Obituary of Patricia Elisabeth Cladis

Personal recollections of Helmut Brand and Pawel Pieranski

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1. Biography

Pat was born on 13 July 1937 in Shanghai and passed away on 3 July 2017 in Summit, NJ. She was married to George H. Cladis, an artist painter (who had an earlier career in aeronautics). They had two twin sons, Franklyn and Harrison and several grand-children. After a three years visit in France (1969–1972), Pat and George returned to the US where they lived in their beautiful house, surrounded by greens and tall trees, in Summit, NJ. Bell Laboratories in Murray Hill (world-wide famous for the invention of the transistor, today Nokia Labs), where Pat worked (Figure 1), was located a few miles away from Summit (about 15 min’ drive on bicycle). George had a studio in Manhattan.

1.2. Academic trajectory

1959: BA combined honours in mathematics and physics, University of British Columbia, Vancouver, Canada
1960: MA in physics, University of Toronto, Canada
1968: PhD in Physics on the superconducting transformer at the University of Rochester, Advisor: Ron Parks,
1969–1972: Post-doc at Physique des Solides, Université Paris XI, Hosts: P.G. de Gennes and M. Kléman
1972–1997: Bell Laboratories, Murray Hill, NJ
after 1997: Advanced Liquid Crystal Technologies, Summit, NJ

1.2. Honors and awards

1983 – … Fellow of the American Physical Society. Honoured member of the ILCS (International Liquid Crystal Society)
1993: Guggenheim Fellowship
1997: Research Prize of the Alexander von Humboldt-Foundation
2001: Invitation Fellowship of the JSPS (Japan Society for the Promotion of Science) for senior scientists

2. Scientific legacy

2.1. Motivation

In the following short overview we focus on Pat’s seminal work, which has stood the test of time. Naturally this selection is based on the taste for science of the two authors, who had the privilege to collaborate with Pat for a total of more than four decades.

We also note that Pat edited two books in collaboration with Peter Palffy-Muhoray: ‘Spatio-temporal Patterns in Nonequilibrium Complex Systems’ and ‘Alfred Saupe: 40 years of research in Liquid Crystals’.

2.2. Re-entrant nematic phase

The discovery of the re-entrant nematic phase by Pat in 1975 \cite{1} attracted immediately a lot of attention because it was in contradiction to a commonly held belief that in phase transitions the higher-temperature phase is less well ordered (more symmetric) than the lower temperature phase \cite{2}.

Indeed, until the discovery of Pat, in all well known phase sequences of mesogenic systems this was the case. For example, the phase sequence of HBAB is ‘crystal $\rightarrow$ Nematic $\rightarrow$ Isotropic’ and that of CBOOA is of the same kind ‘crystal $\rightarrow$ SmA $\rightarrow$ Nematic $\rightarrow$ Isotropic’.

The article \cite{1} in which Pat announced her discovery was titled ‘New Liquid-Crystal Phase Diagram’ because the phase diagram of HBAB-in-CBOOA binary mixtures established by Pat played the role of a counterexample breaking into pieces the common belief. Here, in Figure 2(a) we show a simplified version of Pat’s
phase diagram in which the phase sequence along the \( c = 9\% \) vertical dashed line is: Nematic \( \rightarrow \) SmA \( \rightarrow \) Nematic \( \rightarrow \) Isotropic. Obviously, the transition from the lower-temperature nematic phase to the higher-temperature SmA phase is ‘abnormal’.

Pat stated this in these terms [1]: ‘Although there is, in principle, no a priori reason for a phase of lower symmetry to be the higher-temperature phase rather than the lower-temperature phase, one expects to be one or the other. Thus, this is the first time the smectic phase has been observed to occur both at higher and lower temperature than the nematic phase.’

Six years later Pat, in collaboration with D. Guillon, F. R. Bouchet and P. Finn [2], showed that the same ‘abnormal’ phase sequence occurs on T-vs.-P phase diagram of pure 8OCB.

