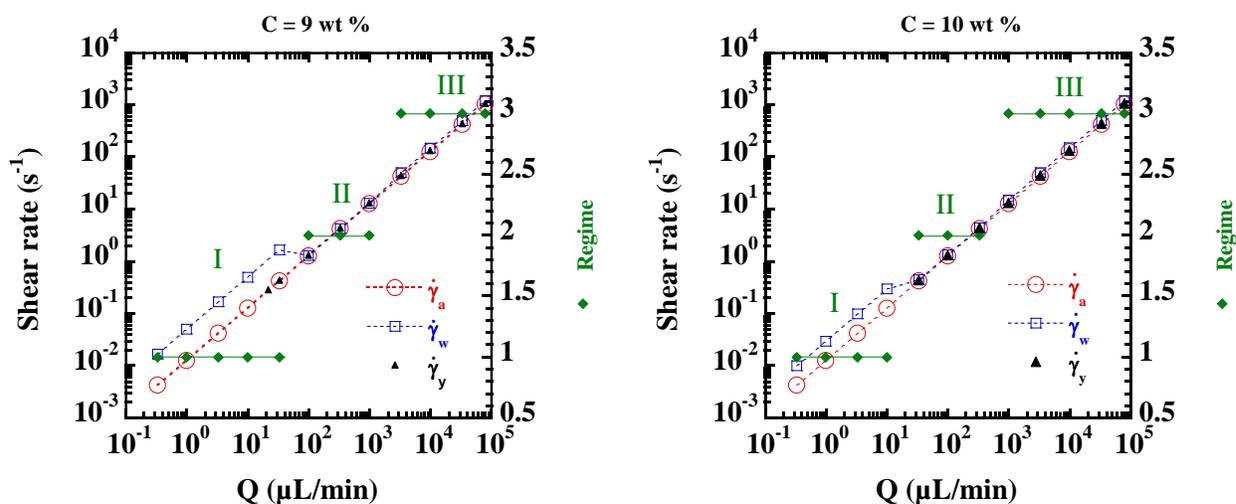


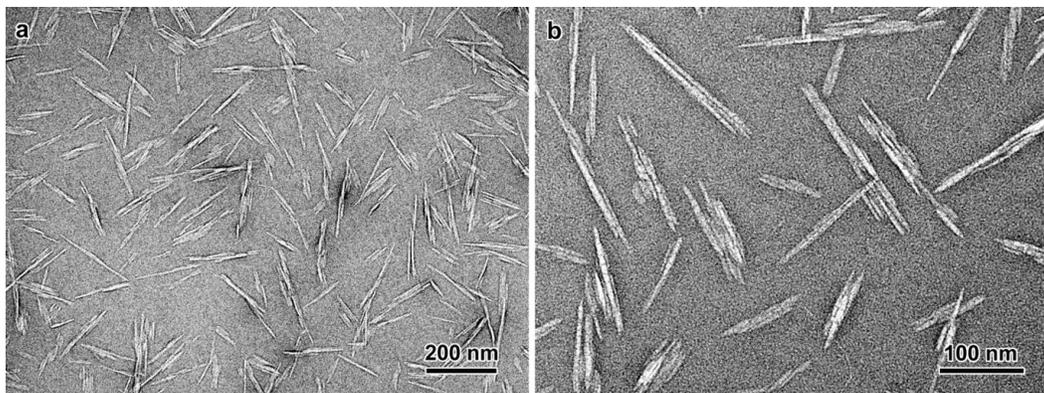
# Breakdown and buildup mechanisms of cellulose nanocrystal suspensions under shear and upon relaxation probed by SAXS and SALS

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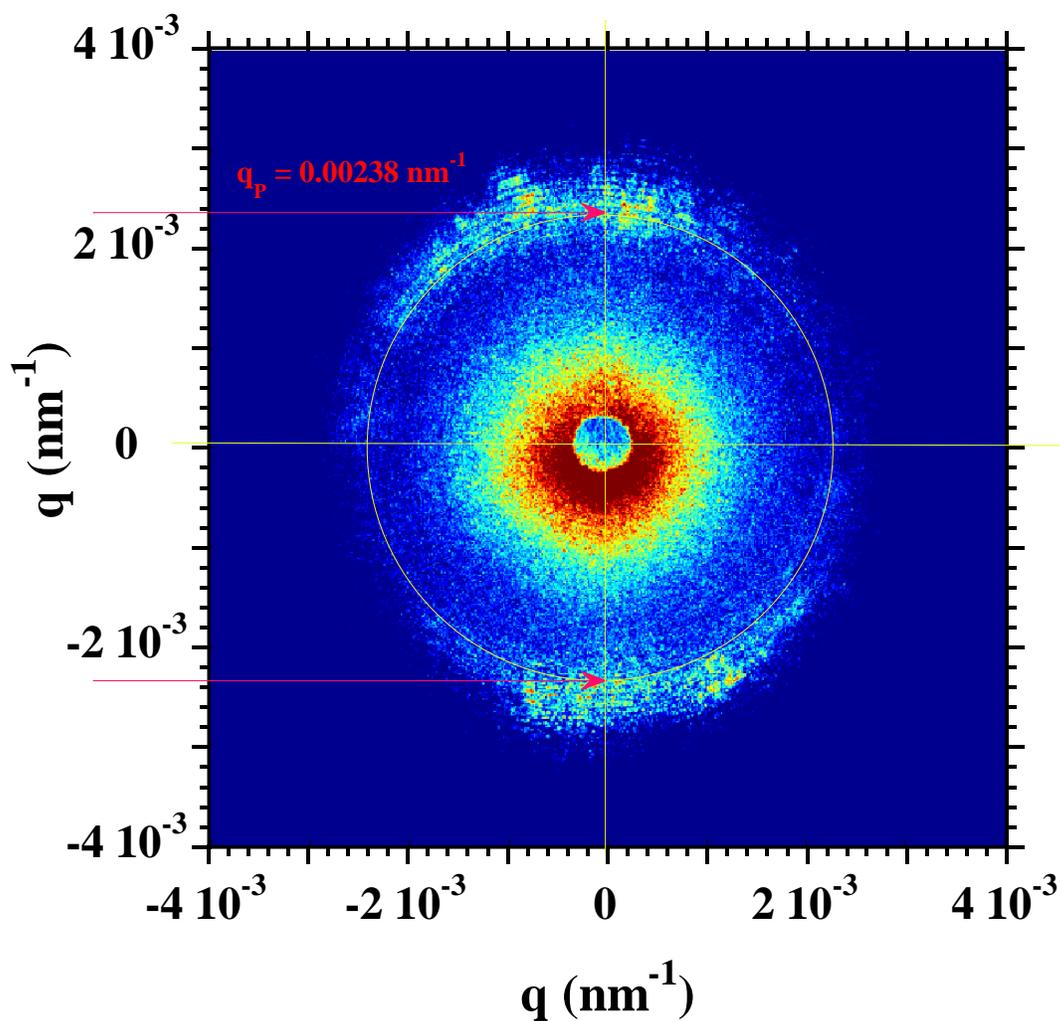
## Supplementary Material



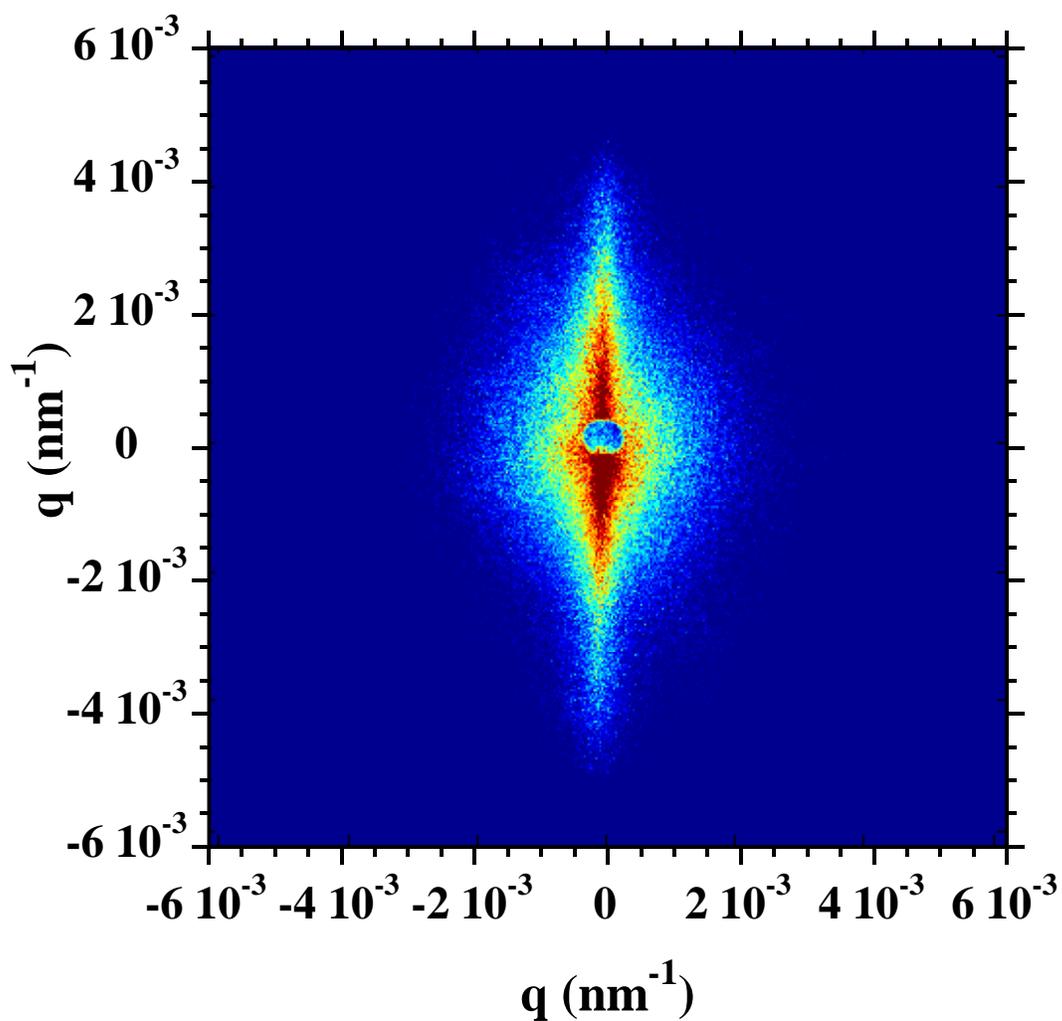
**Figure S1.** Calculation from Son (2007) of mean shear rate  $\dot{\gamma}_y$ , apparent shear rate  $\dot{\gamma}_a$  of a rectangular die and the wall shear rate  $\dot{\gamma}_w$  taking into account the shear thinning behavior (shear thinning index  $n$ ) of the CNCs according to the three different regimes of the flow curve for the two CNC concentrations  $C = 9$  wt% and  $C = 10$  wt%.  $n_1, n_2, n_3$  values of shear thinning index according to regimes I, II and III, respectively ( $C = 9$  wt%:  $n_1 = 0.0940, n_2 = 0.9626, n_3 = 0.6401$ , and  $C = 10$  wt%:  $n_1 = 0.1857, n_2 = 0.8686, n_3 = 0.6228$ ).



