Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

Plenary Lecture. Some pictures of the history of liquid crystals

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To cite this Article Kelker, Hans and Knoll, Peter M.(1989) 'Plenary Lecture. Some pictures of the history of liquid crystals', Liquid Crystals, 5: 1, 19 — 42
To link to this Article: DOI: 10.1080/02678298908026350
URL: http://dx.doi.org/10.1080/02678298908026350

LIQUID CRYSTALS, 1989, VOL. 5, NO. 1, 19–42

Plenary Lecture

Some pictures of the history of liquid crystals

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A short review on the first 80 years of liquid crystal research has been given, beginning with the molecular theories of the 19th century, referring to the early work of Virchow and Mettenheimer, remembering the important pioneer Lehmann and his colleague Reinitzer. The struggle for truth against badly informed people like Tammann is also mentioned, as well as the final breakthrough and complete rehabilitation of Lehmann and Reinitzer. We apologize for having mentioned Vorländer only briefly but in the following review, Horst Sackmann will refer to him in great detail.

One should pay much more attention to the refraction of artificial liquids whether double refraction appears; but, presumably, Newton has already tested all this.
G. Chr. LICHTENBERG (1790), Professor of Physics (Göttingen), under King George III

The twelfth International Liquid Crystal Conference being held in Freiburg, we meet several traces which are related to the early days of liquid crystal research. Even the pavement of the city of Freiburg seems to anticipate mesomorphic textures.

Some roots reach deeply into the 19th century, when Baden was governed by his royal highness, the Grand Duke Frederic I (see figure 1) for a period of 55 years (1852–1907). We mention this expressly, because in these times it was quite usual for the sovereign to be personally present at symposia where reports on liquid crystal research were given, as, for example, in 1905 at the Bunsen-meeting at Karlsruhe [1]. During the reign of Frederic I a schoolmaster lived in Freiburg, named Franz Xaver Lehmann. He wrote the book *The Spiral of Archimedes* and a textbook *Natural Philosophy for Primary Schools* (Freiburg 1862 and Straßburg 1877, respectively): see also [2, 3]. He liked to reflect on the laws governing the boundaries of leaves. The microscope was his favourite tool.

His sole child, Otto Lehmann, was born in 1855 at Constanz, and in 1862 we find him attending the primary school at Freiburg until 1864. Later he went to the grammar schools of Offenburg (1864–1870) and of Rastatt (1870–1872). Figures 2 and 3 show Otto Lehmann at different stages in his life. The young Otto Lehmann shared his father's interests, especially his studies using the microscope, and this passion remained with him for all of his life. The microscope would become the key to the discovery of liquid crystals. In the Lehmann family natural philosophy was the basis of general culture and education. It was the time when Ernst Haeckel published

Figure 1. Grand Duke Frederic I of Baden.

famous books, e.g. *General Morphology*, which became a constant guide for the young Otto Lehmann. Figure 4 shows a characteristic portrait of Ernst Haeckel.

Another book, also of vivid interest at that time, was the nice old picture book 'Microscopical Delectations for Heart and Eyes' (*Mikroskopische Gemüts- und Augenergötzung*) by M. F. Ledermüller (Nürnberg, 1763) [4], see figure 5. We know that the microscope was an already well-developed instrument in the middle of the 19th century, but it was used predominantly by physicians.

To appreciate the high degree of development, a look into the 312 page monograph by Gabriel Gustav Valentin 'The Examination of Plants and Animals Tissues in Polarized Light' (*Die Untersuchung von Pflanzen- und Tiergewebe im polarisierten Lichte*) [5] is recommended. Valentin was a Czech savant who lived in Switzerland, he was awarded in 1835 the prize of the French Academy of Science for a comparative study of the modes and development of animal and vegetable tissues. In 1841 he saw trypanosomes for the first time. His great successes, which were essentially due to the use of the microscope, were completed by the study of muscles and tendons with polarized light. With the Nörremberg instrument, both orthoscopic and conoscopic observations could be carried out, as early as in 1861! A few examples from Valentin's book may demonstrate what perfection polarization microscopy, even with living objects, had been achieved in the middle of the 19th century. Perhaps it is not generally known that J. W. v. Goethe was already strongly interested in optical studies, and that he used a polarization device to investigate 'entoptical colours'

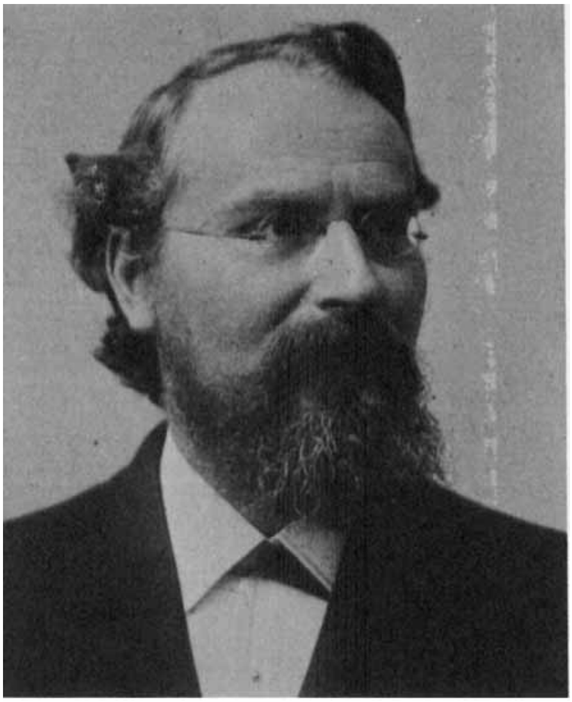


Figure 2. Otto Lehmann.

Figure 3. Otto Lehmann.