The explanation of the re-entrance of the nematic phase upon the cooling of the smectic A phase takes into account changes in the short-range order of molecules. Due to interactions of their large dipolar moments of the nitrile bond, the 8OCB molecules have a tendency to form antiparallel pairs (Figure 3). The reentrant nematic phase is believed to be composed of relatively long-lived association of molecular pairs. The number of associations (and their life time) increases as the temperature decreases […] Such pairing is postulated to neutralize the long-range attractive electrostatic forces necessary to stabilize layering […] [2].
The importance of Pat’s discovery was widely recognised (the seminal article has been quoted 419 times) and confirmed in many other systems.

2.3. Pat’s very first paper on liquid crystals: ferronematics

Pat started to work on Liquid Crystals during her postdoctoral visit at the Solid State Physics Laboratory in Orsay. She arrived there at the best moment – the ‘fat cows’ period – when under the driving force of Pierre-Gilles de Gennes, several teams composed of young researchers started to work frenetically on liquid crystals. As a visitor, Pat was free to work with all of them, besides her main collaboration with Maurice Kléman.

In these circumstances her first paper titled ‘Ferronematics’ was written with Jacques Rault, a PhD student, and Jean-Paul Burger, a senior scientist [3]. This seminal experimental work was inspired by the visionary theoretical considerations of P.-G. de Gennes and Françoise Brochard [4] on the consequences of liquid-crystal-mediated interaction between solid magnetic particles immersed in a nematic phase.

The original aim was to generate a phase, which was truly ferromagnetic and nematic simultaneously. With the nanoparticles available around 1970 the best one could achieve was a ferronematic phase, whose magnetic properties were superparamagnetic with vanishing spontaneous magnetisation. A ferronematic is thus the nematic analogue of magnetic liquids, which are superparamagnetic and thus susceptible to rather small magnetic fields, but not ferromagnetic (Figure 4). In the following four decades many systems, both lyotropic and thermotropic, were shown to have superparamagnetic properties leading to ferronematics and ferrocholesterics.

This situation changed only a couple of years ago when the synthetic group of Darja Lisjak and the...
experimental group of Alenka Mertelj at the Jožef Stefan Institute in Ljubljana were able to generate a truly ferromagnetic nematic liquid crystal [5] with a spontaneous magnetisation, which is parallel to the nematic director thus giving rise to the first liquid multiferroic system. A couple of years later the group of Ivan Smalyuk at Boulder reported the first ferromagnetic cholesteric phase [6]. Quite recently the study of the dynamic properties of ferromagnetic nematics has started [7] and it was shown that dynamic cross coupling terms between the two order parameters, the magnetisation and the nematic director, can play a decisive role in understanding the dynamics of this first liquid multiferroic system.

Thus the early work of Pierre-Gilles, Francoise and Pat has been a precursor of a flourishing and innovative field of soft matter research.

2.4. Nonsingular S = + 1 disclination and nematic monopoles

During her stay in Solid State Physics Lab in Orsay Pat worked principally with Maurice Kleman. By a tradition stemming from Georges Friedel and transmitted by his grand-son Jacques Friedel, Kleman was interested in defects in ordered systems, such as crystals, magnets or superconductors (the former field of interest of P.G. de Gennes). With the outbreak of the research on liquid crystals he focused on their defects.

In these circumstances, generation of $S = + 1$ disclination in capillaries with homeotropic boundary conditions was attempted [8] at LPS in Orsay. As shown in Figure 5(a) the +1 disclination was expected to have a singular linear core similar to the one of the ‘common’ well known $\frac{1}{2}$ disclinations.

Surprisingly, experimental results did not correspond to these expectations because, as shown in Figure 5(b), a non-singular configuration of the $S = + 1$ disclination was obtained. The same observation was reported independently by R.B. Meyer [9].

The singularity was avoided, for energetical reasons, by an escape of the director field into the third (z) dimension [10]. This experiment offered another, even more interesting surprise: instead of the linear singularity, the sample contained point singularities (nematic monopoles) located at junctions between the two possible configurations, ‘up’ and ‘down’, of the ‘escaped’ director field.

