

Supporting Information

Vibrational Sum-Frequency-Generation (SFG) Spectroscopy of Bulk Materials Consisting of SFG-Active Nanocrystals: Experimental and Theoretical Study of Azimuth Angle and Polarization Dependences of SFG Spectral Features of Uniaxially-Aligned Cellulose Crystals

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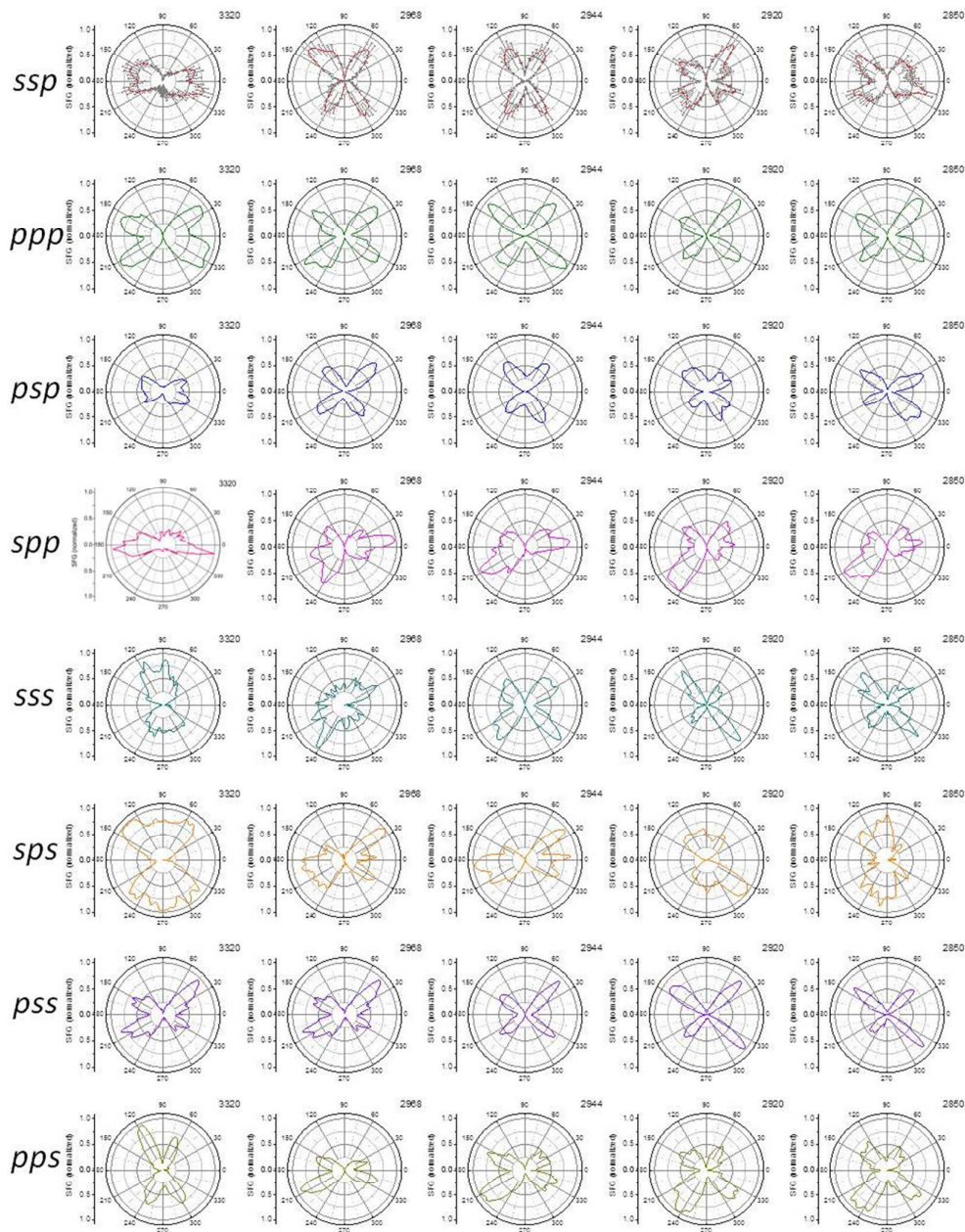


Figure S1. Full data set of the azimuth angle dependence for four SFG peaks in the CH/CH₂ stretch region (2850, 2920, 2944, and 2968 cm^{-1}) and one SFG peak with the maximum intensity in the OH stretch region (3320 cm^{-1}) measured using the ps-scanning system with eight different polarization combinations (*ssp*, *ppp*, *psp*, *spp*, *sss*, *sps*, *pss*, *pps*). The data shown here are the average of signals from multiple locations (4 – 6 spots).

