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Nonequilibrium phase transitions

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Abstract. A brief review is provided of the subject of nonequilibrium phase transitions occurring in various systems in different disciplines. All these systems exhibit a similar kind of ordering phenomena at a macroscopic level and can therefore be classified in terms of appropriate order parameters. Typical order parameter equations are discussed in the light of Landau's phenomenological theory of phase transitions. The instabilities occurring in a few chemical reactions are discussed in some detail and the role of fluctuations pointed out. Mention is also made of the possibility of viewing some metallurgical phenomena as examples of nonequilibrium phase transitions.

1. Introduction

A proper description of phase transitions of different kinds that occur in systems in *thermal equilibrium* may be given within the framework of Gibbsian Statistical Mechanics. Recently however it has been recognised that phase transition or more appropriately, phase transition-like phenomena, i.e., phenomena which describe transitions from disordered states to ordered states, are not just the privilege of physical systems in thermal equilibrium but may occur in a variety of disciplines such as physics, chemistry, biology, sociology, etc. In all these cases the systems are composed of very many subsystems which do not behave independently of one another but act in a controlled, self-organised manner. The "working together" of the subsystems or "joint efforts" exhibited by them has led Hermann Haken to introduce the Greek word "synergetics" for naming a new discipline that deals with the study of co-operative effects in different fields of research (Haken 1973, 1974, 1975, 1977a, b). We shall witness that *albeit* the systems under discussion are composed of quite different subsystems whose mutual interactions are of varied nature, nevertheless, at a certain level of description which might be called "macroscopic", striking similarities appear. Examples of some of these systems and their associated subsystems are listed in table 1.

In a ferromagnet, the subsystem consists of atomic spin-magnetic moments which are in a disordered state of random orientations for temperatures above a critical

Table 1. Examples of systems and their subsystems in various disciplines.

Discipline	System	Subsystem
Physics	Ferromagnet	Spins
	Super conductor	Electrons + Phonons
	Laser	Light + atoms
	Fluids	Atoms (molecules)
Biology	Brain	Neurons
Chemistry	Chemical ensemble	Molecules
Population dynamics	Population	Individuals

temperature T_c (figure 1). Below T_c, the Coulomb exchange interaction between atomic electrons, which favours a parallel alignment of the moments, wins over the entropy term, forcing the system into an ordered state. This example brings to our mind the notion of a *control parameter* which, in the present case, is the temperature, a variation of which makes possible the transition from a disordered to an ordered state.

In a superconductor the electrons are uncorrelated above a certain critical temperature but below that temperature they form Cooper pairs due to an attractive interaction mediated by the phonons. The Cooper pairs are in an ordered state described by a macroscopic wave function in which the electrical conductivity is infinitely high.

In a laser, when the input is low, the atoms emit light in a disorganised manner. The result is a lamp whose emission is totally incoherent. When the input power (the control parameter, in this case) exceeds a certain threshold, laser light becomes extremely coherent (figure 2). At this point, we should like to emphasise that while

Figure 1. Schematic picture of the spin-orientations in the disordered and ordered ($T = 0$) states of a ferromagnet.

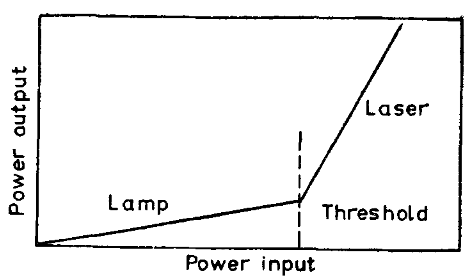


Figure 2. Power output vs. input in a laser which shows that beyond a certa n value of the input power, laser light becomes coherent.

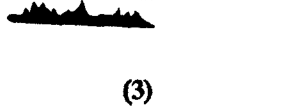
ferromagnets and superconductors are systems in thermal equilibrium in which ordered states are obtained by lowering the temperature, in the laser, a system far from thermal equilibrium, the ordered state can be obtained and maintained only by pumping energy into it. Subsequently, we shall see that there are other examples of cooperative effects in which temperature or energy has at best only a formal meaning or even no meaning at all.

