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General properties of systems with competing interactions

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Abstract. We consider physical systems exhibiting a continuous and monotonic response to their driving fields and study the effects of the interaction between two such well behaved systems. The interaction (cross coupling between the systems) is described by effective fields and is assumed linear. We show that the coupling between the two systems gives rise to a rich variety of phenomena: negative differential susceptivity, phase transitions, bistability and other less usual features. All these effects, absent in the uncoupled systems, cannot be ascribed to an especially 'dangerous' form of coupling between the systems, but must be considered a general property of the cross coupling between two systems.

1. Introduction

A number of 'strange' phenomena, many of which are phase-transition-like (e.g. bistability, re-entrant behaviour, onset of instabilities . . .) considered at first as unusual features exhibited only by some particular system, are now currently observed in an increasing number of physical systems. In short we say that a physical system exhibits a 'strange' effect if we observe any kind of departure from continuity, monotonicity or single-valuedness in the response of the system to its driving field.

The formulation of dedicated theories has accounted for the occurrence of unusual phenomena in several systems and often a good agreement with the experimental data has been obtained. However, dedicated theories start from a detailed description of the interactions and of the processes taking place within the system of interest. This approach in general yields nonlinear equations, so that it is not easy to find analogies among theories formulated for different kinds of physical systems. A relevant idea that is beginning to spread around is that many 'strange' effects are to be ascribed to some general principle and not to the details of the models. This general principle is the interplay between different kinds of ordering within the same physical system. Literature reports where the coupling between order parameters is studied with no reference to a detailed description of some physical system are extremely rare. Perhaps the only remarkable paper of this kind is that by Imry (1975) who studied a Landau model with two order parameters featuring a biquadratic coupling between them. Imry found that such a system (in the absence of an external field) may exhibit several phase transitions as a function of temperature.

In this paper we study a model system which is based on much looser assumptions than Imry's. The model worked out in this paper is of the kind introduced by Micciancio and Vassallo (1982, hereafter referred to as I). In this model system, whose block diagram is shown in figure 1, two subsystems, F_1 and F_2, are driven respectively by their effective fields V_1 and V_2 which are functions of the other subsystem. In this

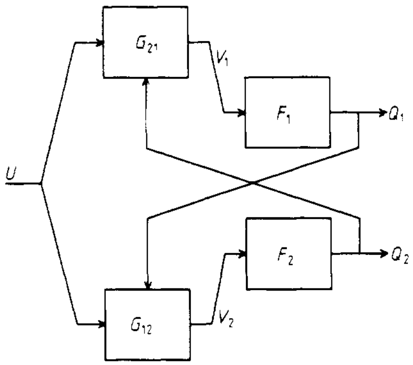

Figure 1. Block diagram of a system exhibiting two responses, Q_1 and Q_2, to the system driving field U. Q_1 and Q_2 are physical quantities providing a macroscopic description of two different processes taking place within the same system. Each process is driven by its effective fields, V_1 and V_2, respectively. Accordingly the system is decomposed in two subsystems (F_1, F_2) driven by effective fields which are functions of the system driving field U and of the response Q_j of the other subsystem. This block diagram is one of the simplest models of a system exhibiting two responses to a stimulus.

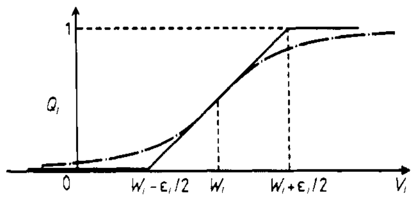

Figure 2. The broken curve is an example of continuous and monotonic response of a subsystem to its input field V_i. The response Q_i exhibits saturation at high and low values of V_i. The full curve is a piecewise linear approximation of the broken curve. This approximation, used in the text to get an analytical solution of the equations of the system shown in figure 1, produces little or no qualitative effects on the system behaviour.

paper we relax the assumption of I that at least one of the subsystems exhibits a first-order phase transition and show that even if the responses $Q_i = F_i(V_i)$ of both subsystems are continuous and monotonic functions of V_i (see the broken curve in figure 2) the system of figure 1 exhibits a variety of phase transitions and other anomalies (like negative differential susceptivity).

