3D Imaging and Understanding the Structure of the Chiral Nematic assembly of Cellulose Nanocrystals in Suspension

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We exploited the inherent confocal effect of Second Harmonic Generation (SHG) microscopy to acquire highly resolved 3D images of the chiral nematic phase of ribbon-like cellulose nanocrystals (CNC) and CNC tactoids in the "isotropic" phase in a label-free manner and in the liquid state. The response to the polarization of the laser beam agreed with a single dominant tensor component along the director axis of the (chiral) nematic liquid crystal planes, showing that the anisotropy of the CNCs dominates their assembly. Rotation of the director results in variations in SHG intensity which can be directly linked to the helical pitch and used to determine directionality of the chiral nematic assembly. This gave us access to high contrast 3-dimensional images of the aligned liquid crystal phase, providing unprecedented detail into their structure in the bulk liquid state. Local alignment, morphology, and the presence of defects are readily visible and can be determined. In addition, through 3D-FFT analysis, we were able to fully characterize the periodicity of the helicoidal structures and allowed for a precise estimate of the helicoidal pitch and identification of preferred directions within the investigated volume. Tomographic images of the "isotropic" phase further enabled us to visualize and measure tactoids in 3D, making it possible to determine a volume range within which tactoids morph from the nematic to cholesteric state and obtain a value for the twist ratio.
In this presentation, we will discuss how SHG microscopy can be used to characterize CNC self-assembled structures in the bulk liquid state (but the fully developed chiral nematic state and tactoids formed in the "isotropic" state). We will further illustrate the numerous benefits SHG microscopy offers and discuss the framework for interpreting the images obtained through this technique.

Keywords:
Second Harmonic Generation Microscopy, Cellulose Nanocrystal Assembly, Chiral Nematic Structure

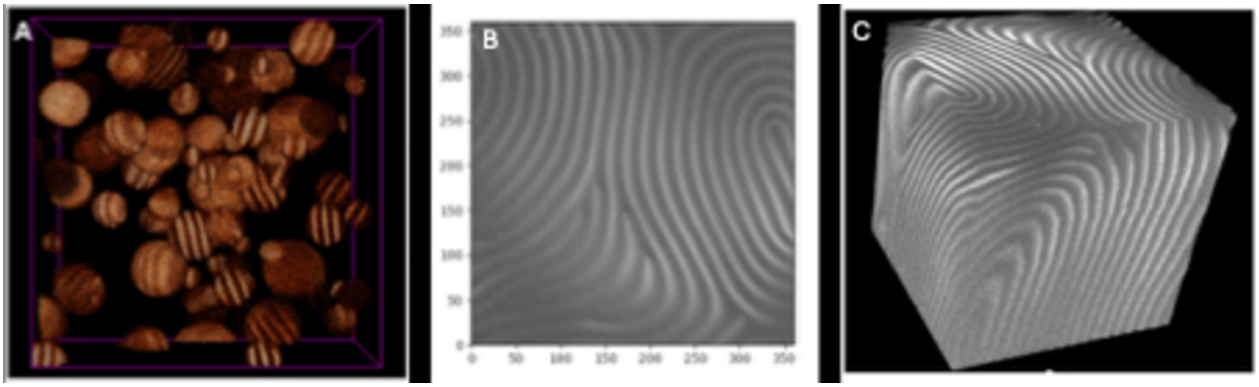

Figure 1. (A) 3D SHG image of an "isotropic" phase of a 2 wt% CNC suspension showing nematic and cholesteric tactoids, (B) 2D SHG image of a plane in a CNC chiral nematic phase (8 wt% CNC suspension), and (C) 3D SHG image of chiral nematic phase (8 wt% CNC suspension) by SHG focus stacking of 2D SHG

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