In fluids, there are examples of a number of ordering phenomena. One of them is the Benard instability concerning a liquid layer heated from below. When the temperature-gradient between the lower and the upper surfaces of the liquid is small, the convection pattern is quite irregular. However when the temperature-gradient increases beyond a certain critical value, a macroscopic convection pattern, in the form of rolls, appears suddenly (figure 3). In this case the temperature-gradient acts as the control parameter. A related phenomenon is the occurrence of the Taylor instability. This concerns a liquid contained between two coaxial cylinders (figure 4). The outer cylinder is kept fixed. When the inner one is rotated at a speed beyond a certain critical value, a macroscopic motion in the form of Taylor vortices appears.

Aside from the foregoing examples of spatial structures in fluids which are formed when a certain external condition is altered, cases of temporal structure or self-organised oscillations are also known to occur. We have already encountered an example of this in the laser. Other instances of oscillatory patterns can be seen in some chemical reactions in which the colour of the reactants changes periodically from red to blue and vice versa. Our third example of ordered oscillations is well-known in population dynamics or ecology. This concerns the so-called Lotka-Volterra cycle in which two kinds of fish, the predator and the prey, " interact " with one another. Initially, the predator population may be low. Assuming that there is a regular food supply for the prey fish, the population of the latter grows. The predator fish now multiplies by eating up the prey fish. Thus the number of the predator increases at the expense of the prey. This situation however cannot continue indefinitely since the food supply for the predator dwindles. There results therefore a famine situation for the predator resulting in a drop in its number. Consequently, the number of the prey fish increases again and the cycle continues (figure 5). Here, to perpetuate the cyclic pattern, the control parameter, viz., the food supply for the prey fish, has to be maintained at or beyond a certain limit.

2. The order parameter

Having introduced the numerous systems in which some kinds of pronounced transitions from a disordered state to an ordered state occur, the question now



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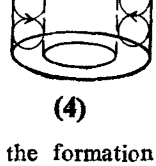

(4)

Figure 3. Benard instability illustrating the formation of a regular convection pattern when a liquid is heated from below.

Figure 4. Formation of Taylor vortices in a liquid kept between an inner rotating cylinder and an outer fixed one.

arises : can there be a unified mathematical description of them ? The language and the mathematical tool necessary for describing the occurrence of instabilities and dissipative structures as observed in the examples mentioned here can be found in the subject of nonlinear thermodynamics discussed by Glansdorff and Prigogine (1971). However, for the purpose of the present discussion we shall confine our attention to a class of phenomena in the realm of dissipative structures whose description bears a close resemblance to that of equilibrium phase transitions. This is the reason why we choose to classify the phenomena mentioned above as "nonequilibrium phase transitions". But we want to stress that the subject of nonequilibrium phase transitions is not the same as that of dynamical critical phenomena, i.e., time-dependent effects near a second order phase transition. Nonequilibrium phase transitions occur in systems in which the concepts of temperature, thermal equilibrium, canonical ensemble, etc., do not have their usual meaning. At the same time we shall see that the familiar notion of the order parameter introduced by Landau for a description of second order phase transitions plays a very useful role also in our understanding of nonequilibrium phase transitions. As a matter of fact, the entire machinery of the modern theory of critical phenomena embodied in Wilson's renormalisation group approach can be brought in to elucidate certain features of nonequilibrium phase transitions. Before we outline how this programme is carried out, we present a list in table 2 of the order parameters associated with the various phenomena under discussion here. The use of the order parameter allows one to immediately circumvent the very difficult many-body problem of having to describe the behaviour of individual entities (e.g., the spins in the ferromagnet); instead, the order parameter provides an understanding of the *relevant* behaviour of the system *as a whole*. As we witness here, the concept of the order parameter is not just restricted to systems like ferromagnets which are in thermal equilibrium; it applies equally well to physical systems far from thermal equilibrium such as the laser.