The main result of this work is that even a linear coupling between two well behaved systems is able to produce a large variety of 'strange' effects.

2. Theory

2.1. The model

The equations ruling the system of figure 1 are formally written as

$$V_i = G_{ji}(U, Q_j) \qquad (1)$$

$$Q_j = F_j(V_j) \qquad (2)$$

$$(i, j) \in (1, 2),\ i \neq j.$$

If the coupling functions $G_{ij}(U, Q_i)$ are strictly monotonic continuous functions of both U and Q_j, according to I equation (1) can be approximated with a Taylor series truncated to linear terms

$$V_i = U + K_{ji}Q_j. \tag{3}$$

The constants K_{12} and K_{21} defined in (3) will be referred to as 'coupling constants' and the plane spanned by them as the 'K plane'.

We assume that the response Q_i of subsystem i, equation (2), is a continuous and monotonic function of the input field V_i. We also assume that $F_i(V_i)$ exhibits saturation at high and low values of V_i, as shown by the broken curve of figure 2. This kind of response is often met in real physical systems and may be ascribed, e.g., to the finite number of particles in the system. For simplicity we assume that $0 \leq Q_i \leq 1$ and that U is defined over all the real axis.

We are primarily interested in the qualitative effects produced by the intersystem coupling present within the system of figure 1; to this end an analytical solution of the system equations (3) and (2) is desirable. However, even in the case that the nonlinear functions $F_i(V_i)$ (broken curve of figure 2) have the simplest algebraic form, a numerical solution of the system equations seems unavoidable. To overcome these difficulties we linearise piecewise the functions $F_i(V_i)$ as shown by the full curve of figure 2 and rewrite (2)

$$Q_i = 0, \qquad V_i < W_i - \tfrac{1}{2}\varepsilon_i, \tag{4a}$$

$$Q_i = \tfrac{1}{2} + (V_i - W_i)/\varepsilon_i, \qquad W_i - \tfrac{1}{2}\varepsilon_i < V_i < W_i + \tfrac{1}{2}\varepsilon_i, \tag{4b}$$

$$Q_i = 1, \qquad W_i + \tfrac{1}{2}\varepsilon_i < V_i. \tag{4c}$$

With no loss of generality, because of the symmetry of the system, we may assume $\Delta = W_2 - W_1 > 0$. The quantities ε_1 and ε_2 are defined to be positive. We introduce for later convenience the following quantities:

$$\begin{aligned} \sigma &= \Delta + (\varepsilon_1 + \varepsilon_2)/2, \qquad & \rho &= \Delta - (\varepsilon_1 + \varepsilon_2)/2, \\ \lambda &= \Delta + (\varepsilon_1 - \varepsilon_2)/2, \qquad & \mu &= \Delta - (\varepsilon_1 - \varepsilon_2)/2. \end{aligned} \tag{5}$$

Looking at the possible combinations of the signs of λ, μ and ρ one realises that only four cases are possible:

(I) $\lambda > 0$, $\mu > 0$, $\rho < 0$;
(II) $\rho > 0$; this implies $\lambda > 0$ and $\mu > 0$;
(III) $\mu < 0$; this implies $\lambda > 0$ and $\rho < 0$;
(IV) $\lambda < 0$; this implies $\mu > 0$ and $\rho < 0$.

The physical meaning of these four cases is readily recognised by means of a superposed plot of the linearised $Q_1(V)$ and $Q_2(V)$, equation (4). In this plot one has to look at the two intervals of V in which Q_1 and Q_2 respectively increase from zero to one. In the four cases above one remarks that:

(I) the intervals of V overlap, but the two rising parts of $Q_1(V)$ and $Q_2(V)$ do not intersect each other;

(II) the two intervals are disjoint;

(III) the two intervals of V overlap; Q_1 and Q_2 intersect each other at a point where $Q_1 = Q_2 > \frac{1}{2}$;

(IV) the two intervals overlap; Q_1 and Q_2 intersect each other; at the intersection $Q_1 = Q_2 < \frac{1}{2}$.

