

Symmetry Breaking Instabilities in Dissipative Systems. II

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HCN. As has been noted, isotopic shifts in the matrix spectra indicate that the far-infrared bands are associated with torsional modes rather than with H-bond stretching. The calculated torsional frequency of 153 cm^{-1} for the HCN dimer is in fair agreement with the doublet band centered at about 141 cm^{-1} . The sharp Raman lines at 173 and 164 cm^{-1} in solid HCN and

DCN are also very probably librational rather than stretching vibrations.

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Symmetry Breaking Instabilities in Dissipative Systems. II

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(Received 12 September 1967)

The thermodynamic theory of symmetry breaking instabilities in dissipative systems is presented. Several kinetic schemes which lead to an unstable behavior are indicated. The role of diffusion is studied in a more detailed way. Moreover we devote some attention to the problem of occurrence of time order in dissipative systems. It is concluded that there exists now a firm theoretical basis for the understanding of chemical dissipative structures. It may therefore be stated that a theoretical basis also exists for the understanding of structural and functional order in chemical open systems.

I. INTRODUCTION

In a recent paper, Nicolis and one of us (I. P.)¹ have discussed instabilities in purely dissipative systems involving chemical reactions and transport processes such as diffusion, but no hydrodynamic motion. Special emphasis was put on the possibility of symmetry breaking instabilities in such systems. An example, due to Turing² was studied in detail, and it was concluded that far from thermodynamic equilibrium such instabilities were indeed possible. Beyond the unstable transition, one may have a situation which may be appropriately called a "dissipative structure" as it is characterized by a structural and functional order and by a low value of entropy.

In this paper, we present a thermodynamic theory of such chemical instabilities. The theory is based on the general evolution criterion due to Glansdorff and one of the authors (I. P.).³ This evolution criterion leads easily to necessary condition for the occurrence of instabilities. A general theory involving both hydrodynamic and chemical instabilities will be published soon.⁴ The thermodynamic approach shows easily the type of autocatalytic or cross-catalytic effects which are necessary to produce chemical instabilities. In this way we may easily construct several examples of kinetic schemes which may lead to instabilities.

The role of diffusion is discussed in some detail. At first it would seem that diffusion can only lead to

homogenization of the system. Here, on the contrary, we see (see also Ref. 1) that diffusion is necessary to obtain a dissipative structure corresponding to a time-independent state in which the concentrations are *space dependent*. This apparent paradox is resolved as we show that diffusion has a double role: on one hand, it increases the stability of the steady state, but on the other hand, it increases the manifold of perturbations compatible with the macroscopic equations of change. We have now to test the stability with respect to inhomogeneous as well as to homogeneous disturbances. It is when the second effect becomes dominant that symmetry breaking instabilities become possible (see Sec. III).

The existence and the stability of chemical structures is investigated in detail in a relatively simple case of a system formed of two boxes (Sec. IV). We show that beyond the instability the concentrations of the intermediate components in the boxes become different indeed.

Finally, we devote some attention to the occurrence of time order (biological clocks) in dissipative systems. As we shall see, there is a remarkable parallelism between the problems of structural dissipative order and time order. Both appear as possible consequences of large deviations from thermodynamic equilibrium in chemical systems undergoing chemical transformations of essentially the same type. The possibility of chemical instabilities and dissipative structures in the case of biochemical processes is illustrated considering a mechanism due to Chance and co-workers.^{5,6}

¹ I. Prigogine and G. Nicolis, *J. Chem. Phys.* **46**, 3542 (1967).

² A. M. Turing, *Phil. Trans. Roy. Soc. London* **B237**, 37 (1952).

³ P. Glansdorff and I. Prigogine, *Physica* **30**, 351 (1964).

⁴ I. Prigogine and P. Glansdorff, *Physica* (to be published).

⁵ B. Chance, A. Gosh, J. Higgins, and P. K. Maitra, *Ann. N.Y. Acad. Sci.* **115**, 1010 (1964).

⁶ J. Higgins, *Proc. Nat. Acad. Sci.* **51**, 989 (1964).