3. Order parameter equations

To demonstrate the usefulness of the order parameter concept, we shall now discuss a few examples of equations for it.

Table 2. The nature of the order parameter in different systems.

System	Order parameter
Ferromagnet	Magnetisation
Superconductor	Pair wave function
Laser	Light field or photon number
Chemical ensemble	Number of molecules
Population dynamics	Number of individuals
Forest	Density of plants

3.1. *Population dynamics (single species)*

We consider an equation which describes the increase of population. This equation holds equally well also for a single-mode laser or for an autocatalytic chemical reaction. The dynamics is described by the equation

$$\frac{dn}{dt} = (a - \beta n)\, n, \tag{1}$$

where n is the number of individuals (human beings, photons, molecules). If the control parameter $a < 0$, the " death " rate is greater than the " birth " rate and the populattion dies out. The solution for the steady state, in that case, reads $n_s = 0$. If $a > 0$, an instability occurs. First, the number of individuals increases exponentially. However, due to limited resources (food, excited laser atoms, reactants), growth saturation occurs as described by the factor $-\beta n$ and a steady state is obtained at $n_s = a/\beta$. Thus a transition from one steady state, $n_s = 0$ to another, $n_s = a/\beta$ takes place as the parameter a is altered.

3.2. *Population dynamics (two species)*

We discussed already an example of this case in the so-called Lotka-Volterra model describing the biological system of two kinds of fish, the predator and the prey. In the absence of the prey fish, the predator fish population, n_1 diminishes and its rate of change with time can be written as $-\gamma_1 n_1$. On the other hand, the presence of the prey provides food for the predator so that the equation for n_1 can be constructed as

$$\frac{dn_1}{dt} = -\gamma_1 n_1 + \beta_{12}\, n_1 n_2. \tag{2}$$

Similarly, for the prey population, n_2, we have the equation

$$\frac{dn_2}{dt} = \gamma_2 n_2 - \beta_{21}\, n_1 n_2. \tag{3}$$

Here the term γ_2 is taken as positive since, as mentioned earlier, it is assumed that the prey fish has a constant food supply.

The solutions for n_1 and n_2 obtained from (2) and (3) have an oscillatory character as shown schematically in figure 5. Equations of the above type are

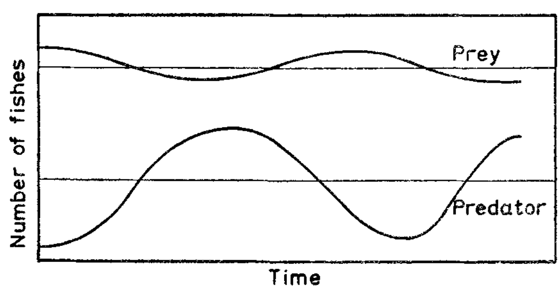

Figure 5. The Lotka–Volterra cycle.

used also for describing certain autocatalytic chemical reactions and we shall have occasion to comment briefly on one such reaction chain later on.

3.3. *More complex systems*

Equations of the types (1)–(3) can be generalised in a straightforward manner for systems with several constituents. They read

$$\frac{dn_j}{dt} = \epsilon_j n_j - \sum_i \beta_{ij} n_i n_j. \tag{4}$$

Such equations may be used for the description of population dynamics, chemical reactions, biological cells, multimode laser, and other non-linear transport problems. If the n's are regarded as pulse rates and the β's are given a slightly more general interpretation, then (4) can be cast into a form that has been used by Cowan to describe neuron networks (Haken 1973).

