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Electroclinic effect amplification, polydispersity and the stability of smectics

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The classical (and highly successful) approach to systematically tailor liquid crystal materials for specific applications is the formulation of optimised mixtures consisting of several mesogenic compounds and non-mesogenic additives such as chiral dopants or UV-stabilizers. While the design of nematic mixtures is highly developed and widely applied, far less is known about the mixing of smectics and the particular effects thereof.

We recently discovered that the electroclinic effect of a chiral smectic A (SmA*) material (consisting of a phenylpyrimidine host and a chiral dopant) was amplified by a factor of three after adding only 5% of another homologous phenylpyrimidine, the molecular length of which was about twice the length of the host molecules [1]. This remarkable electroclinic effect amplification stimulated a more general investigation of how the mixing of smectogens with different molecular length affects the stability, the molecular ordering, and the properties of smectics.

Considering the principle of close packing it seems that the nematic phase – due to the absence of long-range translational order – should be the ideal liquid crystal phase to accommodate molecular rods of different length whereas the periodic layer structure of smectic phases seems to be rather inappropriate for a close packing of rod-like molecules differing in length by, e. g. a factor of two. We thus investigated binary mixtures of several mesogenic phenylpyrimidine homologues (thus making the molecular length distribution in the mixtures extremely bimodal) and expected the smectic A (SmA) and smectic C (SmC) phases to be destabilized whereas the nematic phase should be stabilized (i. e., broadened in the x,T-phase diagrams). The actual results of these experiments are indeed completely reverse to the naive picture drawn above: Whereas the nematic phase (present in the pure homologues of shorter length) completely disappears after the addition of a few percent of the longer homologue, there is no indication of a destabilization of the smectic state at all, i. e., the smectic temperature range in the phase diagram is preserved or even broadened. In addition, the balance between non-tilted SmA and tilted SmC phases is shifted towards the non-tilted SmA. Even in the case where both components only possess SmC phases, SmA is the only liquid crystal phase found in the bimodal mixtures. In conclusion, the bimodal length distribution of smectogenic molecules favours the formation of non-tilted SmA phases on the expense of nematic or tilted SmC phases.

This quite counterintuitive result shades new light on our understanding of the stability of smectic order and will be discussed in terms of current theoretical models. It further opens pathways to a systematic design of interesting new materials such as de Vries-type smectics or low-tilt SmC materials. Finally, some relations to the phase behaviour of lyotropic-lamellar phases will be pointed out.

References

[1] C. S. Hartley, N. Kapernaum, J. C. Roberts, F. Giesselmann, R. P. Lemieux, *J. Mater. Chem* **16**, 2329 (2006)