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NONLINEAR DYNAMICAL PHENOMENA IN LIQUID CRYSTALS *

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I. Introduction

In 1888, one hundred years ago, liquid crystals were first discovered by Reinitzer. As mesophases between liquids and crystals, liquid crystals share the properties of liquids and crystals. This unique quality makes liquid crystals delicate and excellent nonlinear systems. Concretely speaking, (i) liquid crystals are usually organic fluid; the interaction between the molecules is not so strong as it is in crystals; this means that liquid crystals are soft systems on a macroscopic level; (ii) liquid crystals show the long range orientational order so the Frank free energy and the related static and dynamical equation for the director field are substantially nonlinear. The nonlinearities in liquid crystals are of particular interest and importance in physics and biology. So far typical nonlinear phenomena in liquid crystals, such as solitary wave, transient periodic structure, chaos, fractal and viscous fingering, have received close attention. On the one hand, these phenomena bear analogy with those in liquids and crystals; on the other hand, the phenomena show special features which extend and deepen fundamental understanding of the nonlinearities in physical systems.

In this article, we would like to present some nonlinear problems in liquid crystals, which we are concerned with in the past few years.

II. Solitary Waves in Liquid Crystals

A fascinating nonlinear structure in liquid crystals is the wall appearing at the Freedericksz transition. In my recent work, such a nonlinear structure was called the Brochard-Leger[1,2] wall (B-L wall). The relevant nonlinear diffusion equation for describing the dynamical behaviors of the B-L wall was established and a series of theoretical results were obtained. It is pointed out that the B-L wall shows a typical solitary wave in liquid crystals and presents an excellent example of the nonequilibrium phase transition[3]. The scenario of our theoretical approach can be presented as follows:

The starting point is the two-dimensional nonlinear diffusion equation with a boundary condition

$$\gamma_1 \frac{\partial\theta}{\partial t} - k\left(\frac{\partial^2\theta}{\partial x_1^2} + \frac{\partial^2\theta}{\partial x_3^2}\right) = \frac{1}{2}\chi_a H^2 \sin[2(\theta-\phi)]$$

$$\theta(x_1,x_3,t)\Big|_{x_3=\pm\frac{d}{2}} = 0 \qquad (1)$$

Eq.(1) describes the dynamical behavior of director field in nematic liquid crystals(Note that the word nematic comes from "nema",the ancient Greek word for thread. From symmetry point of view,nematics is the simplest liquid crystals.)Here one constant approximation is made.Our aim is to obtain a simple and relevant model for describing the B-L wall. Consider a fine thin layer in the middle of nematic slab.In a normal Freedericksz transition the deformation of the director field may be described by the static equation

$$k\frac{\partial^2\theta}{\partial x_3^2} + \frac{1}{2}\chi_a H^2 \sin(2\theta) = 0 \qquad (2)$$

The nontrivial solution of Eq.(2) is an elliptic function $\theta_0(x_3)$ which gets its maximum value in the middle thin layer of the nematic slab.We consider the solution of Eq.(1) that reads

$$\theta(x_1,x_3,t)=\theta_1(x_1,x_3,t)\ \theta_0(x_3) \qquad (3)$$

Physically we think $\theta_1(x_1,x_3,t)$ is a slow function of x_3 and it may be considered as a modulation function with respect to the known $\theta_0(x_3)$ in the x_1 dimension and in time t. Basing upon this consideration, the required model equation is obtained

$$\frac{\partial\tilde{\theta}}{\partial\tau} - \frac{\partial^2\tilde{\theta}}{\partial X^2} = F(\tilde{\theta}) \qquad (4)$$

where

$$F(\tilde{\theta})= [\sin(2\tilde{\theta}\theta_M)-\tilde{\theta}\sin(2\theta_M)-\varepsilon]/\theta_M^3 \qquad (5)$$

For the definitions of the quantities in the above equation, the reader is referred to Ref.3. Note that Eq. (4) is a one dimensional nonlinear diffusion equation which has the same structure as that of the famous Huxley equation for describing nerve propagation in neurophysics. By expanding the nonlinear function $F(\theta)$ in a power series to the third order, Eq.(4) is indeed the Huxley equation. Such nonlinear diffusion equation was systematically investigated by Aronson and Weinberger in 1975-1979. Adopting their remarkable mathematical results we obtained the corresponding theoretical results presented in in Ref.(3),which are in good agreement with the Leger's experiment and our recent experimental data[4].

