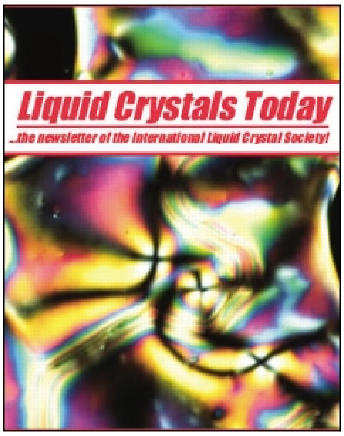

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Professor Sir Charles Frank (1911-1998)

Tim Sluckin[a]
[a] Professor of Applied Mathematical Physics, University of Southampton, UK

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Professor Sir Charles Frank (1911-1998)

Historical Perspectives on the Development of Liquid Crystal Continuum Theory

Tim Sluckin

Professor of Applied Mathematical Physics, University of Southampton, UK

Immortality is granted to those few scientists who give their names to phenomena or constants. In liquid crystal science, one of the best known of these is Sir Charles Frank, who died on 5 April 1998. Sir Charles was much loved and respected by his scientific peers and successors, and those of us who were privileged to know him can still hear his characteristically English incisive tones as he analysed some problem in theoretical physics.

Readers of *Liquid Crystals Today* will be interested in his contribution to the development of the theory of liquid crystals, in the background to this work, and in its subsequent influence. It may well surprise readers that, of Frank's 168 contributions to the scientific literature, only four could remotely be construed as having anything to do with small molecule liquid crystal systems, although there were others devoted to flow alignment in macromolecular systems.

The first of this quartet was a somewhat chatty article written while a postdoc with Debye in Berlin in 1938, in German, entitled *Quasi-kristalline und kristalline Flüssigkeiten*. This youthful contribution, written before the Landau theory of phase transitions elucidated the connection between phase symmetry and the order of phase transitions, gropes somewhat unsteadily toward what eventually became the Maier–Saupe molecular field theory of liquid crystals. Frank cites the standard alloy molecular field theory of Bragg and Williams, and anticipates by many years the insight that small changes in local nematic order can nevertheless be responsible for large changes in global order. The third and fourth papers were somewhat technical discussions written (in 1977 and 1980) after he had already retired. In the former of these papers (in a paper with no references at all!) he points out a rather careless thermodynamic error by de Gennes. In the latter he helped Chandrasekhar to reinterpret some scattering data, which rather than suggesting a hexagonal phase, as Chandrasekhar had previously thought, actually implies a tilted columnar phase.

It is the second paper, however, which immortalized Frank. It appears in the *Discussions of the Faraday Society*, **25**, 19–28, and is entitled simply: *On the Theory of Liquid Crystals*. Most readers

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will not need reminding that these discussions really do take place. The papers are communicated before the event to the participants; the author of the paper then talks for perhaps five minutes in order to highlight the principal points, and then the paper is revisited in a real discussion of the issues raised in the totality of the papers under consideration.

This particular discussion, entitled *Configurations and Interactions of Macromolecules and Liquid Crystals*, took place at the University of Leeds on the 15–17 April 1958. The main focus of the meeting was rather on the newly emergent field of polymers, and only the first few papers were devoted to the study of liquid crystals as we understand the term.

The meeting was chaired by the famous crystallographer and left-wing academic J. D. Bernal, then at the height of his powers, who introduced the meeting. The preamble to the discussion records the attendance of 38 foreign visitors, of whom we might note Professor Stuart Rice, then at the University of Michigan, and Professor Dr W. Maier of Freiburg-im-Breisgau. This is the Maier of the Maier–Saupe theory, who tragically perished in an accident only six years subsequently. Another contributor to the liquid crystal discussion was the young Dr G. W. Gray of the University of Hull, who communicated his discovery of what has come to be called the odd–even effect, and pointed out that some apparent

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hysteresis at the N*-I phase boundary was a misinterpretation of complex data involving surface effects. Professor Maier contrasted his molecular point of view to Frank's continuum approach, publicizing his molecular field theory, which he believed (we now know wrongly!) supported the crucial role of dispersion forces in mesophase behaviour. The hype was necessary, for the Maier–Saupe papers had only appeared in German.

Other contributors to the macromolecular part of the proceedings whose names would later be well known included Andrew Keller of Bristol University, (the late) Rosalind E. Franklin (of Watson–Crick DNA fame) and Aaron Klug (later Sir Aaron, Nobel prize winner and President of the Royal Society) of Birkbeck College, and Michael Fisher of King's College.

