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LETTER TO THE EDITOR

On renormalisation of fluctuations at the onset of a centre manifold

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Abstract. We consider a dissipative reactive system driven far from thermodynamic equilibrium and sustaining a locally attractive centre manifold beyond a symmetry-breaking instability. We derive a functional equation for the renormalised diffusion pressure. This coefficient gives autocorrelations for intrinsic fluctuations and depends on the bifurcation parameter. The system is subject to the following realistic restrictions. (i) The temporal organisation lying beyond the instability can be accounted for by the existence of a centre manifold. (ii) The effective equilibrium energy averaged with respect to the stationary distribution about the centre manifold is independent of the bifurcation parameter.

Although the onset of dissipative structures in open reactors is relatively well understood at the level of phenomenological kinetics, the microscopic aspects of the cooperativity leading to the dominance of certain modes [1] and the localisation of the reactive species [2] are far from clear. In spite of continuous efforts, essentially making use of renormalisation methods, to calculate non-equilibrium fluctuations, the results have not been conclusive in so far as the role of intrinsic fluctuations as triggers of organisation in realistic situations remains elusive [3-5]. Moreover, the implementation of renormalisation methods in symmetry-breaking transitions has been questioned on theoretical grounds [5]. The main objection has to do with the difficulty in showing that the intrinsic lengths in reaction-diffusion systems are independent of boundary conditions. The centre manifold (CM) scaling theory has been particularly successful in deriving the adequate random source term responsible for intrinsic fluctuations even in the case where the characteristic length is dependent on the boundary conditions, as in the case of the Bénard instability [6]. However, the same tools cannot be applied *mutatis mutandis* to open reactive systems unless an adjustable free parameter, the proportionality factor for the scaling of non-equilibrium fluctuations with the bifurcation parameter, is introduced [7, 8]. This bifurcation parameter, denoted 'u', measures the departure from the critical point. The difficulty mentioned above can be circumvented if an additional restriction is included: in the spirit of Klimontovich's approach [9, 10], we shall introduce the realistic assumption that the effective equilibrium energy averaged with respect to the stationary distribution at the onset of a CM is independent of u. We must point out here that under such circumstances, the entropy of the self-organising system gives a measure of the order. In general, this is not the case

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unless the mean energy is fixed [9, 10]. The general derivation presented in this paper is useful in planning for experiments of time-dependent fluctuation spectroscopy aimed at determining non-classical critical exponents. Such experiments are in the spirit of those leading to the elucidation of the stochastic source in the transition to convective structures [6].

The existence of a locally attractive and locally invariant CM in a neighbourhood of criticality is a special case of the subordination of fast-relaxing modes, X_{fj}, to the critical modes, X_{si}. The dissipative structure emerging beyond the instability is contained in the CM and the cooperativity effects leading to fluctuations in the collective modes reflect themselves in the confinement of the probability density $f = f(\boldsymbol{X}_s, \boldsymbol{X}_f, u) = f(\boldsymbol{X}, u)$ to a narrow strip about the CM. The Gaussian width of this strip, w, is a function of u and, to first approximation, independent of the position on the CM. This width is determined from the adequate competition between the drift towards the CM and the diffusion pressure caused by fluctuations. The fluctuation correlation $C = C(\dot{u})$ can be identified with the effective diffusion coefficient. The CM scaling theory requires that the effective diffusion coefficient C be renormalised, that is, $C = C(u)$. This theory successfully gives the critical exponents. However, the exact derivation of the fluctuation-correlation function becomes in general hopelessly complicated [9, 10]. Thus, we shall implement a renormalisation approach subject to the restriction that the average of certain specific functional $R = R(\boldsymbol{X})$ with respect to $f(\boldsymbol{X}, u)$ is a constant:

$$\frac{\mathrm{d}}{\mathrm{d}u}\langle R\rangle = 0. \tag{1}$$

In a realistic situation, this functional is taken to be the effective equilibrium energy of the system (an explicit expression will be given later) [9, 10].

The starting point in our analysis is the factorisation of f in the following form [6–8]:

$$f(\boldsymbol{X}, u) = \delta(\boldsymbol{X}_s - \boldsymbol{X}_{ss}(u)) \prod_j Q_j(X_{fj} \,|\, \boldsymbol{X}_s) \tag{2}$$

where the factors Q_j represent the conditional probability and they determine the statistical subordination of the fast-relaxing degrees of freedom X_{fj} to the order parameters X_{si}. The remaining factor on the RHS is a Dirac delta distribution peaked at the stationary state for the smeared evolution equation in order parameter space. It should be emphasised that the order parameters are viewed as the CM coordinates. The factors Q_j have the form:

$$Q_j = \exp[(N(u) - A_j(X_{fj}, \boldsymbol{X}_s, u))/C(u)] \tag{3}$$

where

$$A_j = A_{0j} - A_{fj} \tag{4}$$

$$A_{0j} = \lambda_j(0)(X_{fj} - \tilde{X}_{fj}(\boldsymbol{X}_{ss}(0)))^2 = \lambda_j(0)^2 X_{fj}(0). \tag{5}$$

Here $\lambda_j(u)$ denotes the damping constant for the fast-relaxing degree of freedom X_{fj}, $X_{fj} = \tilde{X}_{fj}(\boldsymbol{X}_s)$ is the CM equation and A_{fj} accounts for portion of probability shifted due to the departure from the critical point. The CM equation has the general form:

$$X_{fj} = \tilde{X}_{fj} = \sum_{i=2}^{\infty} d_{ij} \left(\prod_{i_1+i_2+\ldots+i_N=i} X_{s1}^{i_1} X_{s2}^{i_2} \ldots X_{sN}^{i_N} \right) \tag{6}$$

where the CM coefficients d_{ij} are determined from the implicit set of equations [7, 8].