Summarizing, we can say that chemical instabilities may lead to highly nonhomogeneous distributions of matter in open chemical systems. The implications for biological processes are evident and are discussed separately in a paper by one of the authors.⁷

II. EVOLUTION CRITERION AND STABILITY OF STEADY STATES

Beyond the linear range of the thermodynamics of irreversible processes, there no longer exists a real thermodynamic potential such as the entropy production, which takes its minimum at the steady state. As a consequence, the stability of nonequilibrium steady states is generally investigated by means of methods which are usual in hydrodynamics and consist in making a normal modes analysis. It is thus verified that the infinitesimal fluctuations of the steady quantities regress in time. However such calculations do not extract and picture clearly the kinetic reasons of the instability. In view of this, it is convenient to consider the evolution criterion introduced by Glansdorff and one of the authors (I. P.).⁸ This criterion is applicable over the whole range of macroscopic physics.⁹ In the case of purely dissipative systems, it can be written in the form

$$Td_X P = \int dV \sum_i J_i dX_i \leq 0, \quad (2.1)$$

where the integration is taken over the volume, and where X_i and J_i are the generalized forces and fluxes of the irreversible processes considered.^{10,8} Thus, (2.1) expresses the fact that the entropy production P at constant fluxes J_i takes its minimum value at the steady state (equality sign). As a consequence, the steady state studied will be stable if⁹

$$\delta_X P \geq 0 \quad (2.2)$$

for all perturbations compatible with the kinetic equations of the system. Passing to a local formalism, we can write (2.2) in the form

$$T\delta_X P = \sum_i \delta J_i \delta X_i > 0. \quad (2.3)$$

The stability theory of steady states may now be conveniently subdivided into two parts:

- (a) the identification of phenomena which may give rise to a negative contribution to $\delta_X P$;
- (b) the discussion of the numerical values of such negative terms and especially the discussion of the pos-

sibility that the sum involved in (2.3) may become negative without violating the kinetic equations.

Concerning $d_X P$, it still must be noted that it is not a total differential. Furthermore, beyond the linear range, no analogous inequality for $d_J P$ is known. It is, therefore, remarkable that in the stability condition (2.3) the forces and the fluxes play again a symmetrical role comparable to the one played in the entropy production

$$P = \sum_i J_i X_i \geq 0.$$

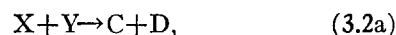
The fact is that $\delta_X P$ appears as an "excess entropy production" (e.e.p.) near the steady state we consider, as in the extension of the minimum entropy production theorem to the nonlinear domain of thermodynamics of irreversible processes.

III. CHEMICAL INSTABILITIES

In the case of chemical reactions, Eq. (2.3) becomes

$$T\delta_X P = \sum_i v_i \delta A_i > 0, \quad (3.1)$$

where v_i and A_i are now the chemical reaction rates and affinities. We now investigate what is the nature of the processes giving negative contributions to (3.1). Let us consider the following two reactions:



As we are mainly interested in situations far from equilibrium we neglect the reverse reactions and for simplicity, put equal to one all kinetic and equilibrium constants as well as RT . It is then easy to see that a fluctuation of the concentration X around its steady state value X_0 gives rise, in the case of (3.2a), to the excess entropy production

$$\delta v \delta A = (Y_0/X_0) (\delta X)^2 > 0. \quad (3.3)$$

Such a fluctuation would, therefore, not bring into danger the stability condition (3.1). On the contrary, with (3.2b) the same type of fluctuation gives a contribution similar to (3.3), except for the sign, which is now negative. And indeed it will be shown that under well-defined conditions such autocatalytic reactions give rise to instabilities. Of course, (3.2b) is meant here only as an example. As we shall see later, there may be more subtle ways involving fluctuations of more than one component to produce negative contributions to the e.e.p.

Before going to the study of specific examples, we want to make the following observation. There are at least two types of chemical instabilities:

- (a) instabilities with respect to homogeneous perturbations;
- (b) instabilities with respect to space-dependent inhomogeneous perturbations.

⁷ I. Prigogine, "Structure, Dissipation and Life," paper given at the International Conference "Physique Theorique et Biologie," organized by L'Institut de la Vie, Versailles, France, 1967.

⁸ S. de Groot and P. Mazur, *Non-Equilibrium Thermodynamics* (North-Holland Publ. Co., Amsterdam, 1962).

