Experimental report

05/09/2021

Proposal: **7-01-511** **Council:** 10/2019

Title: Acoustic phonon dispersion along the chain of fully deuterated cellulose crystal

Research area: Materials

This proposal is a new proposal

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Samples: $(C_6D_{10}O_5)_n$

Instrument	Requested days	Allocated days	From	To
IN6-SHARP	3	3	11/09/2020	14/09/2020

Abstract:

The acoustic phonon dispersion of cellulose will be measured to estimate the ideal longitudinal modulus of crystalline cellulose, which is the principal component of plant biomass, and the tensile load bearing element in many materials. The measurement of inelastic X-ray scattering measurement on hydrogenated sample left high uncertainty in the phonon velocity. We obtained fully deuterated cellulose sample, which should allow coherent inelastic scattering to measure the excitation along the acoustic phonon dispersion to drastically improve the reliability of the elastic constant estimation. The dynamic structure factor will be measured along the chain direction to a Q-range of 30 nm-1 and energy transfer of 100 meV as the modulus is estimated in the 200 GPa range.

Experimental report

7-01-511 Acoustic phonon dispersion along the chain of fully deuterated cellulose crystal

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The elastic properties of cellulose is of interest, as it constitutes the main load bearing element in plant, and also is the most important biomass used mainly for its mechanical properties in structural materials. Due to the nanometric size of the crystals, direct determination of the elastic tensor is impossible. The elastic modulus of cellulose crystal has been studied by uniaxial stretching of uniaxially oriented fiber bundles, or by probing mechanical response of individual nano-objects by atomic force microscope but the interpretation and delicate and the uncertainty of the obtained values are high. Another straightforward measurement was phonon dispersion experiment using X-ray, which gave a transvers modulus of 15 GPa, and a longitudinal modulus of 220 ± 50 GPa. The high dispersion being due to the fact that the energy transfer region was limited to around 30 meV. We thought inelastic neutron scattering would cover this range to obtain a more reliable estimation of the Young's modulus.

Perdeuterated cellulose was produced by acetobacter, acclimated in heavy water medium and fed with deuterated glycerol as carbon source. About 200 mg dry bacteria cellulose sheet having planar orientation was mounted with either film plane