The reliability of the assumption that $\theta_1(x_1,x_3,t)$ is a slow function of x_3 is a crucial point for the validity of our scenario. Here we would like to explain this point. By the definition

$$\theta_1(x_1,x_3,t)=\frac{\theta(x_1,x_3,t)}{\theta_0(x_3)} \qquad (6)$$

and

$$\theta_1(x_1,-d/2,t)=\frac{\theta(x_1,-d/2,t)}{\theta_0(-d/2)}=\frac{0}{0} \qquad (7)$$

Therefore the value $\theta_1(x,-d/2,t)$ should be determined by

$$\theta_1(x_1,-d/2,t)=\lim_{x_3\to -d/2}\frac{\theta(x_1,x_3,t)}{\theta_0(x_3)}=\frac{\left.\frac{d(\theta(x_1,x_3,t))}{dx_3}\right|_{x_3=-\frac{d}{2}}}{\left.\frac{d\theta_0(x_3)}{dx_3}\right|_{x_3=-\frac{d}{2}}} \qquad (8)$$

From Eq.(2)one obtains

$$\left.\frac{d\theta_0(x_3)}{dx_3}\right|_{x_3=-\frac{d}{2}}=\sqrt{\frac{\chi_a}{k}}\,H\sin\theta_M . \qquad \theta_M\equiv\theta_0(0)>0 \qquad (9)$$

The value of $d\theta/dx_3$ is determined by Eq.(1).Here it is worth to note that the directors on the boundary are always in a static state,because of the strong anchoring.So the variation of the directors on the boundary is independent of x_1 and t. Therefore $d\theta/dx_3$ is simply determined by the static equation

$$-k\frac{d^2\theta}{dx_3^2}=\frac{1}{2}\chi_a H^2\sin[2(\theta-\varphi)]$$

$$\left.\theta(x_1,x_3,t)\right|_{x_3=\pm\frac{d}{2}}=0 \qquad (10)$$

One obtains

$$\left.\frac{d\theta(x_1,x_3,t)}{dx_3}\right|_{x_3=-\frac{d}{2}}=\sqrt{\frac{\chi_a}{k}}\,H\sqrt{\sin\theta'_M\sin(\theta'_M-2\phi)} \qquad (11)$$

$$\theta'_M\equiv\theta'(0)>0, \qquad \phi<0$$

So that we get from (9) and (11)

$$\theta_1(x_1,-\frac{d}{2},t)=\frac{\sqrt{\sin\theta'_M\sin(\theta'_M-2\phi)}}{\sin\theta_M} \qquad (12)$$

Furthermore ,noting $|\phi|\ll\theta'_M$,we should have $\theta'_M\approx\theta_M$.

So we get

$$\theta_1(x_1,-\frac{d}{2},t)\approx\sqrt{\frac{\sin(\theta_M-2\phi)}{\sin\theta_M}}\approx 1-\phi/\theta_M \qquad (13)$$

On the other hand,the physical picture of the problem shows

$$-\theta_0(0)\lesssim\theta(x_1,0,t)\leq\theta_0(0) \qquad (13)$$

So

$$-1\lesssim\theta_1(x_1,0,t)\equiv\frac{\theta(x_1,0,t)}{\theta_0(0)}\lesssim 1 \qquad (15)$$

