Experimental Studies of Some Phase Transitions in Nonequilibrium Open Systems

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Self-organization phenomena in nonequilibrium open systems appear when an external power exceeds some threshold value, so that the onset of the ordered state is regarded as a phase transition similar to that observed in equilibrium systems. The talk will review experimental studies of three kinds of phase transitions in nonequilibrium open systems: onset of self-sustained electric oscillation, evolution of vortex around a sink hole and successive transitions in liquid crystals due to an electric field. The points of experiments are focussed on critical fluctuations, the energy transfer through nonlinearity, the evolution of the ordering parameter, an effect of external noise on self-sustained oscillations and successive transitions to turbulence.

§ 1. Introduction

Most of self-organization phenomena or dissipative structures in nonequilibrium open systems appear beyond the threshold of some controlling parameter which governs the state of the system,[1),2)] so that an occurrence of the ordered state is regarded as a phase transition similar to that observed in equilibrium systems. The general aspect of the phase transition in nonequilibrium systems is that when an external power supplied to the system exceeds a dissipated power due to the loss for a certain mode, a disturbance associated with that mode evolves into a macroscopic motion.

If we consider the onset of self-organization as a phase transition, the following questions may arise: 1) Are critical fluctuations and critical slowing down phenomena observed near threshold? 2) What role does the nonlinearity play in nonequilibrium phase transitions? 3) How does the ordering parameter increase above threshold with controlling parameter? 4) What is the effect of an external noise on the phase transitions? and 5) How do successive transitions turn out to be turbulence?

For these five years our group has been concerned with experimental studies of several nonequilibrium phase transitions from the points of view mentioned above. In this report we present three of them: Onset of self-sustained electric oscillation, Evolution of vortex around a sink hole and Successive transitions in liquid crystals due to an electric field.

§ 2. Onset of self-sustained electric oscillation

Onset of self-sustained oscillations in Esaki diode,[3] Gunn diode,[4] Wien-Bridge oscillator[5),6] and so forth has a close similarity to the phase transition in equilibrium systems. Output of these circuits shows critical fluctuations near threshold and in particular the variance and the relaxation time of the output fluctuations of a Wien-Bridge oscillator whose state is controlled by the feedback factor β have been found to become infinite in a manner like $(\beta_c-\beta)^{-1}$ as β approaches the threshold β_c for the oscillation.[5] The aspect is analogous to the Curie-Weiss law in the second kind phase transition.

The Weiss field in spin systems may be regarded as due to a spacial feed-back process where each spin exerts influence on its surroundings and is simultaneously subjected to their backward effect which is proportional to the source itself. The aspect is the same in the oscillatory systems in which the output from an amplifier comes back through a feedback circuit. Hence the concept of the Weiss field approximation can be expanded to the temporal feed-back process where the source is subjected to a backward effect proportional to the source itself.

Another subject of significance in stochastic problems of electric oscillation is the influence of external noise on the transition.[7] An external noise applied to an oscillator, in general, gives rise to amplitude and phase fluctuations. In case of parametric oscillator, however, in which the phase is locked by the pumping oscillator, the external noise gives rise to only amplitude fluctuations. Figure 1 shows a block diagram of a parametric oscillator composed of two ferrite toroidals. A pumping current is applied to the primary circuit and its amplitude J exceeds some threshold, then an oscillation at half the pumping frequency is excited in the secondary circuit. We additionally supplied a white noise in parallel to the pumping oscillator and examined what effect appears in

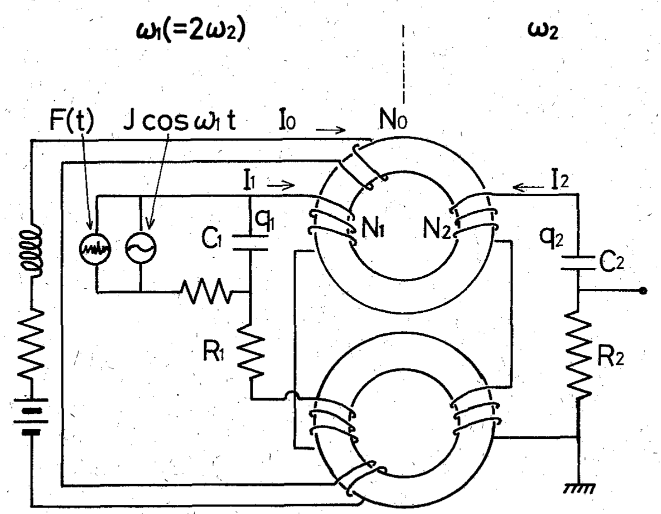

Fig. 1. Schematic block diagram of a parametric oscillator.