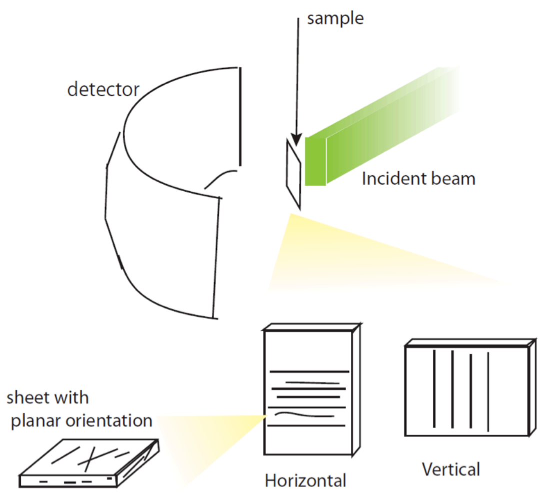

Figure 1. Schematic illustration of sampe geometry

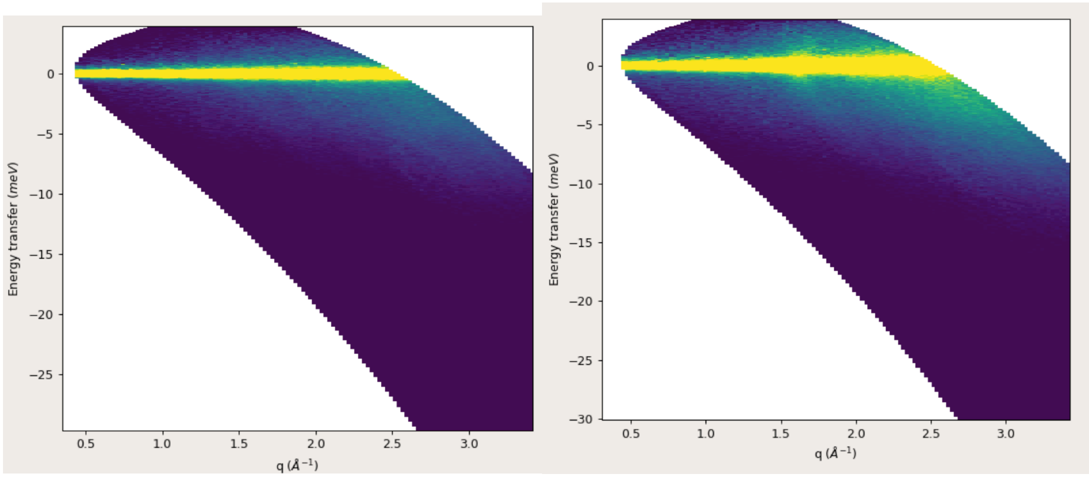

Figure 2. Inelastic neutron scattering with sample mounted, left: horizontally and right: vertically. Some signal of phonon dispersion can be observed at q = 1.6 $Å^{-1}$ (right) corresponding to 2 0 0 reflection but no indication of phonon dispersion could be seen along the fiber direction (left).

horizontal or vertically while the scattered neutron was detected by a banana-shaped detector covering wider horizontal angles (Figure 1). The incident neutron wavelength was 4 Å.

Figure 2 shows the neutron inelastic scattering data binned as function of scattering vector and energy transfer. No clear peak could be observed with horizontal geometry, whereas dispersion can be directly seen close to the 2 0 0 reflection, which also has the largest structure factor. Indeed, the static structure factor along the fiber direction is small, and due to the longer wavelength of neutron compared to the typical inelastic X-ray scattering experiment, the reflections along the fiber direction were buried in the incoherent scattering.

The density of states as function of energy transfer of the per-deuterated bacterial cellulose measured in the current experiment is compared to inelastic neutron DOS measured on flax fibers published in 2000 (1). The peak is clearly shifted to lower energy with per-deuteration from 22.5 meV to 20 meV. This difference would be used in vibration analysis to attribute the corresponding dominant molecular vibration mode.

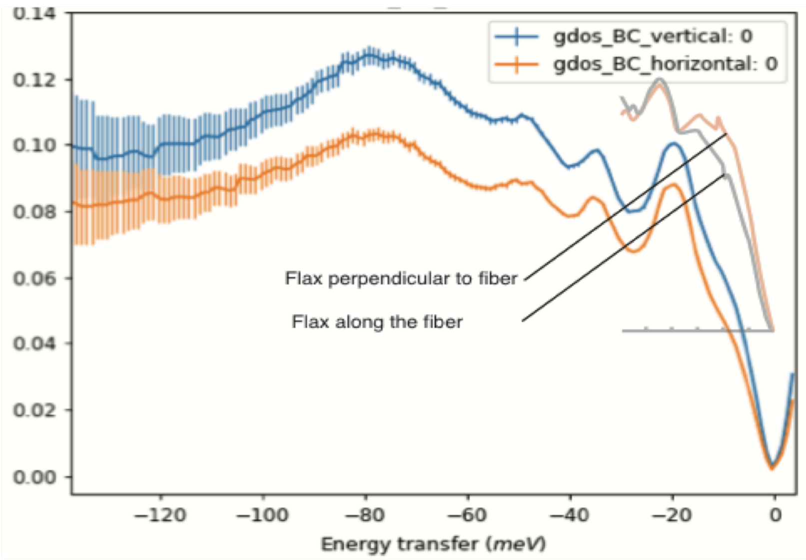

Figure 3. Density of states of per-deuterated bacterial cellulose compared to the flax fiber (undeuterated) samples.

References

1. Müller M, Czihak C, Schober H, Nishiyama Y, Vogl G. All Disordered Regions of Native Cellulose Show Common Low-Frequency Dynamics. Macromolecules. 2000; 33: 1834 - 1840.