⁹ *Non-Equilibrium Thermodynamics, Variational Techniques, and Stability*, R. J. Donnelly, R. Herman, and I. Prigogine, Eds. (The University of Chicago Press, Chicago, 1965).

¹⁰ I. Prigogine, *Introduction to Thermodynamics of Irreversible Processes* (Interscience Publishers, Inc., New York, 1961), 2nd ed.

In case (a), we expect the system to go from a homogeneous steady state to another homogeneous state (which may be steady or not). In case (b) diffusion plays an essential role. Now diffusion appears in this theory in a *twofold* way:

(1) It gives a positive contribution to the e.e.p. similar to that of viscous dissipation in hydrodynamics (Ref. 11); this effect can only stabilize the steady state.

(2) It increases the manifold of perturbations compatible with the macroscopic equations of change. We have to test the stability of the system with respect to a wider class of situations. If the second effect is dominant we may expect symmetry breaking instabilities.¹

Let us now consider, explicitly, some examples. A very simple scheme of reactions is the following:

Scheme I



The over-all reaction is



This reaction scheme is physically unrealistic because of the trimolecular step (3.4b). We shall indicate below other schemes which do not involve such a step. But the discussion of (3.5) can be performed with much simpler algebra as it involves only two intermediate components X and Y. For this reason we shall discuss it in more detail and simply quote the results obtained for the other schemes (for more information see Ref. 12).

The kinetic equations are:

$$\partial X / \partial t = k_1 A + k_2 X^2 Y - k_3 B X - k_4 X + D_X (\partial^2 X / \partial r^2), \quad (3.6a)$$

$$\partial Y / \partial t = k_3 B X - k_2 X^2 Y + D_Y (\partial^2 Y / \partial r^2). \quad (3.6b)$$

We maintain constant the concentrations of the initial and final products (A, B, D, E). The reverse reactions are neglected, and we take diffusion into account. To simplify we assume a one-dimensional medium.

There always exists a time-independent homogeneous solution

$$X_0 = (k_1/k_4) A, \\ Y_0 = (k_3 k_4 / k_1 k_2) (B/A), \quad (3.7)$$

which is the continuation of the equilibrium solution. To investigate its stability, we first investigate the dispersion equation within the framework of a linear

analysis. We consider a perturbed state of the form

$$X = X_0 + x \exp[\omega t + (ir/\lambda)], \\ Y = Y_0 + y \exp[\omega t + (ir/\lambda)], \quad (3.8)$$

with

$$|x/X_0| \ll 1 \quad \text{and} \quad |y/Y_0| \ll 1,$$

and easily see that there exists a critical value B_c of B corresponding to a marginal state $\omega = 0$:

$$B_c(\lambda) = \frac{(k_4 + D_X/\lambda^2)(k_1^2 k_2 A^2 + k_4^2 D_Y/\lambda^2)}{k_3 k_4^2 D_Y/\lambda^2}, \quad (3.9)$$

which separates a root $\omega < 0$ from a root $\omega > 0$. We now have to look for the critical value of the wavelength λ_c at which the instability begins. To do this we have to calculate the wavelength which gives to (3.9) its minimum value. This leads immediately to:

$$(\lambda_c)^2 = (k_4/k_1^2 k_2)^{1/2} [(D_X D_Y)^{1/2}/A], \quad (3.10)$$

and substituting into (3.9) we obtain

$$B_c = \left[\frac{k_1 (k_2 D_X)^{1/2}}{k_4 \sqrt{k_3 D_Y}} \right]^2 A + \left(\frac{k_4}{k_3} \right)^{1/2} \left[\frac{k_1}{k_4} \right]^2. \quad (3.11)$$

We see that λ_c and B_c depend in an intrinsic way on both the various reaction rates and on the diffusion coefficients. If diffusion, as compared to reaction rates, becomes small, instability occurs for short wavelength perturbations. In the inverse case, the instability occurs for long wavelengths. This is quite reasonable and we shall come back to this point below.

We see that we have here a symmetry breaking instability as the system above the instability can no longer be expected to be homogeneous. We shall study this question in greater detail in the next paragraph.

