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# Liquid Crystal Research in China: 1970-1982

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#### IN THE BEGINNING . . .

Liquid crystal was discovered in 1888. However, it was only after the observation of dynamic scattering in nematics in 1968 and the subsequent application in displays that liquid crystal, as a field of research, has obtained its vitality and importance as we know it today.† Not surprisingly, it was in the early years of this modern era of liquid crystals that China ventured into this exciting "new" field. <sup>1-16</sup>

The first project was started in 1970 in the Institute of Chemistry of the Chinese Academy of Sciences (C.A.S.), in Beijing. It involved the synthesis and characterization of many different cholesterics and to use them in the nondestructive testing of metal surfaces. With the collaboration of the Institute of Mathematics (C.A.S.), optimization method was used in the search for the best material. In fact, more than thirty cholesterics were successfully synthesized with a temperature sensitivity of 0.1 degree. This project lasted three years.

Almost at the same time, Qinghua (Tsinghua) University, the Beijing Institute of Reagents and the Shanghai Institute of Organic Chemistry (C.A.S.) also started to work in liquid crystals.

<sup>†</sup>For a brief history of this field see Lin Lei, in Science Yearbook 1981 (Shanghai Science and Technology Press, 1981), p.I.21.

#### 2. IN ACTION

In the past twelve years, in China, research in liquid crystal covers the four areas in application, chemistry, biology and physics.

#### **Application**

Most of the applied research are related to displays and diagnosis of tumors.

In a collaboration between Qinghua University and industries in Beijing and Nanchang, analog voltmeter with liquid crystal display was made in 1977; two years later, large-area matrix display of Chinese characters for weather reporting using negative storage effect was successfully developed.

A large amount of work including numeral and matrix liquid crystal display are also carried out in the Shanghai Institute of Organic Chemistry. For example, using the electrooptic effect of the storage mode of cholesteric-nematic transition, sequential array displays of a matrix addressing model was performed. A liquid crystal page composer of 32 × 32 bits was constructed with a write-in time of 520 ms. When connected with a laser holographic calculating model of 10<sup>6</sup> bits the page composer can function as a coding device for input data. As a joint effort with the Institute of Chemistry an organic conductor photoconductor liquid crystal light valve was constructed. Similar light valve with inorganic conductor is now being developed.

A liquid crystal viewer for three-dimensional x-ray examination of human bodies was built by scientists from Fudan University, East China Institute of Chemical Technology, Shanghai Institute of Dyes and Pigments and Shanghai Huashan Hospital. A liquid crystal storage device with infrared laser write-in and matrix erasure was also studied.

From 1972 to 1975, as a collaboration between Qinghua University and the Beijing Institute of Glasses a reflective-type large television screen using cholesterics as light valve in a sealed electron tube was successfully constructed. 75,74 For the first time liquid crystal material and driving method with response time fast enough to satisfy the requirement for television signals are obtained. The screen is  $2 \times 3$  m<sup>2</sup> in size, resolving power up to 250 lines and contrast ratio 10:1.

In the Enshi Municipal Hospital of Hubei Province a liquid crystal display for x-ray images is being studied. The idea is to replace the current x-ray luminescence screens which are expensive, low in brightness and poor in contrast.

For several years the Shanghai Institute of Organic Chemistry has been engaged in using cholesterics in the nondestructive testing of metal surfaces, the joints of composite materials with honeycomb structures and possible defects in solar batteries.

In the Institute of Physics (C.A.S.), a new liquid crystal hybrid bistable optical device and its use for optical logic elements and ultralinear modulation applications are demonstrated.<sup>67,68</sup> A liquid crystal camera for infrared laser mode examination<sup>69</sup> and a negative lens effect induced by strong laser field in liquid crystal<sup>73</sup> are also reported.

As medical applications are concerned, microencapsulated cholesteric films have been made in both the Institute of Biophysics  $(C.A.S.)^{77}$  and Wuhan University. The diameter of each individual capsule is  $10-30~\mu m$ . The temperature range in which these films change color is  $3-4^{\circ}C$ , with a sensitivity of  $0.5^{\circ}C$ . When used in infrared thermal imaging colored image of the filament of a 500 watt infrared lamp is obtained at a distance of 4 m. In Wuhan University, these films have also been used in tumor diagnosis, laser detection, etc.

