NONEQUILIBRIUM PHASE TRANSITIONS - A REVIEW

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1. Introduction

This is an elementary and tutorial review on nonequilibrium phase transitions. The tremendous progress in understanding equilibrium phase transitions is well known, and even as these advances were being made, concepts like order-parameter, order parameter fluctuations, etc began to find a place in the study of nonequilibrium phase transitions. It is these developments which we shall discuss.

2. Bifurcation

Let us suppose we start with a system initially not coupled to outside world so that it is in a (thermodynamic) equilibrium state. We now switch on the coupling and apply some stationary external 'force' whose strength is at our disposal. The system will naturally respond, as schematically illustrated in Fig.1. For a small applied force, the system is driven from a state of thermodynamic equilibrium to a steady state. The sequence of nonequilibrium states is usually called a branch, (often, as the thermodynamic branch). When the system is driven farther and farther away from equilibrium, nonlinearities begin to creep in, eventually dominating to the point the thermodynamic branch becomes unstable. At this stage one or more totally new options become available to the system, some of which may be stable while others may be unstable. This availability of two or more new states following an instability is usually referred to as bifurcation [1]. An example of bifurcation is given in Fig. 1. We shall mostly be concerned with this type of bifurcation as it has close analogies to a second-order phase transition.

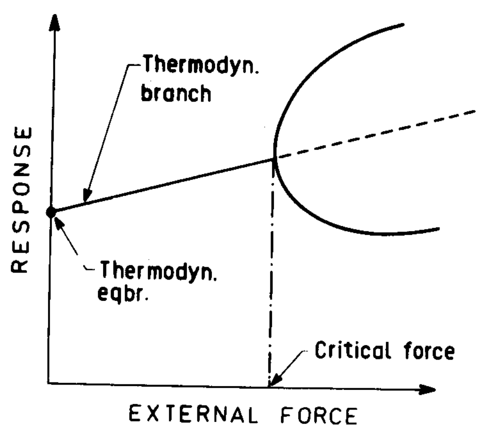

Fig.1 Schematic of a simple bifurcation

The new state that emerges following bifurcation usually has either temporal order, spatial order or even both. The resulting ordered structures are often referred to as dissipative structures [2]. The states on the thermodynamic branch will have the symmetry of the thermodynamic equilibrium state and the applied force. At the point of bifurcation there is a breaking of symmetry [3,4] similar to what happens in phase transitions.

Although we shall be primarily concerned with the type of bifurcation illustrated in Fig.1, we also note that as the external force is increased, the process of bifurcation usually repeats itself many times, resulting in more and more structured and highly complicated nonequilibrium steady states. Eventually, a statistical description of this complicated structure may be necessary for practical reasons.

III. Instabilities

We have noted that when a system is driven far from equilibrium, instabilities set in. We now consider how these instabilities may be analyzed. Initially the description of the process will be deterministic.

To start with, we suppose that our system is described by the macrovariables

$$\bar{X}(r,t) = \left\{ X_1(r,t),\ X_2(r,t), \ldots \right\} \tag{1}$$

and a set of 'control' parameters

$$\lambda = \left\{ \lambda_1,\ \lambda_2 \ldots \right\} . \tag{2}$$

In the problems we shall be interested in, the evolution equations for $\bar{X}(r,t)$ typically have the form

$$\dot{\bar{X}}(r,t) = F(\bar{X}(r,t),\lambda). \tag{3}$$

F is in general a nonlinear operator, and describes the various processes involved energy exchange, matter exchange, transport phenomena, feedback etc. Observe that $\dot{\bar{X}}(r,t)$ depends only on the present state $\bar{X}(r,t)$.

Given below is an example of (1) and (3)

$$\bar{X}(r,t) = \left\{ \rho(r,t),\ V(r,t),\ T(r,t) \right\}$$

where ρ , V and T are respectively the density field, velocity field and temperature field. Equation (3) in this case comprises of the well-known trio, namely, the continuity equation, the Navier-Stokes equation and the heat-conduction equation.