Let us indicate two others reaction schemes which also may lead to a chemical symmetry-breaking instability:

Scheme II



The overall reaction is



There are only one- and two-molecular steps. X and Y catalyze their own formation. It is to be noticed that (3.12) is a modification of a scheme suggested by Lotka¹³ to investigate chemical oscillations (see also Ref. 14). We have simply added the "appendix"

¹¹ W. Strieder, Acad. Roy. Belg., Bull. Classe Sci. **50**, 318 (1965).

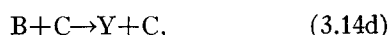
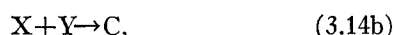
¹² R. Lefever, Acad. Roy. Belg., Bull. Classe Sci. (to be published).

¹³ A. J. Lotka, J. Am. Chem. Soc. **42**, 1595 (1920).

¹⁴ R. Lefever, G. Nicolis, and I. Prigogine, J. Chem. Phys. **47**, 1047 (1967).

(3.12c), (3.12d) involving the intermediate compound V' . Such an appendix also plays a role in the next scheme:

Scheme III



which is a simplified form of Turing's original scheme.^{1,2} We have here the two over-all reactions



and it can be shown that no instability can occur when the affinities of the over-all reactions are smaller than some critical value (see Ref. 1).

Let us now consider the thermodynamic aspects of the chemical instability in terms of the basic quantity (3.1). In case of Scheme I we obtain:

$$\delta_X P = (1-B) \frac{x^2}{A} + \frac{A^3}{B} y^2 + \frac{D_X}{\lambda^2 A} x^2 + \frac{D_Y A}{\lambda^2 B} y^2, \quad (3.16)$$

where the kinetic constants as well as RT have been put equal to one. Diffusion is taken into account (last two terms). Two interesting remarks have to be made:

(1) In agreement with our general discussion, there appears in the e.e.p. (3.16) the negative term $-(B/A)x^2$ due to the autocatalytic action of X . This is the dangerous contribution.

(2) The explicit contribution of diffusion to the e.e.p. is positive and proportional to D/λ^2 . Therefore, if there is an instability, increasing values of D must give rise to increasing values of the critical wavelength. If not, the contribution of diffusion to (3.16) would become dominant and $\delta_X P$ would be always positive. This is in agreement with the formula for λ_c (3.10) derived from the dispersion equation. But the diffusion has, as we already mentioned, a second role: the manifold of perturbations which we may introduce into (3.16) is now increased by the consideration of inhomogeneous systems. It is easy to see¹² that the perturbations (x, y) which satisfy the linearized kinetic equations at the marginal state $\omega=0$ lead to the vanishing of the e.e.p.

$$(\delta_X P)_{B=B_*} = 0. \quad (3.17)$$

There is, therefore, a complete agreement between the

kinetic theory of chemical instabilities based on the dispersion equation and the thermodynamic theory developed in Sec. II.

The situation for the reaction Scheme II described in (3.12) is quite similar. The autocatalytic character of X , Y introduces again negative terms into the e.e.p. It is interesting to comment briefly on the role of the appendix [steps (3.12c) and (3.12b)]. It appears that this role is quite similar to that of diffusion (for more details see Ref. 12): it gives a positive contribution to the e.e.p. However at the same time it increases the range of permissible perturbations. Instability appears if the steady state concentration of V' is larger than some critical value.

The Scheme III leads again to an e.e.p. which contains the dangerous contribution. However here the negative terms are of the form

$$-\alpha y c \quad (\alpha \text{ is a positive constant}) \quad (3.18)$$

and not

$$-(B/A)x^2, \quad (3.19)$$

as in (3.16). This is quite natural, as we have in this scheme a cross-catalytic effect: Y catalyze the formation of C and inversely C the formation of Y .

We believe that the situation for chemical instabilities as compared to that described in our earlier paper¹ has been greatly clarified. We see indeed that:

(1) There are many reaction schemes which may lead to chemical instabilities. In fact, it would be easy to imagine others than those we have listed.

(2) They are all characterized by negative contributions to the e.e.p. due to autocatalytic or cross-catalytic effects.

(3) The essential and unexpected role of diffusion is clearly understood.

Let us now consider the physical situation beyond the chemical instability point.

