

Y. Kuramoto

Chemical Oscillations, Waves, and Turbulence



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Preface

This book is intended to provide a few asymptotic methods which can be applied to the dynamics of self-oscillating fields of the reaction-diffusion type and of some related systems. Such systems, forming cooperative fields of a large number of interacting similar subunits, are considered as typical synergetic systems. Because each local subunit itself represents an active dynamical system functioning only in far-from-equilibrium situations, the entire system is capable of showing a variety of curious pattern formations and turbulencelike behaviors quite unfamiliar in thermodynamic cooperative fields. I personally believe that the nonlinear dynamics, deterministic or statistical, of fields composed of similar active (i.e., non-equilibrium) elements will form an extremely attractive branch of physics in the near future.

For the study of non-equilibrium cooperative systems, some theoretical guiding principle would be highly desirable. In this connection, this book pushes forward a particular physical viewpoint based on the *slaving principle*. The discovery of this principle in non-equilibrium phase transitions, especially in lasers, was due to Hermann Haken. The great utility of this concept will again be demonstrated in this book for the fields of coupled nonlinear oscillators.

The topics I have selected strongly reflect my personal interest and experiences, so that this book should not be read as a standard textbook. Nevertheless, the spirit by which the present theory is guided may stimulate those students in various fields of science who are fascinated at all by the curiosity of the self-organization in nature.

I am particularly grateful to Professor H. Haken who initially suggested that I write a book on this subject. I wish to thank Dr. H. Lotsch of Springer-Verlag for his patience in waiting for my never-ending manuscript. I am also indebted to Mrs. K. Honda for painstaking typing assistance.

Kyoto, February 1984

Yoshiki Kuramoto

Contents

1. Introduction	1
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Part I **Methods**

2. Reductive Perturbation Method	5
2.1 Oscillators Versus Fields of Oscillators	5
2.2 The Stuart-Landau Equation	8
2.3 Onset of Oscillations in Distributed Systems	13
2.4 The Ginzburg-Landau Equation	17
3. Method of Phase Description I	22
3.1 Systems of Weakly Coupled Oscillators	22
3.2 One-Oscillator Problem	24
3.3 Nonlinear Phase Diffusion Equation	28
3.4 Representation by the Floquet Eigenvectors	29
3.5 Case of the Ginzburg-Landau Equation	32
4. Method of Phase Description II	35
4.1 Systematic Perturbation Expansion	35
4.2 Generalization of the Nonlinear Phase Diffusion Equation	41
4.3 Dynamics of Slowly Varying Wavefronts	46
4.4 Dynamics of Slowly Phase-Modulated Periodic Waves	54

Part II **Applications**

5. Mutual Entrainment	60
5.1 Synchronization as a Mode of Self-Organization	60
5.2 Phase Description of Entrainment	62
5.2.1 One Oscillator Subject to Periodic Force	62
5.2.2 A Pair of Oscillators with Different Frequencies	65
5.2.3 Many Oscillators with Frequency Distribution	66
5.3 Calculation of Γ for a Simple Model	67
5.4 Soluble Many-Oscillator Model Showing Synchronization- Desynchronization Transitions	68

5.5 Oscillators Subject to Fluctuating Forces	78
5.5.1 One Oscillator Subject to Stochastic Forces	78
5.5.2 A Pair of Oscillators Subject to Stochastic Forces	80
5.5.3 Many Oscillators Which are Statistically Identical	82
5.6 Statistical Model Showing Synchronization-Desynchronization Transitions	82
5.7 Bifurcation of Collective Oscillations	84
6. Chemical Waves	89
6.1 Synchronization in Distributed Systems	89
6.2 Some Properties of the Nonlinear Phase Diffusion Equation	91
6.3 Development of a Single Target Pattern	93
6.4 Development of Multiple Target Patterns	101
6.5 Phase Singularity and Breakdown of the Phase Description	103
6.6 Rotating Wave Solution of the Ginzburg-Landau Equation	106
7. Chemical Turbulence	111
7.1 Universal Diffusion-Induced Turbulence	111
7.2 Phase Turbulence Equation	114
7.3 Wavefront Instability	120
7.4 Phase Turbulence	127
7.5 Amplitude Turbulence	132
7.6 Turbulence Caused by Phase Singularities	137
Appendix	141
A. Plane Wave Solutions of the Ginzburg-Landau Equation	141
B. The Hopf Bifurcation for the Brusselator	144
References	149
Subject Index	155