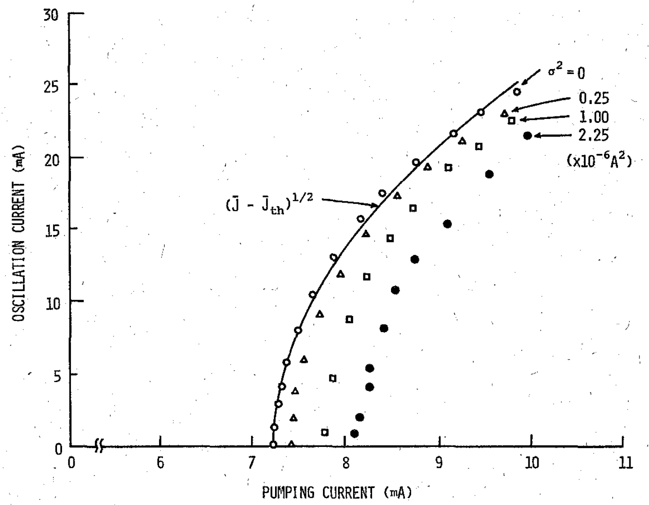

Fig. 2. Oscillation current against pumping current. σ^2 is the intensity of the external noise.

the secondary circuit oscillation.

Figure 2 shows the oscillation current vs the pumping current curve. The open circles represent the noiseless case and the threshold is found to shift to higher values when the external noise is added. Here σ^2 is the noise intensity. If we plot the threshold pumping current against the external noise intensity, we get Fig. 3. It is noticeable that the threshold changes linearly with noise intensity. This also represents a phase diagram of oscillatory and non-oscillatory regions for parametric oscillation.

One may ask a question why the threshold value shifts with external noise. Before our experimental studies, Horsthemke and Malek-Mansour[8] theoretically predicted this behavior on a chemical reaction model. The following treatment is similar to their theories. After some mathematical manipulation taking account of our specific electrical circuit, we get a stochastic equa-

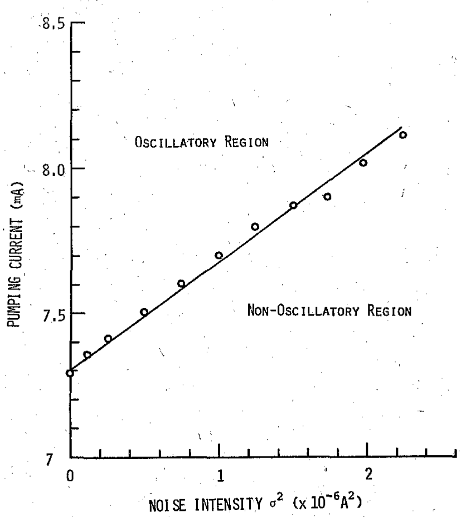

Fig. 3. A change of the threshold pumping current with the external noise.

tion for the amplitude of oscillation current,

$$\frac{db}{dt}=(\alpha-\gamma)b-\beta b^3+bR(t). \tag{1}$$

Here α is proportional to the pumping current, γ comes from the resistance R_1 and R_2 in Fig. 1, β comes from the nonlinearity of ferrite and $R(t)$ is the external noise which is assumed to be white:

$$\langle R(t)R(t')\rangle=2D\delta(t-t'). \tag{2}$$

The crucial point of Eq. (1) is that the random force $R(t)$ is multiplied by the stochastic variable b itself, so that we have to take into account the correlation between b and $R(t)$, in order to derive a Fokker-Planck equation for the probability density from (1). Stratonovich[9] has given a universal method of deriving the Fokker-Planck equation from a general form of stochastic equation, but here we use a conventional method which is only available for our special case. Dividing both sides of Eq. (1) by b, we get

$$\frac{d}{dt}\log b=(\alpha-\gamma)-\beta b^2+R(t), \tag{3}$$

which is rewritten by introducing a new variable

$$x=\log b \tag{4}$$

in the form,

$$\frac{dx}{dt}=\alpha-\gamma-\beta e^{2x}+R(t). \tag{5}$$

Then we can easily get a Fokker-Planck equation for the probability density of x in the form,

$$\frac{\partial\tilde{P}(x)}{\partial t}=-\frac{\partial}{\partial x}[(\alpha-\gamma-\beta e^{2x})\tilde{P}(x)]+D\frac{\partial^2\tilde{P}(x)}{\partial x^2}. \tag{6}$$

From this, we go back to the probability density of the original variable b, using the following relations:

$$\tilde{P}(x)=bP(b) \tag{7}$$

and

$$\frac{\partial}{\partial x}=b\frac{\partial}{\partial b}. \tag{8}$$

Thus we can finally get a Fokker-Planck equation for the probability density of b,

$$\frac{\partial P(b)}{\partial t} = -\frac{\partial}{\partial b}[\{\alpha-(\gamma+D)-\beta b^2\}bP(b)]+D\frac{\partial}{\partial b}b^2\frac{\partial}{\partial b}P(b). \tag{9}$$

Here we are confronted with a problem: Which of the average value $\langle b \rangle$ and the most probable value b_m should we choose as the ordering parameter in the presence of fluctuations? We have no idea of which one we should take, but at least our experimental results shown in Figs. 2 and 3 can be explained using the most probable value b_m. Equation (9) is a convenient form to check the most probable value, because the drift term in (9) gives rise to the most probable value in the stationary state; that is, $b_m = \{(\alpha-\gamma-D)/\beta\}^{1/2}$. Hence we can describe the effect of external noise of the present type: While without external noise, the threshold value of α is γ but in the presence of external noise, it is shifted to $\gamma+D$. In other words, the effective damping coefficient increases linearly with D. That is the reason why the transition point is shifted by the external noise.