Going back to (3), on the thermodynamic branch, the steady states X_s are deduced from

$$F(\bar{X}_s,\lambda) = 0. \tag{4}$$

The emergence of a dissipative structure or self organization as it is sometimes called, is essentially a transition from X_s to a new type of solution. Presently we wish to explore the instability that precedes such a transition. To investigate instabilities, we visualize small disturbances $\bar{x}$ added to the steady state $\bar{X}_s$ and then study the behaviour of the resultant state

$$\bar{X} = \bar{X}_s + \bar{x} . \tag{5}$$

When (5) is introduced into (3) one gets a nonlinear dynamical problem. Linearizing, one obtains

$$\dot{\bar{x}} = \bar{\bar{L}}(\lambda)\ \bar{x}, \qquad (6)$$

where $\bar{\bar{L}}$ is an appropriate linear operator.

To recapitulate, when the control parameter is gradually increased, the system emerges from thermodynamic equilibrium and 'rides' the thermodynamic branch. At each stage, one has a steady state appropriate to the value of λ involved. What we are now trying to do is to perturb each one of these states and see how the system behaves. The behaviour of the perturbed state $\bar{X}(r,t)$ for $t \longrightarrow \infty$ will determine stability of the reference state $\bar{X}_s(r,\lambda)$.

Under the conditions defined above, $\bar{\bar{L}}$ is time independent and (6) admits solutions of the form

$$\bar{x} = x_o \exp(\omega t), \qquad (7)$$

with

$$\bar{\bar{L}}(\lambda)\,\bar{x}_o = \omega\,\bar{x}_o. \qquad (8)$$

Everything now depends on ω. The normal-mode frequency ω will in general be complex i.e.

$$\omega(\lambda) = \mathrm{Re}\ \omega(\lambda) + i\ \mathrm{Im}\ \omega(\lambda) \qquad (9)$$

from which we see that the stability of the system depends on the sign of Re $\omega(\lambda)$. If it is negative, the oscillations induced by the perturbation will decay and the reference state will be stable. Occurence of instability is therefore connected with a situation as in Fig.2 where Re $\omega(\lambda)$ changes sign at a critical value λ_c of the control parameter. For $\lambda > \lambda_c$, the reference state $\bar{X}_s$ is unstable.

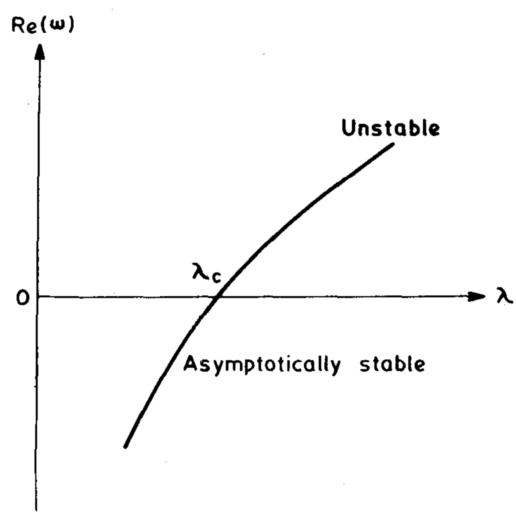

Fig.2 Transition to instability at a critical value of the control parameter.

Directing attention now to Im $\omega(\lambda)$, two possibilities exist:

(i) Im $\omega(\lambda) = 0$ for all λ . (10a)

(ii) Im $\omega(\lambda_c) \neq 0$. (10b)

Instability of type (10a) is referred to as soft-mode instability while that corresponding to (10b) is referred to as hard-mode instability.

In some problems like those involving chemical reactions, the operator $\bar{\bar{L}}$ has a term D ∇^2 to deal with diffusive effects [5]. In such cases, we have (considering a 1-D system for simplicity)

$$\bar{x}(r,t) = \bar{x}_o\, e^{\omega(\lambda,k)t}\, e^{ikr};\ k = \frac{2\pi}{L} n; n = 0,1,2,\ \ldots. \tag{11}$$

where L $\sim$ linear dimension of the system. It is conceivable that the instability is associated with a mode of wave vector $k \neq 0$. If the instability is of the soft-mode type (i.e. $\omega(\lambda, k) = \text{Re}\ \omega(\lambda, k)$; $\omega(\lambda_c, k) = 0$), then a static, spatially inhomogeneous pattern with wave vector k will result. With hard-mode instability, both spatial and temporal oscillations arise.

