## **Supporting Information for Publication**

## Orientation of Cellulose Nanocrystals Controlled in Perpendicular Directions by Combined Shear Flow and Ultrasound Waves Studied by SAXS

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**Figure S1.** a) Anisotropic phase ( $\phi_{LC}$  volume fraction) of CNC suspensions as a function of concentration, b) Kratky plot [corresponding to  $q^2 \cdot I(q)$  vs q] of the SAXS data and corresponding  $q_{\text{peak}}$ , and c) concentration as a function of mean interparticle distance,  $d=2\pi/q_{\text{peak}}$  of the CNC suspensions studied.

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**Figure S2.** Calculation from Son (2007) of mean shear rate  $\dot{\gamma}_{yx}$ , 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 CNC concentrations C = 9.1 wt%. *n*1, *n*2, *n*3 values of shear thinning index according to regimes I, II and III, respectively (C = 9.1 wt%: *n*1 = 0.0940, *n*2 = 0.9626, *n*3 = 0.6401).



**Figure S3.** Transient state of orientation level (*PCA anisotropy*) and corresponding pressure gauge (entry P1) and (outlet P2) measurements in arbitrary units (a.u.) of CNC suspensions undergoing successive push and pull flow-rate inducing a flow in (+V) and (-V) direction respectively, at shear rate of 42.2 s<sup>-1</sup>. C = 9.1 wt%, 0.01 mol L<sup>-1</sup> NaCl.



**Figure S4.** Transient state of orientation level (*PCA anisotropy*) and corresponding pressure gauge (entry P1) and (outlet P2) measurements in arbitrary units (a.u.) of CNC suspensions undergoing successive push and pull flow-rate inducing a flow in (+V) and (-V) direction respectively, at shear rate of 67.6 s<sup>-1</sup>. C = 9.1 wt%, 0.01 mol L<sup>-1</sup> NaCl.



**Figure S5.** Transient state of orientation level (*PCA anisotropy*) and corresponding pressure gauge (entry P1) and (outlet P2) measurements in arbitrary units (a.u.) of CNC suspensions undergoing successive push and pull flow rate inducing a flow in (+V) and (-V) direction respectively at the same shear rate of 92.9 s<sup>-1</sup>. At t = 102 s, upon cessation of flow, the beginning of relaxation of orientation (*PCA anisotropy*) is also followed during time. C = 9.1 wt%, 0.01 mol L<sup>-1</sup> NaCl.



**Figure S6.** Transient state of orientation level (*PCA anisotropy*) and maximum scattering direction  $(\psi_0)$ , and corresponding 2D-SAXS patterns of CNC suspensions successively undergoing step 1: shear flow alone; step2: shear flow (+V) associated to US radiation; step 3: shear flow and US radiation stopped. The applied shear rate was 8.45 s<sup>-1</sup> and the acoustic pressure was  $P \cong 297$ kPa C = 5.2 wt%, 0.01 mol L<sup>-1</sup> NaCl.