We have another experimental evidence for the increase of the effective damping coefficient. Figure 4 shows oscilloscope traces of transient process when the pumping power was rapidly decreased. The upper trace shows the case of no external noise and the lower one shows the case where the external noise is supplied. One can see clearly that the damping is faster when the external noise is supplied.

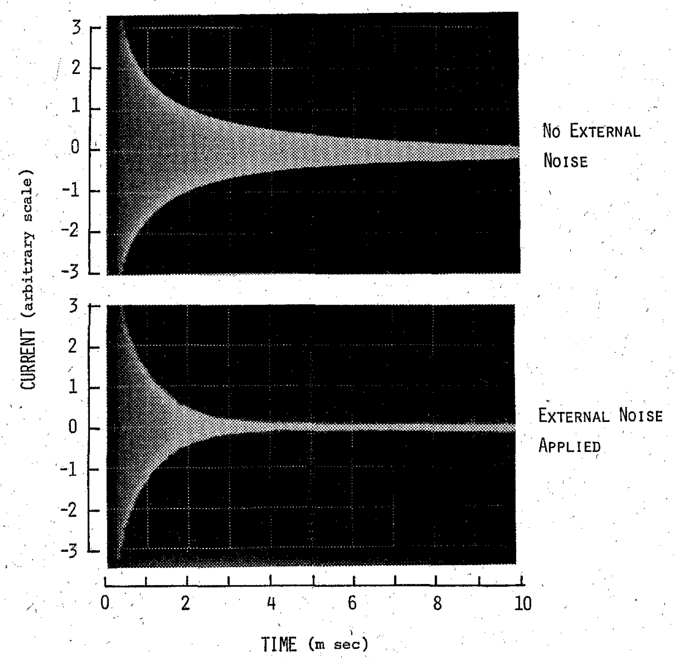

Fig. 4. Oscilloscope traces of decay process in the absence and the presence of external noise. In both cases, the initial pumping current $\bar{J}_i = 9.25$ mA and the final one $\bar{J}_f = 7.20$ mA.

§ 3. Evolution of vortex around a sink hole

It is empirically well known that a fluid flowing centripetally toward a sink hole in the bottom of a vessel has a tendency to form a spiral vortex around it. Large scale vortices of the same kind in meteorology are tornadoes, cyclones and typhoons which are produced by a constant supply of energy from strong local convection. Whether a vortex is produced or not, seems to depend on the amount of discharge from the sink hole, i.e., the vortex is not generated for a small amount of discharge and generated only when the discharge exceeds a certain threshold value.

In order to confirm this threshold bahavior quantitatively, we have carried out measurements of the velocity of vortex flow produced in the presence of a centripetal flow toward a sink hole (8mmϕ) in a vessel, using the method of laser-Doppler velocimetry. Figure 5 shows the velocity of the vortex v_θ at a distance of 4 cm from the center of the sink hole plotted against the amount of discharge Q. The curve indicates that there is a threshold value Q_c for the generation of the vortex. If we take $Q_c=73.5$cc/sec and replot v_θ against $Q-Q_c$ on a logarithmic scale as shown in Fig. 6, the vortex velocity is found to be described as $v_\theta \propto (Q-Q_c)^{0.52}$. We have made a preliminary examination of the r (the distance from the center of the sink hole) dependence of v_θ and got that v_θ is inversely proportional to r, which indicates the angular momentum conservation of the stream. We have to add that the absolute value of v_θ and therefore the threshold value Q_c depend on the boundary condition; i.e., the velocity of the ambient stream. If there is a stream favorable to the generation of vortex, the value of Q_c is lowered.

We cannot give a full explanation of the behavior of vortex but can only give a basic reason why there is a threshold for the generation of vortex. Now let us start with the Navier-Stokes equation expressed in terms of cylindrical coordinates,

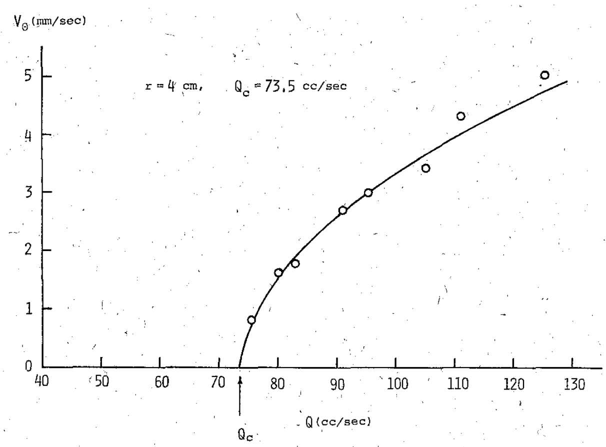

Fig. 5. The velocity of vortex against the amount of discharge through the sink hole.

