

# Alfred Saupe – 40 Years of Research

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A survey of Alfred Saupe's research work over the last forty years is given. Special emphasis is laid on his earlier publications, some of which are in German, while his recent work – still fresh in our memory – is touched only briefly.

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## I. INTRODUCTION

I have divided my talk into four parts representing different main topics in Alfred Saupe's work.

- Structure and order (Maier-Saupe theory, its verification and utilization)
- Defects, dynamics and instabilities (blue phases, undulations, hydrodynamics)
- New phases and phase transitions (polar smectics, lyotropics, biaxial nematics)
- 'Applied' research (optical and electro-mechanical properties)

Of course, this classification of his work is somewhat arbitrary and not quite chronological. During his career Alfred Saupe worked on quite different aspects of liquid crystal science, but never stopped working on earlier topics. So this scheme has to be seen as evolutionary and cumulative rather than sequential.

Another special feature in his work is the close relationship between theory and experiment. Either having started with a theory, the experimental verification follows suit, or an

experimental finding is backed up by a theory, which he developed for interpreting his experiments. This is really a hallmark of Alfred Saupe's work, which is even more remarkable if you know that this kind of doing research is just the opposite of what is common (and appreciated) in Germany. I think this is also the ultimate reason why he came to Kent and stayed there.

Before beginning with the detailed look into his publications, I have a few excuses. Alfred Saupe had a lot of co-authors, whom I cannot mention individually with every paper, but who are listed in Table I (which is hopefully complete). The names in boldface are those who have published with Alfred Saupe over a longer period of time.

TABLE I. Co-authors of A. Saupe 1956 - 1995

Armitage, D.	Arora, S.L.	Boonbrahm, P.	Brown, G.H.	Cantow, H.J.
Chafik, A.	Chang, H.	Chidichimo, G.	Cien, L.C.	Coppola, L.
Doane, J.W.	Dreher, R.	<b>Englert, G.</b>	Ferguson, J.L.	Gault, J.D.
Haven, T.	Holzbach, R.T.	Hui, Y.W.	Imbardelli, D.	Jabben, M.
<b>Jakli, A.</b>	Johnson, D.	Kanekar, C.R.	<b>Khetrapal, C.L.</b>	Kim, D.R.
Klesper, E.	Kumar, S.	<b>Kunwar, A.C.</b>	Kuzma, M.R.	La Mesa, C.
Longeri, M.	Madey, R.	<b>Maier, W.</b>	Martin, A.J.	<b>Meier, G.</b>
<b>Melnik, G.</b>	Nagle, D.E.	<b>Nehring, J.</b>	Olszewski, M.F.	<b>Photinos, P.</b>
Plumley, S.	Povh, A.	<b>Radley, K.</b>	Ranieri, G.A.	Rosenblatt, C.
Schuster, T.M.	Sengupta, P.	Sharma, B.B.	Shenouda, I.G.	Shin, S.
Singletr, H.S.	Spieseke, H.	Stefanov, M.	Taylor, T.R.	Weber, J.-P.
Wedler, W.	Xu, S.Y.	Yeh, H.J.	<b>Yu, L.J.</b>	Zhou, E.
Zhu, Y.K.				

As you can see, I am not on this list. So I will not talk about his personal style of doing science, etc. This will be done by others - but, I am going to talk about his scientific work as it is manifest in his publications. And there are quite many publications (more than a hundred), so I can talk only about *his* papers and I cannot mention anybody else's papers, even if they are related. Sorry, but there is not enough time for this.

## II. STRUCTURE AND ORDER

The first publication of Alfred Saupe [1] was submitted just 40 years ago. It was his diploma thesis dealing with absorption of polarized UV light in PAA in its isotropic, nematic and crystalline state. The aim of this experimental investigation was to compare the intermolecular interaction in the different phases. As a result, no difference was found, i.e. the dielectric constant in the isotropic phase was approximately the mean of those in the nematic phase  $\epsilon_{iso} \approx \frac{1}{3}(2\epsilon_{\perp} + \epsilon_{\parallel})$ . Their conclusion was that there is no special interaction in the nematic phase and the formation of the nematic order must be due to ordinary molecular forces. This was not at all obvious at that time, since all attempts to theoretically explain the nematic to isotropic phase transition correctly had failed [2,3]. Descriptions of experiments with liquid crystals at that time show how difficult it was to handle these materials. One big problem was getting oriented nematic samples. Only rubbing was known as a means for orienting, which, however, worked for thin samples, only.

