

Supporting information

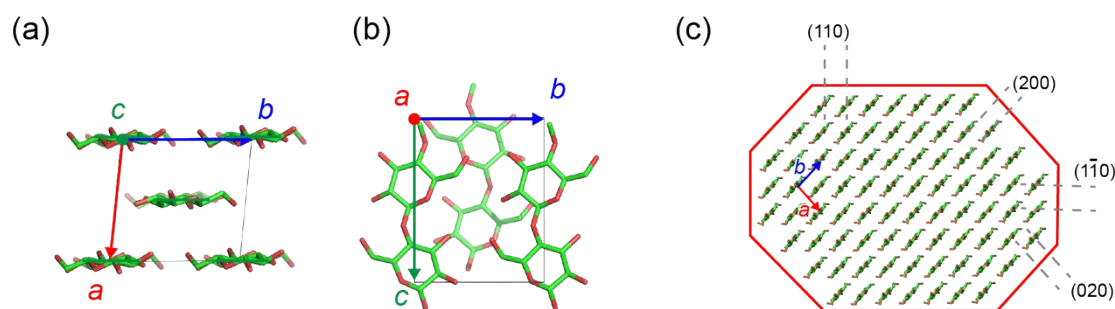


Figure S1. Ultrastructure of a tCNC (a) ab projection of cellulose I β , (b) bc projection of cellulose I β , (c) molecular packing in cross section of the tCNC.