4. Role of Nonlinearities

The linear stability analysis does not obviously tell the full story. We must now extend consideration to nonlinearities and also to another equally important entity namely, fluctuations or noise.

Let us ignore noise for the time being and just consider (3). Displaying explicitly the linear and nonlinear parts,

$$\frac{\partial}{\partial t}\bar{x} = \bar{\bar{K}}\,\bar{x} + \bar{g}(\bar{x}). \tag{12}$$

The nonlinear part will typically have the form

$$g_i(x) = \sum_{\mu} C^{(2)}_{i\mu\upsilon} x_\mu\, x_\upsilon + \sum_{\mu\upsilon\lambda} C^{(3)}_{i\mu\gamma\lambda}\, x_\mu\, x_\upsilon\, x_\lambda + \ldots. \tag{13}$$

When ths system rides the thermodynamic branch, all the $x_i(t)$'s will decay rapidly. Near the point of bifurcation, one mode i = u say, becomes unstable. Since Re $\omega_u(\lambda \sim \lambda_c) \approx 0$, the decay constant of this mode will be very long i.e. this will be a slow mode as compared to all the other modes. Under these circumstances, one may set

$$\frac{\partial x_i}{\partial t} \approx 0 \quad i \neq u. \tag{14}$$

This is quite reasonable on the time scale on which x_u exhibits variations. Using (14) we can write the r.h.s of

$$\frac{\partial x_u}{\partial t} = \sum_i K_{ui}\, x_i + g_u(\bar{x}) \tag{15}$$

entirely in terms of x_u. For instance, in the laser problem one has [6]

$$\dot{E} \approx (G - K)E - C|E|^2 E \tag{16}$$

where G is proportional to the pump power, K is the loss constant and C is a constant depending on the laser material.

The essential physical content of the foregoing steps is that near instability one exploits the fact that there are two vastly different time scales associated with the stable and unstable modes respectively. The stable modes being fast, continuously adjust to the much slower unstable modes. This enables us to write (14) and eventually eliminate the fast modes in the equation for the unstable mode, a process known as adiabatic elimination. It is clear that the mode x_u is connected with the dissipative structure that emerges beyond λ_c and therefore is the analogue of the order parameter of equilibrium phase transition. The slowing down of x_u near bifurcation is thus the analogue of critical slowing down. The dominating role played by x_u is referred to by Haken as the 'slaving principle'. Equation (15) will guarantee the $x_u(t)$ does <u>not</u> increase without bounds.

5. Role of Noise - A First Look

Having identified the analogue of the order parameter, we now ask whether there is the analogue of critical fluctuation also. To discuss this question, we generalize (12) to include fluctuations, i.e., write

$$\frac{\partial}{\partial t} \bar{x} = \bar{\bar{K}} \bar{x} + g(\bar{x}) + \bar{F}(t) \tag{17}$$

where F(t) is the noise term. One must then go through the adiabatic elimination procedure as before, leading eventually to

$$\frac{\partial x_u}{\partial t} = (\text{terms involving } x_u \text{ only}) + \mathcal{F}_u(t) \tag{18}$$

where $\mathcal{F}_u(t)$ is a fluctuating term. In other words, instead of a deterministic equation as in (16), we now have a Langevin equation. If a Langevin equation occurs, can the corresponding Fokker-Planck equation be far behind! We shall say something more about fluctuations later.

6. Phase Transitions of an OPAMP

We now consider an explicit example of a system exhibiting a non equilibrium phase transition, i.e., an operational amplifier with positive feedback. It is instructive to examine first the role of feedback, especially since feedback is important also in equilibrium phase transitions [7,8]. From Fig.3

$$A_{eff} = \frac{v_o(t)}{v_i(t)} = \frac{A}{1-\beta A} \tag{19}$$

(shades of RPA!). Also sketched in Fig.3 are the transfer characteristics (input <u>vs</u> output) for various values of β.