The experiment tells us that the width of the B-L wall is approximately equal to the thickness of the sample.So we have the following estimations:

$$\left|\frac{\partial\theta_1}{\partial x_1}\right|\approx\frac{\theta_1(-\frac{d}{2},0,t)-\theta_1(\frac{d}{2},0,t)}{d}\approx\frac{2}{d} \qquad (16)$$

$$\left|\frac{\partial\theta_1}{\partial x_3}\right| \approx \frac{\theta_1(x_1,-\frac{d}{2},t)-\theta_1(x_1,0,t)}{\frac{d}{2}} \approx \frac{(1-\phi/\theta_M)-1}{\frac{d}{2}} \tag{17}$$

Therefore we get

$$\left|\frac{\partial\theta_1}{\partial x_3}\right| \ll \left|\frac{\partial\theta_1}{\partial x_1}\right| \quad , \quad \left|\frac{\partial^2\theta_1}{\partial x_3^2}\right| \ll \left|\frac{\partial^2\theta_1}{\partial x_1^2}\right| \tag{18}$$

We see $\theta_1(x_1, x_3, t)$ does may be considered as a slow function of x_3. In fact, from intuitive physical opinion, the functions $\theta_0(x_3)$ and $\theta(x_1, x_3, t)$ have nearly same variation behavior in x_3 direction. As a ratio between $\theta_0(x_3)$ and $\theta(x_1, x_3, t)$, $\theta_1 = \theta/\theta_0$ is a slow function of x_3 is a quite natural and clear assumption.

The collision between the solitary waves is an attractive phenomenon in nonlinear science. Recently we have observed such a phenomenon in liquid crystals. Here photographs (a)and (b) in Fig.1 show this collision process in the B-L wall. We see, after the collision, the B-L walls collapse and disappear in the sample. The wall does not show the soliton characters. This is a quite natural experimental result, because the system considered here is a strong dissipative dynamical system. We point out that this collision process can be described by our Eq.(4). Fig.(2) shows the simulation results of the Eq.(4) in a computer, which coincides with the experimental phenomena. Fig.(3) shows the physical picture of the collision. In the process of collision, the directors at the high energy state continuously go down to the low energy state and lose its kinetic energy via dissipative process

Recently, an interesting progress in understanding physical content and mathematical structure of the solitary wave in liquid crystals has been made. Usually, the solitary wave in liquid crystals shows moving wall where a stable state propagates into a region of space occupied by a unstable or metastable state. The physics of such a propagation in dissipative system is different from that of soliton in a hamiltonian system In order to determin the asymptotic velocity of solitary wave in these system, some dynamical selection mechanism must be introduced. In 1975 in their classical mathematical work[5] Aronson and Weinberger pointed out that for the Fisher equation

$$\frac{\partial u}{\partial t} - \frac{\partial^2 u}{\partial x^2} = f(u)$$

$$f(u) \in C^1(0,1), \quad f(u) > 0, \forall u \in (0,1), \quad f(0) = f(1) = 0, \quad f'(0) > 0, \quad f'(1) < 0.$$

the asymptotic velocity should be the slowest one, which can be estimated by

$$2\sqrt{f'(0)} \le C^* \le 2\sqrt{L}, \qquad L \equiv \sup_{0 \le u < 1} \frac{f(u)}{u} \tag{19}$$

The conclusion by Aronson and Weinberger is profound in physics which reveals that there does exist a selection principle in such a dissipative system. Nowadays this principle is called the marginal mechanism, which is originally proposed by the theory of crystal growth. In 1982 we first use this beautiful conclusion to discuss wall propagation phenomena in liquid crystals.[6]

Inspired by the recent progress in physics of liquid crystals and applied mathematics, Cladis and Saarloos have systematically investigated wall propagation in liquid crystals and the related selection mechanism.[7,8] Saarloos pointed out a more general expression for the marginal velocity. We think this is a recent remarkable progress in this aspect.