1. Introduction

Mathematically, a reaction-diffusion system is obtained by adding some diffusion terms to a set of ordinary differential equations which are first order in time. The reaction-diffusion model is literally an appropriate model for studying the dynamics of chemically reacting and diffusing systems. Actually, the scope of this model is much wider. For instance, in the field of biology, the propagation of the action potential in nerves and nerverlike tissues is known to obey this type of equation, and some mathematical ecologists employ reaction-diffusion models for explaining various ecological patterns observed in nature. In some thermodynamic phase transitions, too, the evolution of the local order parameter is governed by reaction-diffusion-type equations if we ignore the fluctuating forces.

One important feature of reaction-diffusion fields, not shared by fluid dynamical systems as another representative class of nonlinear fields, is worth mentioning. This is the fact that the total system can be viewed as an assembly of a large number of identical local systems which are coupled (i.e., diffusion-coupled) to each other. Here the local systems are defined as those obeying the diffusionless part of the equations. Take for instance a chemical solution of some oscillating reaction, the best known of which would be the Belousov-Zhabotinsky reaction (Tyson, 1976). Let a small element of the solution be isolated in some way from the bulk medium. Then, it is clear that in this small part a limit cycle oscillation persists. Thus, the total system may be imagined as forming a diffusion-coupled field of similar limit cycle oscillators.

We now turn to a Navier-Stokes fluid for comparison. The flow may be oscillatory as in, e.g., the Taylor vortex flow (DiPrima and Swinney, 1981). In this case, however, it is apparently impossible to imagine such local dynamical units as persistent oscillatory motion even after isolation. After all, every term on the right-hand side of the Navier-Stokes equation represents "interaction" because it involves a spatial gradient. In this respect, reaction-diffusion systems bear some resemblance to thermodynamic cooperative fields which are also composed of similar subunits such as atoms, molecules, and magnetic spins or, in a coarse-grained picture, semi-macroscopic local order parameters. Furthermore, it may happen in reaction-diffusion systems that the global dynamical behavior is predicted, at least qualitatively, on the basis of the known nature of each local dynamical system, whereas such a synthetic view hardly seems to apply to fluid systems. It is also expected that the bulk behavior is much less sensitive to the boundary effects in reaction-diffusion systems than in fluid systems. This implies that in the former case the study of infinitely large systems would be of primary importance; although different system geometries and boundary conditions may

provide a variety of intriguing mathematical problems, they seem to be of secondary importance at least from the physical point of view which we take in this book.

We noted the analogy between reaction-diffusion systems and thermodynamic cooperative systems. However, the former differ essentially from the latter in that each local subsystem can operate in far-from-equilibrium situations so that it may already represent a very active functional unit. It is this difference that makes reaction-diffusion media capable of exhibiting the wealth of self-organization phenomena including turbulence never met in equilibrium or near-equilibrium cooperative systems. In this book, we will concentrate on the fields of *oscillatory* units which are coupled through diffusion or some other interactions. For a variety of other aspects of reaction-diffusion systems, one may refer to Fife's book (1979a).

It may now be asked what sort of self-organization phenomena are expected in this type of field. In considering this problem, the importance of the concept "synchronization" or "entrainment" cannot be emphasized enough. This simply means that multiple periodic processes with different natural frequencies come to acquire a common frequency as a result of their mutual or one-sided influence. In some literature, the former term is used in the more restrictive sense of the oscillators' phases also being pulled close to each other. In this book, however, we will not be very strict in this respect because the very definition of relative phase between two given oscillators, especially when they represent oscillators of a different nature, is rather arbitrary. The importance of the function of synchronization in the self-organization in nature may be realized from the fact that what looks like a single periodic process on a macroscopic level often turns out to be a collective oscillation resulting from the mutual synchronization among the tremendous number of the constituent oscillators. The human heartbeat may serve as an example of such a phenomenon. Because the component oscillators in nature would never possess identical natural frequencies and, moreover, would never be free from environmental random fluctuations, mutual synchronization appears to be the unique possible mechanism for producing and maintaining macroscopic rhythmicity. The problem of the onset of collective oscillation in oscillator aggregates will be treated in Chap. 5.