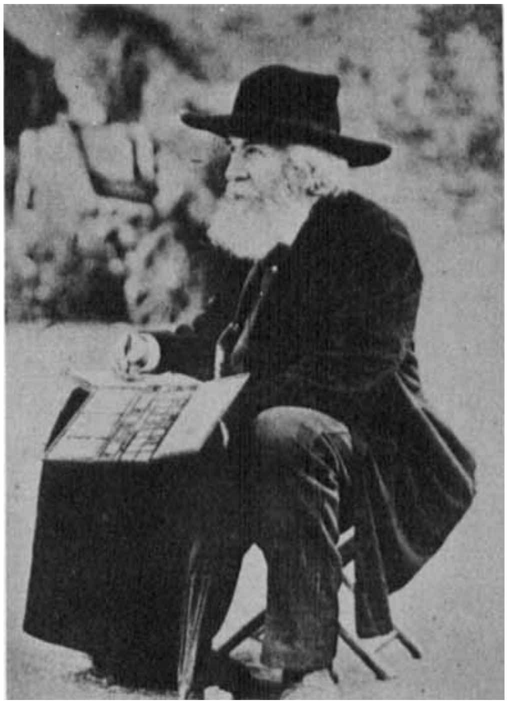


Figure 4. Ernst Haeckel.

('Entoptische Farben') [6]. In this case the double-refracting object consisted of a rapidly cooled glass plate, and polarization was obtained by reflection from mirrors (see figure 6). Figure 7 shows the instrument of Professor Wild, figure 8 the convergence system of Valentin, figure 9 axis pictures with a $\lambda/4$ plate, and figure 10 the tissue of a frog.

At the time that Valentin prepared his splendid monograph, the great physician Rudolf Virchow discovered Myeline [7], one year before Lehmann was born. One year later, in 1855, the ophthalmologist C. Mettenheimer found liquid myeline to be double refracting. It is remarkable that the latter observation was respectfully cited by Valentin. Figures 11 and 12 show Virchow and Mettenheimer [8].

Without doubt, the observations of Virchow and Mettenheimer have to be recognized as the first observations of liquid crystals, lyotropic ones, as we call them today. Even Lehmann himself has clearly pronounced that myeline forms were liquid crystalline [9].

At this time mineralogy, physics and chemistry were already well-developed sciences, but the theoretical background concerning isomerism, phase transitions, isomorphism and polymorphism had just begun to be understood. The most important investigations were made by Faraday, Mitscherlich, Berzelius and Moritz Ludwig Frankenheim, born in 1801 and teaching at Breslau University. Berzelius coined the term 'isomerism' to describe different compounds which have the same elemental composition. Mitscherlich discovered the rules to govern isomorphism, e.g. with phosphates and arsenates. Frankenheim, almost forgotten today, studied the phenomena of phase transitions (polymorphism!) and cohesion [10], which was regarded as a very basic principle of chemistry. Please allow us to cite once more Goethe to demonstrate how deep this phenomenon was embedded in the consciousness


Martin Frobenius Ledermüllers,
Hochfürstlich-Brandenburg-Culmbachischen Justiz-Raths, wie auch der Kayserlichen Akademie der Naturforscher und der Deutschen Gesellschaft zu Altdorf Mitglieds,

Mikroskopische
Gemüths-
und
Augen-Ergötzung:
Bestehend,
in
Ein Hundert nach der Natur
gezeichneten
und mit Farben erleuchteten Kupfertafeln,
Sammt
deren Erklärung.

Verlegt von Adam Wolffgang Winterschmidt,
Kupferstecher in Nürnberg,
gedruckt von Christian de Launoy.
1763.

Figure 5. The cover of Ledermüller's book.

of the early 19th century. In *Wahlverwandtschaften* [11] a scientific discussion is described in which 'cohesion' plays an important role. Note that the work of Frankenheim provided many important inputs into Lehmann's early reflections; see, for example, [12].

Much more could be said about the state of the art in the field of chemical and physical crystal research to understand better the gigantic productivity of Otto Lehmann. After having finished his school education at Rastatt, he went to the University at Straßburg in 1872. For a better understanding of the time and the atmosphere shortly after the 1870–1871 war against France which resulted with a German victory and the re-integration of Straßburg into the German Reich, look at the Siegesdenkmal (Monument of Victory) in Freiburg. The best academic teachers were assembled in this revived place, where Johann Wolfgang von Goethe had, 100 years earlier, studied Law. Lehmann's teachers were A. Kundt (Physics), P. Groth (Crystallography), A. von Baeyer (Chemistry) and Rosenbusch, the petrographer, from whom he learned to deal with heating processes. Lehmann finished his studies in Straßburg at the University in 1876; his doctoral thesis is entitled *Über Physikalische Isomerie* (figure 13). It was published shortly after in its original form, without any changes [13]. Following his father's wishes, Lehmann became a teacher

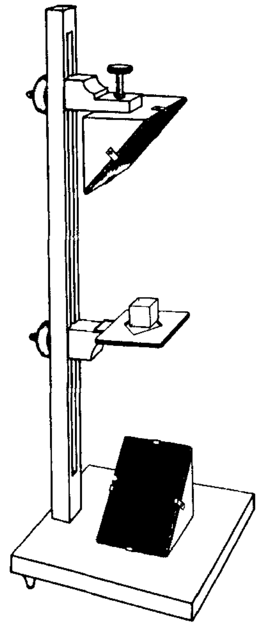


Figure 6. A Seebeck instrument. From R. Matthaei, 1971, *Goethes Farbenlehre*, by kind permission of O. Maier Verlag, Ravensburg.

of natural sciences, and his first workplace was the grammar school at Freiburg. It is worth looking into his dissertation, because here the basis of his future work is clearly visible and the roots of later temptations, errors and priority struggles are also visible.