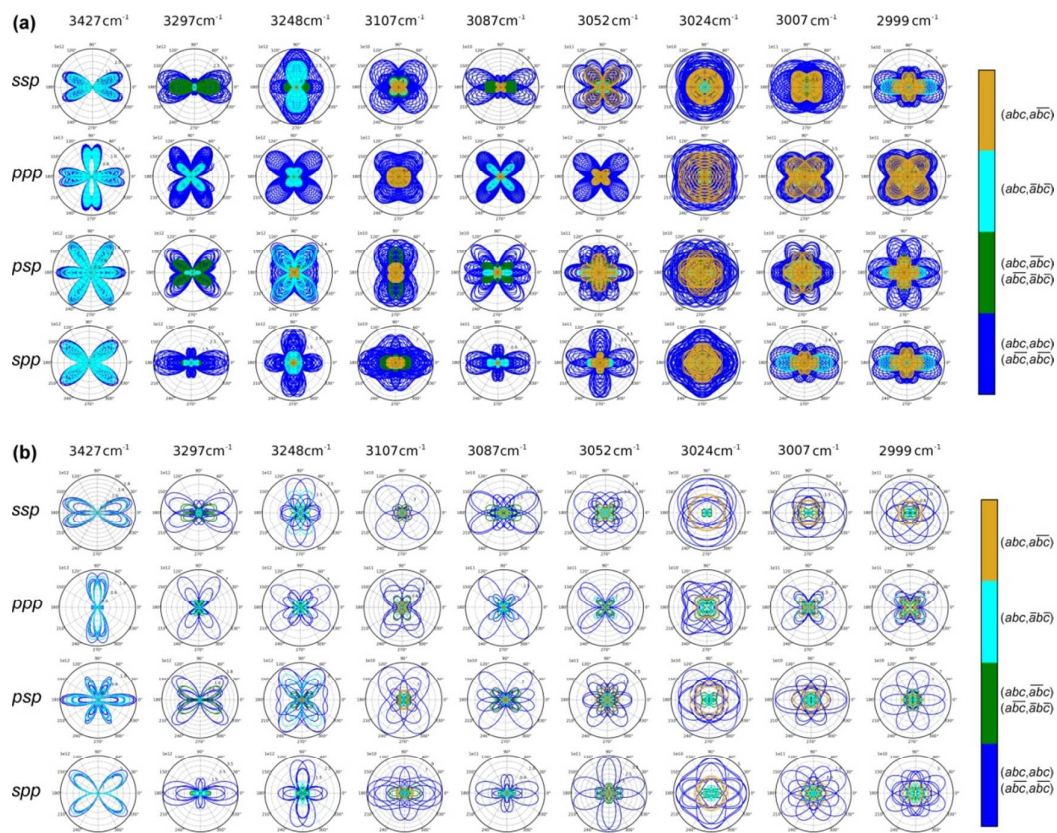


Figure S2. Full TD-DFT calculation results for the azimuth angle dependence of 6 CH/CH₂ modes (2999, 3007, 3024, 3052, 3087, 3107 cm⁻¹) and 3 OH modes (3248, 3297, and 3427 cm⁻¹) for 4 different polarization combinations (ssp, ppp, psp, spp). The random packing and preferential packing of crystallites with respect to the film surface are shown separately in (a) and (b), respectively. In each case, the data show the individual cases of 4 different crystallite packing within the SFG coherence length.

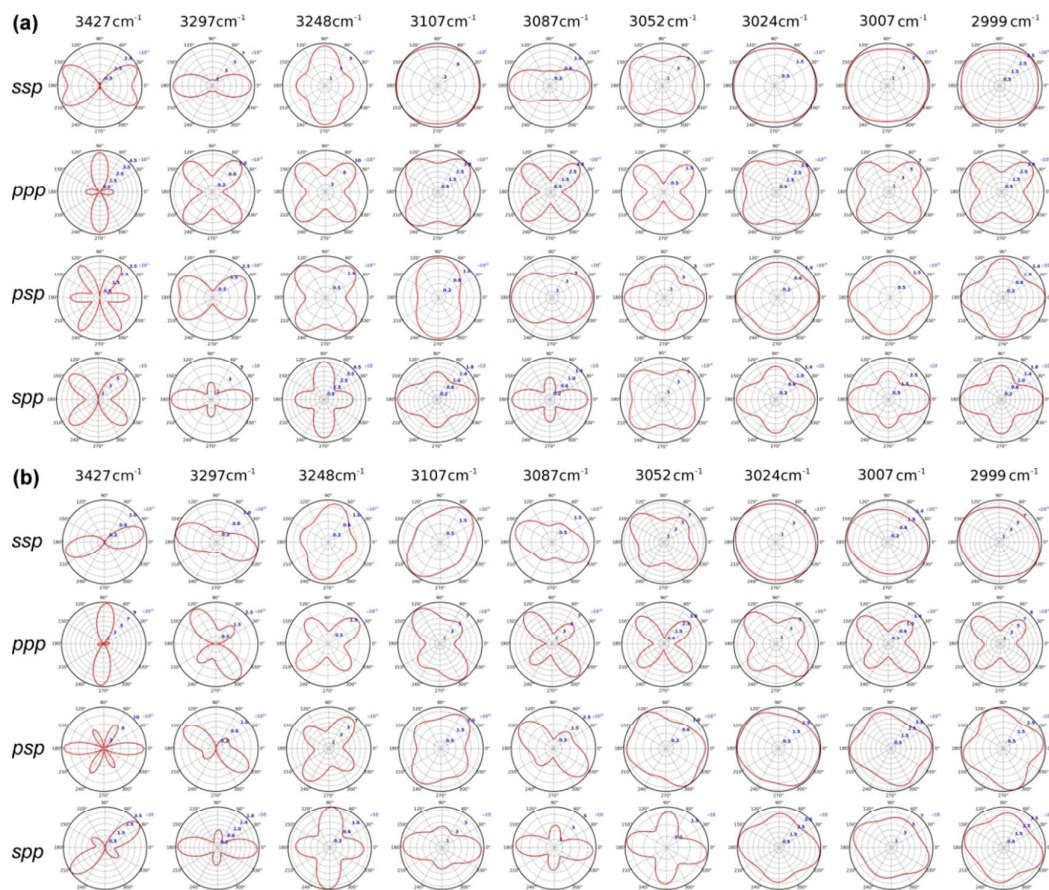


Figure S3. Full TD-DFT calculation results for the azimuth angle dependence of 6 CH/CH₂ modes (2999, 3007, 3024, 3052, 3087, 3107 cm⁻¹) and 3 OH modes (3248, 3297, and 3427 cm⁻¹) for 4 different polarization combinations (ssp, ppp, psp, spp). The averages for the random and preferential packing of crystallites with respect to the film surface are shown separately in (a) and (b), respectively.