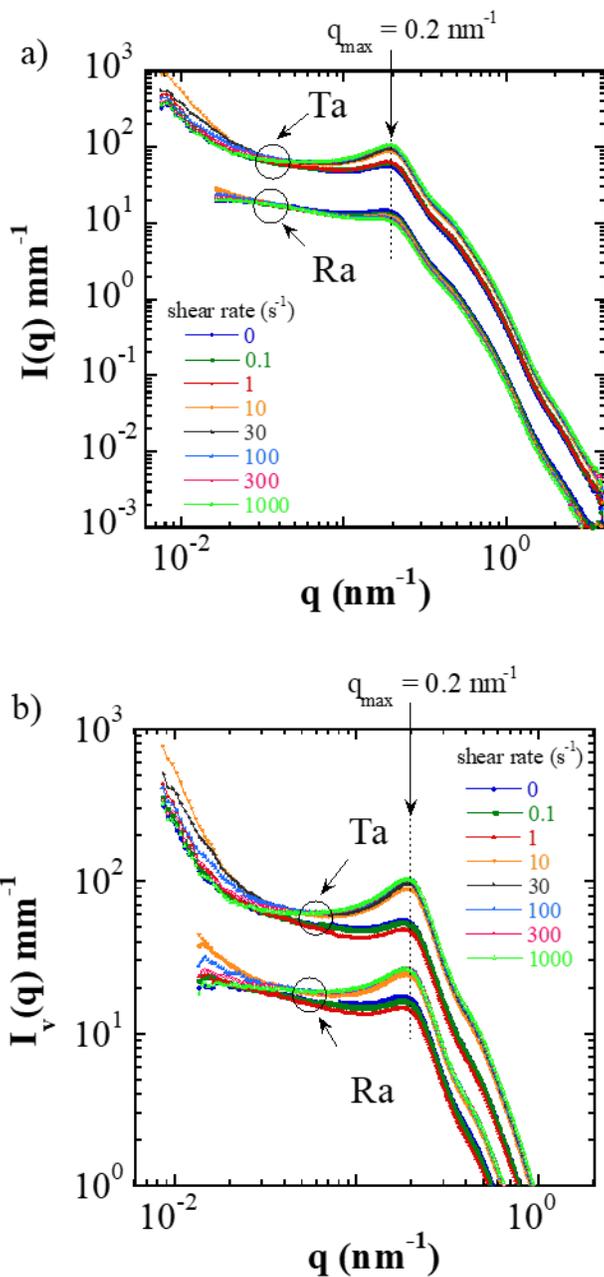
**Figure S2.** a,b) TEM images of negatively stained preparations of CNCs.



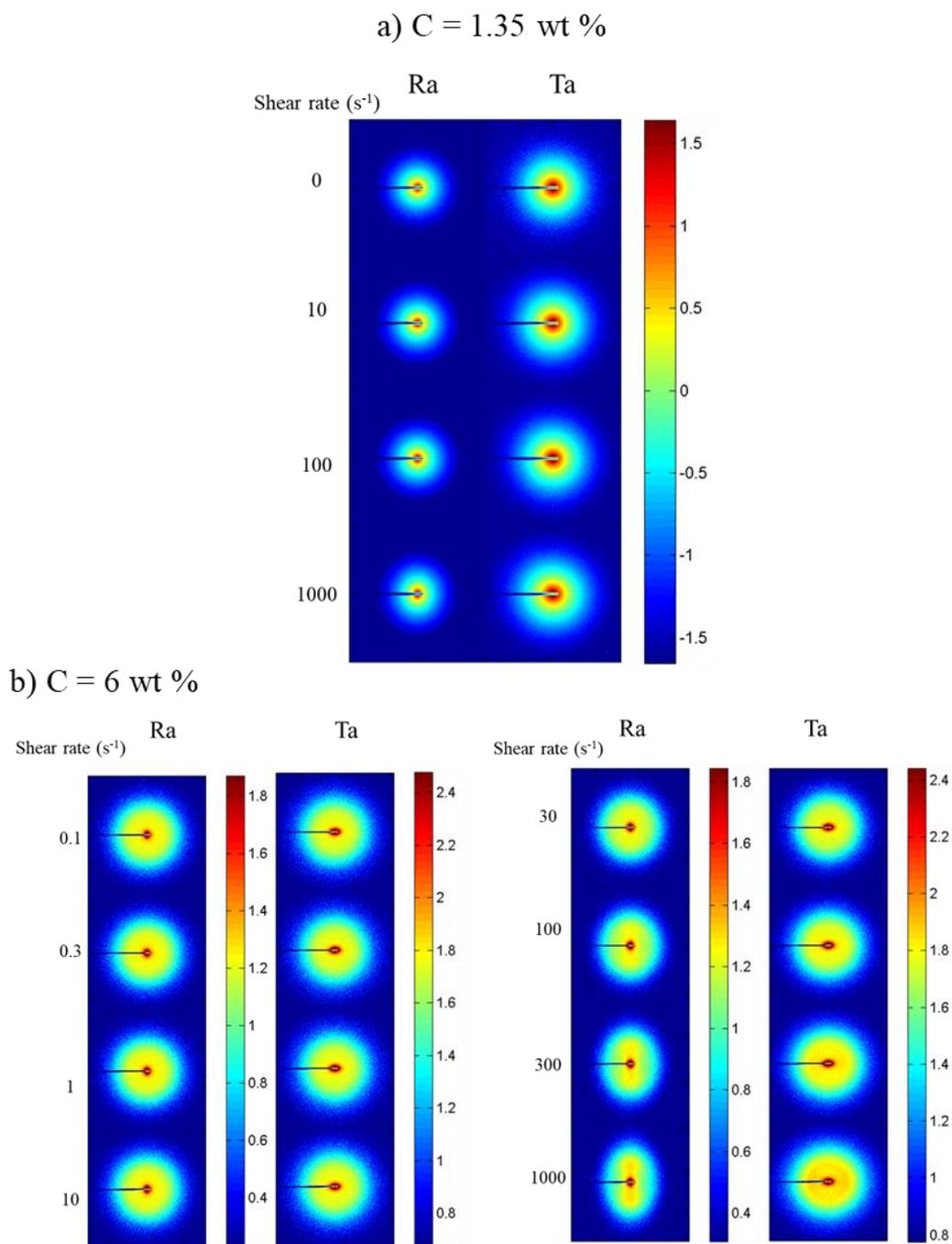
**Figure S3.** SALS pattern of a CNC suspension at rest,  $q_p = 0.00238 \text{ nm}^{-1}$  and determination of the corresponding pitch distance  $P = 5279 \text{ nm}$ .  $C = 9 \text{ wt\%}$  ( $\phi = 5.82 \text{ vol\%}$ ) with  $0.01 \text{ mol L}^{-1} \text{ NaCl}$  at  $T = 25 \text{ }^\circ\text{C}$ .



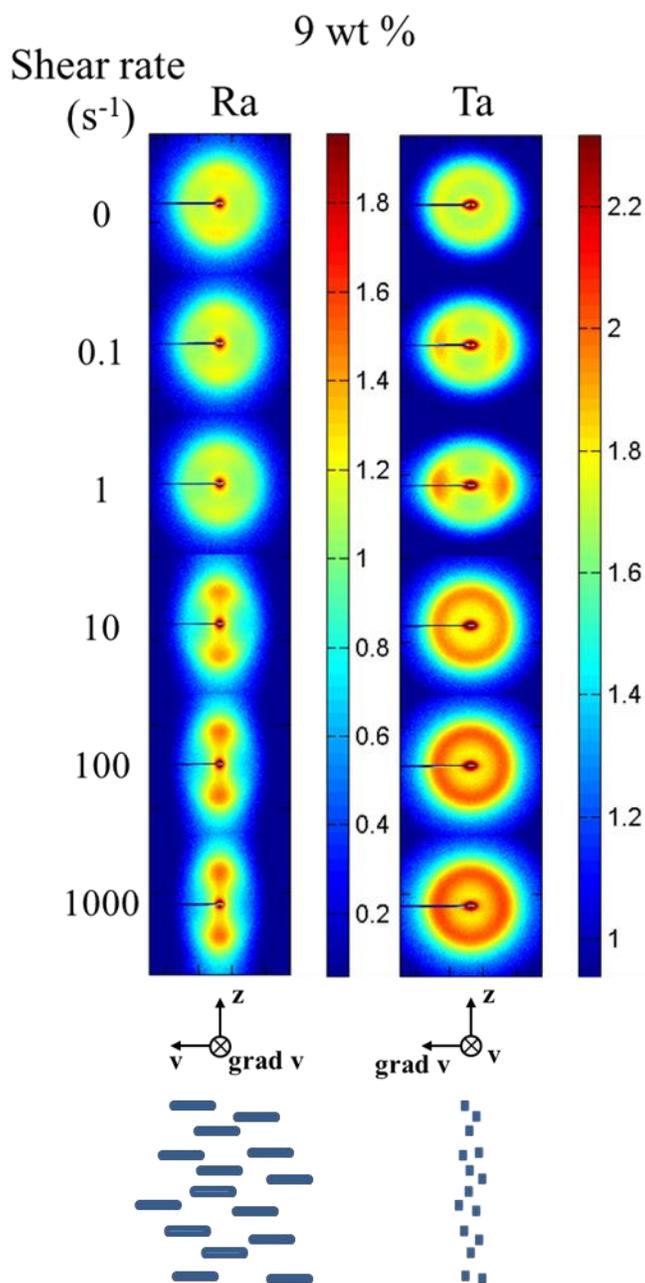
**Figure S4.** *In situ* SALS pattern of a CNC suspension under shear in regime II at a shear rate =  $13.3 \text{ s}^{-1}$  and time  $t = 14.5 \text{ s}$ .  $C = 9 \text{ wt\%}$  ( $\phi = 5.82 \text{ vol\%}$ ) with  $0.01 \text{ mol L}^{-1} \text{ NaCl}$  at  $T = 25 \text{ }^\circ\text{C}$ .