Frank's paper is not so much an original piece of work, as a digest and revitalization of calculations that had been conducted by other workers more than a quarter of a century earlier. He starts in characteristic mode:

> "One of the principal purposes of this paper is to urge the revival of experimental interest in its subject. After the society's successful Discussion on liquid crystals in 1933..."

The paper cites only five references. Reference four is by Frank himself (from 1951) on dislocations, in which he was at the time perhaps the leading theorist in the world. Reference two is to experiments of G. Friedel in 1922, and references one and three are to articles respectively by Oseen and by Zocher in the 1933 Discussion.

Frank's 1958 paper, using the notation that has now become standard and following an argument that is almost standard, introduces what are often now known as the Frank elastic constants. He draws attention to the role of each constant in describing bend, splay and twist, and he is the first that I can find who explicitly uses this terminology. He discusses the K_{24} saddle–splay constant, pointing out its unimportance in bulk problems, because of the disappearance of the K_{24} term in the relevant Euler–Lagrange equation. He explains how the chiral term enters in the description of the cholesteric phase, and hints at the existence of the flexoelectric effect (a point taken up by R. B. Meyer eleven years later). He discusses briefly how the theory might be extended to describe the smectic phase. Finally he goes on to discuss defect lines. He recognizes their essential topological role, and draws attention to the analogy with dislocations, which governs his choice of terminology: *disinclination lines*, he calls them.

> "The nematic state is named for the apparent threads seen within the fluid under the microscope..."

In modern language, Frank's essential point is that these observations, and the characteristic patterns seen in crossed polarizers, are the signature of topological defects in the fluid which are themselves the signature of the type of order parameter in the system.

It is an impressive paper, but even Sir Charles himself would probably have stated that it was a recap in more modern language of points made by Oseen and Zocher in the late 1920s and early 1930s. What I want to do now is to briefly look back to the first classical era, and forward to the second classical era, if by the second classical era we mean the renaissance of liquid crystal research which accompanied the discovery of display devices and which started in the mid to late 1960s. Only in this way can we put Frank's paper in its proper context.

Let us first turn the clock back to 1933. Whereas the 1958 Discussion was held in redbrick Leeds, the 1933 Discussion, by contrast, had been held in the august lecture theatre of the Royal Institution (home of Faraday and Davy) in central London. One hundred and fifty members and visitors attended the meeting on *Liquid Crystals and Anisotropic Melts*. We tend to think that in those days liquid crystals were very definitely a minority interest; it is interesting to note that the 1997 British Liquid Crystal Annual meeting, which I had the privilege to organize, welcomed exactly that number of participants. Among eminent foreign visitors at the meeting were Professors L. Ornstein of Utrecht and L. Zocher of Prague (who, the Proceedings record, brought his wife) and Professor Dr R. Schenk of Berlin, President of the *Deutsche Bunsen Gesellschaft*. Not there, except in spirit, for papers were presented on their behalf, were Professor C. W. Oseen of Uppsala and Professor V. Fréedericksz of the Physico-Technical Institute of Leningrad.

The dominant link between 1933 and 1958 is the ebullient figure of Bernal. In the discussion it is always Bernal who pops up with some point or other. 'Further experimental demonstrations were given on Tuesday by Professor van Iterson and Mr Bernal'. Mr Bernal also contributed a paper in collaboration with a D. Crowfoot (better known as Dorothy Hodgkin, Nobel Laureate in Chemistry 1964), in which they conclude:

> "...the mesophases, far from being an anomalous manifestation, take their place in a regular procession from the disorder of the ideal liquid to the regularity of the ideal crystal..."

But to return to the Discussion itself. The dominant theoretical controversy of the day was between the 'swarm' theorists, represented by Ornstein and Kast, and the 'distortion' theorists, represented by Oseen and Zocher. A sideshow, represented by Rinne, adhered to the 'paracrystal' school. Perhaps the Almighty had a message for Professor Dr Rinne, for the Proceedings record that he had been looking forward to coming to England, and had dutifully sent off his paper, but sadly died on 12 March, just over a month before the meeting.

The two major schools of thought emphasized different aspects of liquid crystalline behaviour. For the swarm theorists the most important property to be explained was the turbidity of the liquid crystal fluid, whereas for the distortion theorists the essential feature was the optical anisotropy. The swarm theorists therefore postulated the existence of swarms of aligned molecules, and the literature of the day is filled with diagrams of these swarms allegedly in Brownian motion. A precise mathematical model of the phenomenon was proving elusive, but Ornstein had used this kind of a picture in his successful description of critical point phenomena in classical fluids.