III. Analogy With Nerve Propagation

The relationship between liquid crystals and living phenomena is a very attractive and important problem in physics of liquid crystals and biological physics. In our former work,[2,3] the analogy between the wall propagation in liquid crystals and nerve propagation in nerve fibers was revealed. Because biomembranes, nerve fibers, parts of the brain are in the liquid crystal state, this analogy is impressive.

Here we would like to address the same point in the smectic ripple phase. The smectic ripple phase exists in hydrated lipid bilayers. This kind of liquid crystals are more closely related with the living phenomena. Carlson and Sethna pointed out that many of the important features of the smectic ripple phase may be described by a frustrated ϕ^4 theory and the minimum energy configuration shows a soliton-like structure.[9] Adopting their expression of the free energy and using time-dependent Ginzburg-Landau model

$$f[\theta(x,t)] = \frac{1}{\lambda}\int_{-\lambda_1}^{\lambda_2}\left[\frac{1}{2}m\theta^2+\frac{1}{4}\theta^4+\frac{1}{2}(d\theta/dx - d\theta_0/dx)\right]dx \tag{20}$$

$$\frac{\partial}{\partial t}\theta(x,t) = -\frac{1}{\gamma_1}\frac{\delta F[\theta(x,t)]}{\delta\theta(x,t)} \tag{21}$$

where γ_1 is the viscosity, we obtain the nonlinear diffusion equation

$$\gamma_1\frac{\partial\theta}{\partial t} - \frac{\partial^2\theta}{\partial x^2} = m\theta + \theta^3 \tag{22}$$

Once again we have seen the similarity between the collective behavior (wall motion) in biological liquid crystals and nerve propagation. In the investigation of the wall in smectic C^* the significance of such a similarity has been noticed.[7] We believe that this similarity is a meaningful fact in physics of liquid crystals and biophysics. In some cases the nonlinear behaviors in liquid crystals can simulate nerve propagation and other biological phenomena. The favorable point of this simulation is that because of the birefringence of liquid crystals, the corresponding phenomena can be observed in real time and space.

IV. Chaos

Liquid crystals are sensitivity nonlinear systems. A small external field can make liquid crystals unstable at a macroscopic level, because of the anisotropic structures and high nonlinear coefficients. The Freedericksz transition in liquid crystals is of fundamental significance. Here we mention chaotic motion in the dynamics of the Freedericksz transition. In the pure twist cases, the motion of the directors in nematics is described by

$$\gamma_1\frac{\partial\theta}{\partial t} - k\frac{\partial^2\theta}{\partial x^2} = \frac{1}{2}\chi_a H^2\sin(2\theta) \tag{23}$$

When a liquid crystal film is subjected to an external field

$H=H_0+\tilde{h}$, $\tilde{h}=h_0\cos\omega t$ and the driving frequency $\omega \sim \tau^{-1} \sim \tau_0^{-1}$, $\tau_0^{-1}=\pi^2 k/(\gamma_1 d^2)$. In this case the directors can follow the changes of the external field and the chaotoc motion of the directors will take place. Such a chaotic motion is due to the competition between the different frequencies.

In order to establish a relevant model to describe the chaotic motion of the directors ,we express the solution of the equation (23) in the fourier series

$$\theta(z,t)=\sum_{m=1}^{\infty}\theta_m(t)\cos\left(\frac{2m-1}{d}\pi z\right) \qquad (24)$$

For the present purpose we need to take the two modes of (24)

$$\theta(z,t)=\theta_1(t)\cos\left(\frac{\pi}{d}z\right)+\theta_2(t)\cos\left(\frac{3\pi}{d}z\right) \qquad (25)$$

Substituting (25) into Eq.(23) ,to the third order, one obtains the following couple ordinary equations

$$\frac{d\theta_1}{d\tau}=\frac{\chi_a H^2}{\gamma_1}\left[\left(1-\frac{H_c^2}{H^2}\right)\theta_1-\theta_1^3-\theta_1^2\theta_2-2\theta_1\theta_2^2\right]$$
$$\frac{d\theta_2}{dt}=\frac{\chi_a H^2}{\gamma_1}\left[\left(1-9\frac{H_c^2}{H^2}\right)\theta_2-\frac{1}{3}\theta_1^3-\theta_2^3-2\theta_1^2\theta_2\right] \qquad (26)$$

The above equation may present chaotic solutions in a computer. It is easy to see that the deduction of equation (26) is analogous to that of Lorentz equation.