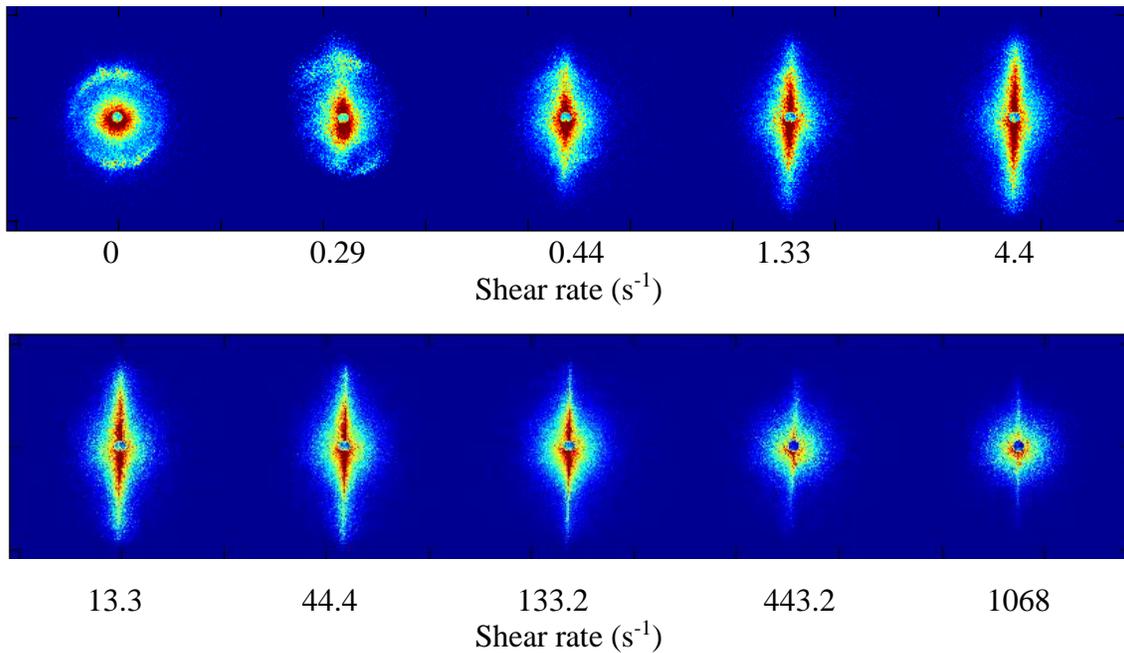
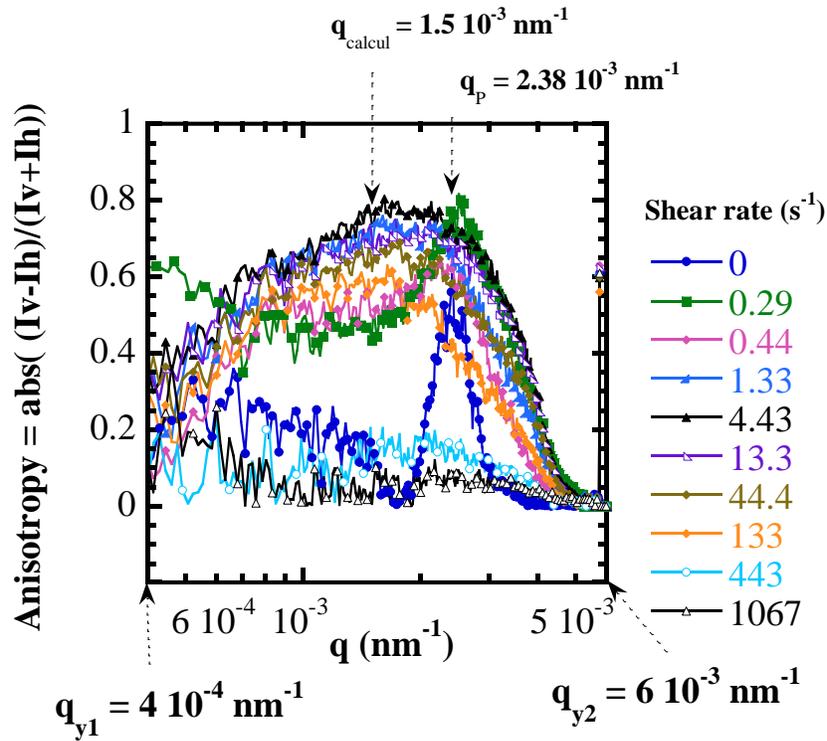
**Figure S5.** Azimuthally averaged  $I(q)$  a) (plotted over a wide range of  $q$ ) and sector averaged  $I_v(q)$  b) (plotted on a narrow  $q$  range) of the Rheo-SAXS measurements from a CNC suspension under steady-state flow, for successive steps of applied shear rates.  $C = 9$  wt% ( $\phi = 5.82$  vol%) with  $0.01 \text{ mol L}^{-1}$  NaCl at  $T = 24.5 \text{ }^\circ\text{C}$ .