Oseen regally ignored the swarm theorists (perhaps he did not attend in order to avoid controversy), and referred readers to his 1929 monograph. In truth Oseen was in a certain amount of trouble. He constructed a continuum theory (similar, but not absolutely identical to Frank's 1958 model), but thought incorrectly that it depended on a pairwise picture of intermolecular interactions. In addition, he could not see why electrostatic interactions could give rise to the cholesteric phase, thought that liquid crystals not only did not obey the Navier–Stokes equations, but even violated Newton's laws! So one reason Frank's paper was more influential than that of Oseen , which it recapitulated in part, is that Oseen's paper contained mathematical and conceptual errors, as well as some essentially peripheral material.

A contribution from Fréedericksz and Zolina was entitled *Forces causing the orientation of an isotropic liquid*, while Zocher's (who can perhaps be truly regarded as the father of the Frank–Oseen theory) talk was entitled *The effect of a magnetic field on the nematic state*. Zocher was seriously grumpy that anyone, anyone at all, was taking these swarm theorists seriously. Zocher's paper reads like a modern theoretical paper on the continuum theory of nematics. He calculates, in business-like fashion, the dependence of the couple transmitted by a nematic between two walls of a cell, as a function of magnetic field. He (or rather his assistant Eisenschimmel) then tests his theory experimentally.

Of course, Zocher cannot admit his irritation, at least not in print. He starts:

> "Two fundamentally different hypotheses have hitherto been used to account for the changes in nematic systems in a magnetic field; these are the swarm theory and the distortion theory (*Verbiegungstheorie*). It is extremely important to decide which is the correct one, not only for the particular problem dealt with in this paper, but for the whole general question of the structure of these phases."

There follows a mainly sober paper, but with occasional digs at Ornstein or Kast, and the inability of the swarm theory to explain this or that piece of data. In the discussion, however, Zocher is more forthright:

> "As to the swarm theory, it is *impossible* (my italics) to find out the diameter of the swarm..."

he averred, pointing out that susceptibility experiments indicated a swarm diameter ~10^{-5} cm, whereas turbidity experiments indicated 10^{-3} cm. No wonder he was frustrated, but actually his battle was won, and all that remained for his theory to be applied was an excuse to revisit the problem: display devices provided that excuse in plenty.

Let us now move forward in time until after Frank's 1958 paper. A measure of its influence can be given by its citation count. I should emphasize, that large as the citation count of this paper appears to mere mortals, it makes up less than 10% of Frank's total count, most of which cite his papers on defects in solids. Over the period 1980–1997, the paper has been cited 497 times, and the citations continue unceasingly. In the period 1975–1979, it was cited 160 times, and in the period 1970–1974, 241 times. As befits a classic paper, citations have increased with time, and indeed we find only 38 citations in the period 1965–1969, and none at all in 1964 or 1963.

The interesting historical question here concerns the way in which Frank's paper came to influence the liquid crystal literature. How did Zocher's distortion theory shade seamlessly into *The* Frank theory? I am aware, of course, that for many, especially outside the English-speaking world, it remains at the very least, the Frank–Oseen, if not the Frank–Oseen–Zocher, theory. But still, the terms Frank elastic constants and disclination lines are recognized by all liquid crystal scientists around the world.

The great explosion of interest in the theory of liquid crystals started in the 1960s. It came in two parallel streams, which came together much later. Jerry Ericksen and his collaborators at the Johns Hopkins University in Baltimore had been edging toward a full hydrodynamic theory of anisotropic fluids. It was, of course, important for a *hydrodynamic* theory that it should be consistent with the known *hydrostatics*, but Ericksen, and later Frank Leslie, were usually scrupulous to refer to the Frank–Oseen theory. But the strain told, because already by 1966, Ericksen was writing:

> "We consider the hydrostatic theory of liquid crystals as presented by Frank..."

The consistency of the statics and dynamics was a problem, as remarked on by Leslie in the first of his two seminal papers on the hydrodynamics of anisotropic fluids (he was commenting why his theory could *not* be applied to liquid crystals!).