In a multimode laser, emission of photons from excited atoms in a cavity is in random directions with the photons distributed randomly over a certain frequency range. This situation is illustrated schematically in figure 6a in which the excited atoms are denoted by circles and the direction of emission of photons are indicated by arrows. The lower half of figure 6a sketches the photon-number n_j versus the "photon-kind" j, selected according to different gains. The dots are thus representative of the gain curve. As the number of photons in different directions and with different energies increases, the non-linear term in (4) (proportional to β_{ij}) starts becoming important. Soon a *stationary state* is reached in which only one kind of photons (in a mode with the highest gain) survives and the rest disappear. Equations of the type (4) are therefore capable of describing *selection* of a mode in a multimode laser system. Indeed, very similar equations have recently been used by Eigen (1971) to explain the operation of selection within the process of evolution which finally led to the genetic code.

4. Discussion of a simple order parameter equation

We now take up a typical order parameter equation and look at it from the familiar angle of Landau's theory of second-order phase transition. The equation for the order parameter q that we want to study is of the form

$$\frac{dq}{dt} = -aq - bq^3 + F(t), \tag{5}$$

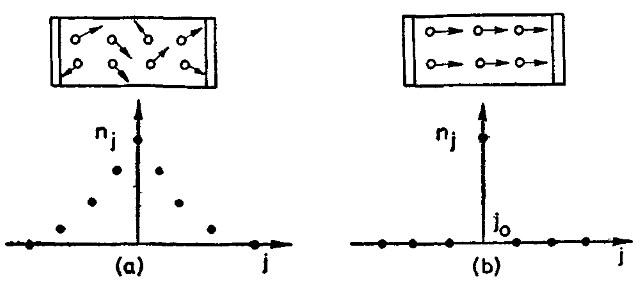

Figure 6. Photon selection in a laser with two end mirrors.

where $F(t)$ is a noise term. Without the term $F(t)$, (5) can be reduced, *via* a simple transformation $q = (n)^{1/2}$, to the form (1) introduced earlier to discuss, for example, the single mode laser. In what context, the noise term $F(t)$ may be viewed as describing the random creation of photons by, for instance, spontaneous emission.

Neglecting $F(t)$ in (5), we obtain as stationary states ($\dot{q} = 0$) the following solutions :

$$q_s = 0 \quad , \quad a > 0,$$
$$q_s = \pm (a/b)^{1/2} , \quad a < 0, \tag{6}$$

where the term b is always assumed to be positive. One may now try to linearise (5) around the stationary solution by setting

$$q = q_s + \delta q. \tag{7}$$

One then obtains, for $a > 0$,

$$\frac{d}{dt} \delta q = - a\delta q,$$

whose solution reads

$$\delta q = q_0 \exp (- at). \tag{8}$$

It is clear that the relaxation-time τ ($= 1/a$) becomes very large as a approaches zero. This phenomenon is a crude description of the critical slowing down associated with the time-variation of an order parameter near a second order phase transition. In a similar manner, for $a < 0$, the solution of the linearised equation reads

$$\delta q = q_0 \exp (- 2 \mid a \mid t). \tag{9}$$

To examine the role of fluctuations in the linearised theory, we reintroduce the noise term $F(t)$ in (5). Assuming the latter to be of Gaussian, white noise nature, i.e.,

$$\langle F(t) F(t') \rangle = C \delta (t - t'), \tag{10}$$

one finds, for $a > 0$,

$$\langle \delta q (t) \delta q (t') \rangle = \frac{C}{2a} \exp [-a (t - t')], \ t \geqslant t'. \tag{11}$$

This implies that

$$\langle (\delta q)^2 \rangle \propto \frac{1}{a}. \tag{12}$$

Therefore, the order parameter fluctuations, measured by $\langle (\delta q)^2 \rangle$, become very large as $a \to 0$. This is reminiscent of critical fluctuations accompanying a second-order phase transition.