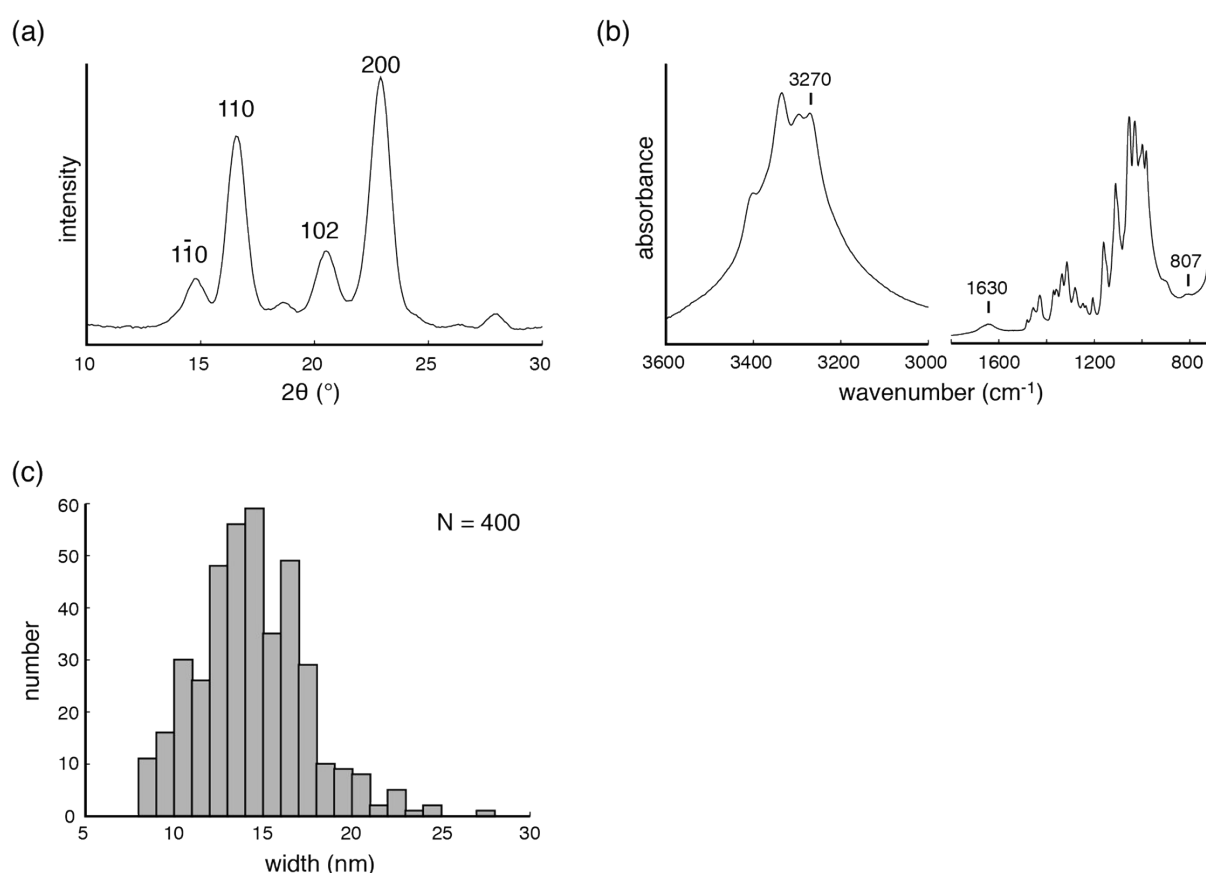


Figure S2. (a) X-ray powder diffraction profile of tCNCs. The indices of characteristic reflections of cellulose I β are indicated upon each peak. (b) ATR FT-IR spectrum of tCNCs. A characteristic O-H stretching band of cellulose I β is visible at 3270 cm^{-1} while the one at 3240 cm^{-1} , characteristic band of the I α allomorph is absent. The bands at 1630 cm^{-1} and 807 cm^{-1} are attributed to O-H scissoring and C-O-S bending vibrations, respectively. The absence of amide II band at around 1540 cm^{-1} indicates that there is no detectable amount of protein in the specimen. (c) Width distribution of tCNCs obtained from cryogenic observations.

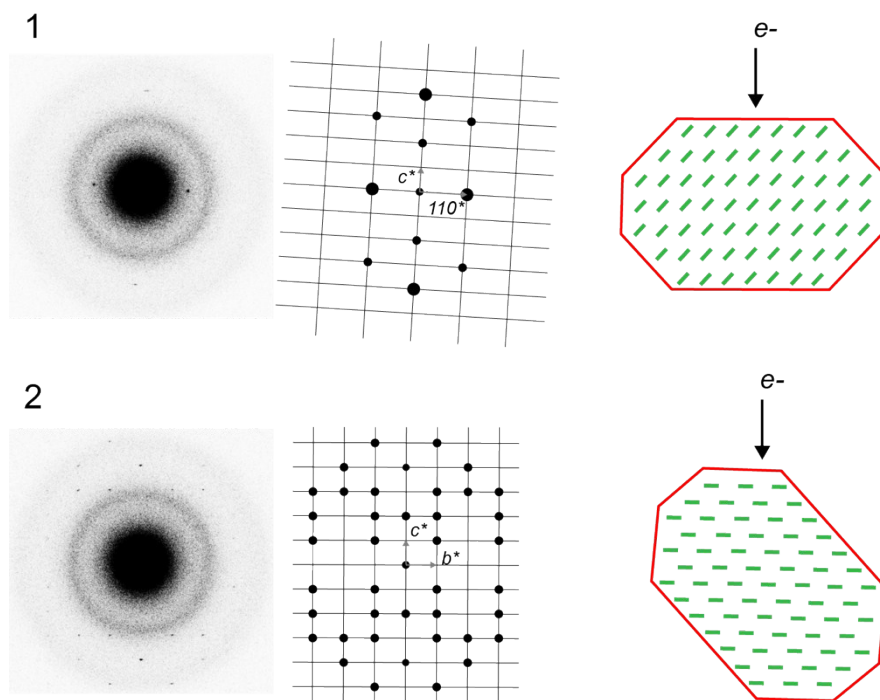


Figure S3. Electron diffraction diagrams, corresponding projections of reciprocal lattice, and cross sectional orientations of cellulose nanocrystals at each acquisition points shown in Fig. 2a in the main text.