As we shall see later a distinction between cases III and IV is necessary, as the system behaves differently in the two cases. The solution of (3) and (4) can be obtained by plain linear algebra, as sketched in the appendix.

2.2. Results

Depending on the values of the coupling constants, the responses of the system to its driving field U may be of two kinds, that we label S (standing for single-valued) and M (for multi-valued). Accordingly the K plane is divided in two main regions, S and M, each containing several subregions. There are four different partitions of the K plane in subregions (maps of the K plane), one for each of the four cases possible for the signs of λ, μ and ρ. The various kinds of response $V_i(U)$ exhibited by the system are described in the next paragraphs, while figure 3 shows typical examples of $V_i(U)$. Figure 4 shows the maps of the K plane for the cases I and IV.

(i) Single-valued response. In this region filling the even quadrants and a part of the odd quadrants of the K plane both effective fields are single-valued for all values

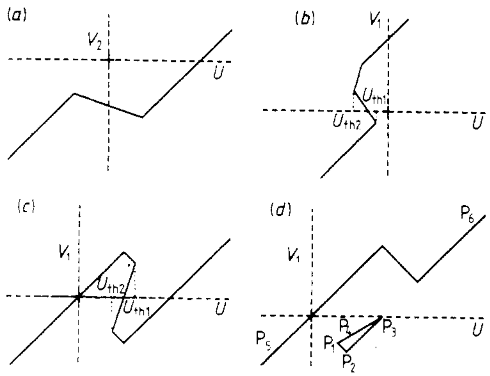

Figure 3. Typical examples of response (effective field versus external field) illustrating different response features caused by the coupling between the subsystems of the system in figure 1. See text for details.

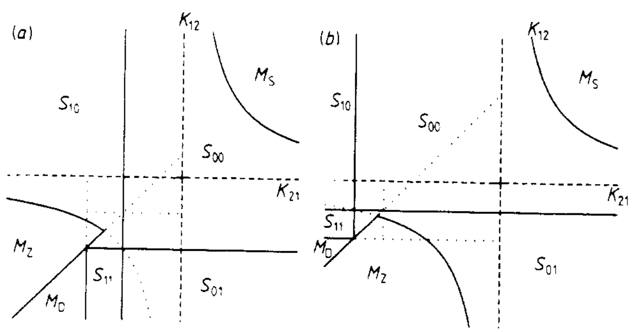

Figure 4. Examples of partitions of the K plane obtained with two different choices of the constants ε_1 and ε_2. (a) $\lambda > 0$, $\mu > 0$ and $\rho < 0$. (b) $\lambda < 0$. See (5) and the paragraph following it. Within each region bounded by broken lines the system exhibits qualitatively similar responses to U, as explained in the text.

of U. Considering the susceptivities of the system

$$\chi_i = \partial V_i / \partial U, \qquad i = 1, 2, \qquad (6)$$

the region S can be divided in four subregions. In fact the χ_i, independently from each other, may become negative in a finite interval of U. In figure 3(a) we show a typical example of a $V_i(U)$ curve exhibiting a region of negative differential susceptivity. The four subregions of the K plane in which the system's response is single-valued will be labelled S_{00} (both χ_i are everywhere positive), S_{10} (only χ_1 becomes negative in a finite interval of U) and so on with S_{01} and S_{11}. In the latter case the two intervals of U in which the susceptivities are respectively negative are different.