To make the connection between an equation of the type (5) and that obeyed by the order parameter (e.g., the magnetisation for a spin system near a critical

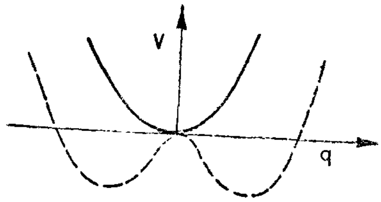

Figure 7. The Landau " Hamiltonian " diagram for positive and negative values of the parameter *a*.

point), more transparent, we cast the former into the form of a generalised Langevin equation (Ma 1976)

$$\frac{dq}{dt} = -\Gamma \frac{\partial \mathcal{H}}{\partial q} + F(t), \tag{13}$$

where the potential term in the Hamiltonian $\mathcal{H}$ is of the Landau form :

$$V = \frac{1}{2\Gamma}(aq^2 + \tfrac{1}{2}bq^4). \tag{14}$$

It is well-known that the instability, which causes the system to make a transition from a stable state $q = 0$ to a new stable state with a nonvanishing q, is associated with the structure of the potential sketched in figure 7. We say that as the value of the parameter a is lowered, the state $q = 0$ becomes unstable and the system goes into a new stable state by having the symmetry (present in the Hamiltonian) spontaneously broken. The concepts of broken symmetry that we are familiar with in ferromagnetism, superconductivity, quantum field theory, etc., acquires a somewhat different meaning in a system exhibiting a nonequilibrium phase transition. Here, broken symmetry means that the structure of the system becomes much richer on a macroscopic level (such as the occurrence of convective patterns observed by Benard in the case of hydrodynamic instability).

5. Chemical instabilities viewed as a critical point

Before we proceed further to exploit the similarities between the nonequilibrium phase transitions of the types mentioned here and the Landau-Ginzburg-Wilson description of an equilibrium second-order phase transition, we should like to discuss in some more detail an example of chemical instability referred to earlier in the paper. This concerns the reaction scheme studied by Zhabotinsky and Belusov —the oxidation in solution of malonic acid in the presence of cerium sulphate and potassium bromate (Glansdorff and Prigogine 1971). The striking feature of this reaction is that it leads to time-oscillation* of the concentrations of Ce^{3+} and Ce^{4+} (like in the Lotka–Volterra cycle). Such oscillations can easily be followed by spectroscopic methods (figure 8).

Even though the detailed nature of the reaction-chain is not known, the following three reactions seem to play an important role :

(i) *Oxidation of malonic acid* :

$$CH_2(COOH)_2 + 6Ce^{4+} + 2H_2O \rightarrow 2CO_2 + HCOOH + 6Ce^{3+} + 6H^+ ;$$

* There are some chemical reactions which exhibit both spatial and temporal oscillations.

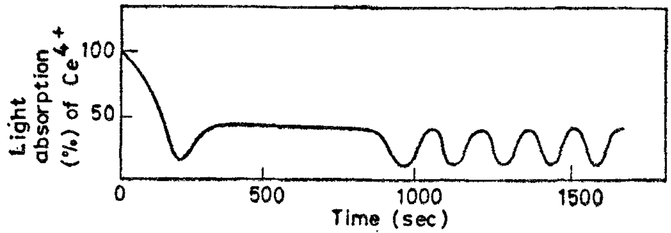

Figure 8. Time-oscillation of the concentration of Ce^{4+} (from 1000 sec onwards) in the Zhabotinsky-Belusov reaction as observed by optical methods.

(ii) *Oxidation of cerium ions* :

$$10H^{+} + 2HBrO_3 + 10Ce^{3+} \rightarrow 10Ce^{4+} + Br_2 + 6H_2O;$$

(iii) *Transformation of malonic acid into bromomalonic acid* :

$$CH_2(COOH)_2 + Br_2 \rightarrow CHBr(COOH)_2.$$

These three reactions give a qualitative explanation of the origin of oscillations. During the period of induction (indicated by the flat plataeu in figure 8), the second reaction (ii) proceeds at the same rate as the first reaction (i). This means that the loss of Ce^{4+} in the transformation