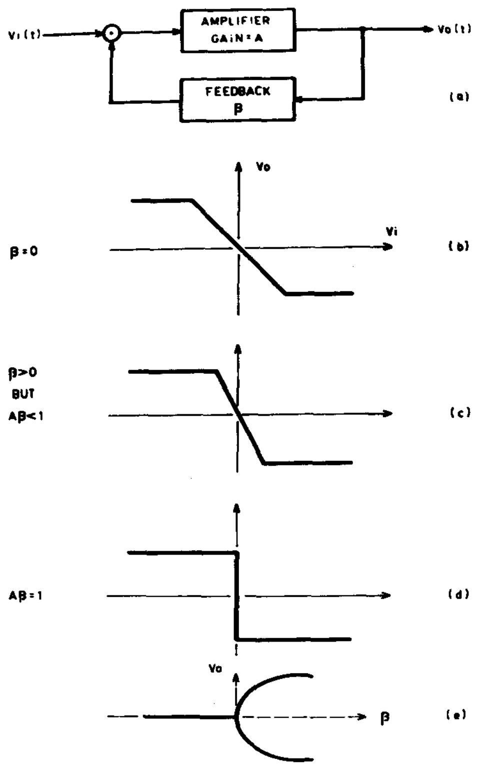

Fig.3 a) Amplifier feedback loop
b) - d) Transfer characteristics for various values of
e) Bifurcation pattern

Suppose v_i is set $\equiv 0$ and we atart the system from the condition $\beta = 0$. From Fig.3 we see that the output voltage will also be zero. A linear stability analysis (which we skip), will show that this in fact is a stable state. Indeed this state is stable even if $\beta > 0$, as long as $A\beta < 1$. When $A\beta = 1$, the system has two new options. When $A\beta$ exceeds unity, the system settles down into one of the available options, leading in effect to the bifurcation pattern of Fig.3e.

The stability of the output voltage v_o depends essentially on what happens to the energy of the OPAMP capacitor, when the voltage changes by a small amount from the previously occupied state. A little reflection shows that the energy stored W is given by.

$$S = \frac{Cv_o^2}{2} - C \int (v_i - \beta v_o)\, A dv_o . \tag{20}$$

The gain A depends on the differential input voltage which, for the OPAMP used, has the form

$$A(v_i) = A_o (1 - \alpha v_i^2) \tag{21}$$

where A_o and α are constants. Hence, for $v_i = 0$,

$$W = \frac{Cv_o^2}{2} (1 - A_o\beta) + \frac{CA_o\alpha\beta^3}{4} v_o^4 \tag{22}$$

which has the familiar Landau form. The manifestation of a second-order phase transition by the OPAMP is immediately understandable.

In Fig.4a we give some observed results. The OPAMP also exhibits a first-order transition when $v_i \neq 0$; see Figs. 4b and 4c. (In the case of the laser too one has

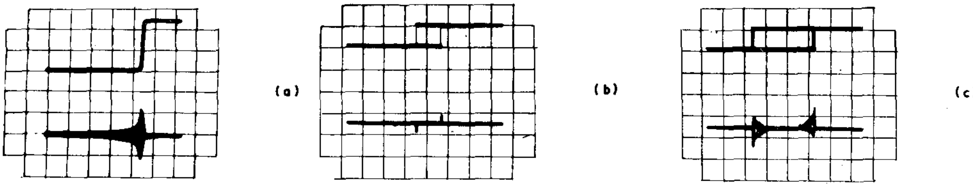

Fig.4 Phase transitions of OPAMP. a) shows the second-order transition. The enhancement of critical fluctuations near the transition is clearly visible. b) and c) show the first-order transition.

a similar behaviour for, with an injected signal, the laser exhibits a first-order transition instead of the usual second-order one). An interesting sidelight relates to the fluctuations. No fluctuations were found in the experiment corresponding to Fig.4b but when the system time constant τ_s exceeds the correlation time τ_c of the input noise, one sees fluctuations.

