No filament

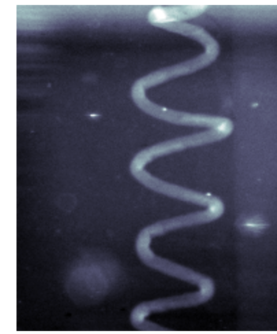


Filament

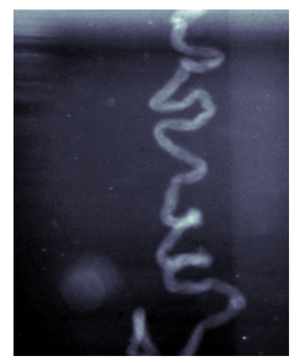
Increasing extension:

$Q_2/Q_1 = 0.69$

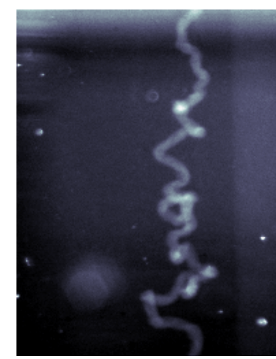
$Q_2/Q_1 = 1.15$



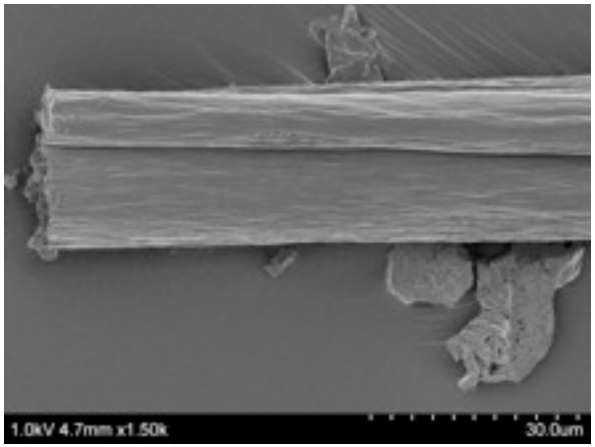
$Q_2/Q_1 = 2.30$

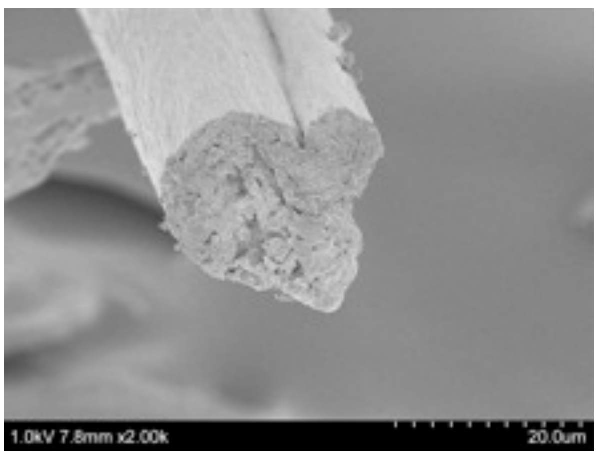