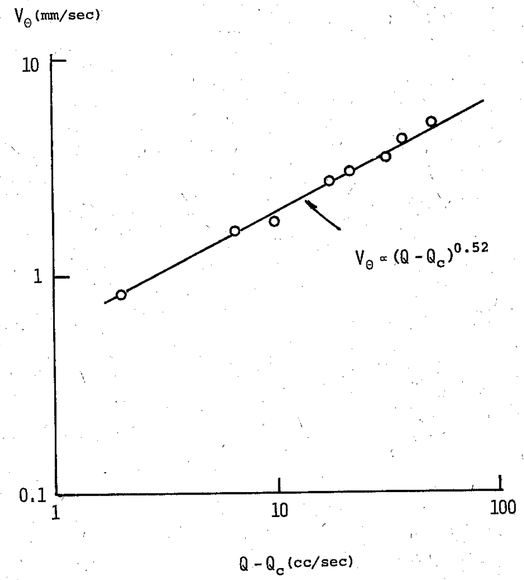

Fig. 6. v_θ vs $Q-Q_c$ curve. Q_c=73.5 cc/sec.

$$\rho\left(\frac{\partial v_r}{\partial t}+v_r\frac{\partial v_r}{\partial r}+\frac{v_\theta}{r}\frac{\partial v_r}{\partial \theta}-\frac{v_\theta^2}{r}+v_z\frac{\partial v_r}{\partial z}\right)$$

$$=F_r-\frac{\partial p}{\partial r}+\mu\left(\frac{\partial^2 v_r}{\partial r^2}+\frac{1}{r}\frac{\partial v_r}{\partial r}-\frac{v_r}{r^2}+\frac{1}{r^2}\frac{\partial^2 v_r}{\partial \theta^2}-\frac{2}{r^2}\frac{\partial v_\theta}{\partial \theta}+\frac{\partial^2 v_r}{\partial z^2}\right), \quad (10)$$

$$\rho\left(\frac{\partial v_\theta}{\partial t}+v_r\frac{\partial v_\theta}{\partial r}+\frac{v_\theta}{r}\frac{\partial v_\theta}{\partial \theta}+\frac{v_r v_\theta}{r}+v_z\frac{\partial v_\theta}{\partial z}\right)$$

$$=F_\theta-\frac{1}{r}\frac{\partial p}{\partial \theta}+\mu\left(\frac{\partial^2 v_\theta}{\partial r^2}+\frac{1}{r}\frac{\partial v_\theta}{\partial r}-\frac{v_\theta}{r^2}+\frac{1}{r^2}\frac{\partial^2 v_\theta}{\partial \theta^2}+\frac{2}{r^2}\frac{\partial v_r}{\partial \theta}+\frac{\partial^2 v_\theta}{\partial z^2}\right), \quad (11)$$

where v_r and v_θ are the radial and tangential components of velocity, respectively, ρ the density, F_r and F_θ the components of force, p the pressure and μ the viscosity. Here we use approximations that the stream is two-dimensional and is isotropic and we omit terms which includes derivatives with respect to z and θ. The radial and tangential forces F_r and F_θ are also omitted. Multiplying (10) by v_r and (11) by v_θ, respectively, we get equations for energy in the form

$$\frac{\partial}{\partial t}\left(\frac{1}{2}\rho v_r^2\right)=-\rho v_r^2\frac{\partial v_\theta}{\partial r}+\rho\frac{v_r v_\theta^2}{r}-v_r\frac{\partial p}{\partial r}+\mu v_r\left(\frac{\partial^2 v_r}{\partial r^2}+\frac{1}{r}\frac{\partial v_r}{\partial r}-\frac{v_r}{r^2}\right), \quad (12)$$

$$\frac{\partial}{\partial t}\left(\frac{1}{2}\rho v_\theta^2\right)=-\rho v_r v_\theta\frac{\partial v_\theta}{\partial r}-\rho\frac{v_r v_\theta^2}{r}+\mu v_\theta\left(\frac{\partial^2 v_\theta}{\partial r^2}+\frac{1}{r}\frac{\partial v_\theta}{\partial r}-\frac{v_\theta}{r^2}\right). \quad (13)$$

We take notice of the second terms on the right-hand side of (12) and (13), which have the same form except for their signs. As will be shown later v_r is negative, hence these terms mean that a power is transferred from the radial motion to the tangential motion. Thus, in the present case, the non-linear term plays a role of a bridge for the transfer of power from one degree of freedom of motion to another degree of freedom of motion.