Physical isomerism: what does it mean? To understand Lehmann's theoretical concepts we have to learn that 'different states of aggregation' are related to and caused by 'different molecules' whatever this may mean. This is the dogma to which he adhered until his life's end! Isomerism, as understood by Lehmann, can be either polymerism or metamerism; either chemical or physical. Chemical isomerism is related to atoms, physical isomerism to molecules. Examples are given in figure 14; these are pictures from Lehmann's *Molekularphysik* [14]. Remember that all of these pictures are purely hypothetical, and a really prophetical sentence of Lehmann means:

> If a new chemical theory should appear which pulls down the barrier between chemical and physical isomerism, we have only to change the terms 'chemical' and 'physical', and suddenly, without all the troubles and confusions, the terms are adapted to a new theory. Isn't it simple and evident?

As a consequence of the 'Lehmann theorem' we read: 'No substance [*Körper*] exists in more than *one* state of aggregation . . .'. He really believed that there are, for example, different molecules existing in the gas phase and the coexisting condensed phase [15]. A note from Lehmann's dissertation (see also [13]) should be added to

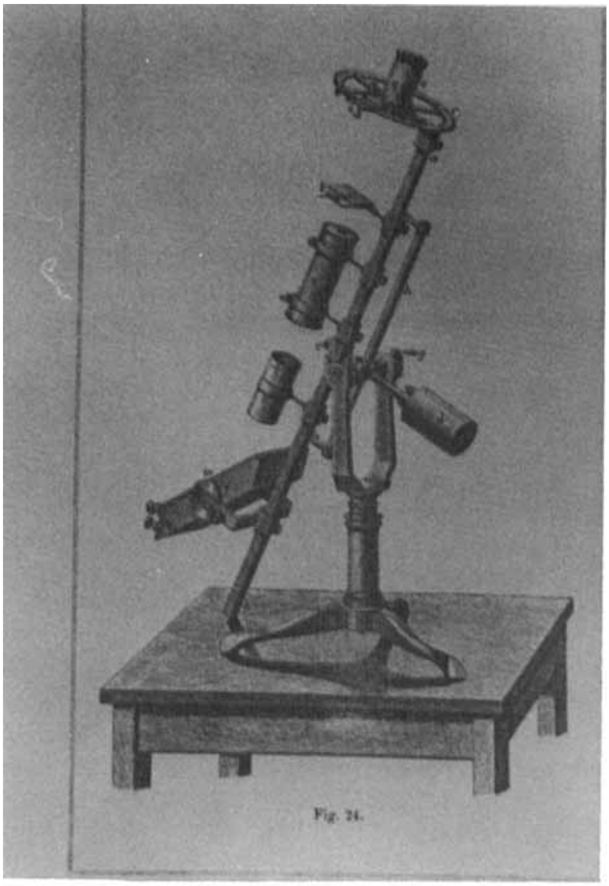


Figure 7. The Wild instrument.

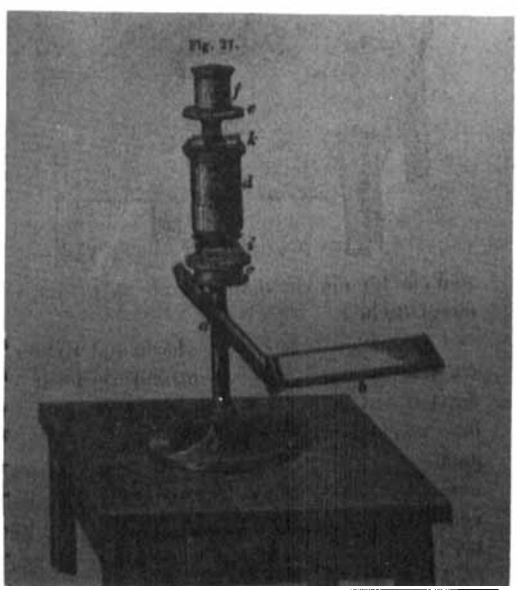


Figure 8. The Valentin system.

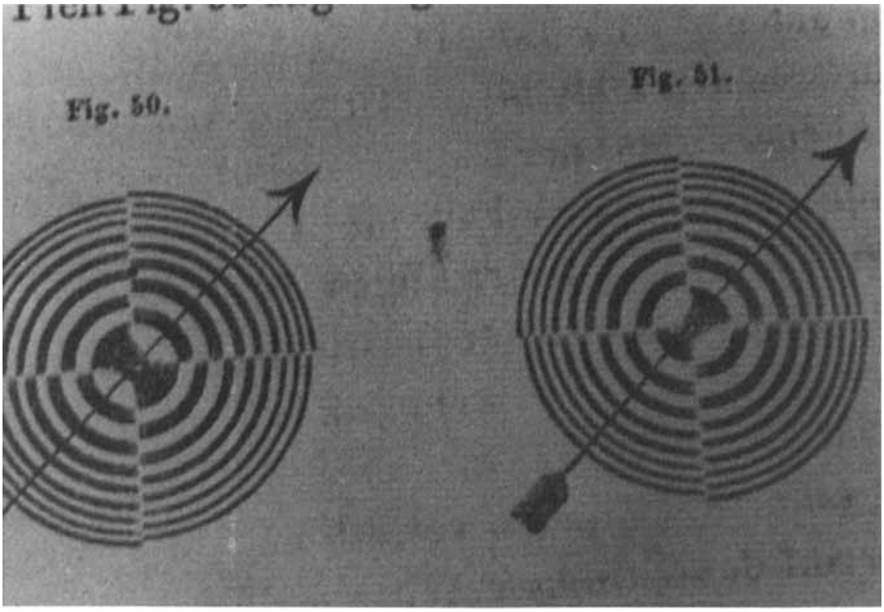


Figure 9. Axis pictures.