Examination of possible tumors in breasts and human skins using liquid crystal thermography were administered systematically to a large public by the Tianjin Institute of Tumors, Wuhan University, Hubei Medical College and Shanghai No. 1 Medical College, respectively. In one example of breast tumor diagnosis, it involves more than one thousand patients and the accurate rate is higher than 80%.<sup>16</sup>

In addition, in quite a number of factories and research institutes in the cities of Beijing, Shanghai, Sian, Zhengzhou, Changsha, Guangzhou, Wenzhou, Hangzhou and Changchun there are on going research in the technology of liquid crystal display and other applications.<sup>16</sup>

In relative short time, liquid crystal displays in watches and pocket calculators are developed and made commercially in China. Progress is also achieved in the research of colored liquid crystal displays. In both Qinghua University and the Shanghai Institute of Organic Chemistry, satisfactory results in negative type color display using guest-host effect are obtained and further study is in progress.

#### Chemistry

A large number of liquid crystal compounds including biphenyls have been synthesized and studied for a long period of time in Qinghua University, 55 Beijing Institute of Reagents, Shanghai Institute of Organic Chemistry, 94 East China Institute of Chemical Technology 53 and oth-

ers,<sup>57</sup> leading to a deeper understanding of the relationship between the structures of thermotropic liquid crystal molecules and the characteristics of phase transitions.<sup>2,4,70</sup> At present, in China, liquid crystal materials qualified for display applications can be manufactured within the country.

In the area of polymeric liquid crystals, <sup>7,50,54,59,60</sup> optical properties and viscosities of poly-p-phenylene terephthalamide (PPTA)/sulfuric acid (99.95%) solution has been studied by the Institute of Chemistry. A transition from nematic to cholesteric-like phase is reported. <sup>59</sup> The supermolecular structure of PPTA films were studied by optical and scanning electron microscopy. The samples were prepared by coagulating crystallization from anisotropic solution of PPTA in 100% H<sub>2</sub>SO<sub>4</sub> which were sheared and then relaxed for different time. Three kinds of morphological structures, viz., shperulitic, shish-kebab and mat like domain aggregates were observed. <sup>60</sup>

In a collaboration with Qinghua University, spherulitic growth of poly-p-benzamide from nematic mesophase were observed.<sup>50</sup>

Besides, in the Institute of Chemistry cholesteryl pelagonate liquid crystal was studied by small angle light scattering. When temperature is lowered to 92°C from the isotropic liquid phase a known cholesteric phase first appears. However, at 85°C a new cholesteric phase was discovered.<sup>56</sup>

In the East China Institute of Chemical Technology, liquid crystals as stationary phase in gas chromatography are extensively studied. 51,52,58,

#### **Biology**

To demonstrate the relationship between the liquid crystalline structure of biomembranes and their functions the ordering and fluidity of rod outer segment (ROS) membranes are studied by the Institute of Biophysics. The diffusion of rhodospin molecules and the birefringence of ROS membranes of frog were measured by microspectrophotometry and polarizing microscopy, respectively. <sup>16</sup> Recently, in the study of ROS membranes of Gekko gekko it was found that the fluidity of ROS membrane was the smallest after it had been pretreated at 45°C. The birefringence turned from positive to negative when the temperature of the treatment was higher than 46.5°C. Some sort of irreversible phase transition in the ROS membrane at about 45°C was suggested to take place. <sup>61</sup> The Institute of Traditional Chinese Medicine and Pharmacology at Jinan participated in this work.

Also, in the Institute of Biophysics, the effect of temperature and glutaraldehyde treatment on early receptor potential (ERP) of the frog

(Rana Nigromaculata) were studied. 65,66 It was found that the amplitude of ERP of isolated frog retinas show enhancement within a certain temperature range. When the retina is treated at higher temperature, the amplitude falls below normal and vanishes eventually. These phenomena are explained to be related to the liquid crystalline state of the ROS membranes.

Presently, the Institute of Biophysics is engaged in the study of artificial membranes to understand the influences of the structures, photoelectric properties and other factors on the electric parameters and functions of these membranes.

The influence of strain on the optical properties of the liquid crystalline state of retina is also studied in Fudan University.

In Wuhan University, liquid crystalline states are found in chickens during their embryonic development, <sup>64</sup> and similarly in some fishes. <sup>62,63,83</sup> There is also interest in the connection between the malignant transformation of the cell and the liquid crystalline phase transitions in biomolecular lipid layer of membranes. <sup>8</sup>

The liquid crystalline properties of water solutions of DNA of fish sperm are studied jointly by the Institute of Chemistry and the Institute of Biophysics. The methods of small angle light scattering, polarizing microscopy, viscometry and ultraviolet spectroscopy are used. Liquid crystal phase is found to appear when the concentration of DNA is greater than 1%, which is related to the double-helix structure of the molecules.