Here let us introduce the amount of discharge Q using the continuity equation

$$Q=-2\pi r v_r \tag{14}$$

and further use the kinematic viscosity

$$\nu=\mu/\rho\,. \tag{15}$$

Then we can rewrite Eq. (11) as

$$\frac{\partial v_\theta}{\partial t}=\left(\frac{Q}{2\pi}-\nu\right)\frac{1}{r^2}v_\theta+\left(\frac{Q}{2\pi}+\nu\right)\frac{1}{r}\frac{\partial v_\theta}{\partial r}+\nu\frac{\partial^2 v_\theta}{\partial r^2}\,. \tag{16}$$

Using this equation, we have carried out numerical calculation in the range of r from 1.11 to 20.09 under the boundary condition of $v(20.09)=\text{const}$ and have got the following result: Whatever initial condition we may start from, when $Q/2\pi>\nu$, the vortex evolves into a macroscopic flow and its profile tends to c/r and when $Q/2\pi<\nu$, the vortex decays. In Fig. 7, the r dependence of v_θ at a computer time of 20000 is shown for three values of $Q/2\pi$. Here the kinematic viscosity ν is chosen as 1 and we took a rather irregular profile as an initial r dependence of v_θ. One can see that the vortex grows in case of $Q/2\pi=6$ and 2 which are larger than unity and decays in case of $Q/2\pi=0.1$ and the asymptotic form of first two cases turns out to be inversely proportional to r. Figure 8 shows the time evolution of v_θ at a distance 2.01 from the center of the sink hole. For larger value of Q, the vortex is found to grow up more rapidly.

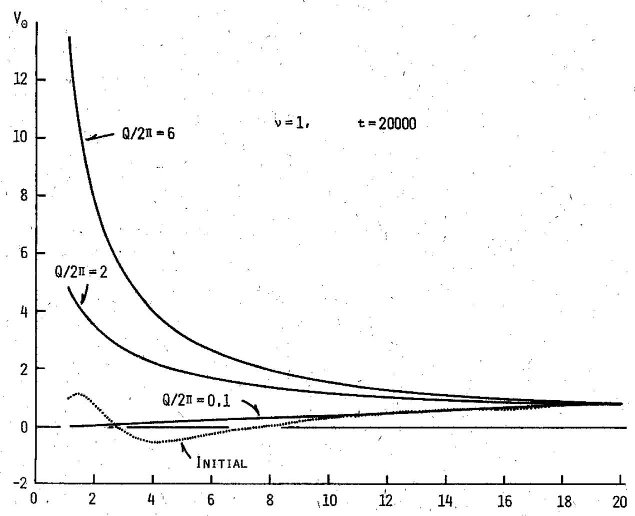

Fig. 7. The r dependence of v_θ computed from (16). The dotted curve is the initial velocity distribution.

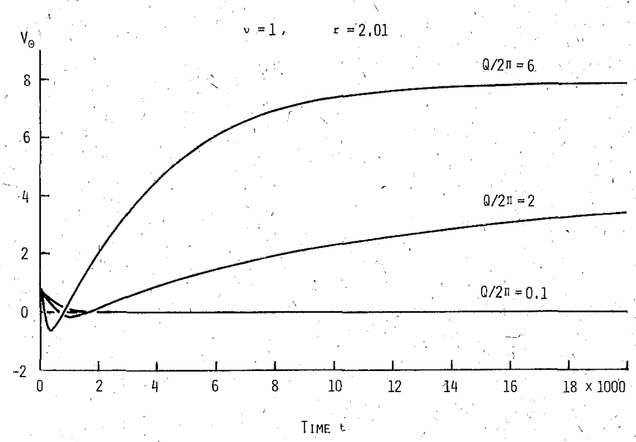

Fig. 8. The time evolution of v_θ at $r=2.01$ computed from (16).

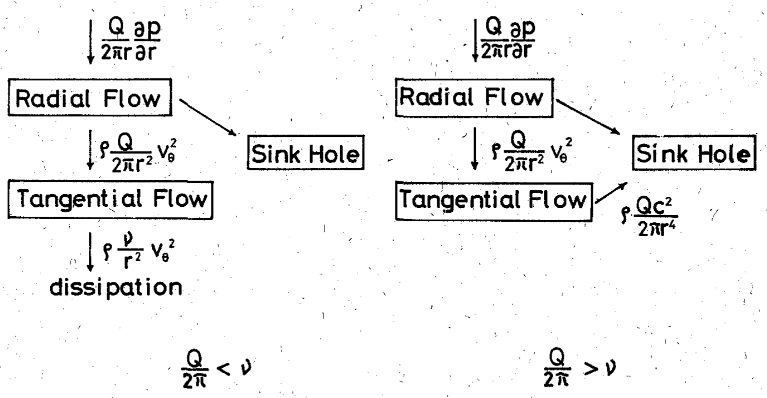

Fig. 9. The energy transfer through the vortex flow system.