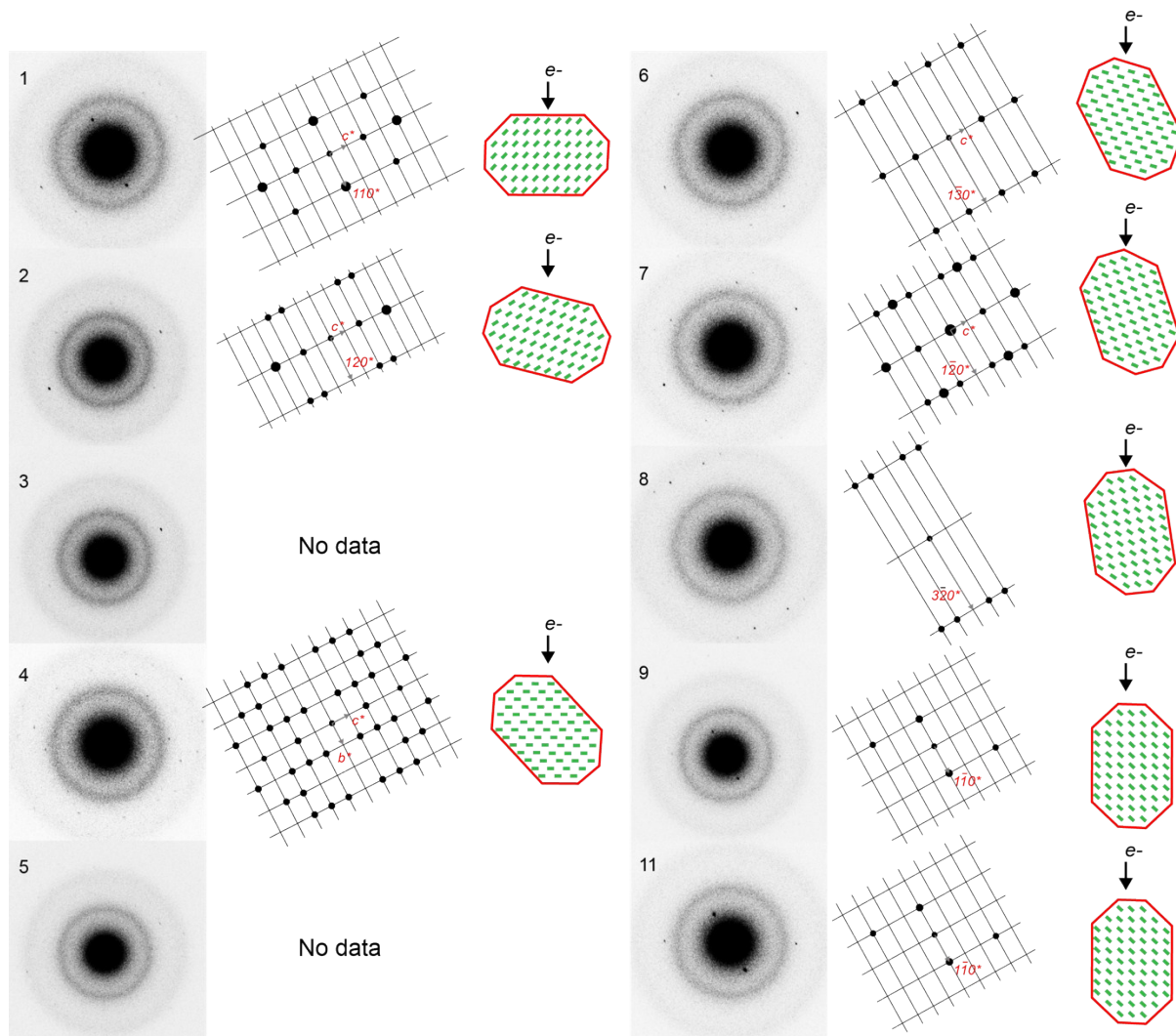
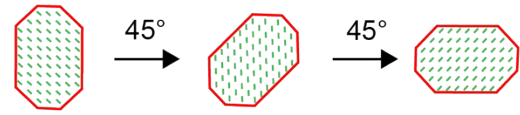
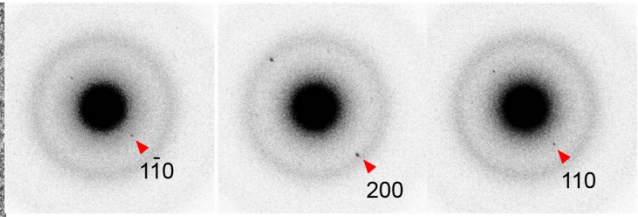
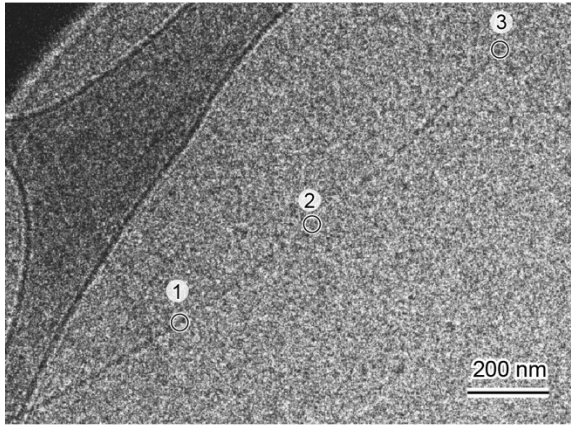