#### **Physics**

In China, research in the physics of liquid crystal did not really begin until 1978. As experiments are concerned, in Beijing University, magnetic birefringence and order parameter of nematic MBBA/nonmesogen binary systems are measured.<sup>28</sup> There is also a study on the internal field parameters for liquid crystals.<sup>29</sup>

In the Institute of Physics, the nonlinear optical four-wave mixing effect was observed for the first time in the isotropic phase of MBBA near the clearing point.<sup>33,44</sup> Relaxation of molecular orientation was also studied.

In a collaboration between Qinghua University and the East China Institute of Chemistry Technology the average viscosity coefficient was measured using a vibrating sphere in small sample of liquid crystal. Recently, in Qinghua University, propagation of director waves in nematics under a mechanical perturbation was observed;<sup>95</sup> there was also an attempt to measure the flexoelectric constants of nematics using a

forced vibrating liquid crystal cell.<sup>88</sup> In addition, there were experimental and theoretical studies on the temperature dependence of the pitch of cholesterics,<sup>85</sup> the pretilt angle of nematics,<sup>86</sup> frequency dependence of dielectric constants,<sup>87</sup> and the influence of high electric electric field on the elastic constants of nematics.<sup>42,84</sup>

In Chiao-Tung University of Taiwan Province, the molecular twisting power of cholesteryl propianate, nonanoate and myristate were measured. For the mixture with smectic mesophase the temperature coefficient of the pitch is anomalous.<sup>31,93</sup>

Most of the theoretical research is carried out in the Institute of Physics. It may be roughly divided into eight parts.

(1) Pretransitional effects—Pretransitional effects in the isotropic liquid phase near the nematic-isotropic (N-I) transition are studied.<sup>43</sup> Correlation functions and the gap exponent are calculated microscopically beyond the mean-field approximation.<sup>46,48</sup> The deviation of the inverse susceptibility from a straight line, an experimental fact observed in 1970 and many times again in recent years in different countries, is explained accordingly. This theory is now generalized (in collaboration with Beijing University) to nematic mixtures with success.<sup>18</sup> For these calculations, the rigorous formula of Suzuki for correlation functions of classical systems and Ising model is generalized.<sup>22,41</sup> Its use in the calculation of order parameters is also demonstrated.<sup>22</sup>

More recently, starting from a modified Kobayashi-McMillan hamiltonian, correlation functions are calculated taking into account the fluctuations of both the nematogenic and the smectogenic order parameters. 81,90 It explains correctly the trend of magnetic birefringence measurements in the homologous series on nCB.

A tentative explanation<sup>20</sup> is offered for the existence of inflexion points observed in the inverse light scattering intensity curve in the isotropic phase of 9CB and 11CB.

(2) Nature of the N-I transition—The contradictions of the tricritical point hypothesis regarding the N-I transition with experimental results are pointed out.<sup>38</sup> Many of the exponents measured experimentally are shown to be apparent exponents only.<sup>17,39,47</sup> For this purpose, three kinds of exponents, viz., critical, apparent and effective, are carefully defined and distinguished.<sup>17</sup> It is shown that the apparent exponents measured on both sides of the N-I (first order) transition can be explained by taking fluctuations of the order parameter into account.<sup>19,25,37</sup> The first order transition model of Landau-deGennes is investigated in the Gaussian approximation; similarities and differences between first and second order phase transitions are compared and

emphasized. For example, it is noted that scaling laws from second order transitions cannot be simply applied in first order transitions.<sup>25,37</sup>

Asymmetry of critical and apparent exponents of correlation lengths on the two sides of the clearing point are predicted<sup>48,47</sup> and subsequently confirmed by NMR experiments abroad. More exponents are then calculated and experiments to confirm them are proposed.<sup>17</sup> Also, more accurate calculation of the gap exponent in better agreement with experiment is carried out.<sup>90</sup>