The scheme of energy transfer through the system will be illustrated by a flow chart in Fig. 9. The power is supplied by the pressure gradient to the radial flow and if there is no tangential disturbance, the whole power is directly transferred to the sink hole. If a tangential disturbance v_θ accidentally occurs, however, a part of the power of the form $\rho Q v_\theta^2/2\pi r^2$ will be transferred to the disturbance, but that power is thermally dissipated as far as $Q/2\pi < \nu$ and the disturbance will decay. On the other hand, when $Q/2\pi$ exceeds ν the disturbance evolves into a macroscopic vortex of the form c/r and the power is transferred through the term $(Q/2\pi r)\partial v_\theta/\partial r$ on the right-hand side of (16) to the sink hole. In that case, three terms including viscosity ν in (16) cancel each other, so that the system behaves like an ideal fluid without viscosity. Thus a dissipative structure in which the vortex has a profile of c/r is realized.

§ 4. Successive transitions in liquid crystals due to an electric field

If a thin layer of nematic liquid crystal MBBA (Methoxybenzylidene

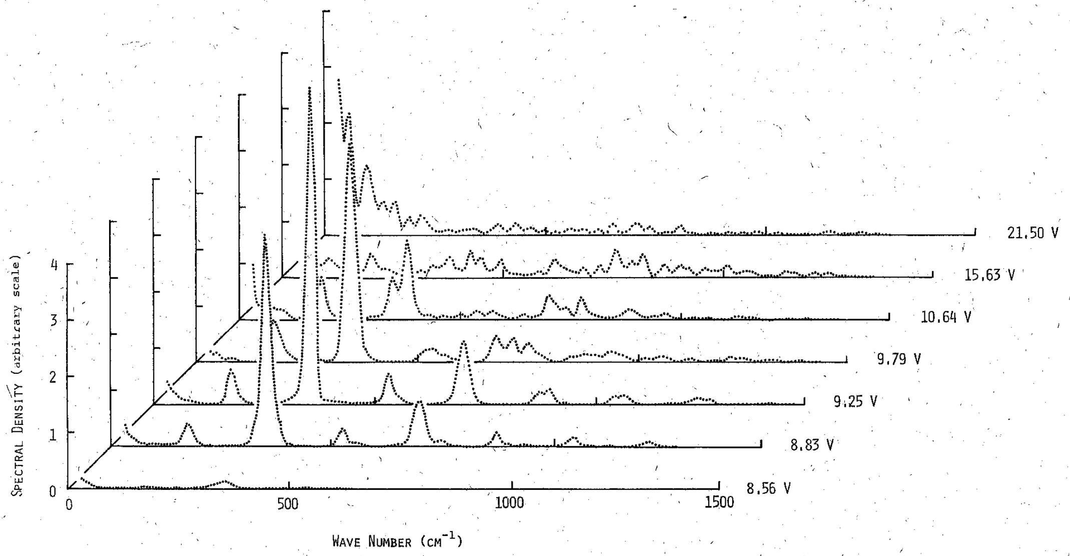

Fig. 10. The wave number power spectrum of the pattern intensity for various values of applied voltage.

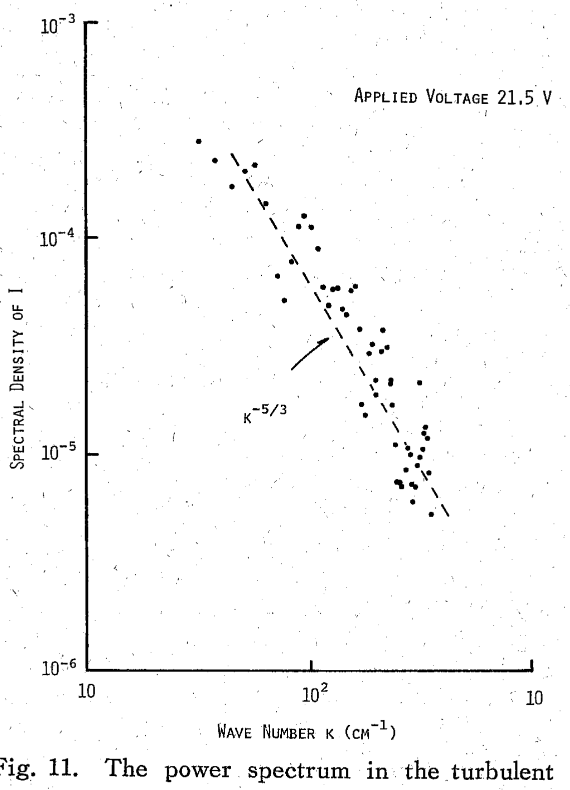

Fig. 11. The power spectrum in the turbulent state.