The work of Ericksen and Leslie was matched by that of a physics-based school centred around Pierre-Gilles de Gennes in Paris. De Gennes himself referred to the Frank–Oseen theory, but his experimental collaborators, the Orsay Liquid Crystal Group, of whom perhaps the most celebrated nowadays is Georges Durand, referred to the 'Frank' elastic moduli in a paper in *Physical Review Letters* early in 1969. It seems that via this source Frank's name became associated with the material parameters of the Frank–Oseen–Zocher theory.

Another interesting puzzle concerns defect nomenclature. Let us recall that Frank introduced the notion of a *disinclination*, but the word that finally entered the canon was *disclination*. Once again the group of de Gennes seems to have had a hand in it. The general study of defects in the condensed state was beginning to get exciting; this work was finally pushed forward most by M. Kléman, who was encouraged by de Gennes to make a general analysis of defects in cholesterics. The crucial paper seems to have appeared in 1968, authored by de Gennes and Jacques Friedel (son of Georges Friedel mentioned earlier in this article), in French in the *Comptes Rendues de l'Academie des Sciences*, entitled *Boucles de Disclination dans les Cristaux Liquides*...Disclination loops in Liquid Crystals. There was no English translation, so disclination could have been simply an *ad hoc* translation of Frank's disinclination.

From the same school in 1969 came a study of defects in cholesterics by Christiane Caroli and Elisabeth Dubois-Violette, which appeared in English in *Solid State Communications*, a year later than the de Gennes–Friedel paper, but still referring to ...'the energy of a disinclination line'. However, other follow-up papers to the de Gennes–Friedel paper which appeared in the *Journal de Physique* in 1969 were hedging their bets. Thus in adjacent papers, the Orsay Liquid Crystal group are referring to *désinclinaisons*, whereas Kléman and Friedel prefer *disinclinaisons*, both surely more natural French than disclinations, which is, however, preferred for the *English* version of the abstracts in both cases. So disclinations seems to have arisen as a result of a certain linguistic ambiguity, perhaps even an uncertain subeditor at *Les Editions de Physique*.

I would like to finish with an appreciation of Frank's other work in physics. He was actually educated as a physical chemist, at Lincoln College Oxford. Soon after his postdoctoral spell with Debye, the war started and Frank, with his excellent knowledge of German, served as deputy to R. V. Jones in Air Ministry Intelligence. This story was told by Jones in his *Most Secret War*; Frank and Jones were a crucial part of the radar war which underpinned the bombing efforts of both sides. It was perhaps his experience in interpreting aerial photographs which predisposed his later career toward crystallography. After the war Frank went to Bristol, firstly as a research fellow, then as reader, and from 1954 until his retirement in 1976 as full professor.

He played an important role in ensuring Bristol remained a major research school in physics. His contributions ranged widely over many topics in theoretical physics. His early papers were on the microscopic origins of dielectric phenomena. In Bristol he collaborated with the group of Nobel prize winner Cecil Powell, which inspired him to write a number of papers on fundamental physics.

In the late 1940s he turned to the properties of dislocations. A set of papers with van der Merwe on dislocation motion, with Cabrera and Burton on the relationship between surface structure and crystal growth, with Read on the detailed role of screw dislocations in crystal growth, and with Eshelby and Nabarro on dislocation structure probably secured his fellowship of the Royal Society. The crucial equation in the paper with van der Merwe has been rediscovered on several occasions in connection with nonlinear physics, and is now probably better known as the Sine–Gordon equation, while his paper with Burton and Cabrera is a seminal paper in studies of what is now called the roughening transition.

He maintained a continuing interest in crystallography, and in all stages of his career wrote papers on the optical interpretation of experimental data within his sphere of expertise. From the late 1950s onwards he was instrumental, in collaboration with Andrew Keller, in establishing a premier division team at Bristol in polymer physics. In the late 1960s he turned his interests to geophysics, and wrote several papers on Earth structure. As recently as 1993 he edited *Operation Epsilon, The Farm Hall Transcripts;* these were the transcripts of the conversations of captured German atomic scientists at the end of the war as the news of the Hiroshima and Nagasaki atomic bombs came through.

Sir Charles received much recognition. He was elected vice-president of the Royal Society in 1979, knighted in 1977, received the Royal medal of the Royal Society in 1979 and its Copley medal in 1994, as well as honours from Sweden and India. The bulk of his work had little to do with liquid crystals. We were but a small sideshow, in which Frank was able to transfer some of his expertise gained elsewhere to us, but we remember his work for the influence it had on a critical period in our history.

Acknowledgement

I would like to thank Dr Mervyn Miles of the University of Bristol Physics Department for providing me with some useful material.

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