In a second publication [4], which was continued later [5],  $\pi$ -electron states of the benzene ring in PAA were investigated by comparison with spectra of other, simpler compounds. The interpretation of these spectra by ordinary methods of molecular physics was a rather puzzling work, as you can see from the tables in [4]. Nevertheless, from these investigations they got convinced that nematic order is a long-ranged orientational order of the molecules' long axes mainly due to *induced* dipole-dipole-interaction, whereas special chemical compounds or structures like permanent dipoles are not necessary for forming a nematic phase.

This is related to the following observation [6]: The larger the dielectric anisotropy  $\Delta\epsilon$ ,

the larger the nematic existence range. This can be explained assuming that  $\Delta\epsilon$  is due to elongated phenyl rings; if you now add alkyl-groups to the phenyl ring, any C-C bond parallel to the elongated axis adds much more to  $\Delta\epsilon$  than a C-C bond perpendicular (or with an angle of  $72^\circ$ ). This is the explanation of the odd-even effect for the nematic existence range, if alkyl-chains are added. This successful interpretation of the odd-even effect must have convinced them that their physical picture was right. So they started, what is now called, the Maier-Saupe theory [7].

The nematic order parameter  $S$  was already known at that time [8,9]

$$S = 1 - \frac{3}{2} \langle \sin^2 \theta_1 \rangle \quad (1)$$

where the angle  $\theta_1$  describes the orientational deviation of some molecule 1 from the mean orientation of all molecules and  $\langle \dots \rangle$  denotes the averaging over all molecules. Within mean-field theory it was also clear, how to calculate this average in analogy to the Weiss theory for ferromagnets. The intermolecular forces that determine  $\theta_1$  are replaced by a mean field,  $D_1$ . Then Boltzmann statistics leads to

$$\langle \sin^2 \theta \rangle = \frac{\int_0^{\pi/2} \sin^3 \theta_1 \exp(-D_1/kT) d\theta_1}{\int_0^{\pi/2} \sin \theta_1 \exp(-D_1/kT) d\theta_1} \quad (2)$$

Of course, the real problem lies in the determination of the (angle-dependent) mean-field potential  $D_1(\theta_1, S)$ , which is a function not only of the individual orientation  $\theta_1$ , but also of the orientations of all the other molecules, described summarily by  $S$  (in mean field approximation). This was the hard core of the problem! It was solved by Alfred Saupe in his PhD-thesis [10] by using a fully quantum-mechanical 2nd-order perturbation theory assuming that *induced dipolar forces* are the only relevant forces with respect to the orientational degrees of freedom. Skipping the lengthy calculations the result was

$$D_1 = -\frac{A}{V^2} S \left(1 - \frac{3}{2} \sin^2 \theta_1\right) \quad (3)$$

which – together with Eqs. 1,2 – leads to a self-consistency relation for  $S$ . The famous graphical solution of this self-consistency problem (Fig. 1 in [7]) has found its way into

various textbooks (e.g. Fig. 2.10 in [11]). At a temperature  $T^*$  the trivial or isotropic solution  $S = 0$  becomes unstable, while below a higher temperature  $T'$  the nematic solution  $S > 0$  is possible. In between these two temperatures one of the two stable solutions is a metastable one and the exchange of stability with metastability (at  $T_c$ ) of the isotropic or nematic phase constitutes a first order phase transition with a finite jump in  $S$ . The universal  $S$  (i.e. independent of the material parameter  $A$  in Eq. 3), which they obtained at the clearing point, had the right order of magnitude.

Now that there was a promising theory a closer look was necessary to actually verify it. Further thermodynamic quantities, like heat of transition, specific heat, and compressibility (and their temperature dependences) were also predicted by the theory and successfully compared with experiments [12]. Another example was the density jump near  $T_c$ , experimentally seen in the thermal expansion coefficient. Not only did the temperature dependence (below  $T_c$ ) fit nicely to the theory, they also detected a pretransitional effect above  $T_c$ , which they interpreted correctly as due to small, random, transient nematic islands [13].