Butylaniline) is subjected to an electric field, a stripes pattern called Williams domain appears at a threshold voltage. If the applied vortage is increased, the stripes fluctuate both spacially and temporally and bifurcate and finally the pattern turns out to be turbulent.[10] We have examined this procedure by measuring the wave number power spectrum of the intensity of transmitted light through the crystal. The intensity pattern photographed on a film was converted into a sequence of electric signals and the power spectrum was computed by the Fast Fourier Transform algorithm from the signals.

The result is shown in Fig. 10. At 8.5V, a sharp peak which corresponds to the Williams domain predominantly grows up and is accompanied by many harmonics. At about 10V, each peak begins to split and the spectrum is changing to be continuous. In Fig. 11, we replot the spectrum at 21.5V, which corresponds to a turbulent pattern, on a logarithmic scale. Though

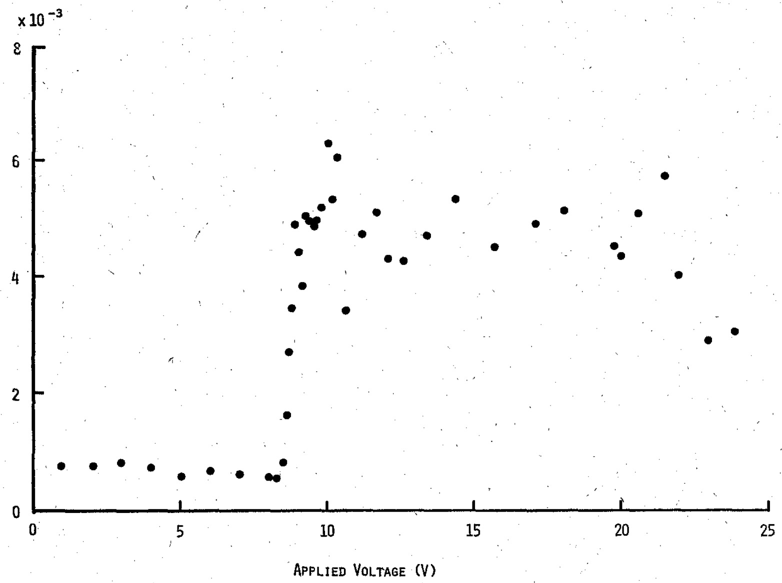

Fig. 12. The total variance which is obtained by integrating the spectral density in Fig. 10.

some fine structures remain in the spectrum but over the wide range of the wave number, the spectrum can be fitted with $k^{-5/3}$ which is characteristic to the turbulence. For the purpose of examining some preexisting phenomena such as anomalous spacial fluctuations below threshold of the formation of Williams domain, the total variance of the pattern intensity has been obtained by integrating the spectral density over the whole range of k. The result is shown in Fig. 12. The variance rapidly increases at the onset of the Williams domain and no evidence for critical fluctuations has been observed.

§ 5. Summary

In this paper, we have considered three kinds of physical systems which appear completely different at first sight. From the standpoint of nonequilibrium statistical physics, however, they exhibit a common behavior: i.e., they continuously take in energy and when the power (the rate of taking in of energy) exceeds some threshold value, they produce macroscopic self-sustained motions. Near threshold of occurrence of the self-sustained motion, the critical fluctuations associated with the instability of the system are expected, and we have succeeded in observing them for electric oscillatory systems. Nonlinearity of the system plays two roles in self-sustained motions: One is a suppression of the order parameter within a finite value, and the other is a transfer of energy among modes or degrees of freedom of motion as has been shown in vortex formation. In cases of vortex and oscillatory systems, the order parameter above threshold has been found to increase obeying the square root law of the form $(\alpha-\alpha_c)^{1/2}$, where α is the controlling parameter, but it is not always true; in general, the form must depend on the nonlinearity. The influence of external noise on the self-sustained motion is an attractive problem because the external noise brings about some reflections in the deterministic motion, but in the present stage general discussions are beyond our grasp.

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Discussion

N. G. van Kampen: In your electronic model you obtained the shift of the critical point in the Fokker-Planck equation by shifting some term from the so-called diffusion term into the drift term. That seems to me a rather arbitrary procedure for justifying a physical result. If you write the coupled equations for both the primary and secondary circuits in your system with the noise generator added, you should have a perfectly well-defined set of equations in which this ambiguity does not enter.

R. F. Fox: In your Fokker-Planck equation, you have changed from

$$-\frac{\partial}{\partial b}(-\gamma bP)+D\frac{\partial}{\partial b}b\frac{\partial}{\partial b}bP$$

to

$$-\frac{\partial}{\partial b}[-(\gamma+D)bP]+D\frac{\partial}{\partial b}b^2\frac{\partial}{\partial b}P.$$

However you could as well write

$$-\frac{\partial}{\partial b}[-(\gamma-D)bP]+D\frac{\partial^2}{\partial b^2}b^2P.$$

This form has the advantage that in looking at $d\langle b\rangle/dt$, you get $-(\gamma-D)\langle b\rangle$ directly because the $\partial^2/\partial b^2$ term yields 0. You also get this from your other form.