$$Ce^{4+} \rightarrow Ce^{3+},$$

is exactly compensated by the second reaction

$$Ce^{3+} \rightarrow Ce^{4+}.$$

Simultaneously, however, bromine is formed in the second reaction which combines with malonic acid in the third reaction to yield bromomalonic and dibromomalonic acids. The latter forms a complex with Ce^{3+} and therefore acts as an inhibitor of the second reaction. Thus the concentration of Ce^{4+} drops (figure 8). The inhibitive complex is however unstable and it decomposes further into carbon dioxide and dibromoacetic acid. Therefore, the second reaction starts again restoring the concentration of the Ce^{4+} ions. The cycle can go on indefinitely as long as enough reactants are present.

It may be possible to make an analysis of the Zhabotinsky–Belusov reaction based on the rate equations for two species (the Lotka–Volterra model) that we looked into earlier. Here however we examine two simpler chemical reaction models whose rate equations are of the single species type. We make an attempt to indicate how the techniques extant in the modern theories of critical phenomena can be applied to study nonequilibrium phase transitions in these reaction models.

In one such model studied by Schlögl (1972) the following chemical reactions between four species A, B, C and X are assumed :

$$A + X \rightleftharpoons 2X$$

$$B + X \rightleftharpoons C.$$

The reactions are assumed to be isothermal and homogeneous in space. The concentration a, b, c of the species A, B, C are held constant by appropriate feeding th reactor; only the concentration n of the species X can vary with time. Under the conditions, the rates of the two reactions can be written as

$$r_1 = k_1 an - k_1' n^2, \tag{15}$$

$$r_2 = k_2 bn - k_2' c, \tag{16}$$

where the k's are rate constants. A steady state is characterised by the vanishing of

$$dn/dt = r_1 - r_2 = 0. \tag{17}$$

By an appropriate choice of the units of time and concentration, we can set

$$k' = 1, \; k_1 a = 1. \tag{18}$$

In addition, introducing

$$k_2 b = b', \; k_2' c = c', \tag{19}$$

we obtain $dn/dt = - n^2 + (1 - b') n + c'$. (20)

This equation is similar to the rate equation for population dynamics of single species we studied before. In the steady state

$$c' = n_s^2 - (1 - b') n_s. \tag{21}$$

If c' is held fixed to zero, we get

$$n_s = \begin{cases} (1 - b'), & \text{for } b' < 1, \\ 0, & \text{for } b' > 1. \end{cases} \tag{22}$$

The situation is therefore similar to the phase transition in a magnet (figure 9). One can even draw an analogy between $(\sqrt{n_s})$ and the magnetisation M; b' and the temperature T ; c' and the magnetic field H. We may recall that in a magnet, the phase transition takes place for vanishing H (i.e., vanishing c' in the present case). As in the Landau model, the critical exponent β associated with the vanishing of $(\sqrt{n_s})$ at the critical point has the value 1/2 [cf, equation (22)] (Stanley 1971).

In contrast to the reaction scheme discussed above in which one finds a nonequilibrium phase transition of second order, there is another model, again studied by Schlögl (1972) in which the phase transition is of first order.* In this, the following chemical reactions are considered

$$A + 2X \rightleftarrows 3X,$$

$$B + X \rightleftarrows C.$$

The rate of the second reaction in this scheme is again given by (16), while that of the first reads

$$r_1 = k_1 an^2 - k_1' n^3. \tag{23}$$

* In the present context, the order of the transition is defined in terms of the behaviour of the order parameter near the instability point. If, for example, the order parameter goes to zero continuously at the instability point, the transition is of second order. On the other hand, if the order parameter shows a jump discontinuity, the transition is of first order.

