

Alexey Eremin, Hajnalka Nádasi

Fascinating World of Vector-Symmetry Nematics: Liquid Magnets and Fluid 3D Ferroelectrics

In the past decade, two significant discoveries have been made in the field of self-assembled soft materials: the discoveries of the ferromagnetic and ferroelectric nematic phases [1-9]. The invariance of the nematic director traditionally marks the cylindrical (quadrupolar) symmetry of the nematic phase, and it can be broken at interfaces but not in bulk. In contrast, the nematics with vector symmetry represent examples of genuine 3D polar fluids, whether electric or magnetic.

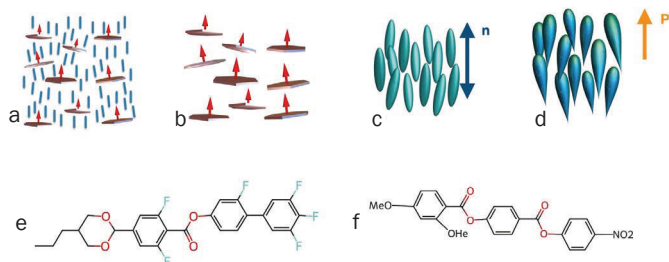


Fig. 1: Structures of vector-symmetry nematics: **a** ferromagnetic nematic dispersion of magnetic nanoplatelets in a nematic host; **b** lyotropic ferromagnetic nematic with an isotropic matrix (the red arrows designate the magnetic moments); **c** structure of a conventional non-polar nematic; **d** structure of a ferroelectric nematic with wedge-shaped molecules (P designates the polarisation). The chemical formula of mesogens exhibiting the N_v phase: **e** DIO, **f** RM734.

In the early seventies, Brochard and de Gennes suggested that dispersing (ferro)magnetic nanoparticles (MNPs) in a nematic host can stabilize a “ferromagnetic” state with a remanent magnetization. However, countless attempts to produce liquid magnets by suspending spherical and rod-shaped MNPs failed. Ferromagnetics turned out to be non-magnetic without an applied external field.

Only in 2013, Mertelj et al. demonstrated that ferromagnetic behaviour is possible in suspensions of magnetic nanoplatelets in a nematic host [1]. The remanent magnetization is stabilized by the coupling between the platelet's magnetization and the nematic director anchored orthogonally to the platelets. Magnetic fields as small as 10 mT allow for the switching of the nematic director, accompanied by nucleation and propagation of the domain walls. The magnetic domains result from the minimization of the stray field energy. Director-magnetization coupling gives rise to a multitude of new effects, such as the

converse magnetoelectric effect, where the magnetization can be controlled by an external electric field. The magneto-optic effect occurs in fields as low as 20 mT. Dissipative dynamics facilitated by the director-magnetization cross-coupling significantly affect the magneto-optic response, resulting in significantly faster relaxation rates.

Chirality, that is easily achieved in liquid crystals, breaks the parity and time-reversal symmetry, giving rise to new topological 2D and 3D soliton structures. Knotted structures of the magnetic director known as skyrmions have been found to form hexagonal lattices in the ferromagnetic cholesterics [3].

An anisotropic LC matrix is not a prerequisite for the stabilization of the ferromagnetic order in a fluid. The platelets alone can form lyotropic nematic even in an isotropic matrix such as *n*-butanol [2, 4]. The magnetic order is attributed to the dipolar chaining of the magnetic platelets, which can be ordered using a small magnetic field. A high degree of dipolar interaction and steric constraints lock the magnetization and stabilize the fluid magnet. Such fluid magnets show a remarkably strong response even to fields comparable to the Earth's magnetic field and complex multimode magnetic dynamics [4].

However, in the case of ferroelectrics, the ferroelectric order was established in chiral smectic phases, exhibiting only 2D fluid order, already in the seventies. The translationally symmetric nematics, 3D fluids, remained apolar. In dielectrics, the first theoretical models of the orientational order developed by Max Born and Peter Debye, as early as 1912, considered dipolar electrostatic interactions as the stabilizing force and predicted nematic phases with a ferroelectric ground state. However, these models were unable to explain the nematic order of nonpolar molecules and the development of quadrupolar order in most nematic phases. Ferroelectric nematics remained elusive until recently. Solely a few lyotropic and polymeric systems showed indications of the polar order in the bulk.

Only in 2017 did Nishikawa et al. and Mandle et al. independently report new LC compounds (DIO and RM734, respectively) exhibiting multiple polymorphic nematic phases [5-6]. Although the high-temperature phases appeared to be nonpolar nematics, the transition to the low-temperature phases was accompanied by a dramatic increase in the dielectric response and the ferroelastic effect, indicating the development of the ferroelectric order. The polar order in this ferroelectric nematic (N_v) phase was evidenced by a large spontaneous second harmonic generation (SHG) signal and the formation of the domains of opposite polarity

Prof. Dr. Alexey Eremin
Dr. Hajnalka Nádasi
Institute for Physics
Otto von Guericke University Magdeburg
Universitätsplatz 2, 39106 Magdeburg
alexey.eremin@ovgu.de

DOI-Nr.: 10.26125/rc5b-bs02

separated by domain walls. Ferroelectric nematics are distinguished by a very high spontaneous polarization in the range of $10 \mu\text{C}/\text{cm}^2$ and remarkable SHG efficiency with the nonlinear optical coefficient in order of $10 \text{ pm}/\text{V}$. These properties make N_F suitable for perspective applications such as electrooptic devices (Pockels effect), nonlinear optics, and sensors.