Figure 5(b,c) explain that the director field around these nematic monopoles, mapped on a sphere, covers it completely. Remarkably, as the space of the nematic order parameter – the projective plane $P_2$ – is equivalent to one hemisphere, this means that the director field of nematic monopoles is covering it twice. This is the reason for which, unlike $\frac{1}{2}$ disclinations, monopoles are scarce and have not been observed (identified?) previously.

Since this precursory work, a lot of progress in the study of nematic monopoles was made (see the review

![Figure 5. S = + 1 disclinations and nematic monopoles: (a) Hypothetic singular S = + 1 disclination generated in a cylinder by homeotropic boundary conditions. The director field lies in the xy plane. (b) Non-singular S = + 1 disclination observed in Pat’s et al. experiments with glass capillaries. It is obtained from the singular one by escapes into the third dimension either in +z (‘up’) or in –z (‘down’) directions. Nematic monopoles are generated at junctions between the up and down configurations. c) Mapping of the director field surrounding the ‘hyperbolic’ monopole onto a sphere. The order parameter space of nematics – one hemisphere – is thus covered twice.](image)
paper by Kleman and Lavrentovich [11]). Beside the cylindrical geometry, other conditions favouring generation of monopoles were found such as colloidal inclusions [12,13] or the pseudo-planar texture (Figure 6) which appears today as a natural metastable universe of nematic monopoles [14].

2.5. Blue phases

At the beginning of the eighties, Pat started to work at Bell Laboratories on the mysterious (at that time) Blue Phases. At the same time colloidal crystals were in focus at LPS in Orsay. In particular, their shear elastic modulus was measured by the method depicted in Figure 7. As periodicities of Blue Phases, belonging to the visible light wave range, were similar to those of colloidal crystals Pat decided to come to Orsay and she brought with her big test tubes filled with low temperature Blue Phases.

After installing a precise regulation of temperature, experiments made with Blue Phase I were successful: 1° a Debye–Scherrer line produced by BPI crystals was visible on the surface of the spherical vessel containing the test tube with the sample; 2° the mean shear elastic modulus ($\approx 10$–$50 \text{ N/m}^2$) was determined from the frequencies of eigenmodes [15].

Close to the BPI–isotropic transition, the polycrystalline samples used in this experiment were made of tiny BPI crystallites hardly visible with the naked eye. However, when observed in a reflecting microscope under monochromatic illumination (Figure 8(a)), these tiny crystallites unveiled their bewitching beauty: they were faceted as it is shown in Figure 8(b–f). The coexistence of two-fold, three-fold and four-fold symmetries, well visible in pictures d, e and f, leaves no doubts as to the crystal symmetry of BPI: it must be a cubic one [16].

Observation of BPI crystals under monochromatic illumination unveils other interesting details such as steps on surfaces of facets. As shown in Figure 8(b), the step has a spiral shape and is connected to a dislocation piercing the crystal. During the growth of the crystal the spiral is rotating (Frank–Read mechanism). Figure 8(c) shows that the growth of BPI crystals can also occur by nucleation and growth on new terraces [17].

**Figure 6.** Point defects – nematic monopoles – in the pseudo-planar texture. (a) A view of a monopole-antimonopole pair seen between crossed polarisers. (b) The corresponding texture of the 2D ‘dowser’ field $d$ [14].

**Figure 7.** Measurements of the shear elastic modulus of Blue Phases. (a) Setup for the excitation of shear oscillation modes of axial symmetry and for their detection from the motion of the Debye–Scherrer line. The polycrystalline sample of BPI is held in a test tube. Illuminated by a He–Ne laser beam it produces a Debye–Scherrer line DS. (b) Bragg reflection from a BPI crystallite contributing to the Debye–Scherrer line DS. Upon a shear deformation of the sample, the BPI crystallites rotate and the whole DS line is shifted. The photodiode PD located at the edge of the DS line detects this motion.
Following this first observation of BP monocrystals in reflecting microscope several other experiments made with perfected setups led to additional discoveries.