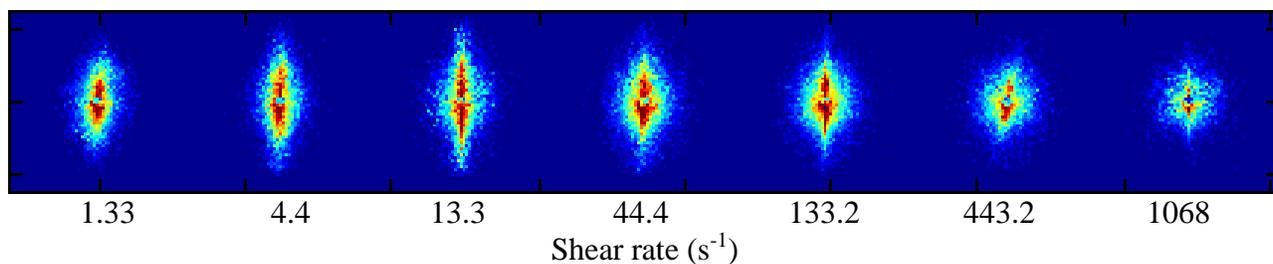
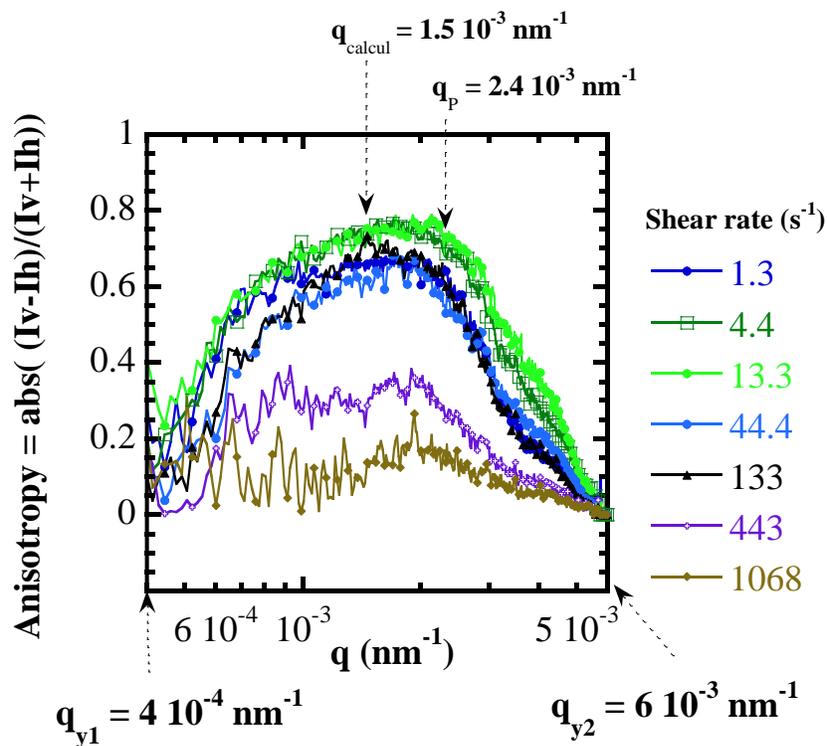
**Figure S6.** 2D-SAXS patterns from CNC suspension under steady-state flow for successive steps of applied shear rates, in radial (Ra) and tangential (Ta) positions, corresponding to observation planes ( $z, v$ ) and ( $z, \text{grad } v$ ). (a)  $C = 1.35 \text{ wt } \%$ ,  $D = 2 \text{ m}$ , and (b)  $C = 6 \text{ wt } \%$ ,  $D = 10 \text{ m}$ ,  $0.01 \text{ mol L}^{-1} \text{ NaCl}$ ,  $T = 24.5 \text{ }^\circ\text{C}$ .



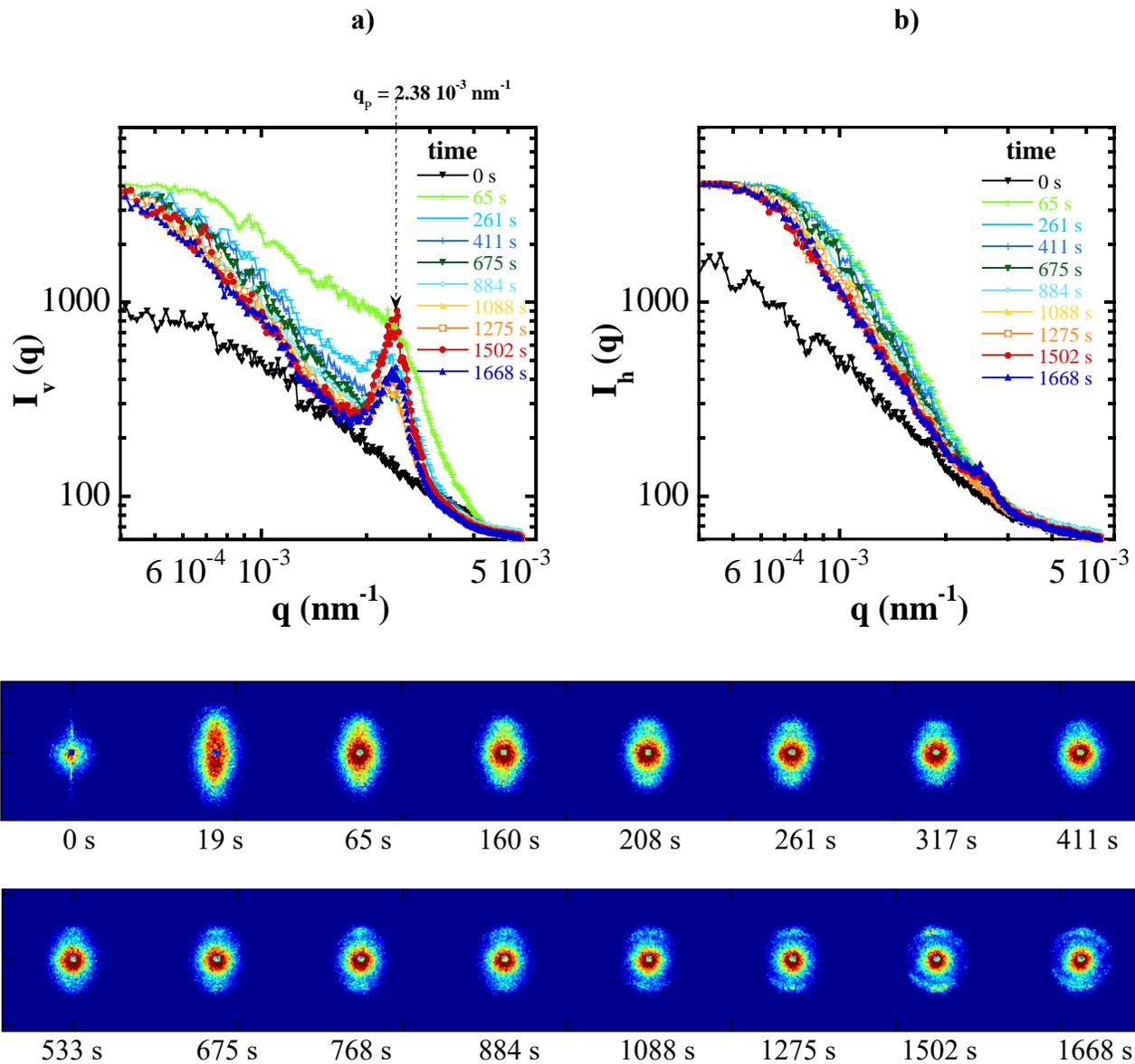
**Figure S7.** 2D-SAXS patterns from CNC suspension under steady-state flow for successive steps of applied shear rates, in radial (Ra) and tangential (Ta) positions, corresponding to observation planes ( $z, v$ ) and ( $z, \text{grad } v$ ).  $C = 9 \text{ wt\%}$ ,  $D = 10 \text{ m}$ ,  $0.01 \text{ mol L}^{-1} \text{ NaCl}$ ,  $T = 24.5 \text{ }^\circ\text{C}$ . Notice that the first 2D-SAXS pattern at rest (shear rate =  $0 \text{ s}^{-1}$ ) in Ra is slightly anisotropic, while not completely relaxed after a precedent step applied at high shear rate of  $1000 \text{ s}^{-1}$ .



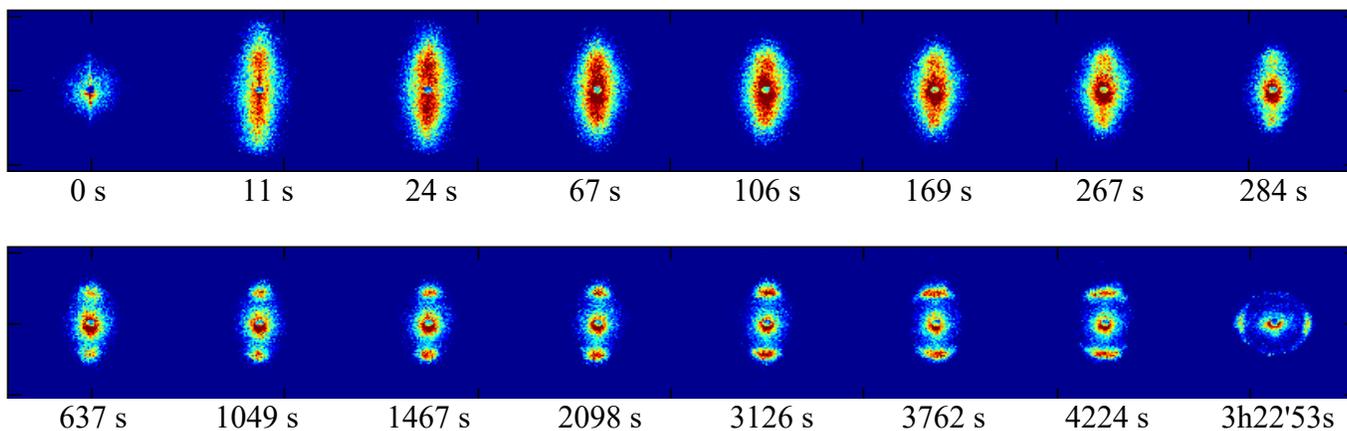
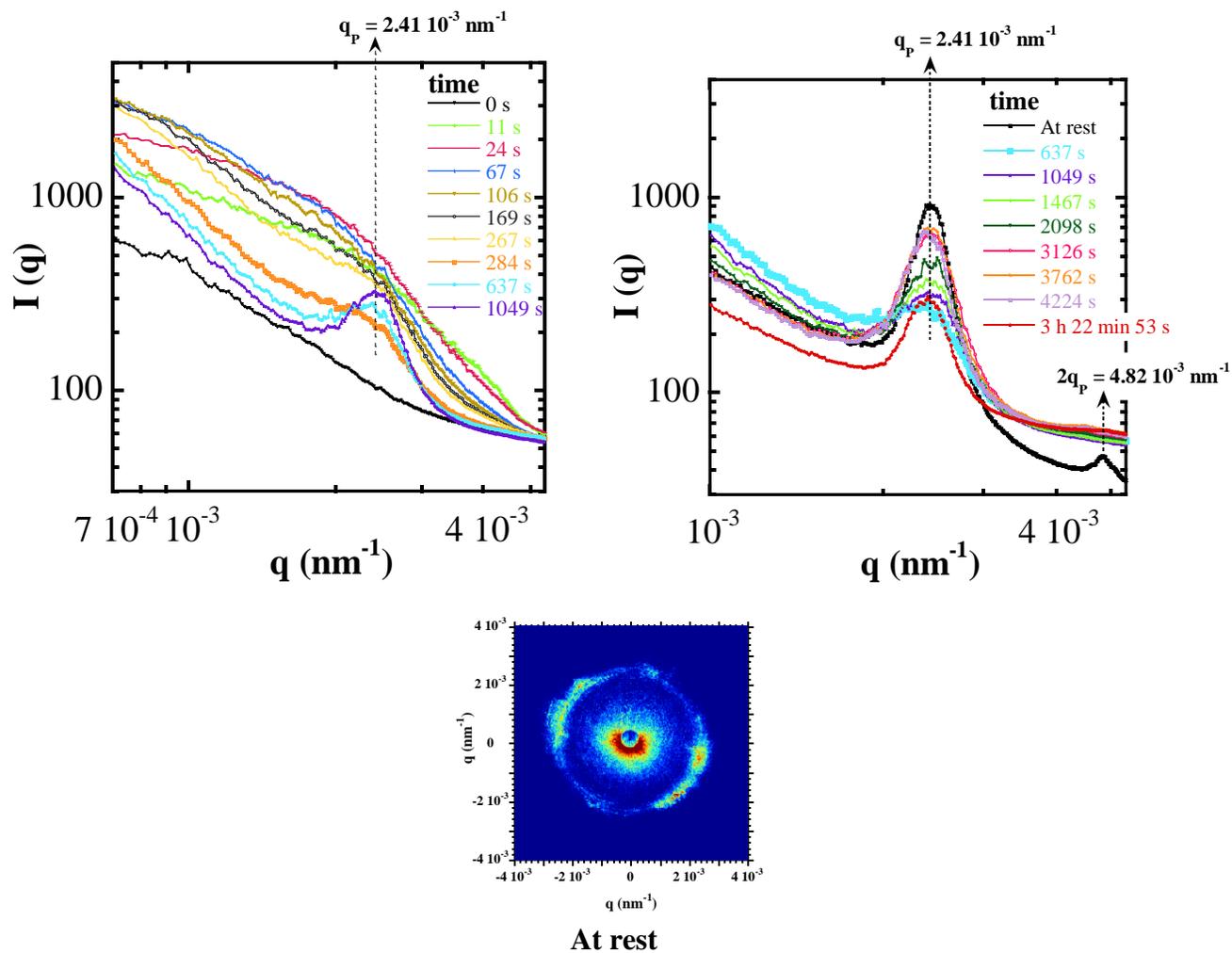
**Figure S8.** SALS patterns and corresponding calculated anisotropy parameter as a function of  $q$  of the CNC suspensions at rest and under shear in steady state for different increasing shear rate belonging to regime I, II and III and pertaining to procedure P2.  $C = 9$  wt% ( $\phi = 5.82$  vol%) with  $0.01 \text{ mol L}^{-1}$  NaCl at  $T = 25 \text{ }^\circ\text{C}$ .



**Figure S9.** SALS patterns and corresponding calculated anisotropy parameter as a function of  $q$  of the CNC suspensions under shear in steady state for different increasing shear rate belonging to regime II and III and pertaining to procedure P2. ( $C = 10$  wt%, ( $\phi = 6.49$  vol%) with  $0.01 \text{ mol L}^{-1}$  NaCl at  $T = 25$  °C. vol%.



**Figure S10.** Transient state relaxation of the CNC suspensions under *in situ* SALS. Scattering patterns and corresponding  $I_v$  a) and  $I_h$  b) sector averaged of the scattered intensity as a function of time, pertaining to procedure P3 with initial shear rates belonging to regime III at  $1068 \text{ s}^{-1}$ .  $t = 0 \text{ s}$  correspond to the cessation of flow.  $C = 9 \text{ wt\%}$  ( $\phi = 5.82 \text{ vol\%}$ ) with  $0.01 \text{ mol L}^{-1} \text{ NaCl}$  at  $T = 25 \text{ }^\circ\text{C}$ .



**Figure S11.** Transient state relaxation of the CNC suspensions under *in situ* SALS. Scattering patterns and corresponding radial averaged of the scattered intensity as a function of time, pertaining to procedure P3 with initial shear rates belonging to regime III at  $1068 \text{ s}^{-1}$ .  $t = 0 \text{ s}$  correspond to the cessation of flow.  $C = 10 \text{ wt\%}$  ( $\phi = 6.49 \text{ vol\%}$ ) with  $0.01 \text{ mol L}^{-1} \text{ NaCl}$  at  $T = 25 \text{ }^\circ\text{C}$ .