(ii) Multi-valued response. In this region of the K plane both system responses are multi-valued in the same finite interval of U, in general. The region M is actually composed of two non-connected areas covering part of the odd quadrants of the K plane. According to the topological properties of $V_1(U)$ and $V_2(U)$ in their interval of multi-valuedness we distinguish three subregions and label them M_S, M_Z and M_D. In the region M_S (within the first quadrant) both the $V_i(U)$ curves are S-shaped (figure 3(b)). In the region M_Z (third quadrant) either or both of $V_i(U)$ are Z-shaped (figure 3(c)). When only one of the $V_i(U)$ is Z-shaped the other is S-shaped. In both the M_S and the M_Z regions the responses exhibit a low-U branch terminating at $U = U_{th1}$, a high-U branch terminating at $U = U_{th2} < U_{th1}$ and a middle branch linking the former. Under the action of the driving field U the system switches from the low-U branch to the high-U one or *vice versa* and exhibits bistable or hysteretical behaviour. The third quadrant contains also the region M_D where the system exhibits an unusual feature, that is a detached pair of branches ($P_1P_2P_3$ and $P_1P_4P_3$ in figure 3(d)). The system may exist in a state of the detached branch provided $U_{th2} < U < U_{th1}$, but switches to the other branch (P_5P_6) as soon as U goes out of that interval. In the subregions M_Z and M_D the system exhibits also negative differential susceptivity. Diagrams similar to that shown in figure 3(d) have been obtained by Uppal *et al* (1976) who studied a system of two rate equations describing a model of reaction kinetics in an open reactor. The rate equations used by these authors yield a block diagram similar to that in figure 1; however, the input–output relationships of some blocks are more complex than in our case.

In figure 4(a) we show the map of the K plane drawn for case I. In cases II and III the maps are very similar to that of case I, the only differences being minor changes of shape of a few subregions. The map of the K plane for case IV is shown in figure 4(b): the major difference with the map of figure 4(a) occurs in the third quadrant. When one or both of the ε_i vanish and the resulting discontinuity of (4) is properly taken into account the model of this paper reproduces the results of I. (Note that equation (3) of I contains a misprint: the sign in front of K_{ij} must be reversed.)

The anomalies described above are exhibited by the system as a function of U when its parameters (ε_1, and ε_2 and the coupling constants) are kept fixed. Transitions from one kind of system response to another may also occur when the system parameters are varied. For example, keeping ε_1 and ε_2 fixed and letting the coupling constants describe a trajectory in the K plane, a phase transition occurs when the trajectory crosses the boundary between two subregions of the K plane.

When ε_1 is let to vanish in either or both subsystems, which hence exhibit a first-order transition, in the subregions M_Z the system, instead of negative differential susceptivity, exhibits re-entrant behaviour in one of its responses, in agreement with the results of I.

This connection between negative differential susceptivity and re-entrant behaviour suggests that, by acting on some suitable parameter of a real physical system, it should be possible to observe a change of the system behaviour from a response exhibiting negative susceptivity to a re-entrant behaviour. This has been indeed observed in La–Al_2 alloys containing Ce impurities ($\geqslant 0.5$ at%). When the pressure is greater than about 6 Kbar this system exhibits a resistivity minimum versus temperature (Kondo effect). At lower pressures the system becomes a superconductor at a pressure dependent temperature T_{c1} and re-enters its finite resistivity state at a temperature $T_{c2} < T_{c1}$ (Ansari *et al* 1981). We believe that a behaviour of this kind is to be expected also in re-entrant nematic liquid crystals. We were not able to find experimental results supporting this hypothesis.

3. Discussion of the model

In order to get a simple analytical solution of the model we made a number of simplifying assumptions that are briefly discussed here.

(1) No time dispersion allowed. We assumed an infinitely fast response of all blocks in the diagram of figure 1, so our results are valid only in the limit $\omega = 0$. Preliminary results show that, for suitable values of the system parameters, the inclusion of time dispersion yields oscillating system responses when U is within some finite interval. This topic will be dealt with more extensively elsewhere.

(2) No fluctuations. Our model exhibits all limitations of mean field theories. When fluctuations are taken into account *a posteriori*, as usual in mean field theories, by means of some equal area rule, in the regions M_S and M_Z the bistability is replaced by a first-order phase transition.

(3) Piecewise linearisation of the subsystem responses. No new or qualitatively different effect is expected if smooth $F_i(V_i)$ functions are used instead of the linearised ones, except for a deformation of the maps of the K plane and a smoothing of the $V_i(U)$ curves. Preliminary numerical results confirm this expectation.