Based equally on mutual synchronization, chemical wave propagation in oscillatory reaction-diffusion systems generates an even more complicated class of phenomena than the mere collective oscillations. Here again, the entire field may be entrained into an identical frequency, whereas the local phases of oscillation may have different values. Such a view, although a little too idealized, enables us to understand the origin of expanding target patterns as observed in the Belousov-Zhabotinsky reaction. As implied from a number of problems in condensed-matter physics and field theory, some field quantity for which the phase is definable is expected to allow for topological defects arising from, e.g., phase jumps and phaseless points. Just as in superfluid helium and plane rotator systems, there exist in oscillatory reaction-diffusion systems, too, vortexlike modes which, in the latter case, develop into rotating spiral waves such as those known in the Belousov-Zhabotinsky reaction. A simple theory of chemical waves from the viewpoint of spatio-temporal synchronization and phase singularity will

be developed in Chap. 6. Granted that mutual synchronization represents a key mechanism in the self-organization in oscillatory media, it would be interesting to ask what is brought about by its breakdown. This will partly be answered in Chap. 7 where we try to relate it to the onset of turbulence-like behavior.

It is unfortunate that only little progress has been achieved in the past towards the understanding of synchronization, pattern formation, and turbulence in nonlinear self-oscillatory media and related many-body systems, in spite of their great potential importance in the future. Although the present theory, too, is far from complete, a particular physical viewpoint at least will be seen to underlie the present book. In Part 1, such a viewpoint will be formulated into some asymptotic methods, while Part 2 may simply be looked upon as their demonstration through a number of specific problems. The underlying physics is closely related to the slaving principle, whose conceptual importance in nonlinear dissipative dynamics in general was emphasized by Haken and first demonstrated by him in laser theory (Haken and Sauermann, 1963; Haken, 1983 a, b). Basically, the slaving principle claims the possibility of eliminating a large number of rapidly decaying degrees of freedom. This principle manifests itself most clearly near the bifurcation points where the system experiences a qualitative change in dynamical behavior. The possibility of a great reduction of the number of effective degrees of freedom and the resulting universality of the evolution law form the physical basis of why the bifurcation theory can serve as a most powerful tool in treating various self-organization phenomena.

It should be noted, however, that the slaving principle is such a general concept that the standard bifurcation theory can embody only a part of this concept on a more or less firm mathematical basis. Thus, the theory developed in this book, although being based on the slaving principle, is not so much based on the standard bifurcation theory. In fact, the kinds of self-organization phenomena and turbulence we want to treat here are rather complicated and require so many effective degrees of freedom that standard bifurcation theory does not seem to be of much help. As a possible alternative, one may think of the bifurcation theory of higher codimensions, which has shown an interesting development in recent years (e.g., Guckenheimer, 1981). Unfortunately, however, the effective degrees of freedom involved are still too few for our purposes. We are rather interested in, so to speak, the bifurcations with *infinitely* high degeneracy, which can in fact cover some physical problems of our concern. Although no rigorous bifurcation theory seems available for such cases, this peculiar kind of bifurcation is of much practical importance. This is because it arises quite commonly in systems with great spatial extension, especially when the instability first occurs for disturbances of sufficiently long spatial scales. As inferred from the fact that the eigenvalue spectrum of the fluctuations around the subcritical state is then almost continuous, the system dynamics can never be confined to a few-dimensional manifold even in the vicinity of the bifurcation point. Or it may be better to say that if the range of applicability of the usual bifurcation theory is measured by some bifurcation parameter, it will be narrowed down to zero as the system extension goes to infinity. Even in such highly degenerate bifurcations, there exist a tremendous number of degrees of freedom which are rapidly decaying and hence follow adiabatically the continuum of long-scale modes. Thus, the

idea of the slaving principle itself is expected to work. Although lacking a rigorous mathematical basis, some practical methods of dynamical reduction appropriate for highly degenerate bifurcations were developed in some fluid-dynamical problems such as the plane Poiseuille flow (Stewartson and Stuart, 1971) and the Rayleigh-Bénard convection (Newell and Whitehead, 1969). In Chap. 2 we apply this kind of approach to simpler systems, i.e., oscillatory reaction-diffusion systems, with a special emphasis on its formalistic contrast to the ordinary Hopf bifurcation theory.

