

## Chaotic nucleation of metastable domains

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We describe a cavitation process that consists of chaotic nucleation of metastable domains. It can be generically observed in spatially extended nonequilibrium systems, whenever they exhibit bistability between a stationary and an oscillatory state, close to the Andronov homoclinic bifurcation, which leads to the disappearance of the former. In the bistable regime, the modulational instability of the homogenous oscillations leads to inhomogenous nucleation of the stationary phase. [S1063-651X(97)51408-5]

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Although the phenomenon of bistability occurs in many different situations, it has received very little attention in the context of spatially extended nonequilibrium systems. In this paper, we analyze the universal properties of a system that exhibits bistability between a homogeneous stationary state and an oscillatory one, close to parameter values where the oscillation disappears through an Andronov homoclinic bifurcation. It is one of the simplest bistable systems that has no equilibrium counterpart, since sustained oscillations are ruled out at equilibrium.

In zero space dimension, one of the generic transitions from a limit cycle to a fixed point can be generically studied by considering a one parameter family of planar dynamical systems [1]. Varying a typical parameter (see Fig. 1), the limit cycle disappears through a codimension one homoclinic bifurcation [2], where the frequency of the oscillations vanishes.

In one or more space dimensions, the homogeneous oscillatory state can be modulationally unstable. This instability breaks the phase coherence of the oscillations and leads to a form of spatiotemporal disorder [3–6] which acts as an intrinsic noise. Above a parameter threshold value, this intrinsic noise leads to the nucleation of the stationary state. Depending upon the relative stability of the stable stationary state (see Fig. 2), and the disordered state, the domains nucleated either grow or shrink. In the case where they shrink, i.e., when the stationary phase is metastable [7], a “chaotic nucleation of the metastable state” is observed in some parameter range see Fig. 2). This provides a simple example of spatiotemporal intermittency [8] that has been observed in a large class of dynamical systems, including the Maxwell-Bloch equations which describe an array of lasers submitted to an injected signal, a variant of the complex Ginzburg-Landau equation [9], reaction-diffusion equations which describe the pigmentation patterns molluscs [10], and some chemical reactions [11]. All these models have in common the existence of an Andronov homoclinic bifurcation for a limit cycle that leads to a fixed point, in the synchronous situation, i.e., when spatial effects are suppressed.

The particular model we have chosen has been constructed as the unfolding of a degenerate bifurcation: the normal form of the Bogdanov-Takens bifurcation [12] with reflexion symmetry  $U \rightarrow -U$ , in the presence of an imperfection which preserves the stationary solution. It reads

$$U_{tt} + \mu_1 U_t + \epsilon_1 U U_t + U^2 U_t + \mu_2 U + \epsilon_2 U^2 + U^3 = 0, \quad (1)$$

where  $\epsilon_1$  and  $\epsilon_2$  measure the effects of the imperfection. In order to analyze the transition in more detail, we study this one:

$$U_{tt} + \nu(U) U_t + \frac{\partial V}{\partial U} = U_{xx} + \kappa U_{xxt}, \quad (2)$$

where  $\nu(U) = -\mu + U^2$  and  $V = U^2/2 + aU^3/3 + U^4/4$  (see Fig. 3). When the terms of the right-hand side are set to 0, this equation becomes a homogeneous dynamical system that is a variant of the Van-der-Pol–Duffing equation. It possesses a flow similar to that of Fig. (3). The terms on the right-hand side of Eq. (2) represent propagation and diffusion respectively.

In the range of parameter “ $a_{SN} < a < a_M$ ,” the homogeneous dynamical system possesses three stationary solutions:  $U = 0$ ,  $U = -a/2 + \sqrt{\Delta}/2$ ,  $U = -a/2 - \sqrt{\Delta}/2$ , respectively noted  $\mathcal{A}$ ,  $\mathcal{B}$ , and  $\mathcal{C}$  (see Fig. 2), where  $\Delta = a^2 - 4$ .  $a_{SN} = 2$  stands for the value of  $a$  at which  $\mathcal{B}$  and  $\mathcal{C}$  disappear through a saddle node bifurcation and  $a_M = 2.12132$  stands for the value at which  $V_{\mathcal{A}} = V_{\mathcal{C}}$  (the Maxwell point of the potential  $V$  in phase transition terminology).

For negative  $\mu$ , both  $\mathcal{A}$  and  $\mathcal{C}$  are stable. When  $\mu$  crosses zero, a Hopf bifurcation renders  $\mathcal{A}$  unstable and leads to a

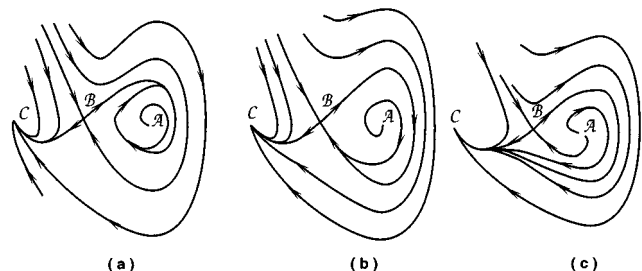


FIG. 1. Phase portrait of a typical second order oscillator that exhibits bistability between a limit cycle and a fixed point, as a typical parameter is varied. (a) Prior to the Andronov bifurcation, there are two stable attractors: a fixed point  $\mathcal{C}$  and a limit cycle surrounding the unstable focus  $\mathcal{A}$ . (b) At the bifurcation, the limit cycle becomes an homoclinic orbit. (c) After the bifurcation, the fixed point  $\mathcal{C}$  becomes a global attractor.