For example, in spite of their cubic structure, BP crystals were oriented by the electric field with their fourfold axis parallel to $E$. The explanation of this effect involves the third order nonlinear polarisability expressed by a fourth rank tensor which is anisotropic for cubic crystals $^{[18]}$. From the structural point of view, the nonlinear polarisability stems from deformation of the Blue Phase structure by the electric field, i.e. electrostriction, which was studied later extensively $^{[19,20]}$.

Another, even more striking effect observed in strong enough fields consisted in the induction of tetragonal (Figure 9) $^{[21]}$ and hexagonal Blue Phases. Remarkably, this electric-field-induced polymorphism was predicted by Dick Hornreich and Mula Shtrikman in terms of their Landau-de Gennes mean field theory of Blue Phases $^{[22]}$.

2.6. Banana/bent-core liquid crystal phases and biaxial orthogonal smectics

Stimulated by the observation of an orthogonal biaxial fluid smectic phase in side-on side-chain liquid crystal polymers by Hartmann Leube und Heino Finkelmann (Freiburg) $^{[23]}$, Pat, Harald Pleiner and one of us (HRB) investigated the symmetry properties and the defects shown by such an orthogonal fluid biaxial smectic phase $^{[24]}$. This phase had been hypothesised before in the book by de Gennes and called $C_M$ phase. Later on many authors called it a biaxial smectic A phase. It turns out that this phase has defects for the director in the layer planes of strength $S = 1/2$, in contrast to the case of ordinary
tilted smectic C phases, for which the defects of the in-plane director $c$ can only be of integer strength. After this prediction it took almost a decade before the group of Carsten Tschierske (Halle) could confirm this prediction [25].

In addition, Ref. [24] contained another amusing prediction. At the time it was a commonly held belief in the liquid crystal community for well over a decade that chiral molecules were necessary to generate interesting electric properties beyond dielectric behaviour. In Ref. [24] it was argued (see Figure 10 below) that bent-core or banana-shaped molecules could give rise to an in-plane polar order and thus to a macroscopic electric polarisation – even for non-chiral molecules. Shortly thereafter Pat and HRB [26] pointed out that antiferroelectricity and ferri-electricity could occur along the same lines. In the next couple of years Pat talked about this possibility at various institutions including a visit of the groups of Junji Watanabe and Hideo Takezoe at the Tokyo Institute of Technology. A few years later the TIT group reported the observation of a biaxial fluid smectic phase with interesting macroscopic electric properties [27]. After that the field of liquid crystal phases formed by non-chiral bent-core (or banana-shaped) molecules expanded rapidly and continues at present. We refer to Ref. [28] for an early review of the field.

Thus once again Pat was pioneering the development of a new field in liquid crystals.

2.7. Pattern formation in freely suspended smectic $c$ films: the influence of topology

Following the prediction of the possibility of flow alignment in smectic C [29], Pat and HRB discussed the possibility to observe this phenomenon. It was clear that only freely suspended films were the geometry of choice to avoid focal conics and defects in the layering entering the picture. How to generate flow and coupling to the in-plane director was less clear at the time (1984), because previous experiments on freely suspended smectic films had focused on light and X-ray scattering. The solution came with a visit of Pat and HRB of Yves Couder’s laboratory at ENS (Paris) in 1984. Yves was working on turbulence in large area soap films at the time [30,31] and used a running needle as an external force.

As a result of this visit, Pat, Yves and HRB decided to collaborate on the pattern formation of thick freely suspended smectic C films in a circular geometry and use as an external force to apply a torque a rotating needle piercing the centre of a freely suspended film.

It turns out that the phase (azimuthal angle $\Phi$) of the in-plane director $c$ and its associated boundary conditions at the rim of the film and at the needle played a decisive role in the determination of the resulting spatio-temporal patterns [32]. The observations were carried out using a polarising microscope. In addition to the phase winding regime, corresponding to a winding-up of the phase of the director as a function of time (target pattern) also spirals were observed when changing the boundary conditions at the boundary of the needle. Pulling out the needle for a target pattern led to the relaxation of the phase and allowed to extract from the relaxation time at the centre of the film the determination of $\gamma_1/K$. From these experiments it was clear that the boundary conditions of the director phase at the rim and at the surface of the needle were crucial for the nature of the stationary patterns observed.