7. Role of External Noise

We come back again to the question of noise. Firstly we note that near an instability, noise plays a delicate role in switching the system to the 'other phase' even as it does in equilibrium phase transitions. Once the system acquires a dissipative structure, noise loses its dominant role. (For example, the laser emits only monochromatic radiation).

The role of noise in triggering a switching action deserves a closer look when noise enters in a multiplicative way. The rate equations now have the form

$$\dot{x} = \alpha(x) + \beta(x)\,\eta(t) \tag{23}$$

where $\eta(t)$ is the random force. Particularly interesting examples of (23) are [9]

$$\dot{x} = \gamma x - gx^2 + x\,\eta(t) \tag{24a}$$

$$\dot{x} = \gamma x - gx^m + x^m\eta(t) \tag{24b}$$

An experiment to study the role of external noise has been done by Kabashima and Kawakubo [10]. These authors used a degenerate parametric amplifier connected to an external noise source having a flat spectrum between 0.01 and 10^5 Hz. The pumping frequency was 50 KHz and the subharmonic 25 KHz. Figure 5 sketches how the transition was affected by the noise frequency. Kabashima and Kawakubo have also performed several other experiments clarifying various aspects of the role played by external noise.

To paraphrase, multiplicative noise produces nontrivial changes in the behavioural pattern of the system concerned. Such effects are of particular interest

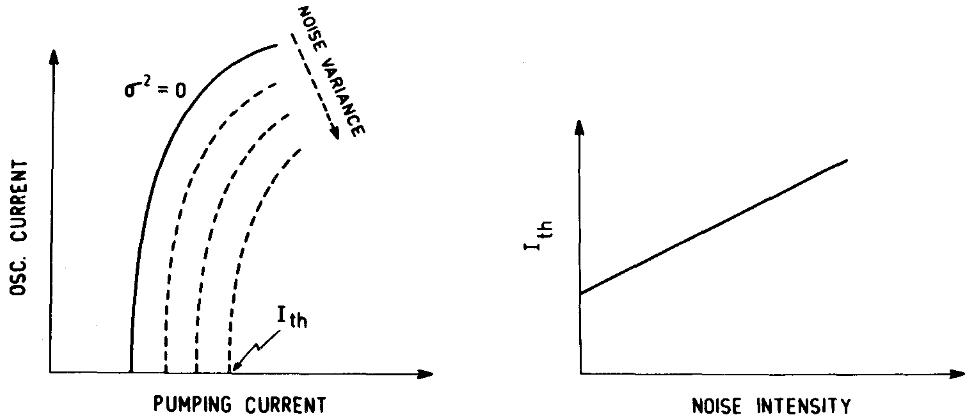

Fig.5 Effect of multiplicative noise on the behaviour of a parametric amplifier. The sketches are schematic. (After ref.[10])

in many biological and ecological problems. We ourselves are interested in external noise with reference to a metallurgical phenomenon known as serrated yielding.

8. Chaos

Although in this review we have been primarily concerned with the very first bifurcation, it is perhaps worth making a few remarks about the random behaviour usually exhibited under high excitation. Such random phenomena often called chaos, are not only seen in hydrodynamical systems (which are the ones usually cited) but also in several other systems like the laser. We shall introduce the concept of chaos via the driven anharmonic oscillator, especially as it is amenable to study in the laboratory.

The oscillator we consider consists of a LCR circuit driven by a sinusoidal voltage, the capacitance C being that of a varactor diode. The special characteristic of C is that it is nonlinear, the law of variation being

$$C(V) \quad \frac{C_o}{(1+V/\phi)^{r}}$$

where V is the voltage across the diode, and ϕ and r are suitable parameters.

The experiment consists in tuning the signal generator to the fundamental frequency f_1 of the LCR circuit and gradually increasing the amplitude of the applied sine wave [11]. For low applied voltages, the normal mode frequency f_1 alone is excited (labelled 0 in Fig. 6). On increasing the driven voltage, the first subharmonic ($f_1/2$) appears (marked 1 on the figure). With further increase in the excitation, there are further manifestations of period doubling. The asymptotic situation is shown in Fig. 6d. The spectrum corresponds to chaotic behaviour, and is definitely not a white noise spectrum.

