Multistage switching in a novel bent-core smectic

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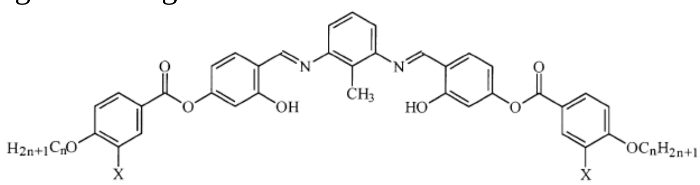
In our presentation we report a novel type of switching behaviour, where in addition to an antiferroelectric ground state and a ferroelectric switched state, there is an additional intermediate ferrielectric state [1]. In our experiments we used bent-core mesogens shown in Fig. 1. The experimental research has been done using polarizing microscopy and Second Harmonic Generation techniques. Using a triangular waveform applied to a sandwich cell, we found an unusual five-peak current response and an unusual optical response that we designate by multistage switching.



No.	X	n	$T/°C$ (ΔH/kJ mol^{-1})	layer spacing d (nm)
I	H	10	Cr 121 [61.5] SmCP[*] 191 [18.5] I	3.49
II	H	12	Cr 123 [71.0] SmCP[*] 193 [19.5] I	3.84
III	H	14	Cr 123 [82.0] SmCP[*] 193 [19.0] I	4.02
IV	F	12	Cr 105 [63.0] SmCP[*] 206 [20.5] I	4.10

Figure 1: Chemical structure and the phase sequence of the investigated compounds.

The first stage is marked by a continuous increase of the apparent tilt angle from 0 to nearly 45° with increasing field strength *E* (Fig. 2). The initial field-free state is non-polar (antiferroelctric) and anticlinic. Further increase of the field *E* leads to strong discontinuous change of the birefringence by one order of magnitude and is accompanied by a sharp current response. When the field exceeds the second threshold, there is a second discontinuous switching stage with corresponding change of the birefringence and the current-response peak. The two values of the switched polarization relate as 1:2. The measurements of the SHG intensity have confirmed an antiferroelectric ground state and discontinuous changes of the polarization in both switched states in agreement with our electro-optic data (Fig. 3).

These experimental results indicate that between the antiferroelectric ground state and the ferroelectric state there is an intermediate ferrielectric state. This behaviour, however, cannot be explained using a simple single-layer structure of tilted bent-core mesogens. Analysis of the birefringence changes during the switching suggest that a "unit cell" of the intermediate state structure consist of two synlinic ferroelectric layers adjacent to a layer which is oppositely tilted and polarized (Fig. 4).

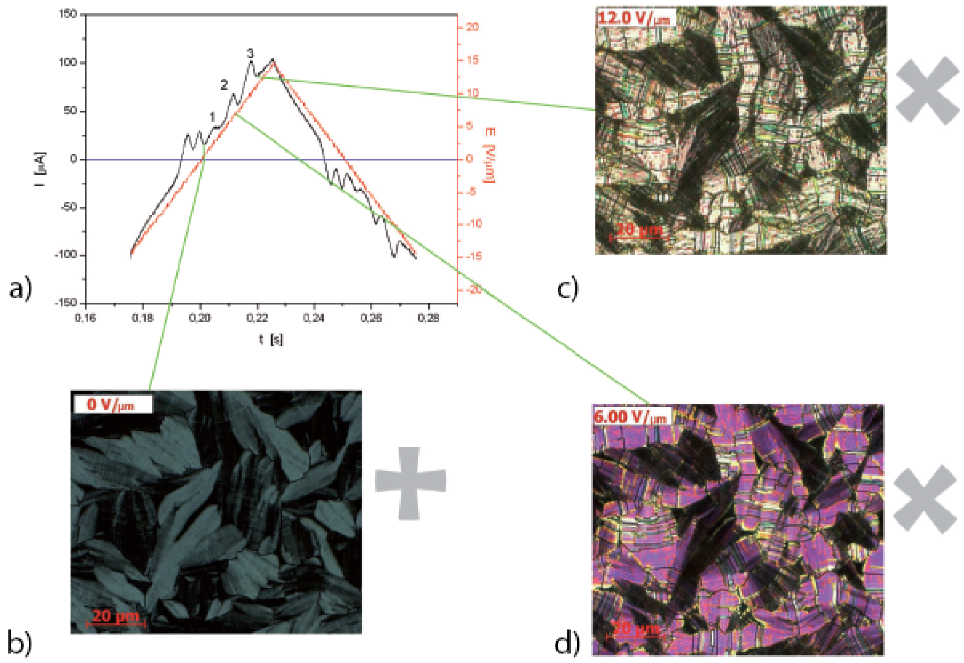


Figure 2: a) Current response curve in a triangular wave electric field and the microscopic textures of the $E = 0$ state b); the intermediate ferrielectric state $E_{th1} < E < E_{th2}$ c); and the ferroelctric state d). $T = 145°C$, the polarizers are vertical and horizontal.