Figure S4. Electron diffraction diagrams, corresponding projections of reciprocal lattice, and cross sectional orientation of cellulose nanocrystals at each acquisition points shown in Fig. 2c in the main text.

(a)



(b)

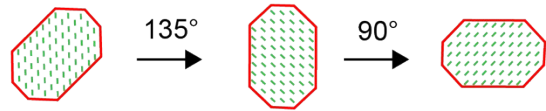
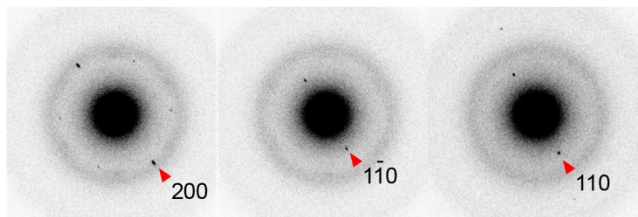
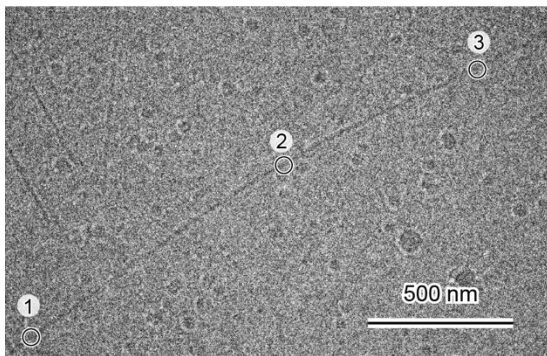
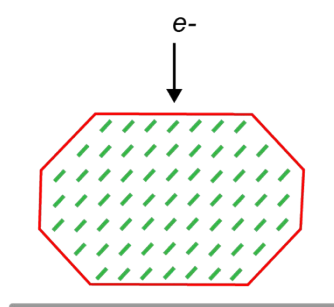
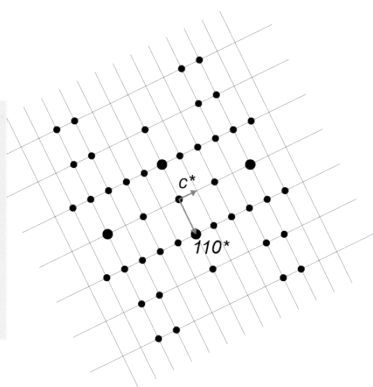
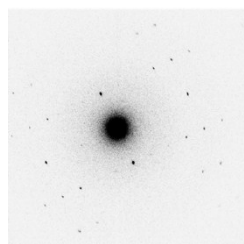


Figure S5. μ ED analysis of tCNCs embedded in vitreous ice. Twist pitches were (a) $10^\circ/100$ nm and (b) $13^\circ/100$ nm.

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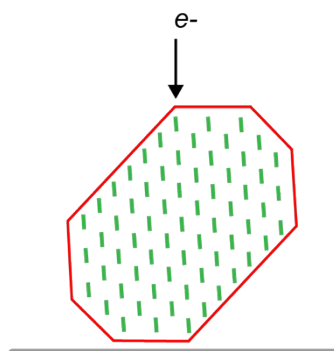
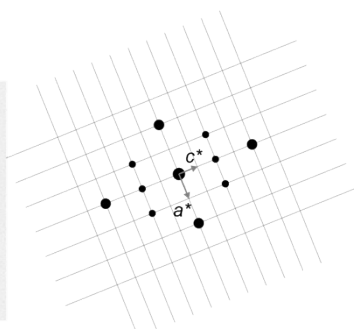
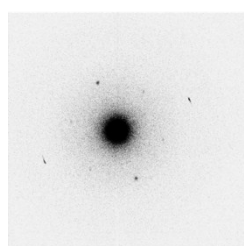


Figure S6. Electron diffraction diagrams, corresponding projections of reciprocal lattice, and cross sectional orientations of cellulose nanocrystals at each acquisition points shown in Fig. 3a in the main text.