$Q_2/Q_1 = 3.45$

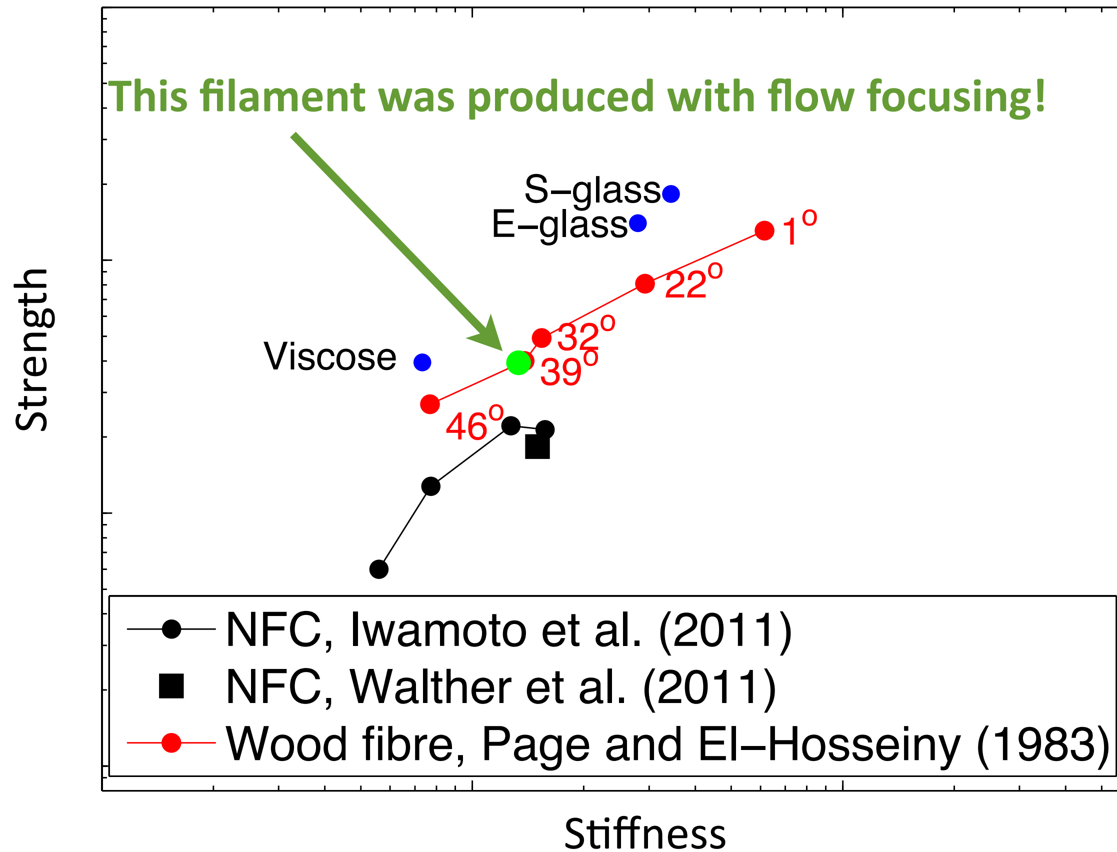


A smooth, homogenous and strong filament!°





Material properties



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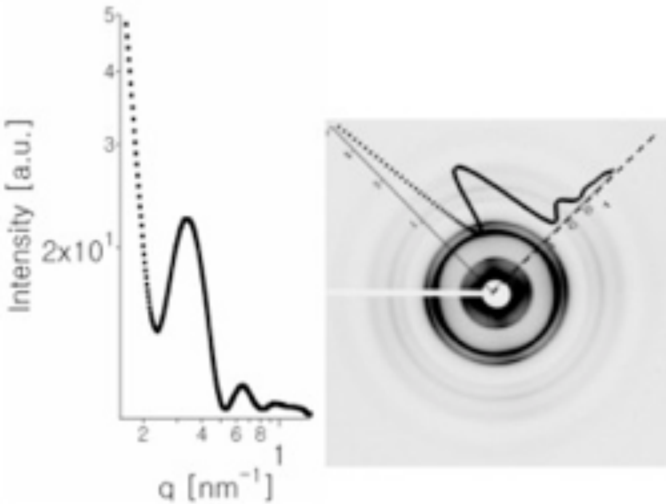
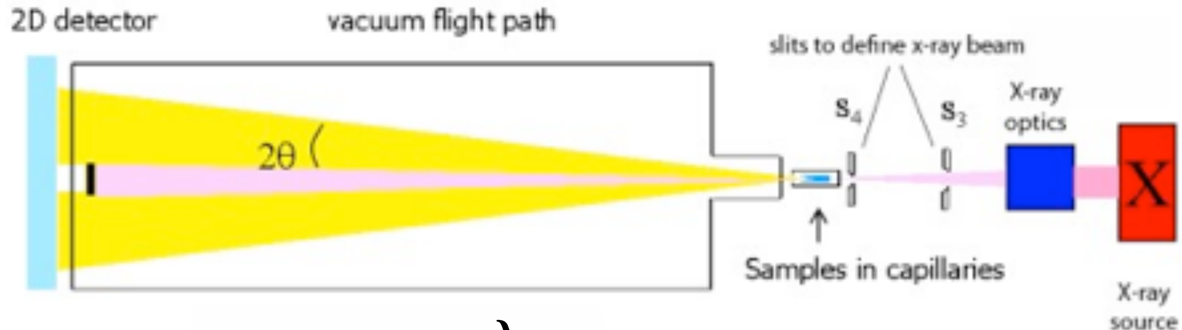
X-ray diffraction