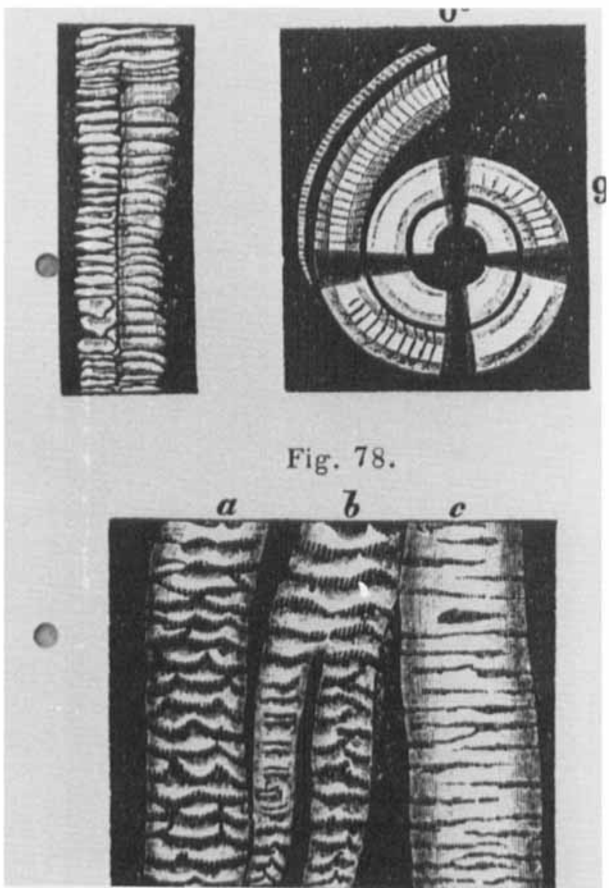


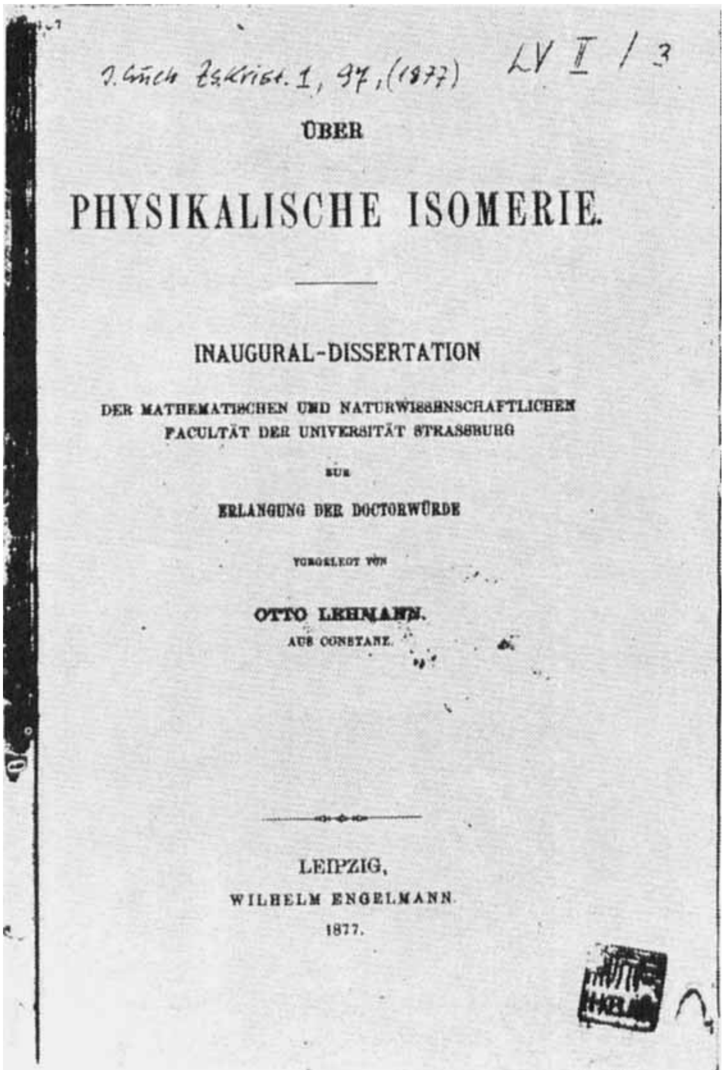
Figure 10. Tissue of the frog.

Figure 11. Virchow.

Figure 12. Mettenheimer (1824–1898).

demonstrate how Lehmann was obliged to revise his opinions fundamentally:

> A liquid is not a body with no cohesion, as it is sometimes declared, but it is a body with a cohesion being too weak for being able to hold the molecules in a fixed position against the movements caused by heat; *such a liquid will never be able appearing with crystalline structure* [13, p. 121].



ÜBER

PHYSIKALISCHE ISOMERIE.

INAUGURAL-DISSERTATION

DER MATHEMATISCHEN UND NATURWISSENSCHAFTLICHEN
FACULTÄT DER UNIVERSITÄT STRASSBURG

ZUR

ERLANGUNG DER DOCTORWÜRDE

VORGELEGT VON

OTTO LEHMANN.

AUS CONSTANZ.

LEIPZIG,
WILHELM ENGELMANN.
1877.

Figure 13. Lehman's doctoral thesis.

With this statement in mind, Lehman entered his professional career at Freiburg. After some months he had already moved; although the reason is not clear, he went to Mülhausen im Elsaß, not far away from Freiburg. He stayed there for 6 years, developing 'his' polarization microscope.

The development of the polarization microscope played an important role in the history of the discovery and development of liquid crystals and is strongly associated with the name of Otto Lehmann. It is his merit that it reached a high standard of development within only a few years. Since Otto Lehmann was one of the best-known and most experienced scientists in crystallography, Reinitzer asked him for his opinion and his help for his discovery.

The first publication by Otto Lehmann in which a detailed description of the instrument was given was his doctoral thesis [13]. His hand sketch of the microscope is illustrated in figure 15. In the following years he built altogether more than 25 different types of the instrument. One of the most interesting samples is the

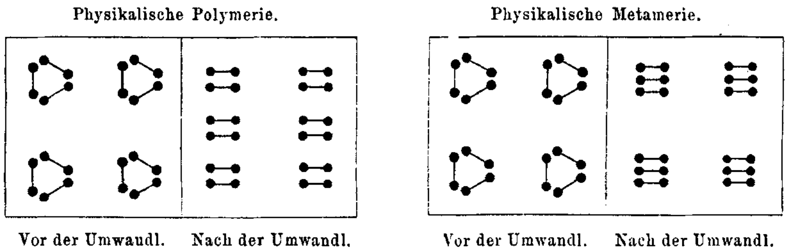


Fig. 545. Fig. 546.