(4) Unbounded range of variability of U, V_1 and V_2. For simplicity all the fields were allowed to span over all the real axis. When some field is to be identified with a positive defined physical quantity, additional constraints in the form of inequalities must be added to the system equations. The more immediate effect of these additional constraints is a deformation of the maps of the K plane. In certain cases which may bear a relevant physical interest, it may happen that no solution for V_1 and V_2 is found in some finite interval of U. This simply means that in that interval there is no response of the kind implied by assumption (1), i.e. a response constant in time. In fact if the equations are modified so as to allow time dispersion, in these cases we obtain oscillating responses.

The physical meaning of the block diagram of figure 1 is clearly evident. This block diagram, or eventually a more elaborate variant of it, can be adapted to several real physical systems whose macroscopic state is defined by two *a priori* uncorrelated physical quantities or order parameters. We say that two quantities measured in the same physical system are *a priori* uncorrelated when they are related to different processes and there are, or there may be in principle, physical systems exhibiting only either. Of course when the two processes occur within the same physical system a cross coupling between the two is usually expected. One of the more impressive situations of this kind is met when the two properties considered are superconductivity

(SC) and ferromagnetism (FM). Materials exhibiting either FM or SC have long been known; recently several workers have reported on the existence of materials exhibiting both FM and SC (see e.g. Sinha *et al* 1982). Among the several kinds of unusual features exhibited by these materials are hysteresis and re-entrant phase transitions. The dedicated theories formulated for these systems invoke a competition between the SC and FM order parameters (see e.g. Jaric and Belic 1979).

In other kinds of physical systems the recognition of the two *a priori* uncorrelated quantities may be less immediate than in the case of the ferromagnetic superconductors. Here are a few examples.

Some lyotropic liquid crystals, composed by alternately stacked hydrophilic and hydrophobic layers, exhibit thermal hysteresis at a transition between mesophases (Giammarinaro and Micciancio 1981). In this case there are spatially segregated subsystems (the two kinds of layer) coupled by molecular interactions at the layer interfaces. Because of the different molecular composition the 'order' within each kind of layer is *a priori* uncorrelated with the 'order' within the other.

The re-entrant behaviour of nematic liquid crystals may be challenging because of the spatial homogeneity of these systems. To explain the re-entrant behaviour of a class of liquid crystals Longa and De Jeu (1982) invoke a dynamic monomer–dimer equilibrium and define separate order parameters for the monomers and the dimers. The two subsystems (the monomeric and the dimeric ones) are coupled by a temperature dependent equilibrium kinetics ruling their relative abundances.

In all the above examples the system driving force is always temperature and the systems can always be assumed closed and in their thermodynamic equilibrium state. Block diagrams featuring two interacting subsystems can also be drawn for non-equilibrium and open systems. For example the two rate equations used by Landsberg *et al* (1979) and Robbins *et al* (1981) to describe the complex behaviour observed in some materials exhibiting threshold switching and negative differential resistivity correspond to a block diagram very similar, although somewhat more complex, than that shown in figure 1. In this case the two subsystems are, respectively, the positive and negative current carriers. Also the block diagram drawn from the three equations of the oregonator (a mathematical model for chemical bistability and chemical oscillations in a continuously stirred tank reactor, see e.g. Tyson (1981)) can be read in terms of two interacting subsystems. In this case the coupling between the subsystems (two reaction equilibria) is far more complex than in the case considered in § 2. An even more complex block diagram describes optical bistability.

The model system worked in this paper can be considered an oversimplification of the physical systems referred to in the preceding paragraphs: no phase transition nor other potentially 'dangerous' trend was assumed in the interacting subsystems and the coupling was assumed strictly linear. In spite of these oversimplifications the model exhibits a rich variety of 'strange' responses to its driving field U as well as transitions between different kinds of responses when the system parameters are varied. Our results show that a number of 'strange' effects arise quite naturally when two well behaved systems are brought to interact.