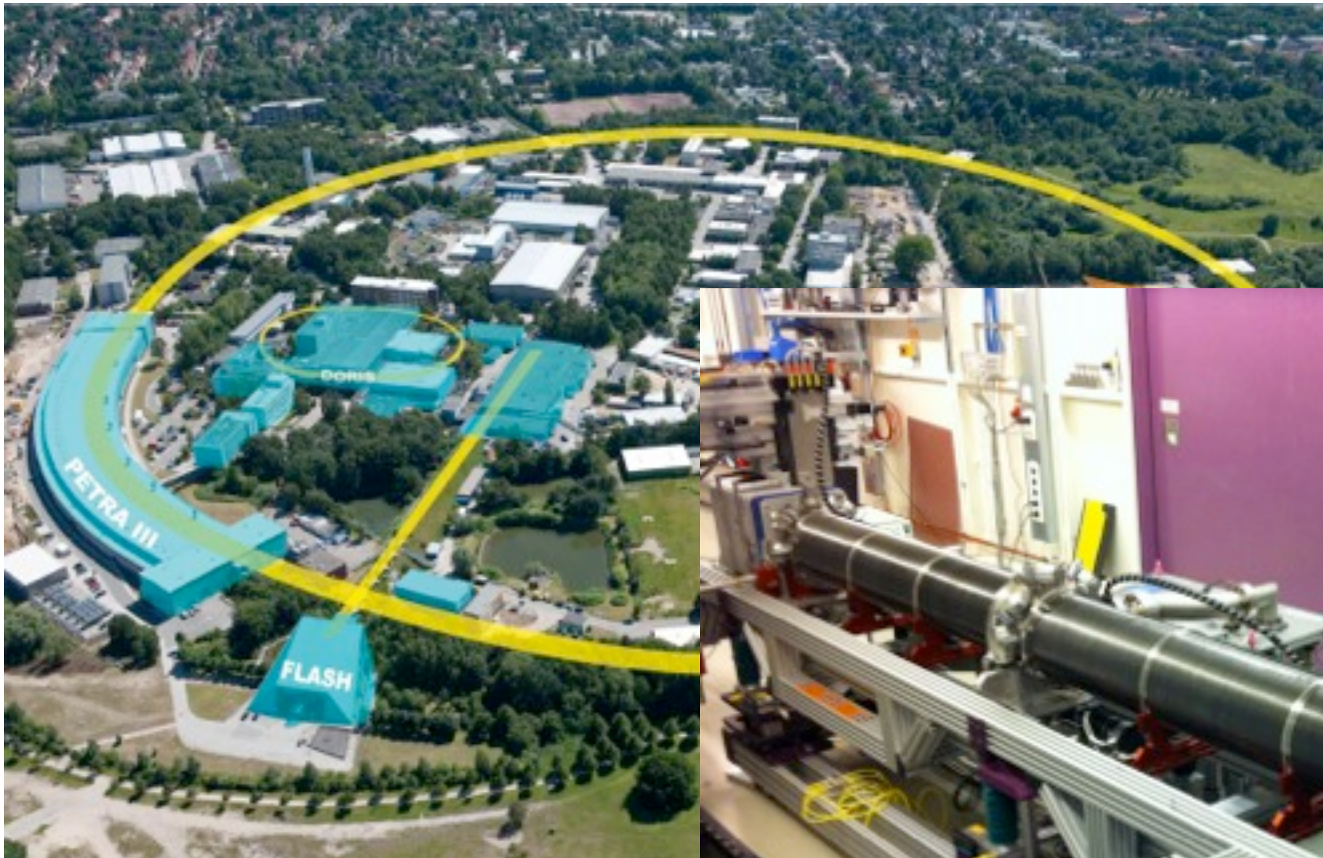


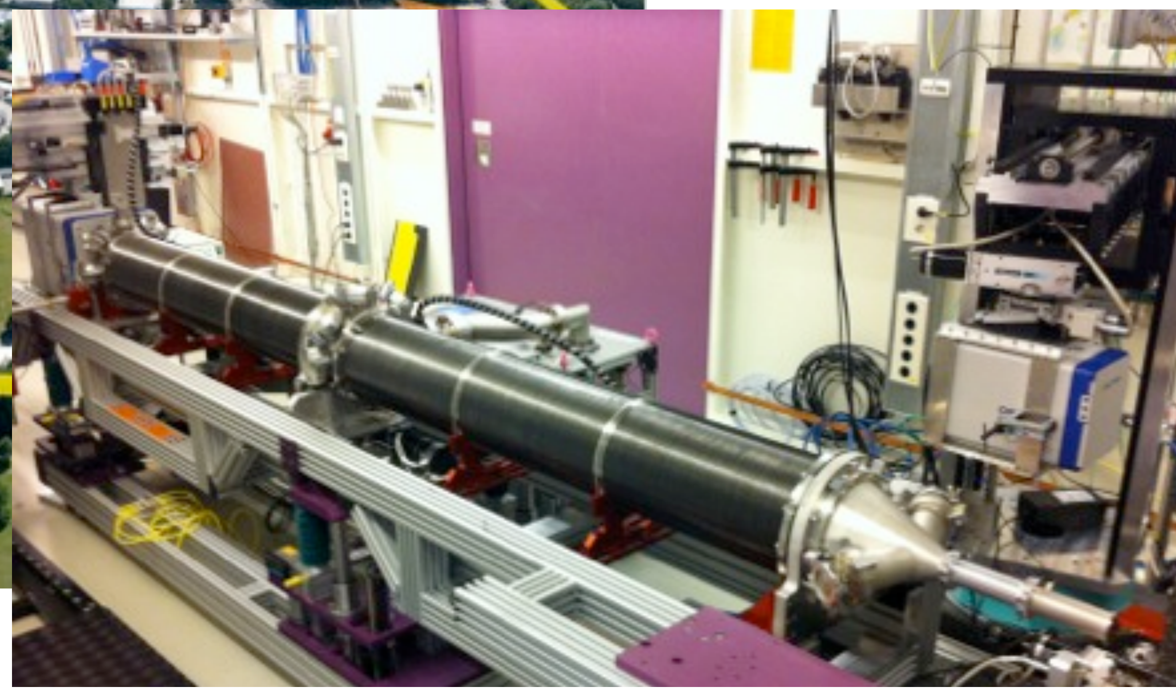
$$q = \frac{4\pi \sin\theta}{\lambda}$$