In der That findet sich das merkwürdige Resultat, dass sich thatsächlich die bis jetzt beobachteten Erscheinungen von physikalischer Isomerie mit sehr wenig Ausnahmen in zwei Gruppen zusammenstellen lassen. Bei der einen Gruppe, die ich enantiotrope genannt habe, lässt sich die Umwandlung durch entsprechende Temperaturänderung rückgängig machen, bei der anderen, der monotropen, findet sie (wenigstens im festen Zustande) immer nur im einen Sinne statt.

Figure 14. Pictures from Lehmann's *Molekularphysik*.

microscope which was derived from the Merz instrument, a gift from his father, and which was developed in 1888–1889, a period which coincides with the anniversary of the discovery of liquid crystals. Indeed, we may assume that most of the early observations of liquid crystals, perhaps even of the Reinitzer's samples were made with this instrument. It is illustrated in figure 16. Later this microscope was equipped with a supplementary photographic unit to be swung in front of the ocular. Illumination was made with the help of a thread of magnesium (q), even a little ashtray (s) for collecting the burned magnesium was provided. It was also this instrument which, in his later period at Karlsruhe, was extended to include a strong magnet to observe the behaviour of liquid crystals under the influence of a magnetic field.

A later development which was derived from the microscope of Voigt and Hochgesang was built in larger numbers (see figure 17). The particular instrument which was used by Otto Lehmann is still existing. It is probably one of the ten instruments which were exhibited for demonstrations on the occasion of Lehmann's famous talk at the Sorbonne in 1909. A more detailed description of the history of the crystallization microscope is given in [2].

In 1883 Lehmann followed a call to the Polytechnical School at Aachen to become an assistant of Professor Wüllner. He continued his studies with the polarization microscope, but his main job was to give lectures on experimental physics and on electrotechnical problems such as ignition of firedamp by heat or by spark. In 1885 he was named 'Extraordinary Professor'. During this time he was very busy writing textbooks on physics, and he collected material for a large collective work, namely *Molekularphysik*: two volumes, with more than 2000 citations; volume I, from 1888, has 852 pages and 375 figures; volume II, from 1889, comprises 697 pages and 245 figures. In this book Lehmann summed up the whole knowledge about the physics of matter in his time, but Wilhelm Ostwald has criticized it: 'The author has collected an uncommon and widespread material, and he reported in a fullness which affects confusion. We receive infinite instruction, but no evident picture of the whole field'.

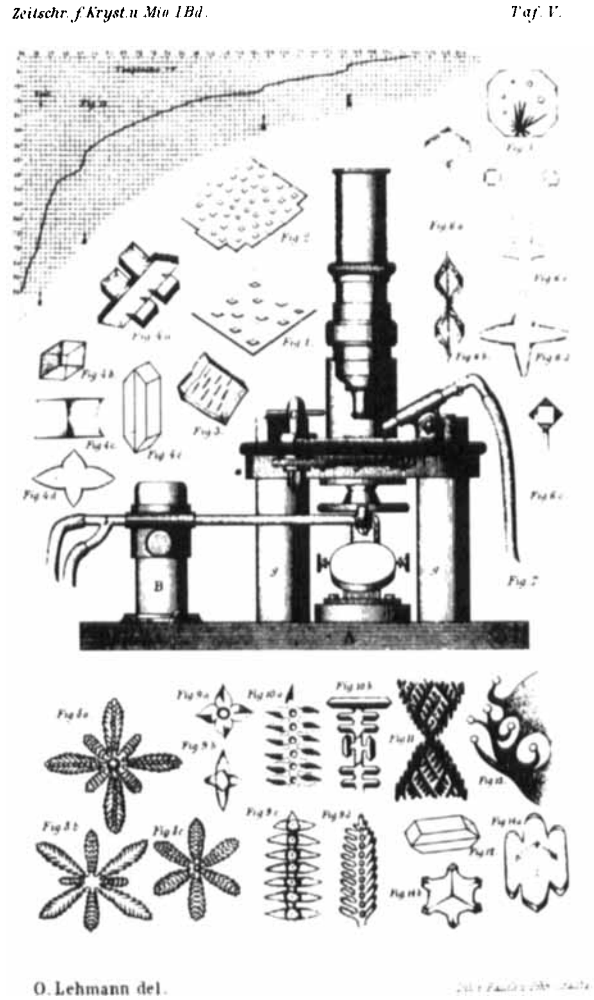


Figure 15. Sketch of Lehmann's first microscope.

It is remarkable that already in volume I, pp. 521–523, 'contact movements and myeline forms' are briefly described, referring to Quincke (see figure 18) and E. Brücke [16]. A nice woodcarving is presented (see figure 19), and most remarkably, he refers to Brücke's observations on myeline forms which 'generate in polarized light beautiful colour phenomena'. They are produced from nerve core and a solution of soap in oleic acid. However, there is no reference to Virchow/Mettenheimer who had described the phenomenon 25 years earlier.

The year is 1888! The greatest suprise has broken through, the reason to celebrate *this* year, the centennial of the discovery of thermotropic liquid crystals! Let us report exactly the story of these important events in the right succession.

14 March 1888: First letter from F. Reinitzer, Prague (see figure 20) to O. Lehmann, who was at Aachen at this time (see the letter detailed in [2]).