$$\frac{\partial \tilde{X}_{fj}}{\partial X_s}\dot{X}_s = \dot{X}_{fj}(X_s, \tilde{X}_f(X_s), u) = \left.\frac{\partial X_{fj}}{\partial t}\right|_{X_f=\tilde{x}_f}. \tag{7}$$

The probability distribution defined via the A_j adopts the form of a Gaussian peaked at the CM. The Gaussian widths $w_j = w_j(u)$ are, to first approximation, given by: $(C(u)/\lambda_j(u))^{1/2} = w_j(u)$; i.e. they are given by the competition between the drift towards the CM and the diffusive pressure given by the intrinsic fluctuations. The second term in (4) adopts the general form

$$A_{fj} = \sum_{i=0}^{2} g_{ij}(u) X_{fj}^i \tag{8}$$

with

$$\begin{aligned} g_{2j}(u) &= \lambda_j(0) - \lambda_j(u) \\ g_{1j}(u) &= 2\tilde{X}_{fj}(X_{ss}(u)\lambda_j(u) - 2\tilde{X}_{fj}(X_{ss}(0))\lambda_j(0) \\ g_{0j}(u) &= \lambda_j(0)\tilde{X}_{fj}^2(X_{ss}(0)) - \lambda_j(u)\tilde{X}_{fj}^2(X_{ss}(u)). \end{aligned} \tag{9}$$

In order to evaluate $C(u)$ explicitly, our probability distribution f is subject to the condition

$$\int \left(\sum_j A_{0j}\right) f \, \mathrm{d}X = ct \qquad \text{(independent of } u\text{)}. \tag{10}$$

This condition corresponds to fixed averaged effective equilibrium free energy

$$\frac{\partial}{\partial u}\left(\sum_j \langle A_{0j}\rangle\right) = 0. \tag{11}$$

Making use of the fact that f is normalised for every value of u in a neighbourhood of the critical point, we obtain

$$N'(u) = -\sum_j \sum_{i=0}^{2} h_{ij}\langle X_{fj}^i\rangle + \left(N(u) - \sum_j \lambda_j(u)\langle \Delta^2 X_{fj}(u)\rangle\right)\frac{C'(u)}{C(u)} \tag{12}$$

where

$$h_{ij} = \frac{\partial}{\partial u} g_{ij}. \tag{13}$$

From equations (10) to (12), we obtain

$$\int \mathrm{d}X \left(\sum_j A_{0j}\right) f \left[\sum_j \left(\sum_{i=0}^{2} h_{ij}\bar{\Delta}X_{fj}^i + \frac{C'(u)}{C(u)}\lambda_j(u)\bar{\Delta}(\Delta^2 X_{fj}(u))\right)\right] = 0 \tag{14}$$

where we have introduced the simplifying notation $\bar{\Delta}G = G - \langle G\rangle$ for any arbitrary functional G.

Making use of the explicit expression for A_{0j}, as given in equation (5), we obtain from equation (14) the following functional differential equation for $C(u)$:

$$C'(u)F(u, C(u)) + C(u)H(u, C(u)) = 0 \tag{15}$$

with

$$F(u, C(u)) = \sum_{j,k} \lambda_k(0)\lambda_j(u)\langle \Delta^2 X_{fk}(0)\bar{\Delta}\Delta^2 X_{fj}(u)\rangle \tag{16}$$

$$H(u, C(u)) = \sum_{j,k} \sum_{i=0}^{2} \lambda_k(0) h_{ij}\langle \Delta^2 X_{fk}(0)\bar{\Delta}X_{fj}^i\rangle. \tag{17}$$

We need to warn the reader that (15) is not an ordinary differential equation for $C(u)$ since an average of the form $\langle G \rangle$ is always a functional of $C(u)$.

Thus, $C(u)$ satisfies the following functional integral equation:

$$C(u) = \Gamma(C(u)) \tag{18}$$

where

$$\Gamma(C(u)) = \exp\left(-\int_0^u \frac{H(u', C(u'))}{F(u', C(u'))}\,du'\right). \tag{19}$$

The solution of equations (18) and (19) can be constructed for a specific case by noting that it is given by the limit of the convergent sequence of functions $\{C_j(u)\}_{j=1,\ldots}$. This sequence is defined inductively, by the following recursive relation:

$$C_{j+1}(u) = \Gamma(C_j(u)). \tag{20}$$

The first step in the induction process consists of defining $C_1(u)$. This poses no additional problems since the CM reduction introduces, to first approximation, scaling laws of the form $C_1(u) \sim u^{\alpha}$. For example, in the case of a Hopf instability, $\alpha = 2$. The proportionality factor is an adjustable parameter.

Thus, once the initial gauge has been made, the solution can be constructed recursively and it will be parametrically dependent on the proportionality factor for the CM scaling. At this point it should be stressed that this is a rigorous derivation of the fluctuation autocorrelations under non-equilibrium conditions in a self-organising system. In a forthcoming publication, this recursive method will be implemented to derive the stochastic source in the transition to a convective structure [6] and to make comparisons vis-a-vis the experimental findings. The CM scaling approach has already given very satisfactory agreement in this case but an analytical expression has never been obtained.

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