$$\sin\theta = \frac{\lambda}{2d}$$

www.mrl.ucsb.edu

P03 beamline @ PETRAIII





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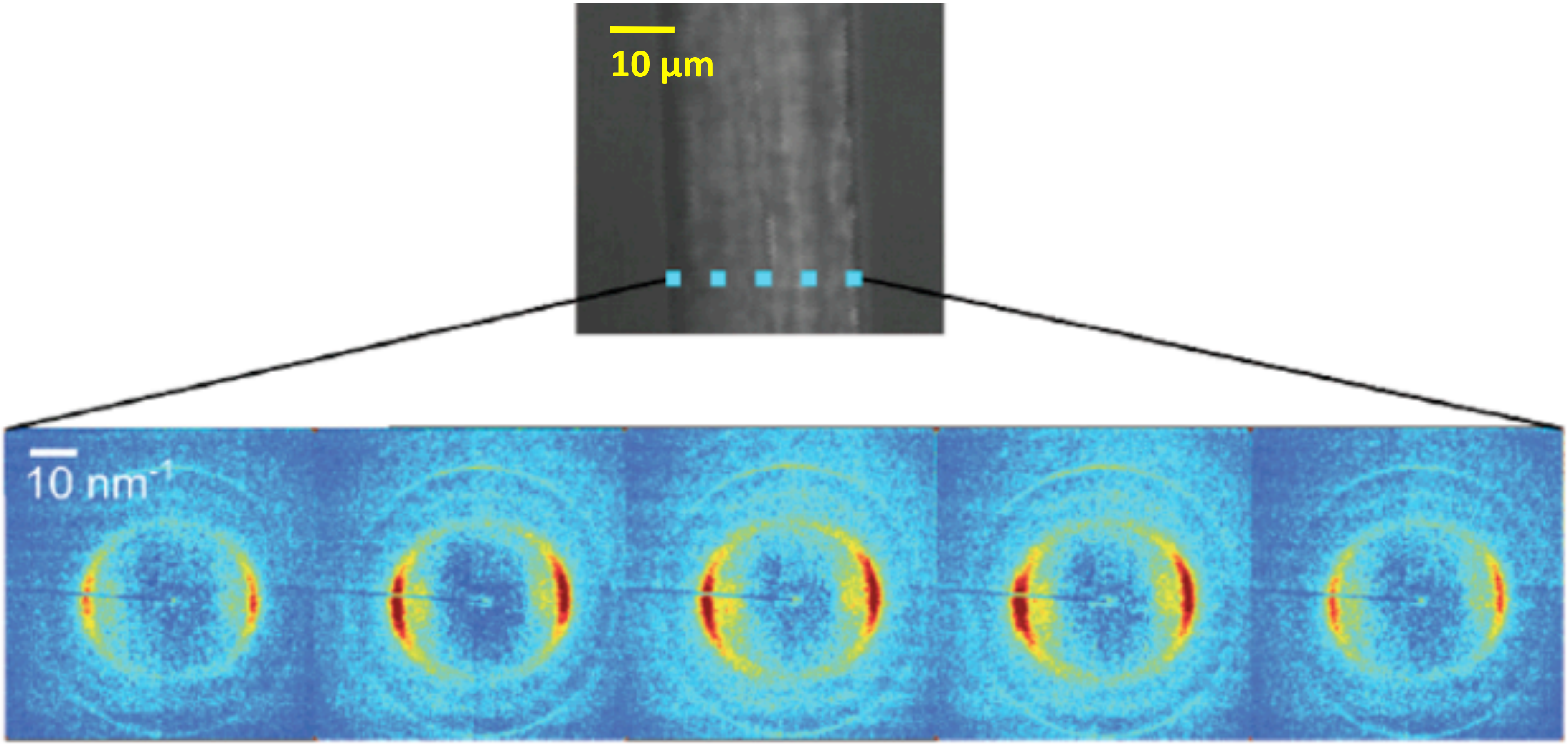
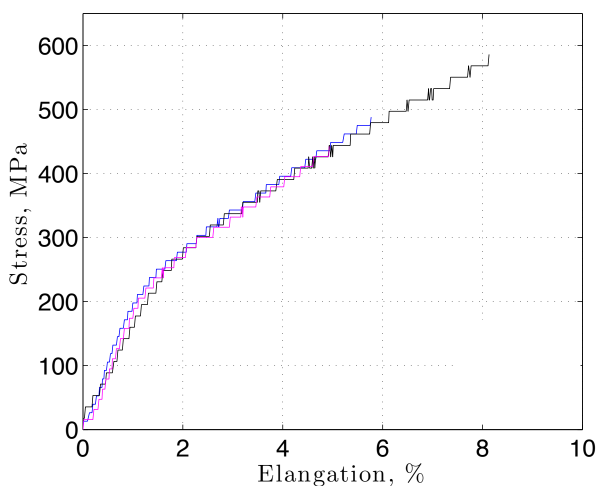
WAXS across the filament (Wide Angle X-ray Scattering)