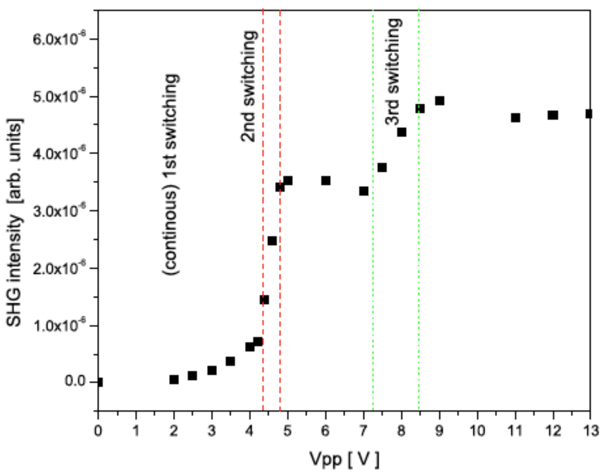


Figure 3: Dependence of the SHG intensity on the applied vOltage ($T = 145°C$, $10\mu m$ cell).

To explain a possible mechanism for such unusual behaviour as multistage switching, we employ an axial next-nearest-neighbour Ising model (ANNNI). In this model we consider long-range electrostatic interactions which give nearest-neighbour and next-nearest neighbour couplings. Additionally, we take into account fluctuation-driven short-range interaction favouring anticlinic interface between the adjacent layers. In tilted SmCP-like phases of bent-core smectics, the helical superstructure has not been unambiguously detected, therefore, we can restrict our discussion to a 1D model with scalar two spins p for polarization and σ for tilt.

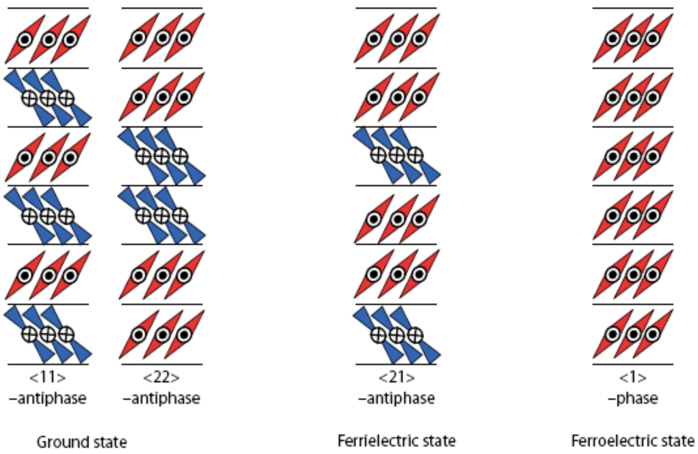


Figure 4: Proposed structure of molecular organization of the layers: The initial state is antiferroelectric (either bilayer or monolayer), the intermediate state is ferrielectric and has two synclinic parallel in p layers and an oppositely tilted and antiparallel aligned layer; the final state is ferroelectric synclinic.

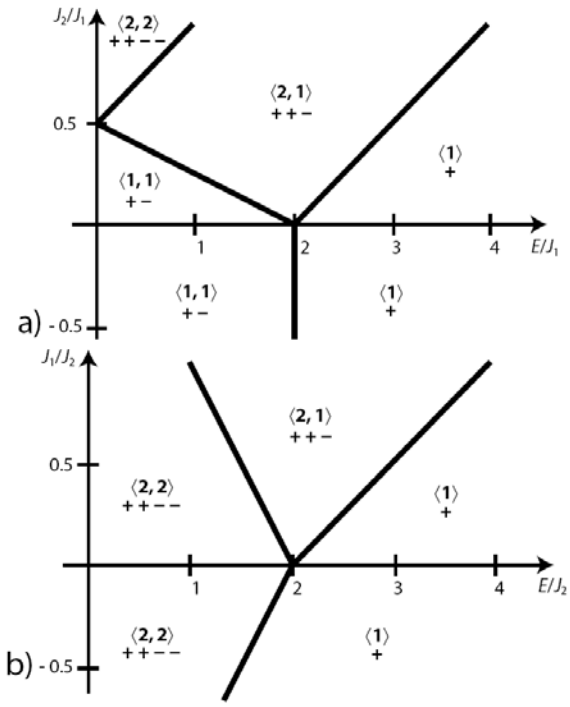


Figure 5: Phase diagram of the model at $T = 0$, focusing on regions of a) $J_1 \geq J_2$ and b) $J_2 \geq J_1$

We can also assume that the switching occurs by rotation of the molecules on a cone around the layer normal. This would mean that a flip of p is nessesarilly accompanied by a flip of σ. Thus, this leaves us with one spin variable, say, *p*. The Hamiltonian of our model reads:

$$H = -J_0 \sum_{\langle NN \rangle} p_{ij} p_{i,j+1} + J_1 \sum_{\langle NN \rangle} p_{ij} p_{i+1,j} + J_2 \sum_{\langle NNN \rangle} p_{ij} p_{i+2,j} - E \sum_{i} p_{ij}$$

where the term with J_0 ensures ferroelectric order within a smectic layer, J_1 >0 and J_2 >0 and nearest- and next-neares neighbour couplings respectively. The last term ist he coupling to the external electric field E. The phase diagram of the model is given in Fig. 5. The NN-interaction favours an antiferroelectric mono-layer state <1,1>. On the other hand, the NNN-term favours a bilayer antiferrelectric state <2,2>. The competition between these two interactions can give rise to an intermediate triple-layer state <2,1> which is ferrielectric. This simple model shows that such intermediate states and multistage switching can be expected in bent-core compounds since they possess a strong spontaneous polarization and very anisotropic shape, which renders the character of fluctuation-mediated interactions.

References (ACS format)

(1) S. Findeisen-Tandel et al, *Eur Phys. J. E.*, **2008**, *25*, 395