In the case of the optical Freedericksz transition,one can modulate the intensity of the laser beam and require the modulation frequency $\omega \sim \tau^{-1}$,so the chaotic motion of the directors is also possible .In this situation,the output of the optical beam will show a complicated behavior.This is because of the chaotic motion of the directors in the sample.[10] Starting from the equation for the optical Freedericksz transition one can also easily obtain the similar ordinary equation to describe the chaos dynamics of the directors in the optical field.The similar phenomena may also appear in the cholesterics and smectics.This interesting phenomenon forms the "director chaos" .Recently such a chaotic motion has been observed in our experiments.

V. Dendritic Formation

A fascinating ohenomenon in crystal growth is dendrite formation.Dendrites growing from a supercooled melt or supersaturated solution display beautiful symmetry and fine structure distribution.The phenomenon has raised challenging nonlinear problems of physics and mathematics. Nowadays it has been recognized that surface tension acts as a singular perturbation in dendritic growth .In the last few years,although considerable progress has been made in understanding this graceful nonlinear structure.yet there remain some very important unsolved questions.Noticing the nature of liquid crystals and inspired by the recent progress in this research line,we put a question:whether a process similar to the to the dendritic evolution in crystal growth can take place in liquid crystal growth? Clearly if such a process happens in a liquid crystal , the corresponding phenomena may give out very useful information on the dendritic formation from another angle. It is easy to guess that proper candidates which are to present dendritic behavior should be discotic liquid crystals. From X-ray diffraction the discotic phases exhibit a two- dimensional long range order, which appear to consist of an array of columns parallel to each other and the axes of which are regularly positioned on a two- dimensional hexagonal or rectangular lattice. Clearly such structural characters in favor to the existence of the dendritic liquid crystals. Basing upon this motivation, we have carried out an interesting research.[11]

Put a small amount of a thermotropic discotic liquid crystal RHO on a clean glass. At 107°C the RHO shows a very viscous discotic liquid crystal which shows birefringence effect. By raising the temperature, the liquid crystal becomes a clean isotropic liquid at 127.5°C. This RHO isotropic liquid is carefully expanded as a thin film by shearing. The thickness of the film is about 10 μm. The RHO fluid wets the base material By decreasing the temperature of the sample, the RHO liquid is in supercooled state at a definite temperature, which tends to transform the RHO into a more stable discotic phase. In this case , the nucleation transition is to take place in the sample. In the microscope we have directly observed many beautiful dendrites formed by RHO liquid crystal. Here

photographs (a) and (b) in Fig.4, which are taken by the automatic micrograph , show the images of the two formed dendrites respectively. The dendrite (a) is obtained at a cooling rate of 1.5°C/min. The growth time is about 90 seconds. The dendrite (b) is obtained at 10°C/min. The growth time is about 50 seconds. The dendrite (a) shows the sixfold symmetry structure with elegant sidebranches. The dendrite (b) presents a pine -leaf pattern which shows interesting self-similarity structures reminiscent of fractals. Note that there are only five main branches in the pattern (b). Clearly, the other one is screened.

We should make the following notions : (i) in a supercooled RHO liquid which has not been expanded as a thin film, one cannot observe dendrites; this fact shows that the pre-alignment order in the surface induced by shearing is important for the dendritic formatio; (ii) if the thickness of the film is too small (<3μm) or too large (>40μm), no dendrites appear in the sample; in a free suspended film, the dendrite does not exist.; it reflects that there is a critical thickness d of the film; the influence of the basal wall on the dendrite formation is of significance; (iii) there coexist dendrites and other fan-shaped defects in the sample.

