## Dynamics of ordered colloidal particle monolayers at nematic liquid crystal interfaces: Supplementary Information

Wei-Shao Wei,\*<sup>a</sup> Mohamed Amine Gharbi,<sup>b,c</sup> Matthew A. Lohr,<sup>a</sup> Tim Still,<sup>a</sup> Matthew D. Gratale,<sup>a</sup> T. C. Lubensky,<sup>a</sup> Kathleen J. Stebe,<sup>d</sup> and A. G. Yodh<sup>a</sup>

<sup>a</sup> Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

<sup>b</sup> Department of Physics, McGill University, Montreal, Quebec, Canada

<sup>c</sup> Department of Chemistry, McGill University, Montreal, Quebec, Canada

<sup>d</sup> Department of Chemical and Biomolecular Engineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA



Supp. Fig. 1 (a) Dispersion curves for transverse (red) and longitudinal (black) modes along one of the two high-symmetry directions,  $\Gamma$ -M. The blue solid lines are the linear fits for linear dispersion (*i.e.*, low frequency) regimes, whose slopes give the sound velocities  $c_T = 23.6 \pm 0.3$  mm/s and  $c_L = 62.8 \pm 0.8$  mm/s, respectively. (b) The same process is applied to direction  $\Gamma$ -L and gives  $c_T = 24.4 \pm 0.9$  mm/s,  $c_L = 57.6 \pm 0.3$  mm/s. The measurement was performed at room temperature, 21.8 °C. Inset: High-symmetry directions in reciprocal space.



**Supp. Fig. 2** Our temperature-dependent study shows that as the system temperature decreases, the range of eigenfrequencies becomes wider, which arises from the distribution broadening of effective spring constants as shown in the plot. The effective spring constant becomes larger with decreasing temperature, since the elastic constants increase with decreasing temperature. This effect, in turn, leads (at least in part) to a shifting (and some broadening) of the  $\kappa_{eff}$  distribution and thus to a broader width for the DOS with respect to frequency. Moreover, several mechanisms may also induce such disorder, including non-uniform wetting (caused by uneven DMOAP functionalization or uneven surface roughness of colloidal particles) and slowly-relaxed contact lines of colloids at interfaces.<sup>1</sup> The hypothesis is that, since the relaxation is slower at lower temperature due to a larger LC viscosity,<sup>2-8</sup> the contact line needs more time to relax and would be slightly more "zigzag" (caused by non-uniform wetting) when we observed. The resultant heterogeneities could then contribute to a broadening of the  $\kappa_{eff}$  distribution.



Supp. Fig. 3 (a) One of the candidate approaches for interparticle interaction is described as follows. The interface with air prevents dipolar distortions within the semi-infinite air-exposed region. Due to the strong homeotropic anchoring at the air-NLC interface, and by analogy with the method of image charges in electrostatics, the texture in our system is equivalent to the director around two particles (with the same topological charge; one real and the other virtual), which locate symmetrically about the interface. For this situation, the leading asymptotic term in the potential is thus given by the "electric" quadrupole term.<sup>9</sup> Two particles would then interact elastically in the far field with the pair potential  $U_q(r) = 36\pi\beta^2 R^6 K/r^5$ , where  $\beta$  is a material-dependent unitless parameter.<sup>9,10</sup> A corresponding form for the effective spring constant can be derived within the harmonic approximations used in the displacement covariance matrix method by taking the second derivative of  $U_a(r)$  to find  $\kappa_a(r) = 1080\pi\beta^2 R^6 K/r^7$ . To compare this form to experiment, a curve of power  $r^{-7}$  and its 99% confidence band are fitted and plotted together with the data (consult Fig. 5 in main article). The expected quadrupolar form does not agree well with our experimental data. (b) Fully immersed colloids with homeotropic anchoring typically form dipolar defects in aligned bulk NLC.<sup>10</sup> We compare the power law corresponding to interacting dipoles with our data. We express the pair potential as well as effective spring constant as  $U_d(r) = 4\pi \alpha^2 R^4 K/r^3$  and  $\kappa_d(r) =$  $48\pi\alpha^2 R^4 K/r^5$ , where  $\alpha$  is again a unitless parameter. The dipolar fit of a curve of power  $r^{-5}$  is shown; while the fit is improved, the uncertainties in the data preclude us from asserting that the particles interact as elastic dipoles. Inset: The binned data is re-plotted with both axes logarithmically scaled. The best linear fit gives a power law of  $-3.43 \pm 2.41$  (slope of grey dashed line); for reference, the expected power law for quadrupole and dipole (slope of -7 and -5) are also presented respectively.

## References

- D. M. Kaz, R. McGorty, M. Mani, M. P. Brenner and V. N. Manoharan, *Nature materials*, 2012, 11, 138–142.
- [2] M. Cui and J. R. Kelly, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 1999, 331, 49–57.
- [3] A. G. Chmielewski, *Molecular Crystals and Liquid Crystals*, 1986, **132**, 339–352.
- [4] S. V. Letcher, The Journal of the Acoustical Society of America, 1973, 53, 307–307.
- [5] F. Kiry and P. Martinoty, J. Phys. France, 1977, 38, 153–157.
- [6] K. Skarp, S. T. Lagerwall and B. Stebler, Molecular Crystals and Liquid Crystals, 1980, 60, 215–236.
- [7] H. Herba, A. Szymanski and A. Drzymala, *Molecular Crystals and Liquid Crystals*, 1985, 127, 153–158.
- [8] R. Orr and R. A. Pethrick, Liquid Crystals, 2011, 38, 1169–1181.
- [9] M. Oettel, A. Dominguez, M. Tasinkevych and S. Dietrich, *The European Physical Journal E*, 2009, **28**, 99–111.
- [10] T. C. Lubensky, D. Pettey, N. Currier and H. Stark, Phys. Rev. E, 1998, 57, 610-625.