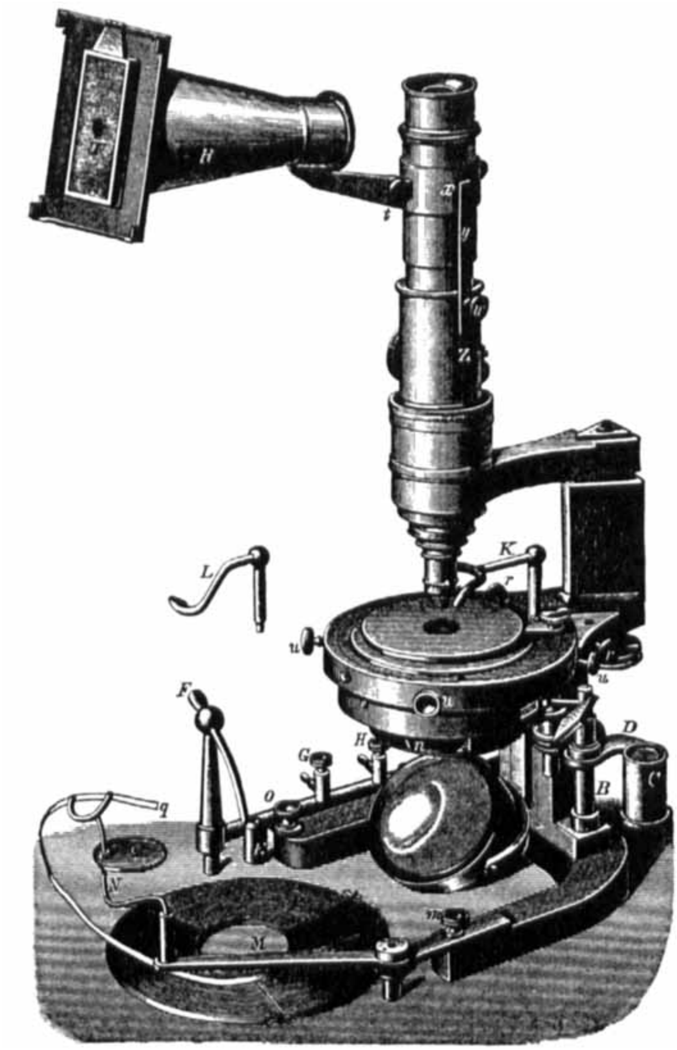


Figure 16. The Merz microscope.

27 March 1888: Answer from Lehmann. He regrets not being able to engage himself with the matter thoroughly, but he found 'three modifications of the cholesterol benzoate as well as with the acetate'.

2 April 1888: Reinitzer discusses, in his letter to Lehmann, the optical activity of the cholesteric esters. The main object of their discovery is the so-called 'Gries', a provisional name for the anisotropic crystal lammellae. This discussion is governed by the question: where might the optical activity come from?

4 April 1888: Lehmann to Reinitzer: Lehmann asks for more material, because he intended to occupy himself during the summer holidays with the cholesterics. He adds a sketch showing droplets with the polarization cross (see figure 21)!

10 April 1888: Reinitzer to Lehmann: Reinitzer reports different textures if the cholesteryl benzoate is heated, starting with the solid phase, or, on

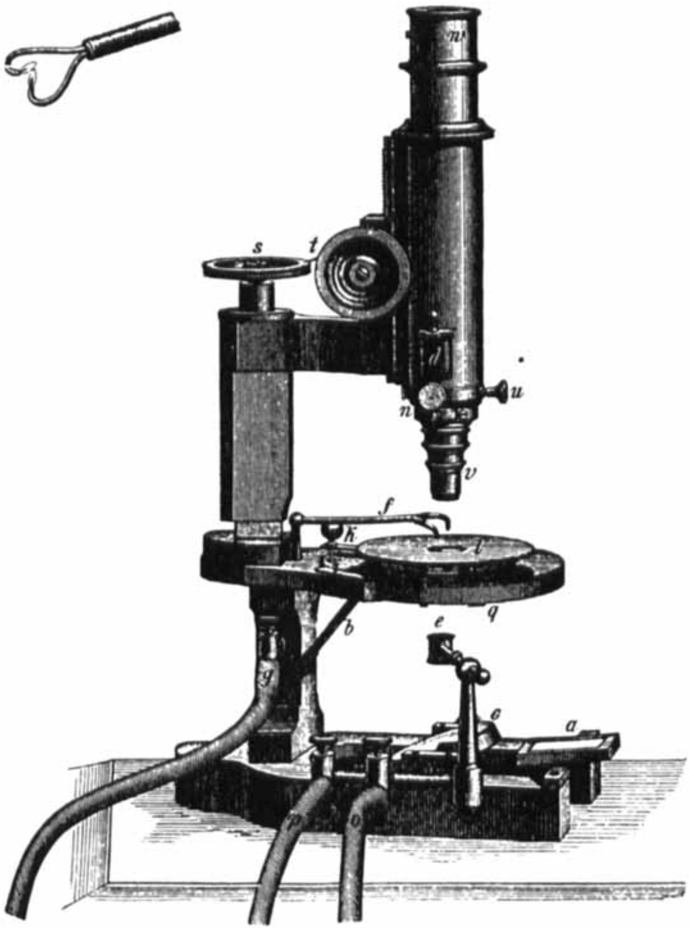


Figure 17. Crystallization microscope of Otto Lehmann.

the other hand, if the isotropic melt is cooled. He means that an 'isotropic fluid' should exist between the star-like aggregates. Later Lehmann remarks that this 'isotropic' liquid is 'pseudo-isotropic' and chemically identical to the droplets: see also figure 3.5 in [2].

13 April 1888: Lehmann answers. Droplets or crystals? 'Like real crystals but vanishing at their borders'. 'Observations during working hours' together with Wüllner, but by no means a clear explanation.

20 April 1888: Reinitzer discusses the behaviour of the melt after sudden cooling. Blue colour phenomena! [42].

24 April 1888: Lehmann discusses the 'oily streaks': crystal or not? Laminae which look like liquid. The substance looks quite strange, really ghostly (*unheimlich*).