A combination of fluidity and 3D polar order results in new astonishing properties of N_F materials such as superscreening, interfacial instabilities, fibre formation and active locomotion [8, 9]. The superscreening effect allows the electric field to be guided through curved microchannels, confining the N_F liquid [8]. Without any layer structure, N_F forms freely suspended ferroelectric fibres and even films. At the free surface, the polarization charges interacting with external fields can drive interfacial instabilities, inducing bursting and finger formation in droplets on ferroelectric substrates. When placed between two electrodes and exposed to AC voltage, microscopic N_F bridges display complex dynamic behavior, including active propulsion. This movement is powered by the piezoelectric and electrostriction effects, causing the contact line of the asymmetrical bridges to undergo local stick-slip motion.

The fascinating properties of vector symmetry nematics provide us not only with new, unexpected physics but also with exciting novel applications. The continued exploration of these materials and their unique properties is essential for unlocking their full potential and realizing their practical applications.

References

- [1] Mertelj A, Lisjak D, Drogenik M, Copic M. Ferromagnetism in suspensions of magnetic platelets in liquid crystal. *Nature*. 2013, **504**, 237-241.
- [2] Shuai M, Klitnick A, Shen Y, Smith GP, Tuchband MR, Zhu C, Petschek RG, Mertelj A, Lisjak D, Čopič M, Maclennan JE, Glaser MA, Clark NA. Spontaneous liquid crystal and ferromagnetic ordering of colloidal magnetic nanoplates. *Nature Communications*. 2016, **7**, 10394.
- [3] Mertelj A, Lisjak D. Ferromagnetic nematic liquid crystals. *Liq Cryst Rev*. 2017, **5**, 1-33.
- [4] Nádasi H, Küster M, Mertelj A, et al. Role of ionic surfactant in magnetic dynamics of self-assembled dispersions of nanoplatelets. *J Mol Liq*. 2023, **382**, 121900.
- [5] Nishikawa H, Shiroshita K, Higuchi H, Okumura Y, Haseba Y, Yamamoto S, Sago K, Kikuchi H. A Fluid Liquid-Crystal Material with Highly Polar Order. *Adv Mater*. 2017, **29**, 1702354.
- [6] Mandle RJ, Cowling SJ, Goodby JW. Rational Design of Rod-Like Liquid Crystals Exhibiting Two Nematic Phases. *Chem - European J*. 2017, **23**, 14554-14562.
- [7] Chen X, Korblova E, Dong D, et al. First-principles experimental demonstration of ferroelectricity in a thermotropic nematic liquid crystal: Polar domains and striking electro-optics. *PNAS* 2020, **117**, 14021-14031.
- [8] Caimi F, Nava G, Fuschetto S, et al. Fluid superscreening and polarization following in confined ferroelectric nematics. *Nat Phys.*, 2023, **19**, 1658-1666.
- [9] Jarosik A, Nádasi H, Schwidder M, et al. Fluid fibres in true 3D ferroelectric liquids. *PNAS*. 2024, **121**, e2313629121.

Prof. Dr. Alexey Eremin



Dr Alexey Eremin is a deputy Professor at Otto von Guericke University Magdeburg (Germany). He is leading the Department of Nonlinear Phenomena, with a research focus on multifunctional soft materials, soft matter physics and biophysics. Having graduated from Moscow State University in Biophysics, he pursued his Ph.D. in Physical Chemistry at Martin Luther University in Halle (Germany), studying the physics of ferroelectric liquid crystals. After receiving his Ph.D., he went to Otto von Guericke University, where he became an assistant professor in the Department of Nonlinear Phenomena. His research topics included liquid crystals in restricted geometry, thin films and fibres, the dynamical properties of liquid crystals and colloids, magnetic colloids and liquid crystals, photoswitchable interfaces, pattern formation, signal transduction and intercellular communication in biological systems. Currently, he is a deputy professor at the Institute of Physics, Otto von Guericke University. Since 2021, he has been a member of the German Liquid Crystal Society board.

Dr. Hajnalka Nádasi



Dr. Hajnalka Nádasi is a research associate at Otto von Guericke University Magdeburg (Germany). Her research focuses on multifunctional soft materials, such as ferrofluids, liquid crystals and their nanocomposites. Having graduated from Eötvös Loránd University in Budapest in chemistry, she pursued his Ph.D. studies in Physical Chemistry at Martin Luther University in Halle (Germany), studying the structure-property relationship of bent-shaped liquid crystal mesogens. After her maternal leave she joined the Department of Nonlinear Phenomena at the Otto von Guericke University. Her research topics have been included photomanipulation of liquid crystals, (ferro)magnetic colloids along with the study of dynamical and magneto-optical properties of magnetic nanocomposites.