How does the pattern emerge from the supercooled liquid and evolve into a complicated dendrite? How does the growth speed of the dendrites depend on the cooling rate? Our further investigation is concerned with these dynamic problem. We have carefully observed the total process of the dendritic formation. Here the photographs in Fig.5 show such a growing process.

At the beginning t=0 a nucleus stochastically appear in the sample,which evolves very quickly into a beautiful rose pattern with sixfold symmetry.The photograph (a) illustrates this pattern.The radius of the pattern is 12 um.The macroscopic sixfold symmetry of the pattern just corresponds with the microscopic symmetry of the hexagonal discotic phase structure. The growth speed of the tip of the pattern is 1.5 um/sec.At t=40 sec.,the diameter of the rose pattern has increased six times the original size,which is shown by the photograph (c).We think that this photograph is meaningful.Notice that the small seed sidebranches have already appeared randomly in two of the six main branches of the pattern (c). We see that there appears the other pattern which is developing towards the pattern (c).The two branches which the seed sidebranches appear in are just closed to such a new pattern.This phenomenon strongly states the fact that the sidebranches emerging randomly in the main-branches are excited by latent heat disturbance released by the new pattern.The photograph (c) contributes a physical understanding of the interfacial instability. At the end of the process, the pattern has evolved into a very complicated structure which is no longer topologically equivalent to the original pattern.

In order to obtain more information, the time evolution of the phenomenon has been videotaped.From the videotape, we have measured the time dependence of the growth speed of the tip on the cooling rate,which

is shown by Fig.6. Surprisingly,we found that the velocity V of the tip is nearly proportional to the cooling rate η

$$V \propto \eta \qquad (27)$$

This experimental result is in contrast with the current standard theory of dendritic growth,which predict that the tip velocity of a dendrite should be proportional to the supercooling.That is to say, while continuously cooling the liquid ,the tip velocity will be accelerated and not have a constant velocity.[12] To our knowledge,no theory can explain this result at present.

We think the significance of this experiment is twofold First it reveals the beautiful forms of dendritic liquid crystals and shows the influence of the free surface and basal on the plane dendrites.A very interesting quantitative result is obtained.Second,the experiment provides a direct visual evidence to show that the influence of the thermal noise plays an important role in the sidebranching process. In general,the existence of noise can deeply alter the macroscolic behavior of nonlinear nonequilibrium system. From the point of view of physics,the emergence of the side-branches excited by the thermal noise favors thermal diffusion of the interface but increases the energy of the interface. The initial sidebranches compete with each other.The small sidebranches are restrained or swallowed up in the stochastic diffusion field. This process will favor decreasing the interfacial energy. The realistic structure of the dendrites should be determined by interplay between the interfacial energy and thermal diffusion in a fluctuation environment. In a practical process, the selection mechanism seems to be influenced by noise. In a relevant theory on dendrite growth, the noise should substantially enter the related equations.

It also should be emphasized that the phase behavior, transition temperature, and orientational ordering of liquid crystal system can be altered by a free interface or basal wall. The experiment shows that the plane dendritic formation in the RHO liquid crystals is sensitive to the free surface and wall. We believe that the dendritic liquid crystals have raised very attractive nonlinear problems.

Finally we mention that there exist other interesting nonlinear phenomena in the RHO liquid crystals, for example, fractals and bubble growth. The investigation of these nonlinear structures is currently under way.

VI. Summary

Liquid crystals provide an excellent system for nonlinear science. Because of the existence of the orientational order and anisotropy, strong nonlinear phenomena and singular bevaviors can be excited by a very small disturbance, which can be directly observed in the time and space. These phenomena and behaviors are in connection with physics, biology and mathematics. In this sense, liquid crystals are particular suitable for nonlinear researches. We believe that the fascinating phenomena in liquid crystals will attract more and more attention and interest of physicists, biologists mathematicians and others.