Another testing ground was the temperature dependence of the Frank constants. First Alfred Saupe made the theory for this temperature dependence using the same approach as for  $S(T)$ , then he developed a method how to measure it and then he actually did the measurements [14]. To measure  $K(T)$  he used the (known) Frederiks instability, where director rotations are induced by an external magnetic field. However, since at the onset of the instability there is still no director rotation (2nd-order-like transition) one must go beyond the critical field to see anything. So he made the theory for Frederiks deformations *above* the critical field and then he made a theory how to detect these deformations by measuring the phase difference between two orthogonally linear polarized light beams crossing the Frederiks-deformed nematic. Thus from the measured phase difference at a given magnetic field he could get the Frank constants (by the initial slope in Fig. 4 of [14]) and doing this for different geometries (to probe the different Frank constants) and for different temperatures, he got experimentally the temperature dependence of all the Frank constants.

His theory predicted that the temperature dependence is equal for all three Frank constants, since  $K \sim S^2$ , which was confirmed extremely well by his experiments. On the other hand the absolute values of, and the relative magnitudes between, the Frank constants were very poorly given by this theory. This problem never came to rest and some years later Alfred Saupe tried again [15,16] to calculate the Frank coefficients using a molecular model, when he found the famous (or should I say ‘infamous’) extension to the Frank-Oseen-Zöcher theory introducing the  $K_{13}$ -term, which still today gives a lot of headaches to theorists and experimentalists alike. He found  $K_{13}$  to be of the same magnitude as the bulk Frank coefficients. He also characterized correctly what is now known as the flexo-electric terms as ‘curvature-electric’ (in contrast to ‘piezoelectric’ as what they were known at that time).

But let’s come back to the nematic order parameter  $S$ . The Maier-Saupe-Theory proved to be very successful and the temperature dependence of  $S$  was investigated [17] using lots of different methods, e.g. via the electrical polarizability, the anisotropies of any second order tensorial material parameter, linear dichroism, NMR (to which I will come back soon) and the dielectric relaxation in nematics, for which also the theory had to be given first [18,19]. Of course, the simple version of the Maier-Saupe theory had some shortcomings, e.g. the nematic short range interactions were not considered. So there was a need for some improvements, which he did later [20], although it is very difficult to really improve such a nice, simple and successful theory.

Another string of experiments by Alfred Saupe were then concerned with a kind of utilization of the nematic order by NMR. First he gave a theory of NMR in the nematic state [21] and then he used NMR to investigate organic compounds that are dissolved in a nematic liquid crystal. The spectra (NMR, UV, IR) of the molecules so obtained are simpler than those of the crystalline phase, because of motional narrowing. On the other hand these spectra contain much more information than those taken in the isotropic state, because of the orientational order. Thus, intermolecular forces, quadrupolar interaction, electronic structure, proton distances, anisotropic chemical shifts can be obtained. A host

of new information on various organic molecules was so obtained utilizing the nematic state [22–45].

### III. DEFECTS, DYNAMICS AND INSTABILITIES

Alfred Saupe never wrote a book on liquid crystals, but from time to time he gave reviews of what was known about liquid crystals (or a certain aspects of them), e.g. [46] and later [47]. Such a retrospective must have inspired him to also look into the future. That brings me to my favorite publication of Alfred Saupe. In a MCLC in the late 60's [48] he set the stage for his future work; this paper contains four rather uncorrelated topics and is highly speculative – no referee would let pass such a paper today! – but it is full of good ideas. First it dealt with smectic morphology: Known were at that time nematic, cholesteric and the simple smectic phases A,C (also B,D, which did not play a role). First he noted that the tilted smectic C phase has a nematic-like degree of freedom, he gave the biaxial gradient energy for it, and noticed that the smectic C phase should show Schlieren textures because of its nematic-like degree of freedom. Shortly after that he used this for identifying possible smectic C phases [49–51].