The utility of the slaving principle is by no means restricted to near-bifurcation situations. In fact, in connection with the present concern, the slaving principle is also applicable to systems of *weakly* coupled limit cycle oscillators in general. A theoretical framework particularly suited for weakly coupled oscillators and systems analogous to them will be presented in Chaps. 3 and 4. Here the simplification of the dynamics comes essentially from the fact that the amplitude disturbances decay much faster than the phase disturbances. In conclusion, the slaving principle enables us to contract the original dynamics to a much simpler one which still retains a sufficiently large number of effective degrees of freedom to admit a variety of self-organization and turbulent phenomena.

Finally, one should keep in mind the severe limitation of the present methods in that they apply only to those phenomena associated with sufficiently long space-time scales. It is under this restriction that the present theory can enjoy its coherent character. A number of important phenomena in self-oscillating fields, especially those for which the coexistence of short-scale and long-scale spatial variations are important, are omitted. An important problem of this kind would be the propagation of trigger waves in reaction-diffusion systems, i.e., waves which are typical in non-oscillating excitable media but may arise also in systems of highly distorted oscillations or relaxation oscillations. Some simple classes of phenomena related to trigger waves may be dealt with by a method quite different from the present ones, for which the reader may refer to Ortoleva and Ross (1975) and Fife (1976a, b, 1979b).

2. Reductive Perturbation Method

Small-amplitude oscillations near the Hopf bifurcation point are generally governed by a simple evolution equation. If such oscillators form a field through diffusion-coupling, the governing equation is a simple partial differential equation called the Ginzburg-Landau equation.

2.1 Oscillators Versus Fields of Oscillators

Many theories on the nonlinear dynamics of dissipative systems are based on the first-order ordinary differential equations

$$\frac{dX_i}{dt} = F_i(X_1, X_2, \dots, X_n; \mu), \quad i = 1, 2, \dots, n,$$

which include some parameters represented by μ ; a more convenient vector form

$$\frac{dX}{dt} = F(X; \mu) \tag{2.1.1}$$

is sometimes preferred. As a specific example, we mention the dynamics of chemical reaction systems which are maintained uniformly in space. In this case, X usually represents a set of concentrations of the chemical species involved, and μ may be taken to be the flow rate at which certain chemicals are constantly fed into the system so that their consumption due to reactions may be compensated.

For some range of μ , the system may stay stable in a time-independent state. In particular, this is usually the case for macroscopic physical systems which lie sufficiently close to thermal equilibrium. In many systems, such a steady state loses stability at some critical value μ_c of μ , and beyond it (say $\mu > \mu_c$), gives way to periodic motion. In the parameter-amplitude plane, this appears as a branching of time-periodic solutions from a stationary solution branch, and this phenomenon is generally called the *Hopf bifurcation*. For various mathematical aspects of the Hopf bifurcation, one may refer to the book by Marsden and McCracken (1976). In chemical reactions, the corresponding phenomenon is called the onset of *chemical oscillations*. Besides chemical reactions, one may point out many examples from electrical and mechanical engineering, optics, biology, biochemistry, and possibly some other fields, for which ordinary-dif-

ferential-equation models form a natural basis for mathematical analysis, so that the appearance of oscillations may be understood in the way stated above.

As μ increases further, the system may show more and more complicated dynamics through a number of bifurcations. It may show complicated periodic oscillations, quasi-periodic oscillations or a variety of non-periodic behaviors. For instance, we know of the recent discoveries of fantastic bifurcation structures in the spatially homogeneous Belousov-Zhabotinsky reaction, see Hudson et al., 1979.