T. Kawakubo: As Prof. van Kampen and Prof. Fox commented, there is an arbitrariness about the way how to redistribute the terms in the Fokker-Planck equation into the drift and the diffusion terms. But the behavior of the system which the equation represents as a whole must be the same. The problem is not how to change the form of the Fokker-Planck equation but what kind of quantity should be taken as a deterministic value.

If we take $\langle b \rangle$ as an ordering parameter, the transition point shifts to lower values, since the effective damping coefficient decreases from γ to $\gamma - D$ as Prof. Fox mentioned. When a variable associated with the ordering parameter is fluctuating, I have no idea which of the average value and the most probable value should be taken as the ordering parameter. But our experimental result can be explained only by the most probable value, and I think that the envelope curve of transient process may represent the time variation of the most probable value. Our final form

$$\frac{\partial P}{\partial t} = -\frac{\partial}{\partial b}[(\alpha - (\gamma + D) - \beta b^2) bP] + D\frac{\partial}{\partial b} b^2 \frac{\partial}{\partial b} P$$

is more convenient for looking at the most probable value in the steady state. It is given by $b_m = \{(\alpha - \gamma - D)/\beta\}^{1/2}$. I would like to stress again that the problem is not how to change the Fokker-Planck equation but which of the average value and the most probable value of the stochastic amplitude should be taken as the ordering parameter of the electric oscillation.

R. Kubo: If the noise in your circuit plays the role of a random frequency modulation, then the effective increase of damping is just the same effect as is familiar in spin resonance problems, that is the transverse relaxation. According to your equation, the noise should work as negative damping. So the physics is rather hard to understand.

H. Hasegawa: What is the reason why you need to multiply the stochastic force in your Langevin equation by 'b'? It causes a difference between Itô and Stratonovich integrals for the solution. I do not think it is a very important point, but still we do not know which should be adopted in the phenomenological treatment, which is mentioned as the problem of modeling in Arnold's book.

T. Kawakubo: It is simply a circuit problem why the external random force is multiplied by the amplitude b. Our treatment of $bR(t)$ in deriving the Fokker-Planck equation seems to be equivalent to the Stratonovich integral. But I don't know either why a treatment based on the Stratonovich model should be adopted to our electrically oscillatory system.

H. Haken: The difficulties arising in Langevin equations with variable-dependent fluctuating forces stem from the fact that physical noise is replaced by white noise. Thus when one assumes non-white noise first and that the variables of the system change much more slowly in time, one can certainly justify Prof. Kawakubo's procedure and avoid a formulation of Itô's equations.

T. Kawakubo: In the treatment of stochastic processes, we started with the Langevin equation for macroscopic variables. I would like to know if we can deduce cooperative behavior such as an onset of oscillation in the electric circuit from the microscopic level.

N. Nicolis: I believe that when cooperative behavior sets up in the form of an asymptotically stable dissipative structure, there is no major difference between microscopic processes before and after the dissipative structure formation. However, the situation may change in the immediate vicinity of bifurcation point. There, fluctuations may get amplified and attain a macroscopic range. This should imply a change in the behavior in the microscopic level. In your example of the electronic circuit this could be reflected by a change in the current density and other properties of the plasma.

G. Ahlers: Your last figure seemed to indicate that $(\Delta I)^2$ has a jump for $V \simeq 8.5$ volts. Does this mean that the amplitude of the Williams domains does not grow continuously from zero?

T. Kawakubo: No, I do not think so. It grows up rapidly but continuously.

P. C. Martin: Does the total intensity satisfy a square root law at the threshold?

T. Kawakubo: $\langle \Delta I^2 \rangle$ is the total variance of the intensity which is integrated over the whole range of the wave number. So it grows up more rapidly than the square root law. If we plot the height of the dominant peak which corresponds to the Williams domain, it may obey the square root law.

H. Haken: Prof. Ruelle, is there any similarity between bifurcations in hydrodynamics and phase transitions?

D. Ruelle: There are certain similarities between hydrodynamics and equilibrium statistical mechanics, and in particular between bifurcations and phase trnsitions. I would like to express my opinion that these similarities are rather superficial. At the level considered, hydrodynamics is not a variational problem which statistical mechanics is. In hydrodynamics several attractors may coexist, but it is now believed that in statistical mechanics systems with short range forces do not have true metastable states. These remarks do not preclude the existence of relations between statistical mecahnics and hydrodynamics at a deeper level.

P. C. Martin: I agree that bifurcations and phase transitions are essentially different. With phase transitions, infinitely many degrees of freedom are necessary. But in both phase transitions and bifurcation theory there can be jumps and smooth onset phenomena. We only wanted to know which occurred here.