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Figure Captions

Fig.1,The collision of the Brochard-Leger walls.

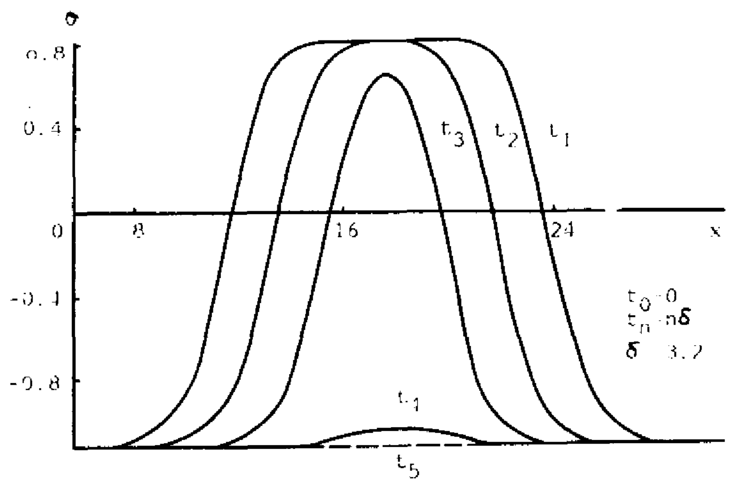
Fig.2,The simulation results of the collision of the B-L wall in a computer.

Fig.3,The physics picture of the collision of the B-L wall.

Fig.4,Dendritic liquid crystals,photograph (a) is obtained at a cooling rate of 1.5^{o}C/min;(b) is obtained at 10^{o}C/min..

Fig.5,The growth and evolution of dendritic liquid crystals.

Fig.6,The time dependence of the tip velocity on the cooling rate.


σ
0.8
0.4
0
-0.4
-0.8
8
16
24
x
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t2
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$t_0=0$
$t_n=n\delta$
$\delta=3.2$

Fig.1

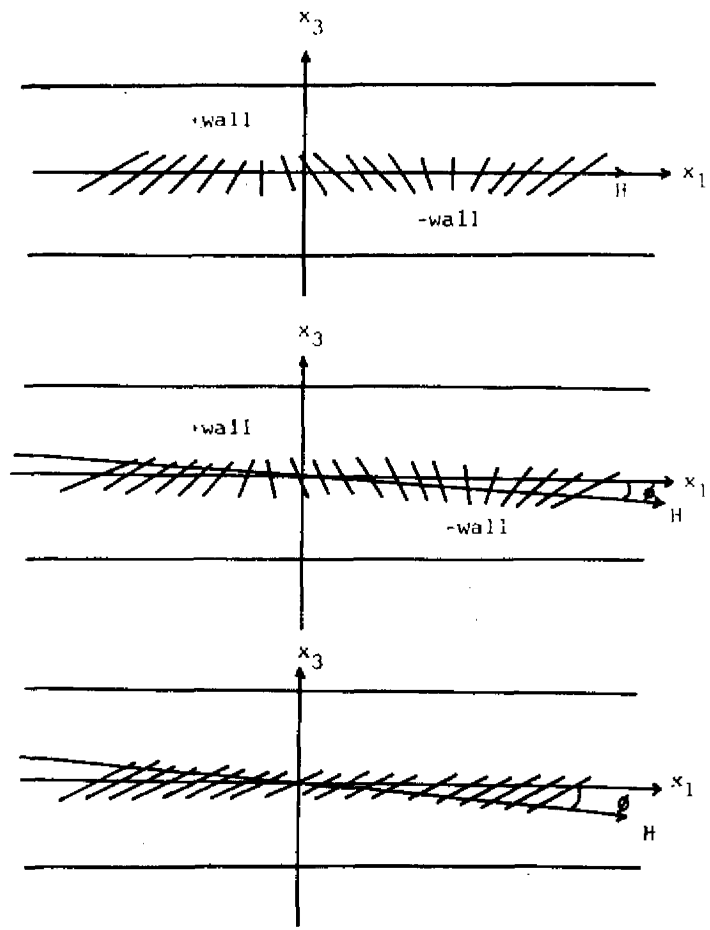
x3
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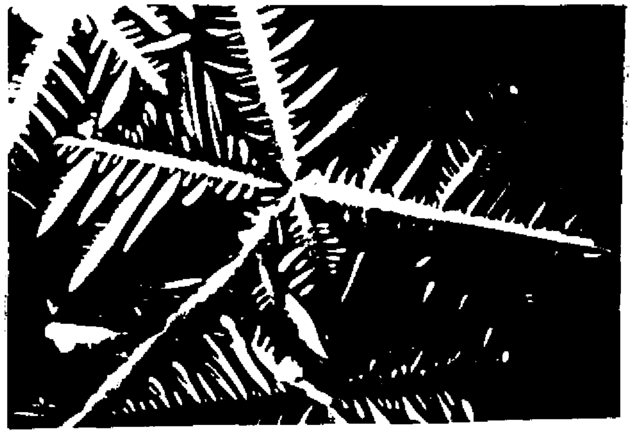
Fig.2