Coming back to limit cycle oscillations shown by systems of ordinary differential equations, this simple mode of motion still seems to deserve some more attention, especially in relation to its role as a basic functional unit from which various dynamical complexities arise. This seems to occur in at least two ways. As mentioned above, one may start with a simple oscillator, increase μ , and obtain complicated behaviors; this forms, in fact, a modern topic. However, another implication of this dynamical unit should not be left unnoticed. We should know that a limit cycle oscillator is also an important component system in various self-organization phenomena and also in other forms of spatio-temporal complexity such as turbulence. In this book, particular emphasis will be placed on this second aspect of oscillator systems. This naturally leads to the notion of the “many-body theory of limit cycle oscillators”; we let many oscillators contact each other to form a “field”, and ask what modes of self-organization are possible or under what conditions spatio-temporal chaos arises, etc. A representative class of such many-oscillator systems in theory and practical application is that of the fields of diffusion-coupled oscillators (possibly with suitable modifications), so that this type of system will primarily be considered in this book.

In any case, we should begin with some investigation of the component systems, i.e., limit cycle oscillators. Although the specific feature of limit cycle oscillations (e.g., orbital forms, oscillation patterns, etc.) may vary greatly from system to system, there exists one remarkable universal fact, namely, that all systems come to behave in a similar manner sufficiently close to the onset of oscillations. Mathematicians may say that this is a consequence of the *center manifold theorem*. More physically, we are left with only a couple of relevant dynamical variables close to criticality, whose time scales are distinguishably slower than those of the remaining variables, so that the latter can be eliminated adiabatically. As a result, (2.1.1) is contracted to a very simple universal equation which is sometimes called the Stuart-Landau equation. In fact, Landau was the first to conjecture the equation form (Landau, 1944), and Stuart was the first to derive it through an asymptotic method (Stuart, 1960). In quite a different context, specifically in laser theory, Haken and Sauermann (1963) derived a similar but more general equation. We shall outline in Sect. 2.2 how the Stuart-Landau equation is derived. The fact that dynamical systems can be reduced to some simple universal systems is by no means restricted to this particular bifurcation type. However, we do not intend in this book to present theories from such a general viewpoint. The method employed in Sect. 2.2 is a well-known multi-scale method, although there may be some possible variants leading to identical results. A practical use of the theory in Sect. 2.2 lies in the fact that it enables

us to calculate explicitly a certain constant (called the Landau constant) appearing in the Stuart-Landau equation, whose sign determines the stability of the bifurcating periodic solution. Otherwise, the Stuart-Landau equation itself is not likely to arouse much theoretical interest, although it may have some value in serving as an ideal nonlinear oscillator model.

So far, the discussion has been concerned with systems of *ordinary* differential equations. In many physical problems, *partial* differential equations describing processes in the space-time domain prove to be a more useful mathematical tool. For instance, one may mention the Navier-Stokes fluids, chemical reactions including diffusion, some ecological systems with migration, etc. Suppose that oscillatory motions occur in any of these continuous media as some control parameter is varied, and consider how to describe them. It is true that if the system is confined within a finite volume, the governing partial differential equations can, in principle, be transformed into a discrete set of ordinary differential equations, which describe the evolution of the amplitudes of the basis functions satisfying prescribed boundary conditions. Although the system then involves an infinite number of degrees of freedom, a mode-truncation approximation is usually allowed. Thus, as far as the onset of oscillations is concerned, there seems to be nothing theoretically new, compared to the bifurcation theory for systems of ordinary differential equations. Specifically, the application of a multi-scale method will lead to a Stuart-Landau equation again. (For a mathematical theory of the Hopf bifurcation for systems of partial differential equations in bounded domains, see Joseph and Sattinger, 1972; bifurcation analyses of reaction-diffusion systems have been developed by Auchmuty and Nicolis, 1975, 1976, and Herschkowitz-Kaufman, 1975.)

There may be some situations, however, where keeping to formal bifurcation theories easily makes us overlook a fact of considerable physical importance. The situation of particular interest in this connection seems to be when the system size is very large. Then, formal bifurcation techniques applied near μ_c cannot claim full validity except in an extremely limited parameter range about μ_c . This is basically because the eigenvalue spectrum obtained from the linearization about the reference steady state is almost continuous for large system size, so that, in addition to the couple of modes which are becoming unstable, a large number of degrees of freedom come into play as soon as μ deviates from μ_c (a more detailed description will be given in Sect. 2.3). Thus it is desirable that the Stuart-Landau equation be generalized so as to cover such circumstances. People in the field of fluid mechanics have developed theories in this direction, which proved to be very useful in understanding instabilities (not restricted to the Hopf type) arising in systems with large dimensions at least in one or two directions. Typical examples are the Newell-Whitehead theory (1969) on a fluid layer heated from below with infinite aspect ratio, and the Stewartson-Stuart theory (1971) on plane Poiseuille flow. In these theories, one works with partial differential equations throughout, not transforming them into ordinary differential equations. A method was contrived to reduce the equations to a generalized form of the Stuart-Landau equation, thereby admitting slow spatial and temporal modulation of the envelope of the bifurcating flow patterns. We call that equation the Ginzburg-Landau equation (named after a similar equation appearing in super-