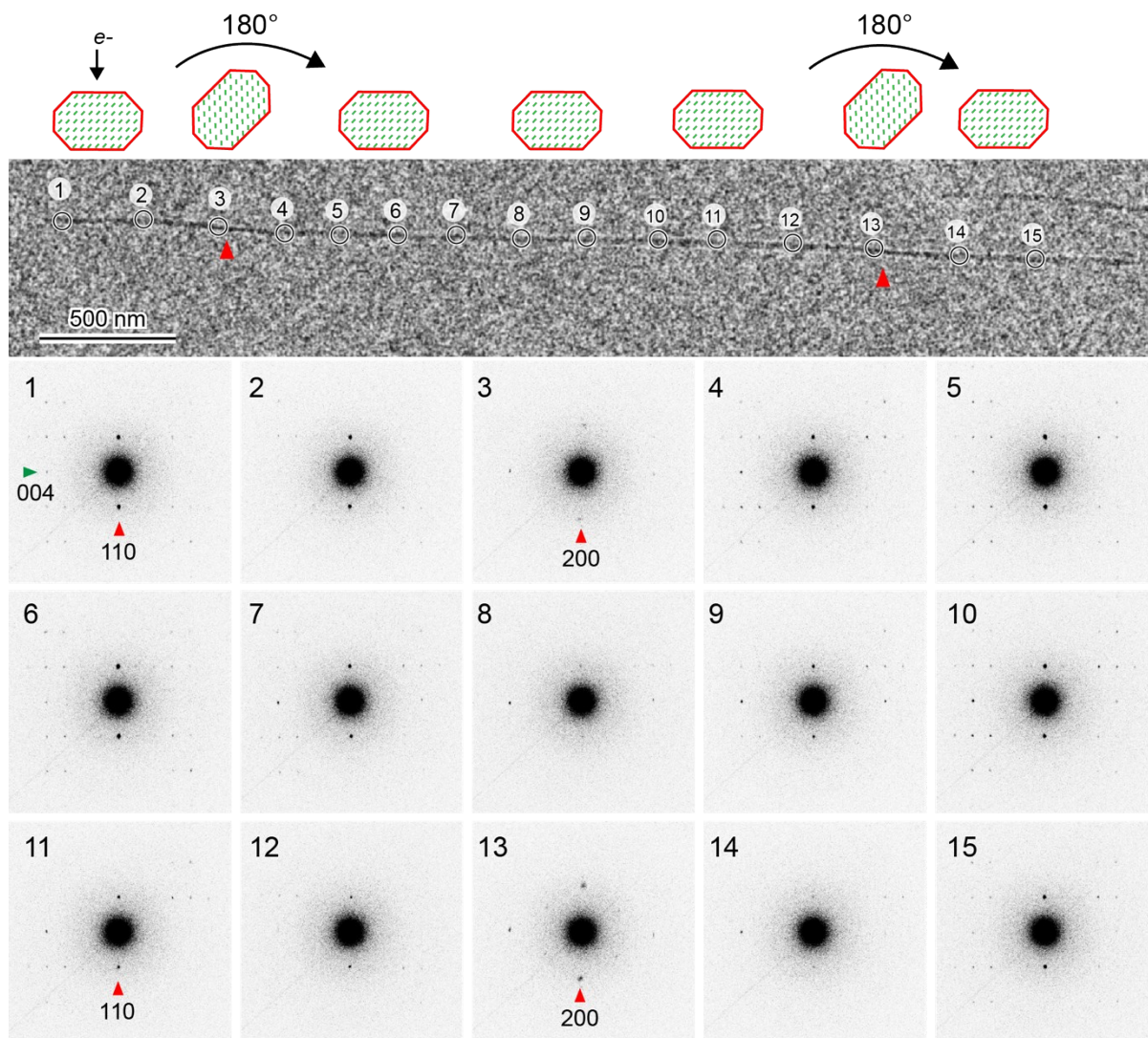


Figure S7. μ ED analysis of a tCNC dried on flat carbon film. Apparent twist regions were indicated by red arrowheads.