With this letter the correspondence was interrupted for 16 months! On 1 October Lehmann moved to the Technical High School, Dresden, where he remained for only six months, but he completed his giant work in two volumes, namely *Molekularphysik*. In *both* volumes Lehmann presents himself as 'Professor der Elektrotechnik am Königlichen Polytechnikum zu Dresden'. It is interesting that Lehmann had published

Figure 18. G. Quincke.

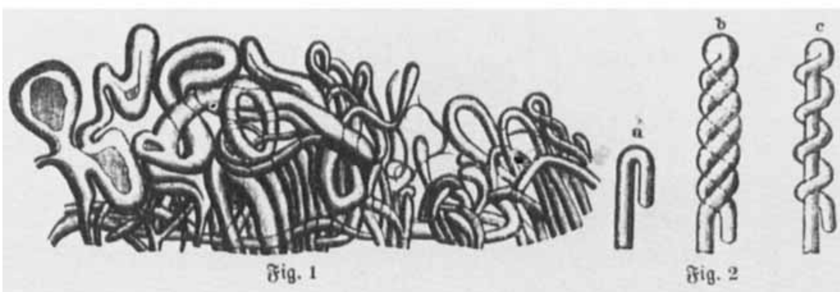


Figure 19. Myeline forms, original printing block from 1888 (design by Lehmann).

already, early in 1889 (January–March) in *Molekularphysik II*, his own observations with cholesterol esters in an electrical field, and in the supplements he gives rather long descriptions of the 'chromatic polarization', including the colour phenomena and their temperature dependence. However, the term 'liquid crystals' is not yet used. The correspondence with Reinitzer was continued when Lehmann became the successor of Heinrich Hertz in Karlsruhe in the chair of experimental physics.

20 August: 1889: Lehmann writes: my new observations confirm your formerly expressed opinion, that the 'Gries' consists of very soft crystals that should be regarded as a physically isomeric modification. No foreign liquid is present . . . . The letter closes: 'It is of a high

Figure 20. F. Reinitzer.

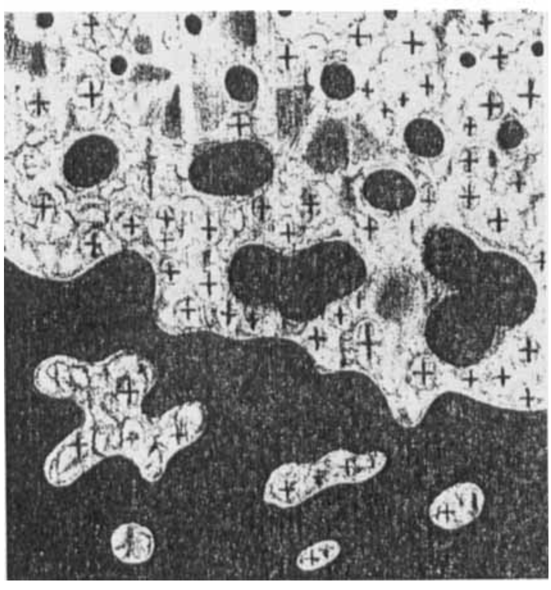


Figure 21. Droplets of cholesteric.

interest for the physicist that crystals exist with a softness, being so considerable *that one could call them nearly liquid.*' Ten days later,

30 August 1988: Reinitzer answered, giving some more details, concerning the twofold appearance of colours during heating. *The same day* Lehmann sent his paper 'Über fliessende Kristalle' to the *Zeitschrift für Physikalische Chemie* [17].

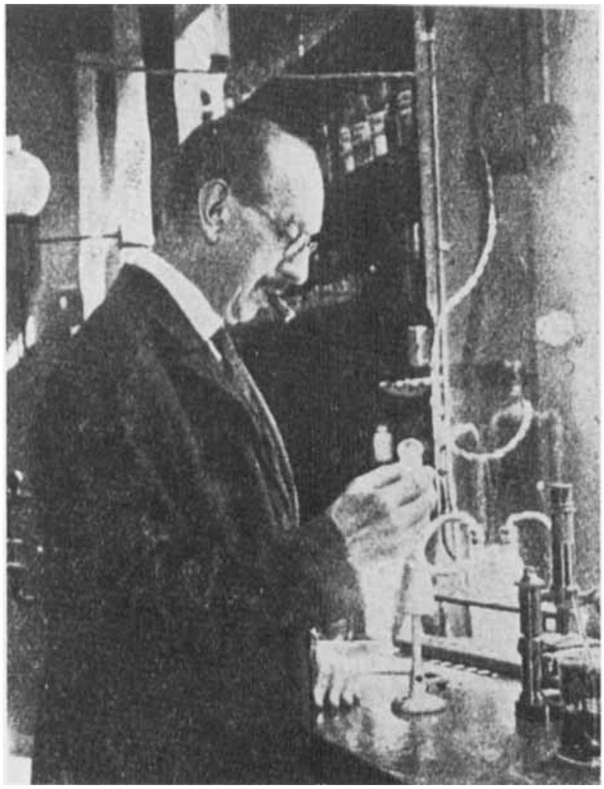


Figure 22. Gattermann.

Lehmann has found *the* theme for his further life! The events to come confirmed Lehmann's results. The part of Reinitzer was, nevertheless, the decisive step. One is very sorry about the struggle for priority which broke out at the beginning of the 20th century, when Lehmann misunderstood intentionally an objective remark in D. Vorländer's textbook from 1908 [18–20].

In the following we give only the most important events, names and fragments of texts.