conductivity) or the Stewartson-Stuart equation. In this book we adopt the former name.

Independently of the hydrodynamical context, the Ginzburg-Landau equation was derived by Graham and Haken (1968, 1970) in multimode lasers as a further development of the Haken-Sauermann theory (1963); it should be noted that fluctuations are included in most of their series of works. For various non-equilibrium phase transitions described by the Ginzburg-Landau-type equation, see the review article by Haken (1975b) and his more recent monograph (1983).

The derivation of the Ginzburg-Landau equation usually involves the method of multiple scales (in space and time), and again there are some variants in technical details. For convenience, we sometimes call all the related techniques involving the use of stretched space-time coordinates *the reductive perturbation method*, a term originally coined for a systematic method of deriving various nonlinear wave equations mainly in dissipationless media (Taniuti and Wei, 1968; Taniuti, 1974). It is now widely known that the Ginzburg-Landau equation is not only related to a few fluid mechanical or optical problems but that it can be deduced from a rather general class of partial differential equations (Newell, 1974; Haken, 1975a; Gibbon and McGuinness, 1981; Lin and Kahn, 1982). Chemical reactions with diffusion form a simple and particularly interesting class of systems in this connection (Kuramoto and Tsuzuki, 1974; Wunderlin and Haken, 1975), and we shall derive in Sect. 2.4 the Ginzburg-Landau equation for general reaction-diffusion systems. Just as the Stuart-Landau equation describes the simplest nonlinear oscillator, so the Ginzburg-Landau equation describes the simplest *field* of nonlinear oscillators. In later chapters, this equation will be frequently invoked in discussing chemical waves and chemical turbulence.

2.2 The Stuart-Landau Equation

In this section, we outline how a small-amplitude equation valid near the Hopf bifurcation point is derived from the general system of ordinary differential equations (2.1.1).

Let X and F be n -dimensional real vectors and μ a real scalar parameter. Let $X_0(\mu)$ denote a steady solution of (2.1.1) or

$$F(X_0(\mu); \mu) = 0.$$

We express (2.1.1) in terms of the deviation $u \equiv X - X_0$ in a Taylor series:

$$\frac{du}{dt} = Lu + Muu + Nuuu + \dots, \quad (2.2.1)$$

where L denotes the Jacobian matrix whose ij th element is given by $L_{ij} = \partial F_i(X_0)/\partial X_{0j}$; the abbreviations Muu and $Nuuu$, etc., indicate vectors whose i th components are given by

$$(Mu u)_i = \sum_{j,k} \frac{1}{2!} \frac{\partial^2 F_i(X_0)}{\partial X_{0j} \partial X_{0k}} u_j u_k,$$

$$(Nu u u)_i = \sum_{j,k,l} \frac{1}{3!} \frac{\partial^3 F_i(X_0)}{\partial X_{0j} \partial X_{0k} \partial X_{0l}} u_j u_k u_l,$$

and the higher-order terms in u may be expressed similarly. We shall later use quantities like Muv and $Nuvw$ for different vectors u , v and w , and their definitions may be understood as an obvious extension of the above. Note, in particular, that Muv and $Nuvw$ are symmetric functions of u , v and w . Note also that the expansion coefficients, which are symbolically expressed by M , N , etc., generally depend on μ at least through $X_0(\mu)$.