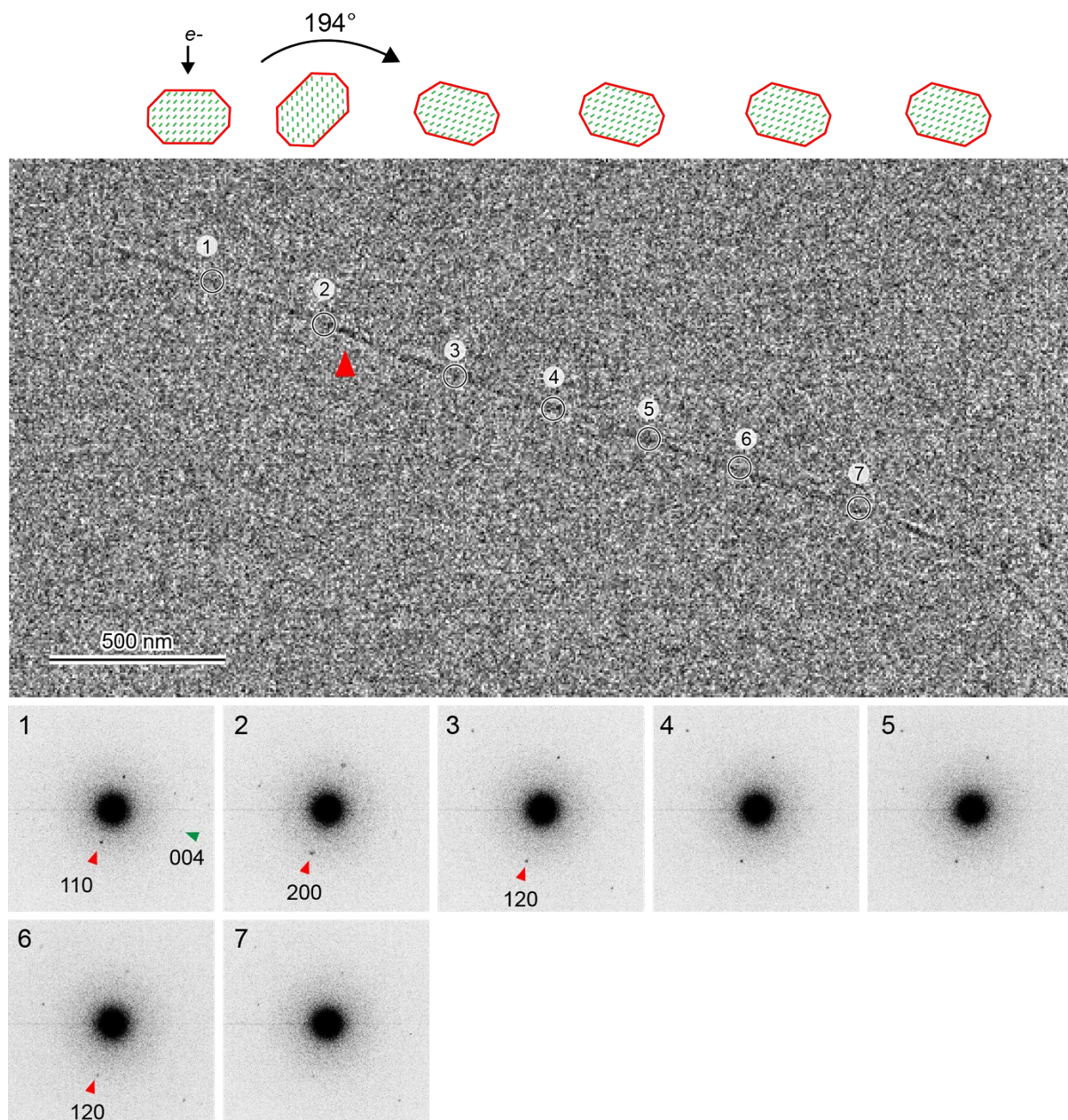


Figure S8. μ ED analysis of a tCNC dried on flat carbon film. An apparent twist region is indicated by red arrowheads.