(a)



(b)

Fig.3

(a)

(b)

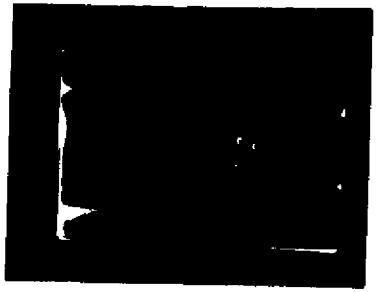
(c)

(d)

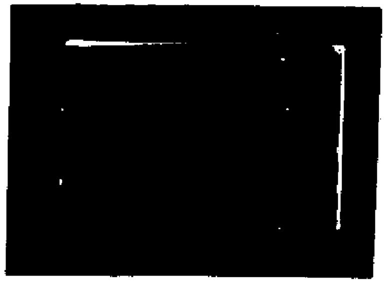
(e)

(f)

Fig.4



(a)



(b)

Fig.5

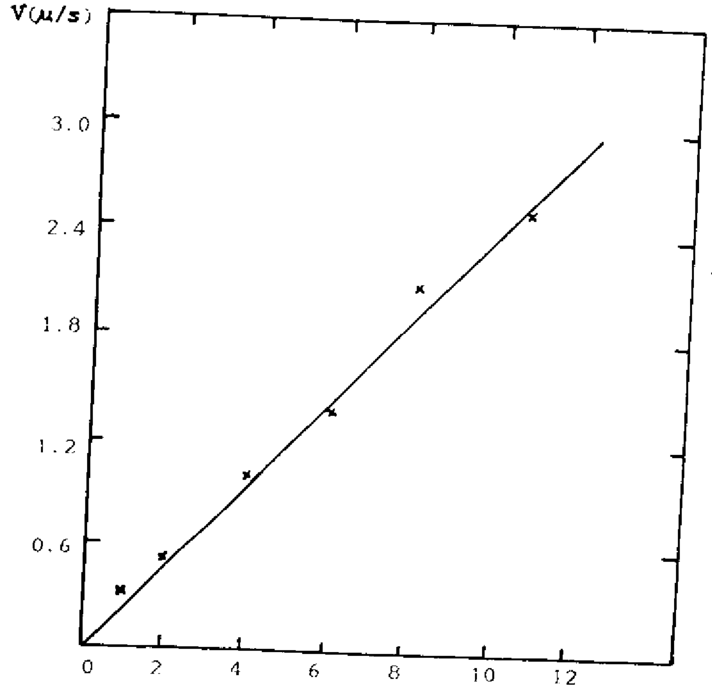


Fig.6

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del Centro Internazionale di Fisica Teorica