IV. CHEMICAL DISSIPATIVE STRUCTURE

Again, Scheme I provides us with a relatively simple example. However, instead of considering disturbances of arbitrary wavelength, we shall now consider a system of two boxes, within which the concentrations of the intermediate products X , Y may be different. Instead of (3.6) we now have the 4 equations:

$$\begin{aligned} dX_1/dt &= A + X_1^2 Y_1 - B X_1 - X_1 + D_X (X_2 - X_1), \\ dX_2/dt &= A + X_2^2 Y_1 - B X_2 - X_2 + D_X (X_1 - X_2), \\ dY_1/dt &= B X_1 - X_1^2 Y_1 + D_Y (Y_2 - Y_1), \\ dY_2/dt &= B X_2 - X_2^2 Y_2 + D_Y (Y_1 - Y_2), \end{aligned} \quad (4.1)$$

where we have put all the kinetic constants equal to one.

We have a single time-independent homogeneous solution

$$X_i = A; \quad Y_i = (B/A) \quad (i=1, 2), \quad (4.2)$$

which becomes unstable when

$$B = B_c = (1/2D_Y)(A^2 + 2D_X A^2 + 2D_Y + 4D_X D_Y). \quad (4.3)$$

The basic difference with (3.11) is that here instability arises only for a finite range of values of the diffusion coefficients. Both for $D_X, D_Y \rightarrow 0$ and for $D_X, D_Y \rightarrow \infty$ we have $B_c \rightarrow \infty$. Clearly, this is related to the fact that the wavelength here is artificially imposed by the size of the system.

To pursue the calculations, we make the following choice of numerical values:

$$A = 2, \quad D_X = D_Y = 1; \quad (4.4)$$

therefore,

$$B_c = 9. \quad (4.5)$$

And if we look for the time-independent solutions of the kinetic Eqs. (4.1) we find that:

(a) for $B \leq B_c$ only the homogeneous steady-state solution is accessible to the system. All other solutions of the Eqs. (4.1) are devoid of physical meaning.

(b) for $B > B_c$ there exists always two different acceptable solutions: one is the continuation of the previous case homogeneous solution and the other corresponds to a new steady state which is space dependent, and is reported in Fig. 1.

In this figure, we have a striking example of a dissipative structure as defined in the introduction.

In one compartment, say 2, the concentration of Y is larger than in the other, while at the same time the concentration of X is smaller. If we refer to the reaction scheme (3.4), we see that the consumption of B

will essentially proceed in compartment 1. The system begins to look like a kind of "factory" with characteristic structural and functional order.

The stability of the dissipative structure may in turn be analyzed using Eqs. (4.1). The study of the corresponding dispersion equations indicates that it is stable for

$$9.8 \leq B \leq 11.2. \quad (4.6)$$

Outside of this range, at least one root of the dispersion equation has a positive real part.

We do not know yet what happens there; it seems likely to us that this additional instability arises from the artificial division of the system into two homogeneous boxes. The real steady state may correspond to a much more complicated distribution of matter. Certainly the behavior of matter beyond the instability deserves a much more careful investigation. However the existence of dissipative structures arising beyond symmetry breaking chemical instabilities seems to us to be established with a reasonable degree of certitude.

V. CHEMICAL OSCILLATIONS

The last paragraphs were mainly devoted to the study of structural order. Let us now consider the problem of order in time and especially the problem of oscillations around steady states. We go back to the general evolution criterion which, due to the fact that we have to consider complex normal modes $\omega = \omega_r \pm i\omega_i$, must now be written in the form

$$T(d_X P/dt) = \frac{\omega_r}{2} \sum_i (\delta v_i^* \delta A_i + \delta v_i \delta A_i^*) + i \frac{\omega_i}{2} \sum_i (\delta v_i^* \delta A_i - \delta v_i \delta A_i^*) \leq 0. \quad (5.1)$$

It is easy to see that equilibrium stability conditions imply that each of these two terms is separately negative (see also Ref. 11). As a consequence, we have two inequalities: the first is the same as we discussed in connection with the stability of steady states, and the second gives us the direction of rotation around the steady state (see also Ref. 10).

Very little is known about the general conditions for the appearance of rotations. Essentially we may say that it is a typical far from equilibrium phenomenon^{10,14} and it is only possible in the case of systems where the kinetic laws are nonlinear. The evolution criterion shows that rotations are only possible if

$$\sum_i (\delta v_i^* \delta A_i - \delta v_i \delta A_i^*) \neq 0. \quad (5.2)$$

A necessary and sufficient condition is that the matrix which relates, near the steady state, the rates δv_i to the affinities δA_k is not symmetric.

