

Twist and shout: chirality amplification and fluctuation modes in the cholesteric phase of short DNA

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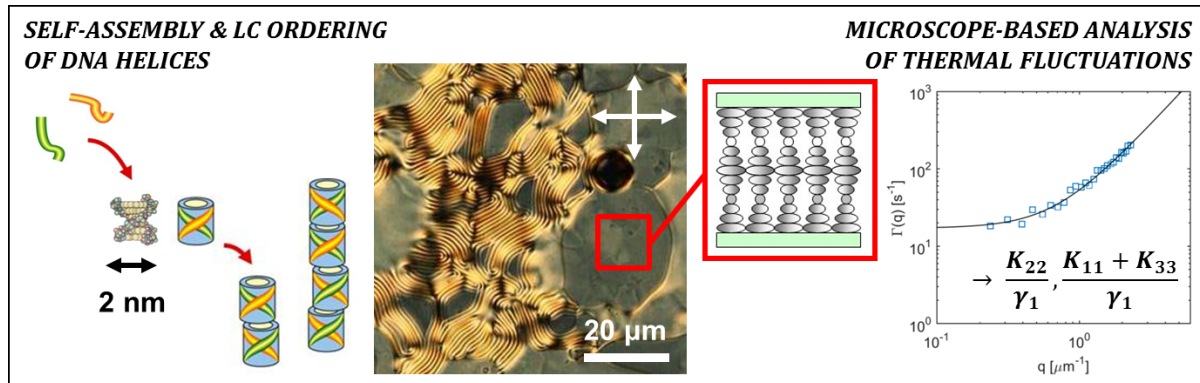


Figure. Short DNA helices self-assemble and order into Cholesteric phase, whose viscoelastic ratios can be extracted from Fourier analysis of fluctuations in microscopic textures.

Predicting how the molecular chirality propagates up to the mesoscale remains a challenge even for systems in which the molecule has the simple form of a straight helix. This is the case of DNA short double strands, whose concentrated aqueous solutions show Cholesteric (N^*) liquid crystalline (LC) ordering, mediated by the reversible end-to-end aggregation of duplexes into linear aggregates. Right-handed DNA helices show an extremely diverse behaviour, with both left- and right-handed N^* phases and pitches from the visible range up to several μm [1]. The N^* behaviour depends on the length and sequence of the oligomers, and on their aggregation mode [2]. The combination of these parameters determines the concentration range of the N^* phase, setting the balance between electrostatic and steric interactions and thus phase handedness.

Measuring viscoelastic properties of N^* can provide important information on chiral torque and the delicate interplay of underlying molecular interactions. However, traditional light scattering techniques cannot be used in samples like DNA because of the lack of suitable alignment strategies. To study thermal fluctuation of N^* textures, we thus exploit the unique space-resolving capabilities of polarized Differential Dynamic Microscopy (p-DDM), recently demonstrated for Nematic LCs [3].

Here, we extend p-DDM to the more complex, coupled N^* modes [4], and validate it with a thermotropic LC, finding good agreement with literature data. Similar measurements performed on DNA N^* enable extracting splay-bend and twist viscoelastic constants as a function of temperature. Surprisingly, we also find an additional mode with non-diffusive relaxation, possibly related to the end-to-end aggregation.

[1] G. Zanchetta et al., PNAS 107, 17497 (2010).

[2] M. Rossi et al., PRL 110, 107108 (2013).

[3] F. Giavazzi et al., Soft Matter 10, 3938 (2014).

[4] J. Yoshioka, Y. Takanishi, J. Yamamoto, EPL 98, 16006 (2012).