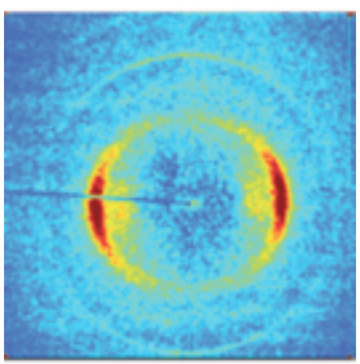


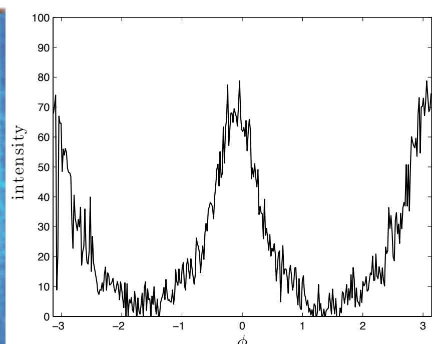
Summary of the filament properties

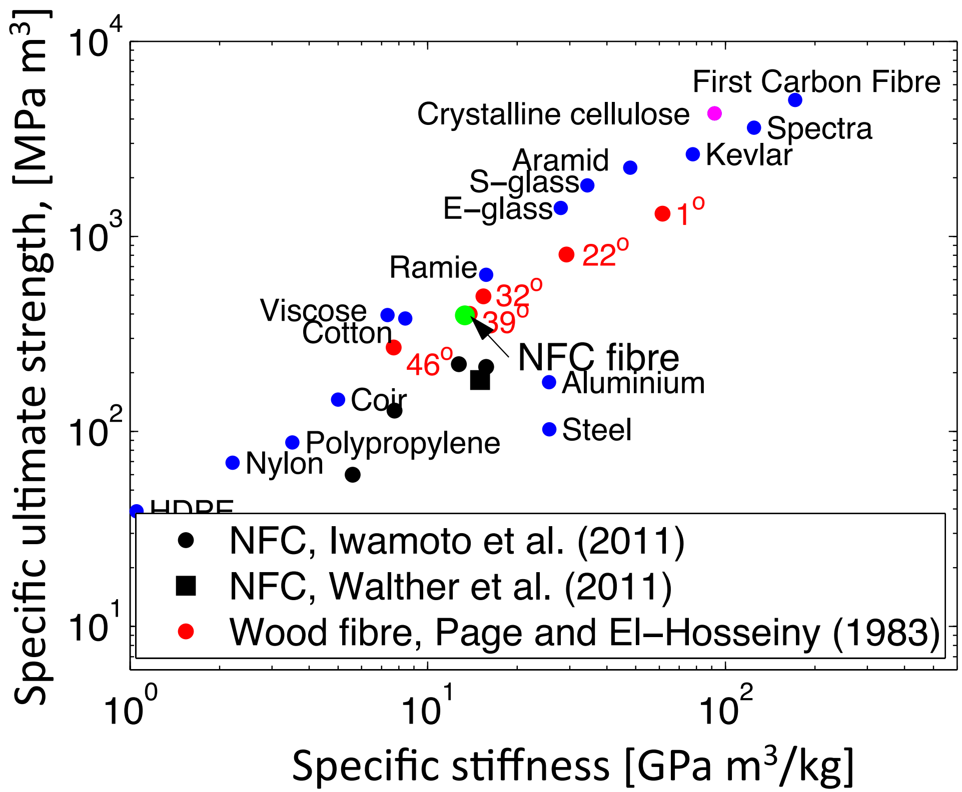
Tensile test



WAXS results







S = 0.5 corresponds to a mean angle of 35°!