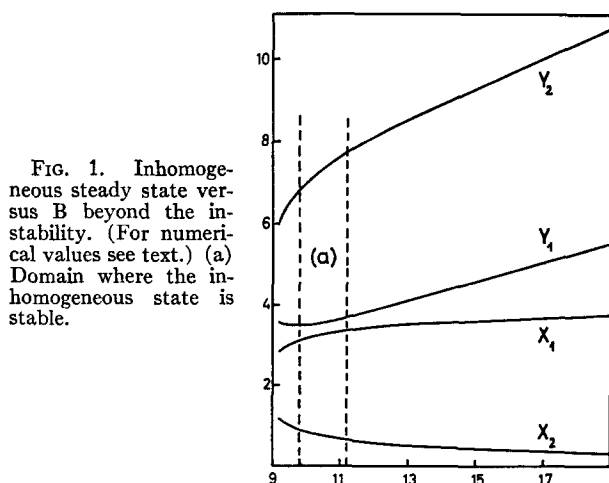


FIG. 1. Inhomogeneous steady state versus B beyond the instability. (For numerical values see text.) (a) Domain where the inhomogeneous state is stable.

Moreover, if a system presents undamped rotations, the first term of (5.1) must vanish

$$\sum_i (\delta v_i^* \delta A_i + \delta v_i \delta A_i^*) = 0. \quad (5.3)$$

This condition is very similar to that used in the stability theory (Secs. II and III): it can only be satisfied if there appear negative contributions to the e.e.p. Therefore, most of our discussions of Sec. 3 apply as well to the problem of rotations: For an oscillatory behavior to occur, it is necessary to have steps involving autocatalytic or cross-catalytic reactions. This is in complete agreement with the conclusions reached by Chance and his co-workers.^{5,6}

In fact, all the schemes we discussed in Sec. III lead to rotations for some range of the constraints involved. However, the converse is not true: even undamped rotations may occur on the thermodynamic branch and are not necessarily related to instabilities.¹⁴

VI. CHEMICAL INSTABILITIES AND BIOCHEMICAL MECHANISMS

We believe that the discussion presented in the preceding paragraphs establishes firmly the existence of chemical instabilities and consequently of chemical dissipative structures. It may therefore be stated that a theoretical basis exists for the understanding of structural and functional order in chemical systems.

Are these considerations relevant for biological systems? Clearly, the answer can only come from biologists. However, we would like to present two general arguments in favor of an affirmative answer:

(1) The general picture of a dissipative structure as presented, for example, in Sec. IV has a striking similarity with the description of biological order as has emerged from the progress in molecular biology.¹⁵

(2) The chemical mechanisms considered in molecular biology are often precisely of the type considered in Secs. III and IV and may indeed lead to chemical instabilities.

As an example, let us discuss more in detail the Chance-Higgins mechanism for oscillatory reactions.^{5,6}

¹⁵ A. Lwoff, *Biological Order* (M.I.T. Press, Cambridge, Mass., 1965).

This mechanism may be written in the form



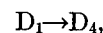
C_1 , C_2 are intermediate compounds; D_1 , D_2 , D_3 , V , V' are enzymes. A is the initial compound (glucose) and E the final (glyceraldehyde phosphate).

This scheme does not lead to chemical instabilities under this form. The reason is easy to understand: we have two resultant reactions



While the Reaction (1) is taking place in nonequilibrium conditions (its affinity may be taken as infinite), the Reaction (2) may be considered at equilibrium as the steady-state conditions lead to $V_4 = 0$ for the rate of step (4) of scheme (6.1). In a similar situation¹ it has been shown that this prevents the occurrence of chemical instability.

If we now add to (6.1) the step



where D_4 is some other form of the enzyme D_1 , Reaction (6.1b) goes out of equilibrium and chemical instability may occur. However, we have not yet investigated what the state of the system beyond the chemical instability is.

In any case, we see that the idea of chemical instabilities is certainly compatible with the type of chemical reactions presently investigated in biochemical processes.

ACKNOWLEDGMENTS

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