Choosing $k' = 1$, $k_1 a = 3$, one obtains

$$dn/dt = r_1 - r_2 = -n^3 + 3n^2 - b'n + c'. \tag{24}$$

For fixed values of b' and c', there are three real positive values of n at which dn/dt vanishes. Two of them are stable while the third one is unstable. A plot of c' versus n for various values of b' corresponding to the equation

$$n^3 - 3n^2 + b'n - c' = 0, \tag{25}$$

is shown in figure 10. For $b' = 3$ and $c' = 1$, the three roots of (25) coincide and one obtains a critical point. The phase diagram in figure 10 is similar to that of a Vander Waals gas if n', c' and b' are identified respectively with v^{-1} (v = specific volume), the pressure P and the temperature T.

6. Inhomogeneity effects in chemical reactions: The time-dependent Ginzburg–Landau model

Thus far in Schlögl models discussed above, we have neglected the spatial variation of the composition. In an actual situation, such inhomogeneous fluctuations need to be included. This can be done by having a Fick's law-type diffusive term in the rate equation (Dewel *et al* 1977). One then writes

$$\frac{d}{dt} n(x, t) = D\nabla^2 n(x, t) - \mu_1 n(x, t) - \mu_2 n^3(x, t) + \zeta(x, t), \tag{26}$$

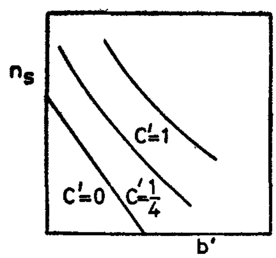

Figure 9. "Phase diagram" for the order parameter n_s vs. the field variable b' for various values of c'. The critical point occurs for $c' = 0$ and $b' = 1$.

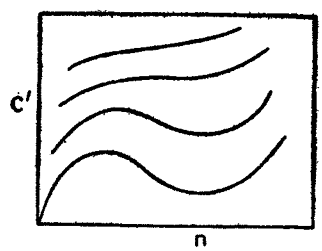

Figure 10. The analogue of a p-v diagram in a chemical reaction model (Schlögl 1972).

where D is a phenomenological diffusion constant. In (26) ζ is a noise term which takes into account various spatial and temporal fluctuations. As usual it is assumed (Ma 1976) that

$$\langle \zeta(x,(t)) \rangle = 0, \tag{27}$$

and $$\langle \zeta(x,t)\,\zeta(x',t') \rangle = 2\Gamma\delta(x-x')\,\delta(t-t'). \tag{28}$$

In the Fourier-space, the rate equation assumes the structure

$$\frac{d}{dt} n(k,t) = -\Gamma \frac{\partial \mathscr{H}}{\partial n_{-k}} + \zeta_k, \tag{29}$$

where the "Hamiltonian" $\mathscr{H}$ can be cast into the Ginzburg-Landau (Ma 1976) form :

$$\mathscr{H} = \int d^d k\,(a_2 + ck^2)\, n_k n_{-k} + \int\int\int d^d k d^d k'\, d^d k''\, a_4 n_k n_{k'} n_{k''} n_{-k-k'-k''}, \tag{30}$$

d being the space-dimensionality. As is well-known, equations such as (28) are commonly used for studying dynamical effects near an ordinary second-order phase transition (Ma 1976).

The question may be raised : what does one achieve by making a formal connection, as outlined above, between a nonequilibrium phase transition in a chemical reaction on the one hand, and the dynamical critical point phenomena? The answer seems to be the following. It may be possible, by making use of dynamical scaling (Ma 1976) to group a variety of chemical reactions under one "universality class". One is also able to employ the established machinery of ϵ-expansion to determine the critical exponents associated with the non-analytic behaviour of certain quantities in a chemical reaction near some points of instability (Dewel *et al* 1977; Nitzan 1977). It is possible, for example, to establish the so-called Ginzburg criterion for the size of the critical region, expressed in terms of a feeding rate of chemical reactants as a control parameter inside which the mean field theory fails (Nitzan 1977).