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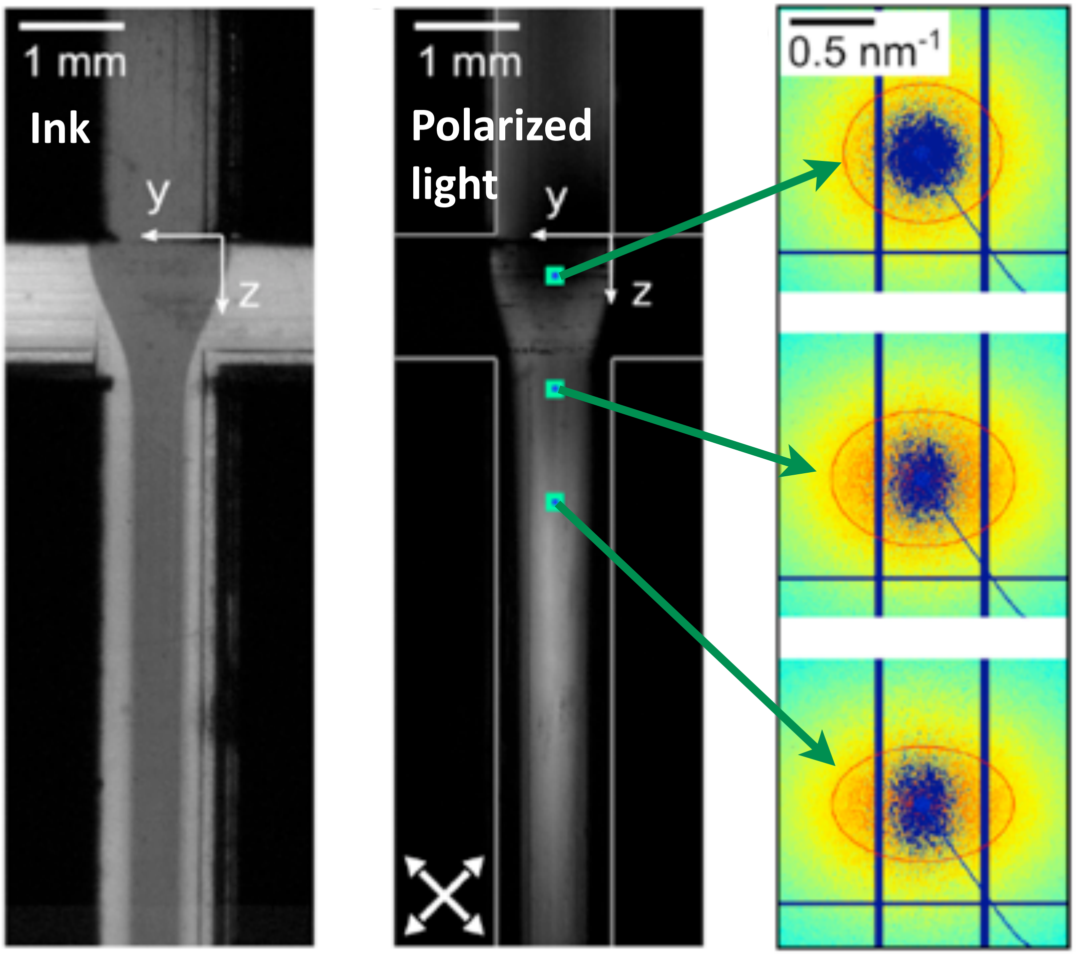
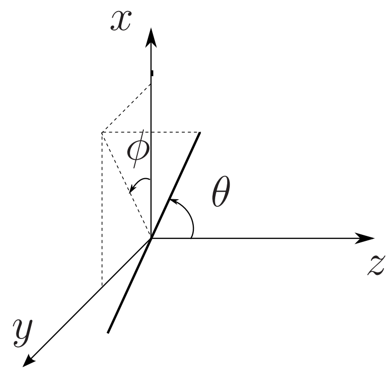
Alignment during production: SAXS: *Small* Angle X-ray Scattering