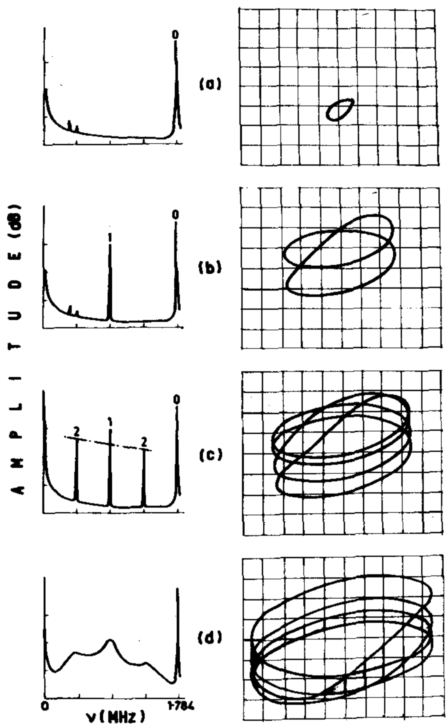

Fig.6 Response of the anharmonic oscillator. On the left are shown the power spectra obtained by Linsay (ref.11). On the right are the corresponsing phase-space plots obtained by us.

We have studied the approach to chaos by observing the phase-space plots electronically. Initially, the phase-space plot has just one loop. As successive period doublings occur, more and more loops get added; after n doublings, there are 2^n loops. When n becomes infinite (which happens when λ exceeds λ_c), we no longer see individual loops but just a blur. In this situation, the solutions $x_i(t)$ of (3) are nonperiodic. Despite this feature, we notice that the representative point always stays around a given region of phase-space. For this reason this region is called an attractor.

We now turn to universalities in nonlinear systems exhibiting period doubling. The benchmark work in this area is due to Feigenbaum [12-14]. In the problems he considers, the value assumed by the variable x at some step (j+1) in its evolution depends on the value it had at the previous step j, i.e.

$$x_{j+1} = f(x_j, \lambda) \tag{25}$$

Making rather general assumptions about the nature of f, Feigenbaum deduced that the control parameter λ should asymptotically satisfy the recurrence relation

$$\frac{\lambda_{n+1} - \lambda_n}{\lambda_{n+2} - \lambda_{n+1}} = \delta$$

where index n refers to the nth period doubling and δ is universal constant that depends only on the value of f near an extremum. For a quadratic extremum, Feigenbaum deduced $\delta = 4.699 \ldots$. In the case of the anharmonic oscillator, Linsay [11] measured δ by observing the voltage thresholds for the successive bifurcations. One value he obtained for δ was 4.5 ± 0.6 which is in reasonable agreement with theory.

Another universal feature deduced by Feigenbaum related to the amplitude of the Fourier components of the oscillations. Let $x^n_{(2k+1)}$ be the (complex) amplitude of

(2k+1)th Fourier component of x(t) when it is periodic with period $T_n = 2^n T_o$. Feigenbaum showed that there is relationship connecting the amplitudes of the odd components x^n_{2k+1} featuring a universal constant $\alpha = 2.5029 \ldots$. Translated to the anharmonic oscillator experiment, what this means is that if a line is drawn joining the peaks labelled 2 (see Fig.6), then the line is drawn joining the n = 3 and n = 4 peaks occur respectively 8.2 and 16.4 dB below it. Linsay found that the third and fourth generation peaks are in remarkable agreement with the theoretical prediction. Gollub [15] has noted a similar feature in his hydrodynamical experiments.

To sum up :

(i) Chaos is a situation where a seemingly random behaviour is exhibited by a deterministic system.

(ii) The chaotic state arises due to the existence of nonlinearities.

(iii) The power spectrum of the chaotic state is quite different from that of thermal noise.

(iv) Experiments have confirmed certain universalities when the route to chaos is through bifurcations involving period doubling.

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