7. Conclusions

In this article, we have discussed the occurrence of nonequilibrium phase transitions in myriad of problems scattered over many disciplines. We have also outlined, in a rather qualitative manner, an analysis of these phenomena based on the established methods of equilibrium second-order phase transitions. Such analyses have been carried out, in addition to the examples cited here, for the instabilities in tunnel diode (Landauer 1971), the Gunn effect (Landauer 1975), parametric oscillator (Landauer and Woo 1972) and the Wien bridge oscillator (Horn *et al* 1976). More recently, Bishop and Trullinger (1978) have looked into the behaviour of the thermal noise voltage near the threshold current in a dc Josephson junction in the light of a second-order phase transition. Here, the mean thermal-noise voltage plays the role of the order parameter while the applied current takes the role of the temperature.

In metallurgy too, it is not uncommon to find the appearance of well-organised spatial and temporal structures in systems that are nonlinear and dissipative. We mention here the highly ordered serrated pattern seen in the stress-strain diagram in, for example, brass when the strain rate (the control parameter, in this case) is increased beyond a certain limit (Ardley and Cottrell 1953). Another example of the formation of ordered spatial structure is the so-called void lattice in irradiated metals. The voids, which result from aggregation of mobile vacancies generated during irradiation at high doses and at high temperatures, generally appear to be randomly dispersed in the medium (Cawthorne and Fulton 1967). However, under favourable experimental conditions, voids can grow at regularly distributed sites, forming a three-dimensional array of extremely high spatial coherence (see for example Evans 1971). The rate equations governing the agglomeration of vacancies may be written in the form of (4) discussed before and the formation of voids of a certain size may be viewed as the outcome of a selection process (similar to the one in a multimode laser) brought about by the nonlinear terms in the equations. The subsequent arrangement of the voids in a regularly faceted superlattice must of course have to be the result of an interaction between the voids themselves, all of the same size now. Attempts are underway now in our laboratory to investigate the problems of serrated yielding and void lattice mentioned here, on the basis of rate equations of the types (1)–(4), and to view these phenomena as the outgrowth of some kinds of instabilities associated with a nonequilibrium phase transition.

We conclude by remarking again that a great many systems in various disciplines, which exhibit some kinds of ordered structures, can be treated by similar concepts and mathematical tools, and that at a macroscopic level, there exist fascinating analogies between them. This is useful because it may enable one to study the behaviour of complex systems by establishing analogies with more clearly understood ones. Another important point which needs additional remarks is the one concerning the role of fluctuations—both spatial and temporal—in the various systems discussed here. One may adopt the point of view that in a system like the Lotka–Volterra model of population dynamics, fluctuations play no manifestly important role and therefore attempts to understand this system by making an analogy with critical point phenomena may be completely unnecessary. While it is true that the stable solutions of nonlinear equations such as (2) and (3) can be obtained directly without invoking any fluctuation term in the equations, the question as to which stable solution the system chooses cannot, in general, be answered without studying the nature of fluctuations. Also a proper understanding of the role of fluctuations may allow one to forecast the occurrence of a spontaneous transition of the system from one state to another. Recall that in a ferromagnet the ordinary mean field theory of Landau is quite adequate to predict that below the Curie point, the system develops an instability and goes into an ordered state. However, it is precisely the fluctuations which trigger this instability and tip the balance in favour of one state than another. This is of course what necessitates the introduction of a "diffusive" term or a "kinetic energy" type term in the Ginzburg–Landau model [cf., equation (30)], and an additional noise term [cf., equation (26)], for a time-dependent description. These terms are well-known to have very important effects in determining certain properties of the system within the so-called critical region. True, it may be diffcult to estimate the size of the "critical region" for a model of population dynamics; nevertheless, the appearance of fluctuation terms in the rate equations for such a model is quite physical.

For example, a realistic model for population study *within* a certain locality has to take into account sudden influx or outflux of the population (i.e., population-diffusion) due to random " extraneous " events. So again in a certain sense, it is the interaction between the subsystem (the population within a certain region, in this case) and the surrounding world at large (the latter playing the role of a " heat bath ") that gives rise to fluctuations.

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