- (3) Phase diagrams—The influence of molecular length on the shape of isotropic-nematic-smectic A phase diagram is studied. 90 Existing phase diagrams are explained 21 (in collaboration with Qinghua University) and new ones predicted. The stepwise increase of length and width of molecule with the number of carbon atoms in the end chain in a homologous series is proposed as the origin of the odd-even effect (in contrast to the theory of Marcelja). Odd-even effects of phase transition temperature, supercooling temperature and order parameter are calculated and agree well with experiments. 90
- (4) Pressure effects and reentrant phenomena—The Landau-deGennes theory is generalized to explain, in a unified way, all the existing pressure experiments of nematic PAA. Reentrant isotropic phase and other results are predicted. Equation of state shows interesting novel features. For example, in contrary to the case of simple liquids, the intersection of the supercooling and superheating curves does not coincide with that of the two coexistence curves. 82,89,91,92
- (5) Elasticity and flexoelectricity—A simple derivation of the Oseen-Frank elastic constants is given.<sup>36</sup> This method is further used to obtain a macroscopic expression for the quadrupole flexoelectricity (introduced microscopically by Prost and Marcerou) in nematics and cholesterics. Similarly, influence of high electric field on the elastic constants are also derived macroscopically (in collaboration with Qinghua University).
- (6) Viscosity and instability—Using the dissipation function formalism nonlinear viscous stress tensor is derived. The one dimensional theory of Dubois-Violette, deGennes and Parodi is extended to treat the case of planar nematics under the action of an inclined electric field. For MBBA, electrohydrodynamic instability occurs only when the angle between the field and the normal of the liquid crystal cell is less than a critical value. 80
- (7) Soliton propagation—Nematic under uniform shear is shown to be capable of carrying solitons.<sup>24,96</sup> Properties of these solitons and their

appearence under monochromatic or white light are studied. Some recent experiments are interpreted accordingly. Solitons are also generated experimentally (in collaboration with Qinghua University) by applying pressure gradient along the liquid crystal cell.

(8) Mesophases of bowl-like molecules—Three dimensional bowl-like molecules (which break the up and down symmetry in contrast to the case of disc-like molecules) are predicted to be capable of forming liquid crystal phases. A phase intermediate between the nematic (all bowls in one direction) and columnar (bowls stack on each other as in disc-like case) should exist. The intermediate phase is analogous to the micellar phase (between the ideal solution and the hexagonal phase) in soap-water system and may also exist in the case of disc-like molecules. If exist, these mesophases of bowl-like molecules are very likely to be ferroelectrics.

In the past few years, we saw many fruitful collaborations between scientists from China and foreign countries. Molecular theory of the N-I transition using the orientation-averaged-pair-correlation approach is studied<sup>26,27</sup> (Institute of Physics/University of California at San Diego); four-wave mixing and optical-field-induced helical structure in liquid crystals are investigated theoretically<sup>32</sup> (Institute of Physics/University of California at Berkeley); a unified theory of NMR and dielectric relaxations of nematics in the isotropic phase is proposed and compared with experiments (Institute of Physics/Naval Research Laboratory). Experimentally, propagating shear waves and melting behavior in smectics, <sup>34,35,45</sup> flexoelectric induced second-harmonic generation in nematics<sup>49</sup> (Institute of Physics/Northwestern University), and refractive index measurements using the surface plasmon technique<sup>30</sup> (Qinghua University/University of California at Berkeley) are studied. All these experiments are performed abroad.

#### 3. IN SHORT . . .

The research in liquid crystals in our country in the past twelve years is not unsuccessful. On the other hand, in spite of the wide range of topics studied there are still quite a number of spots untouched (e.g., ferroelectric smectics, mesophases of disc-like molecules, etc.). The efforts in lyotropic systems are relatively limited. In the future, we hope that there will be more creative work in both basic research and application studies. In short, since 1970, a team of scientists and workers of some size and level of expertise in the field of liquid crystals has been gradu-

ally built up. In recent years, the progress is more noticeable. In order to promote scientific exchange both within and without China and to push forward the development of this interdisciplinary field of research in our country, the Chinese Liquid Crystal Society† was founded on July 18, 1980 in Beijing.

The International Conference on Liquid Crystals held in December, 1979 at Bangalore, India is recorded as the first one ever attended by Chinese liquid crystalists, nine years after the beginning of research in this field in our country.

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Even though the research in liquid crystals in China was started more than a decade ago most of the research results were not published until 1978. They are scattered in different journals. A partial list of the published papers are given below, which is not meant to be complete but to give the readers an idea about the scope and areas of interest in the research activities in this field. Those marked by an asterisk are in English; otherwise, in Chinese.

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