To investigate the pattern formed as a competition between the boundary conditions at the rim and an external bulk force, we investigated the effect of a rotating electric field on the patterns formed in a freely suspended smectic film [33] giving rise to an electric torque in the bulk. Stationary spatio-temporal patterns observed [33] in freely suspended thick films included (a) target patterns, (b) spirals with $S = +1$ at the centre and (c) The coexistence of an orbiting $S = +1$ defect and a target acting as a phase sink leading to a stable stationary bound state with an indefinite lifetime, at least months in a system for which the longest time scale is about 100 s (Figure 11).

We emphasise the importance of the boundary topology, which must be compatible with targets, spirals and the spiral-target bound states, for which the spiral acts as a phase source and the target as a phase
sink reflecting the coupled spiral-target dynamics. We note that the boundary topology can change while the handedness is externally imposed.

3. Pat and the scientific community

3.1. International aspects

Pat was an internationally well connected scientist. Not only did she spend 3 years in Orsay (1969–1972) as a post-doc, she also had many international collaborators. These included at a senior and post-doc level: Daniel Guillon (Strasbourg, France), John W. Goodby (Hull and York, UK), Harald Pleiner (MPI Polymer Research, Mainz, Germany), Shoichi Kai (Kyushu University, Fukuoka, Japan), Heino Finkelmann (Macromolecular Chemistry, Freiburg, Germany), as well as the two authors.

3.2. Frequent visitors in the Cladis’ Mansion

Pat had many visitors staying extended periods of time in her house in Summit. This included senior colleagues, as well as post-docs. We are, for example, thinking of Peter Palffy-Muhoray, Daniel Guillon, R. Shashidar, Harald Pleiner and the two authors.

3.3. Service to ILCS

Pat was contributing to the international liquid crystal society in a number of ways. Her major formal position was clearly to be the chair of the Honours and Awards Committee for 8 years (1996–2004).

3.4. Pat and women in physics

Undoubtedly one of the major professional efforts of Pat was to encourage female students to pursue scientific research in physics. To promote physics research Pat gave many lectures at colleges across the United States. At Bell Laboratories this was visible by Pat having many female summer interns. Her efforts in this direction were also recognised early on in her career – 1975 – by the award of the Kreeger Wolf Distinguished Visiting Professorship at Northwestern University.

We know from Pat that this activity was inspired by her own personal experience. Indeed, Pat was always pleased to tell the story of her encounter in 1967, as a PhD student, with Christiane Caroli, visiting US. Pat was very impressed by this young female French physicist speaking with a lot of authority to an almost exclusively male audience. She decided to follow her example with positive results exposed here.

4. Epilogue

This obituary would be incomplete without a few words about the human qualities of Pat and of her husband George. All who had the chance to stay in their house know how warmhearted they were, how big was their hospitality.

George was a very gifted cook (grand chef). Their kitchen (immense) was equipped with a very big stove which in French is called ‘piano de cuisson’. One can thus say that George, beside being a painter, was also an artist cook preparing delicious meals on this
instrument. Stemming from a Greek family, Georges loved to cook vegetables on olive oil (of course). By the way, PP reminds very well astonishment of George hearing the word ‘vegetable’ pronounced with a polish-french accent. He laughed loudly and said: you should say ‘wędzGab(a)’! Olive oil, exclusively, was used generously by George. It seems that when he had less then one gallon of it in reserve he was getting nervous.

Besides the ‘piano de cuisson’, there was also a true piano in the house of Pat and George: a black concert grand piano exposed in the lounge. The sound of ‘Moonlight sonata’, played late in the evening by Pat on this sonorous instrument, was getting through walls and ceilings to ears of visitors logged in the guest room on the top floor. Visible through the open window, the spectacle of the rising Moon, stars and fireflies was beginning...

Here come real stars to fill the upper skies,
And here on earth come emulating flies,
That though they never equal stars in size,
(And they were never really stars at heart)
Achieve at times a very star-like start.
Only, of course, they can’t sustain the part.
Robert Frost, ‘Fireflies in the garden’

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