1890:	Gattermann (figure 22) and Ritschke synthesize the azoxyethers [21].
1893:	Foundation of the 'Deutsche Bunsengesellschaft' at Freiburg; first president: J. H. van't Hoff.
1902:	Meyer and Dahlem synthesize the first smectic mesogen, the azoxybenzoic acid ester [22].
1901:	van Romburgh: on *p*-methoxycinnamic acid [23]. Habilitation R. Schenck at Marburg (see figure 23).
1904:	The large monograph *Flüssige Kristalle sowie Plastizität von Kristallen im allgemeinen, molekulare Umlagerungen und Aggregatzustandsänderungen* by Lehmann [24], Volume I of Frick's *Physikalische Technik*, editor: Lehmann [25].
June 1905:	Bunsen-Symposium at Karlsruhe, Schenck and Lehmann present their experiences with liquid crystals. Confrontation with G. Tammann.

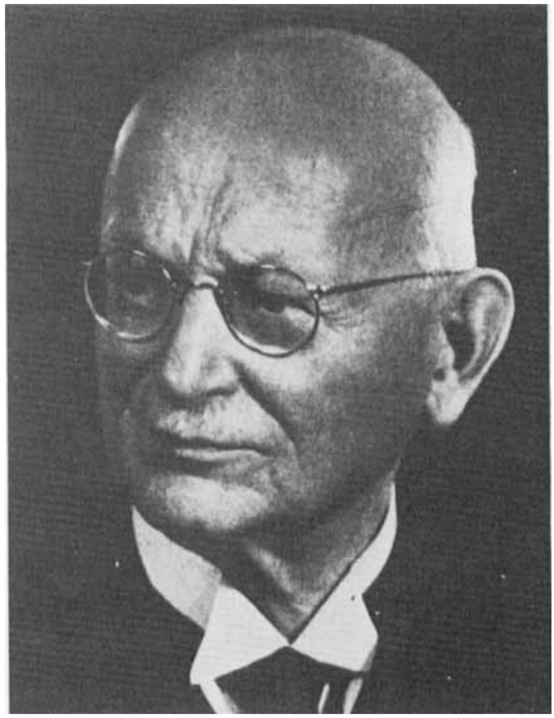


Figure 23. Schenck.

Figure 24. Vorländer.

Figure 25. Voigt.

Figure 26. Riecke.

Figure 27. Ernst Haeckel's *Kristallseelen*.

Figure 28. G. Friedel.

(*a*)

(*b*)

Figure 29. Beetles in (*a*) natural and (*b*) polarized light.

1905:	R. Schenck's work *Kristalline Flüssigkeiten und Flüssige Kristalle* [26]. Continuing the struggle against Tammann.
Sept. 1906:	78th Assembly Deutscher Naturforscher und Ärzte at Stuttgart. Lehmann: *Flüssige Kristalle und die Theorien des Lebens*; D. Vorländer: *Neue kristallin-flüssige Substanzen.*
1906:	VIth International Congress for applied chemistry at Rome. W. Nernst presents his new theory: liquid crystals are to be regarded as mixtures of tautomeric molecules.
1907:	Lehmann gives a lecture in Wien, 30 December!
1907–09:	E. Bose publishes his papers on swarm theory [27].
1908:	The first monograph of D. Vorländer (figure 24) comes out [28].
1909:	O. Lehmann gives lectures at the Sorbonne in Paris and in Geneva.
1910:	W. Voigt, Göttingen (see figure 25) has included a short article on liquid crystals in his textbook *Lehrbuch der Kristallphysik* [29] and a complete rehabilitation of Lehmann who plans for Hundsbach.
1916:	Another theory of liquid crystals (electric dipoles) is presented: M. Born [30], see also [31], Riecke (figure 26).
1917:	F. Stumpf reviews all papers on optical effects in liquid crystals until 1917. Stumpf left Göttingen, he worked together with Born, he is now in Kiel [30a, 32].
1917:	E. Haeckel's *Kristallseelen* (souls of crystals) is published. An exaggerated acceptance of Lehmann's 'Theorien des Lebens'. See also [2, 33]; figure 27. Intensive correspondence between Haeckel and Lehmann, which had begun in 1906, see [33].
1921:	*Versuch einer kinetischen Theorie der kristallinen Flüssigkeiten* by C. W. Oseen: see [34], a really basic analysis and a classical theory, still valid today.
1922:	Death of O. Lehmann and the year of his last papers [35, 36].
1922:	G. Friedel's (see figure 28): famous publication, where he coins the terms 'nematic', 'cholesteric' and 'smectic': 'Les Etats mésomorphes . . .' [37].
1924:	Another monograph of Vorländer: correct explanation of the homeotropic state, relations between molecular structure and mesomorphic behaviour elucidated, and a counter order concerning Lehmann: *Chemische Kristallographie der Flüssigkeiten* [38]. P. Gaubert observes circular polarization with beetles (see figure 29), [43].

During the 1920s the science of crystalline fluids flowered more and more, but no applications were seen, as Vorländer mentioned explictly at the end of his monograph [38]. Symposia did not yet exist; the first took place in 1933 with The Faraday Society [39], but the first written discussion was organized by the famous crystallographer P. P. Ewald in Stuttgatt [40]. All of the experts participated, among them were some personalities whom we have just mentioned. A listing of the names may be enough: G. and E. Friedel (Strasbourg), D. Vorländer (Halle), L. S. Ornstein (Utrecht) H. Zocher (Berlin-Dahlem), K. Herrmann and A. H. Krummacher (Charlottenburg), W. Kast (Freiburg), C. W. Oseen (Uppsala), C. Hermann (Stuttgart), W. Ostwald (Leipzig), V. Freedericksz and V. Zolina (Leningrad). As discussion contributors R. Schenck, J. J. Trillat, G. Foex, M. Jezewski, J. Errera and, of course P. P. Ewald took part.

About the following events we are well informed by an abundant literature, for a nearly complete collection of papers until 1980 let me mention our *Handbook of Liquid Crystals* [41].

The authors wish to dedicate their article to Dr. Gert Noll, Frankfurt-Höchst, for his contributions to applied ophthalmology. He saved the sight of Professor Dr. H. Kelker and, for many years, has been a good friend to him.

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