In [48] Alfred Saupe also speculated on possible additional smectic phases, when the molecules are polar. So you can see in Fig. 1 of that paper the smectic  $A_2$  phase (which he called antiferroelectric) and a truly ferroelectric smectic phase, which unfortunately never has been found in nature. This opened the discussion on polar smectic mesomorphism. Later he also did a mean field study of such polar smectic phases utilizing the polar order parameter  $P_1$ , additionally [52]. There is another interesting speculation on this figure: A chiral smectic C phase, which he correctly assumed to be conic-helically twisted. He also stated that this conic helical structure would be quite similar to the cholesteric structure, optically. He assumed commensurability between the pitch and the layer spacing (1 to 8 in this picture), but – unfortunately – he did not realize the helielectricity of that phase. Nevertheless, obviously this was the starting point for the experimental search of this phase.

I will come back to Alfred Saupe's work on phases and phase transitions in the next Chapter.

In a third topic of this paper he investigated theoretically the possibility of linear twist or bend waves in nematics. He gave the dispersion relation, but noticed immediately that there are no propagative (linear) waves, because of the smallness of the molecular moment of inertia. Instead he proposed director relaxation experiments, which have proven so fruitful.

In another section of this paper he speculated on the nature of the Blue Phase (I in today's notation) (Fig. 2 of [48]). Convinced that this Blue Phase is really a thermodynamically stable phase he proposed a structure that explained the optical isotropy of this phase. This is a cubic lattice of disclinations with a large unit cell, filled with a twist structure. Although this structure is not exactly what we now believe to describe the Blue Phases, his idea of a cubic superstructure of defect lines is still valid. Let's stay with defects and textures. Next he tackled the problem of an explanation of the Schlieren textures in nematics, cholesterics and smectic C [53]. Careful optical studies of the brushes were followed by the discussion of possible molecular structures, and calculations of core energies and the interaction between point defects were performed. A comprehensive review on textures and defects in nematics and cholesterics was given shortly thereafter [54], which not only contained the well-known topological classification of bulk defects, but also considerations on defects in restricted geometries, e.g. surface structures in cholesterics, as well as optical observations of spiral defect structures at free droplet surfaces and cholesteric fan textures. Again this paper is remarkable for its close connection between experiment and theory.

There are a few papers on dynamics and instabilities. The first [55] dealt with electrohydrodynamic convection in nematics, where numerical calculations were performed in order to study stability diagrams as a functions of relaxation times. In another paper [56] he looked into the thermally induced undulation instability of smectic C phases. Changing the temperature of a smectic C phase leads to a change of the tilt angle, which is nothing but a change of the layer spacing. If the smectic liquid crystal is put between fixed plates, a change in the layer spacing leads to an external mechanical force on the smectic, which above a certain

threshold gives rise to the undulation instability. Not only the experiments were done, also the theory for the threshold and the pattern above threshold was given. Then he discussed how to detect this undulation pattern above threshold by optical means. To this same string of papers belongs his rather well-known dynamic, Leslie-Ericksen-like theory on elasticity and flow (and its stability) in biaxial nematics [57], which he did just after he had discovered such a phase in lyotropics. But now I am already a little bit ahead of chronology.

#### IV. NEW PHASES AND PHASE TRANSITIONS

Let's come back to the seventies. Liquid crystal phases are not only found in man-made substances, but are also part of nature, e.g. cholesterics are found in the human bile [58], as discussed in a Nature article. Then his interest turned to lyotropic or micellar systems. Here, the composition – and related to that – the shape of the aggregates comes into play. First a nematic phase formed by bilayer micelles was investigated by NMR and microscopy [59,60]. Not only the formation of a liquid crystalline phase, but also the actual shape of the micelles was a major topic of the investigations. When chiral compounds were mixed in, cholesteric states occurred as can be seen by the pretty texture of Fig. 3 in [61]. It is the distortion of the micellar shape due to optical active compounds that determines the twisting power and the range of the liquid crystal phases.

