Twisted Cellulose

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Lignocellulosic materials often display twisted structures at many length scales. Tree trunks, wood sections, paper strips, wood cell walls, cellulose microfibrils, nanocellulosic materials and cellulose derivatives all display a spontaneous twist under appropriate conditions. In many cases, the twist is chiral, displaying a single handedness. The cellulose molecular structure of natural polysaccharides such as cellulose is also chiral, but the relationship between molecular chirality and higher expressions of chirality in natural materials is poorly understood.

A truly twisted tree, *Leptospermum laevigatum,* in San Francisco's Golden Gate Park

The twisted organization of chiral nematic phases in suspensions of cellulose nanocrystal (CNC) suspensions [1] should provide a tractable *in vitro* system linking molecular and macroscopic chiral structures. Difficulties remain. Most theoreticians now agree that crystalline natural cellulose should form nanofibrils with a right-handed helical twist, but experimentalists have difficulty providing solid evidence for this twist in nanocellulose samples from cotton or wood pulp. Conversely, experimentalists have strong evidence that suspensions of CNC normally form a left-handed helicoidal arrangement in suspension, but theoreticians have a hard time explaining why. It is also clear that the twisting inherent in chiral nematic phase formation is in competition with the tendency of concentrated solutions and suspensions to undergo kinetic arrest due to gelation and glass formation [2, 3]. While many questions remain, there has been significant recent progress in understanding the ordering and twisting tendency of chiral natural materials.

[1] J.-F. Revol, H. Bradford, J. Giasson, R.H. Marchessault and D.G. Gray, Int. J. Biol. Macromol., 14, pp. 170-172, 1992.

[2] D.G. Gray, Nanomaterials, 6, 11, p. 213, 2016.

[3] C. Honorato-Rios, A. Kuhnhold, J.R. Bruckner, R. Dannert, T. Schilling and J.P.F. Lagerwall, Frontiers in Materials, 3, 21, 2016.