The diffraction pattern is deformed thanks to fibrill alignment!

Orientation modelling (Smoluchowski eq.)



1D Smoluchowski equation:

$$^{*}\frac{\partial \Psi}{\partial z^{*}} = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\hat{D}_r^{*}\sin\theta\frac{\partial\Psi}{\partial\theta} - \sin\theta\dot{\theta}\Psi\right)$$

Order parameter:

$$S = \int_0^{\pi}\Psi(\theta)\left(\frac{3}{2}\cos^2\theta - \frac{1}{2}\right)\sin\theta d\theta\int_0^{2\pi}d\phi$$

Rotational diffusion coefficient, Doi & Edwards (1986):

$$\hat{D}_r = \boxed{\frac{3k_BT(2\ln(2r_p)-1)}{16\pi\eta_s a^3}}\boxed{\beta(nl^3)^{-2}}\boxed{\left[\frac{4}{\pi}\int d\mathbf{p}'\left|\mathbf{p}\times\mathbf{p}'\right|\Psi_s(\mathbf{p}')\right]^{-2}}$$

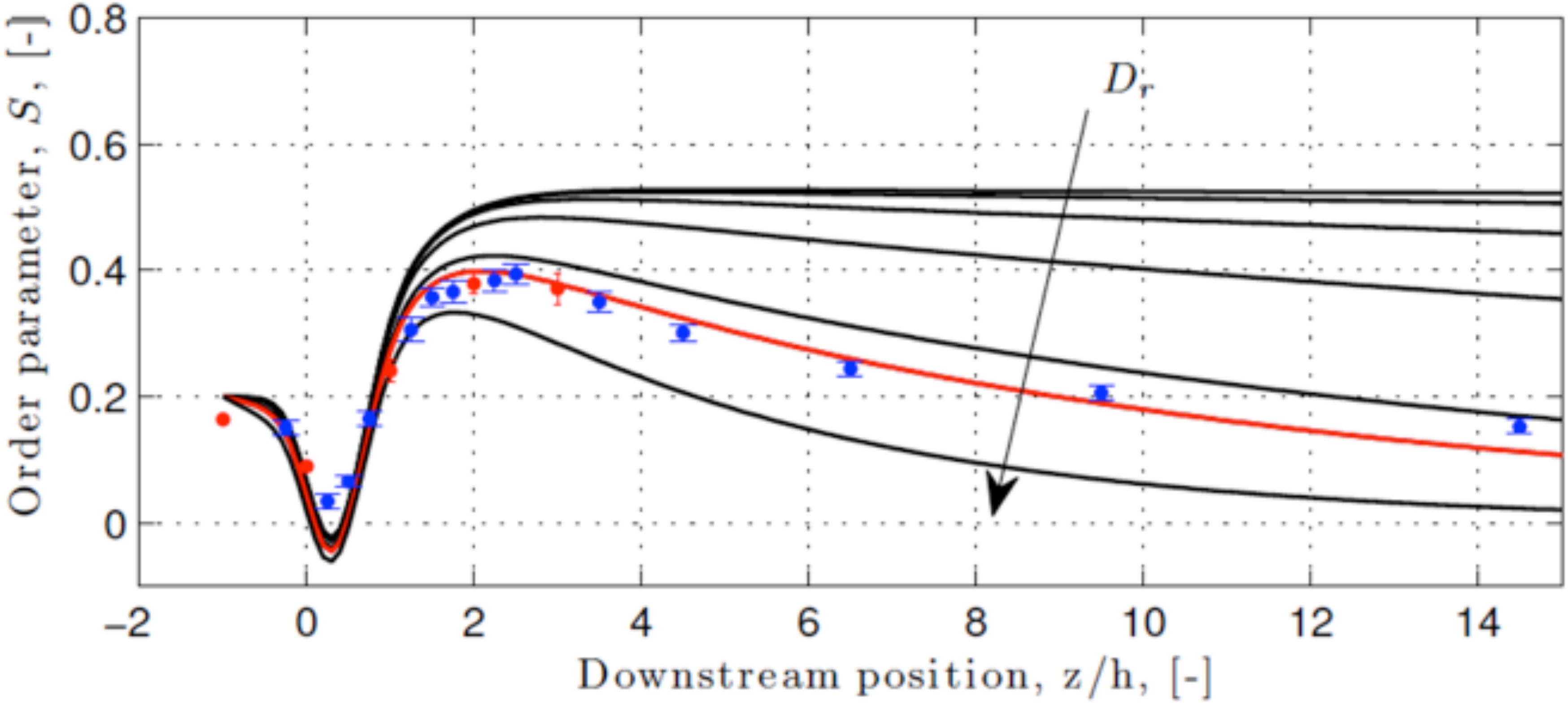
Brownian diffusion　Conc　Orientation

Angular velocity from Jeffery (1922) for a biaxial flow:

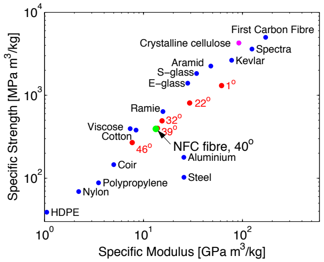
$$\dot{\theta} = \frac{\partial\theta}{\partial t^{*}} = -\frac{\partial w^{*}}{\partial z^{*}}\left(\frac{r_p - 1}{r_p + 1}\right)\frac{3}{2}\cos\theta\sin\theta$$

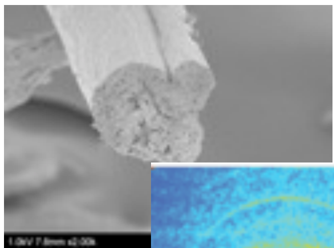
Rotational diffusion coefficient, $\hat{D}_r^{*}$
Aspect ratio, r_p
Velocity in z-direction, w
Orientation distribution, Ψ

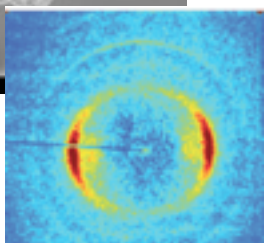
Modelling of fibrill order: *alignment vs. diffusion*

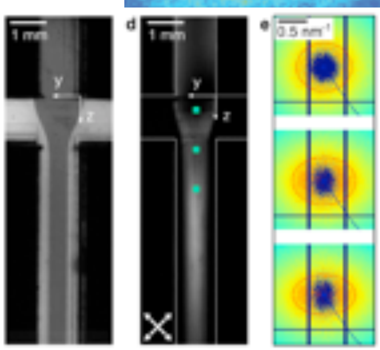


Summary









- Cellulose filaments with excellent properties seem to be within reach
- Detailed knowledge of the chemistry and physics of the process is necessary
- The knowledge is obtained by combining visualizations, computations and X-ray diffraction measurements