Theoretical results similar to ours to some extent have been reported by Imry (1975) for a Landau model with two interacting order parameters. The effects predicted in Imry's paper are produced by a nonlinear coupling between the order parameters, while our results show that just a linear coupling is enough for the occurrence of a complex phase transition behaviour as well as other kinds of anomalies. Our approach is more general than Imry's and evidences conceptual similarities among systems described by different theoretical formalisms. A comparison between Imry's results and ours may raise an intriguing question, how a linear coupling might be able to produce a richer variety of 'strange' effects than a nonlinear one. The following argument may help to solve the apparent paradox. Let ξ_0 be a set of values of the control parameters of a nonlinear model. ξ_0 belongs to some space Ξ which will be divided in a number of regions, each defined by some feature exhibited by the solution of the model. If we fix ξ_0 we may linearise the model in a neighbour of a solution. The linearisation algorithm will yield a new set of (effective) values for the control parameter, so that the linearised model will be individuated by some point ξ_0' of the space Ξ'. The linearisation is expected to give correct results in some neighbour of $\xi_0 \in \Xi$ to which corresponds a neighbour of $\xi_0' \in \Xi'$. Now we believe that, in general, it is not granted that there is a one-to-one correspondence between the points of Ξ and Ξ'. That is, we believe that, depending on the model and on the kind of nonlinearity, it may happen that the linearised model predicts, in a neighbour of ξ_1' (far from ξ_0') some feature that is not exhibited by the exact model. In other words $\xi_1' \in \Xi'$ cannot be mapped in a point of Ξ. Conversely all points of Ξ are expected to possess an image in Ξ', unless some pathological situation prevents the application of a linearisation algorithm (e.g. in isolated critical points of Ξ).

Since our assumptions concerning the subsystems and their couplings are very loose and general one may expect that 'strange' effects can be observed, at least in principle, in almost all real physical systems. However, the experimentally accessible range of control of certain parameters of real physical systems is limited or even vanishingly small. For this reason some 'strange' effects are unusual and often can be observed only at the expense of a relevant experimental effort.

Acknowledgments

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Appendix

We eliminate Q_1 and Q_2 between (3) and (4) and obtain the equations of two surfaces in the space UV_1V_2:

$$V_2 = U, \qquad V_1 < W_1 - \tfrac{1}{2}\varepsilon_1, \tag{α}$$

$$V_2 = U + K_{12}(\tfrac{1}{2} + (V - W)/\varepsilon_1), \qquad W_1 - \tfrac{1}{2}\varepsilon_1 < V_1 < W_1 + \tfrac{1}{2}\varepsilon_1, \tag{β}$$

$$V_2 = U + K_{12}, \qquad W_1 + \tfrac{1}{2}\varepsilon_1 < V_1, \tag{γ}$$

and

$$V_1 = U, \qquad V_2 < W_2 - \tfrac{1}{2}\varepsilon_2, \tag{α'}$$

$$V_1 = U + K_{21}(\tfrac{1}{2} + (V_2 - W_2)/\varepsilon_2), \qquad W_2 - \tfrac{1}{2}\varepsilon_2 < V_2 < W_2 + \tfrac{1}{2}\varepsilon_2, \tag{β'}$$

$$V_1 = U + K_{21}, \qquad W_2 + \tfrac{1}{2}\varepsilon_2 < V_2. \tag{γ'}$$

Each surface is a single connected sheet and is composed of three plane regions: two parallel half-planes (respectively α, γ and α', γ') linked by a strip of plane (β and β'). The intersection of the two surfaces is a broken line composed of two straight half-lines and up to seven straight segments. Each of these nine possible parts (partial intersections) comes out from the intersection of one of the unprimed parts of the plane, equations (α)–(γ), with one of the primed ones, equations (α')–(γ'). The projection of this broken line onto the plane UV_1 yields a plane broken line, that is $V_1(U)$. Substituting $V_1(U)$ in (4) yields $Q_1(U)$. $V_2(U)$ and $Q_2(U)$ are obtained in a similar way.

In general when one studies the conditions for the existence of each of the nine possible partial intersections one finds two kinds of conditions (inequalities). The first kind involves only the sign of λ, μ and ρ, while the second kind involves the coupling constants and ε_1, ε_2, Δ. The conditions of the second kind generate lines (straight lines or hyperbolae) each dividing the K plane in two parts. Since the allowed combinations of signs of λ, μ, ρ are four, there are four distinct partitions of the K plane.

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