Suppose that μ is varied in some range about $\mu = 0$. We assume that up to $\mu = 0$ the solution X_0 remains stable to sufficiently small perturbations, while it loses stability for $\mu > 0$. Consider the linear eigenvalue problem associated with (2.2.1), i.e.,

$$Lu = \lambda u. \quad (2.2.2)$$

The stability of X_0 is related to the distribution of the eigenvalues λ in the complex plane. By assumption, this distribution changes with μ in the following way: all λ stay in the left half-plane if $\mu < 0$, and at least one eigenvalue crosses the imaginary axis at $\mu = 0$. Since the eigenvalues are given by the zeros of an n th-order polynomial with real coefficients, we have the following two general possibilities: (a) one eigenvalue on the real axis crosses the origin (Fig. 2.1 a), (b) a pair of complex-conjugate eigenvalues cross the imaginary axis simultaneously (Fig. 2.1 b). In each case, the eigenvalues are assumed to have nonzero transversal "velocity" when crossing the imaginary axis, or

$$\left. \frac{d \operatorname{Re}\{\lambda(\mu)\}}{d\mu} \right|_{\mu=0} > 0.$$

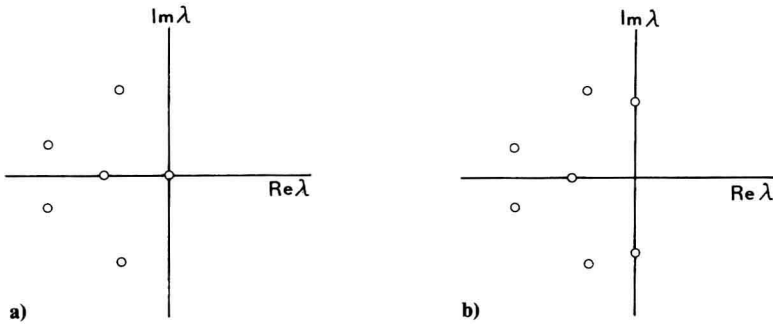


Fig. 2.1 a, b. Two typical distributions of the eigenvalues at criticality

Furthermore, the rest of the eigenvalues are assumed to remain at a nonzero distance from the imaginary axis. In the following, we shall restrict our attention to case (b), since this corresponds to the Hopf bifurcation.

Near criticality, the matrix L may be developed in powers of μ :

$$L = L_0 + \mu L_1 + \mu^2 L_2 + \dots \quad (2.2.3)$$

To save notation, let $\lambda(\mu)$ denote a special eigenvalue which is becoming critical rather than denoting a general one, and $\bar{\lambda}(\mu)$ its complex conjugate (we use a bar to signify a complex conjugate throughout). We assume a power-series expansion for λ also:

$$\lambda = \lambda_0 + \mu \lambda_1 + \mu^2 \lambda_2 + \dots, \quad (2.2.4)$$

where λ_ν are generally complex, or $\lambda_\nu = \sigma_\nu + i\omega_\nu$. By assumption,

$$\sigma_0 = 0, \quad \sigma_1 > 0.$$

Let U denote the right eigenvector of L_0 corresponding to the eigenvalue $\lambda_0 (= i\omega_0)$:

$$L_0 U = \lambda_0 U, \quad L_0 \bar{U} = \bar{\lambda}_0 \bar{U}.$$

Similarly, the left eigenvector is denoted by U^* :

$$U^* L_0 = \lambda_0 U^*, \quad \bar{U}^* L_0 = \bar{\lambda}_0 \bar{U}^*,$$

where $U^* \bar{U} = \bar{U}^* U = 0$, and these vectors are normalized as $U^* U = \bar{U}^* \bar{U} = 1$. Note that λ_0 and λ_1 are expressed as

$$\lambda_0 = i\omega_0 = U^* L_0 U, \quad (2.2.5a)$$

$$\lambda_1 = \sigma_1 + i\omega_1 = U^* L_1 U. \quad (2.2.5b)$$

It is convenient to define a small positive parameter ε by $\varepsilon^2 \chi = \mu$, where $\chi = \text{sgn } \mu$; ε is considered to be a measure of the amplitude to lowest order, so that one may assume the expansion

$$u = \varepsilon u_1 + \varepsilon^2 u_2 + \dots \quad (2.2.6)$$

The expression in (2.2.3) now becomes

$$L = L_0 + \varepsilon^2 \chi L_1 + \varepsilon^4 L_2 + \dots \quad (2.2.7)$$

Similarly, for some higher-order expansion coefficients in (2.2.1), we write symbolically