And the lyotropic systems revealed some surprises. First a nematic to nematic phase transition was found by NMR and microscopy [62], which showed that there are bilayer or disk-like micelles in the low temperature phase,  $N_L$ , and cylindrical or rod-like one in the high temperature phase,  $N_c$ , with opposite electrical and magnetic susceptibility anisotropies. After chiralization an appropriate cholesteric to cholesteric phase transition was obtained.

There was only one further step to get the biaxial phase in between the  $N_L$  and the  $N_c$  phase [63]. By careful microscopic investigations as well as by NMR studies the biaxiality of this nematic phase was clearly demonstrated. Observations of Schlieren textures and the

possibility of a uniform alignment in a magnetic field ruled out a tilted smectic phase, which would also be biaxial. There was a lot of doubt at the beginning, because there had been so many false alarms concerning biaxial nematics before, but Alfred Saupe was absolutely sure about this finding – and he was right [64].

Now with so many different phases, phase transitions in lyotropic systems were a natural topic to investigate in the following. Critical phenomena at the second order uniaxial-biaxial phase transition close to the transition temperature were seen by measuring the biaxial order parameter (and the susceptibility) via birefringence [65,66], revealing at least partially the expected x-y model critical exponents. A Landau point – a point in phase space, where the cubic term in a Ginzburg-Landau description of a phase transition vanishes – was found in a lyotropic nematic to isotropic phase transition [67] utilizing the composition as an additional degree of freedom for tuning the system. At such a point large order parameter fluctuations occur. A slow extra relaxation at a second-order nematic to lamellar phase transition was seen in a lyotropic system [68], which has no counterpart in thermotropic systems. This was interpreted as a slow structural change from the micellar cylindrical shape to a lamellar structure with defects or holes in it.

In the 80's measurements on lyotropic systems took a great part of Alfred Saupe's activities. Among them were measurements of material properties, like elastic constants, diffusion constants, resistivity, magnetic birefringence, diamagnetic susceptibilities, density and expansion coefficient, rotational viscosity and relaxation times, electric conductivities etc. [69–80]. Of course, also the theoretical side was involved with calculations of the electric conductivity and the Cotton-Mouton constant in nematic and defective lamellar phases [81–84]. Great efforts went into the investigation of the relation between micellar structure, chemical composition and existence range of liquid crystalline phases [85–90].

## V. ‘APPLIED’ RESEARCH

I will close this talk with a brief glance of what I have called the ‘applied’ research activities of Alfred Saupe. Of course, also these publications deal with purely scientific questions, although somewhat nearer to application-related problems.

One of the longer standing topics of this kind were the optical properties of cholesterics, which are clearly of great interest for conventional liquid crystal devices. Alfred Saupe looked into the dielectric properties of selective reflection by cholesterics [91], into the absorption in cholesteric fibers and contrast of dichroic liquid crystal displays [92] and into reflection bands of Grandjean textures [93,94].

Quite recently he worked on the electro-mechanical properties of smectic C\* phases to get a better understanding of devices and displays. Here defects and textures come into play again, like the transition from a chevron to a striped texture [95] and uniform bookshelf textures [96,97]. Electro-optical effects [98], electro-clinic effects [99] and electro-mechanical vibrations [100] were investigated.

Polymeric liquid crystalline systems came into the focus of applied research originally because of their enhanced mechanical properties. In this field Alfred Saupe had made some early experience with NMR of non-liquid crystalline copolymers [101], but in the 90’s he started measurements on the interaction of polymeric with liquid crystalline properties. Among them were the effect of dissolved polymers on the rotational viscosity [102], the alignment and switching behaviour [103] of thermotropic low molecular weight nematics, and the nematic to lamellar transition in lyotropic systems [104].

In addition he discussed the phase behaviour of liquid crystalline side-chain homo- and copolymers with large C\* phases [105] and the switching behaviour of some polymeric smectic C\* phases [106] thus combining his interest in polymeric systems with that on the electro-mechanical behaviour of smectic phases.

About his newest topic ‘atomic force microscopy on liquid crystalline surfaces’, which resumed his longstanding interest in surface properties and surface defects [107,108], Alfred

Saupe talked just before the coffee break.

At the end there is nothing left for me but to express my deep admiration for his impressive and enduring work and to ask for your applause for him.

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