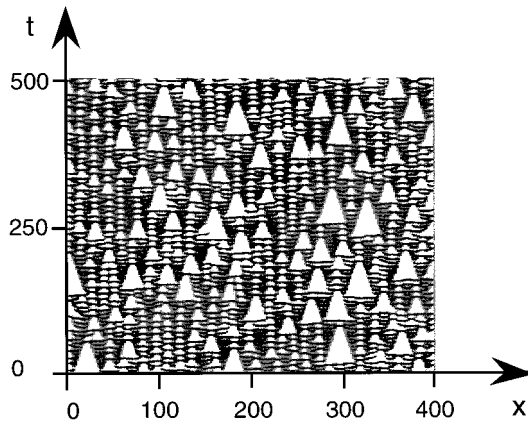


FIG. 2. Spatiotemporal record of the chaotic nucleation of the metastable phase (white areas).

stable limit cycle  $l$ . When  $\mu$  increases, the amplitude of this limit cycle  $l$  increases (like  $\sqrt{\mu}$  for small  $\mu$ ) until it disappears through the Andronov homoclinic bifurcation for  $\mu = \mu_A$ . For slightly greater values of  $\mu$ ,  $C$  becomes the global attractor of the dynamical system.

Spatially inhomogeneous perturbations modify this simple scenario. First, the homogeneous oscillation about  $A$  becomes unstable. Qualitatively speaking, the presence of the saddle point  $B$ , in the local phase portrait of the homogeneous dynamical system renders the oscillation soft: i.e., its frequency decreases as its amplitude increases. This property conflicts with the spatial coupling, which tends to increase its frequency as the wave number of the oscillation increases and leads to modulational instability [4,5]. This phenomenon can be understood in the following way: close to the birth of the oscillations ( $\mu \approx 0$ ), Eq. (2) can be reduced to the complex Ginzburg-Landau equation for the complex amplitude of the homogeneous oscillations, using standard asymptotics methods [4,13]

$$A_\tau = \mu A - (1 + i\alpha)|A|^2 A + (1 + i\beta)A_{xx}, \quad (3)$$

where  $\alpha = 10a^2/3 - 3$ ,  $\beta = -1$ ,  $\tau = t/2$ ,  $\kappa$  has been chosen equal to the unity for the sake of simplicity and

$$U = A e^{it} + \bar{A} e^{-it} - 2a|A|^2 + \frac{a}{3}(A^2 e^{2it} + \bar{A}^2 e^{-2it}) + \dots \quad (4)$$

Note that the homogeneous limit cycle oscillates with a non-zero mean value, which is consistent with the asymmetry of the ‘‘potential’’  $V$ . When  $a = 0$ , the Benjamin-Feir criterion ( $1 + \alpha\beta > 0$ ) for the stability of the homogeneous oscillations  $A = \sqrt{\mu} e^{-i\alpha\mu\tau}$  is satisfied. The modulational instability occurs for  $a \approx 1.095$ . It takes place before the saddle node bifurcation leading to the appearance of homogeneous solutions  $B$  and  $C$ . Equation (3) generally displays phase or defect turbulence [14,15].

In the following we choose a typical value of  $a = 2.08$  in the bistable regime such that  $V_A < V_C$ , and we increase the parameter  $\mu$  which controls the oscillatory instability. The first nucleation events occurs for a parameter value  $\mu = 0.12$  where the limit cycle has already disappeared ( $\mu_A = 0.0761$ ).

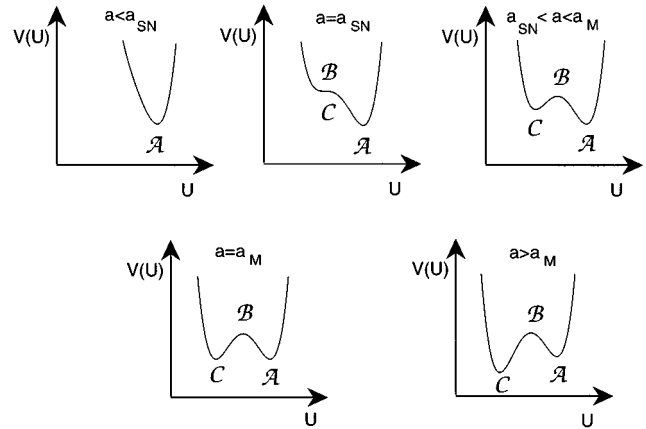


FIG. 3. ‘‘Potential’’  $V(U)$  for various values of  $a$ .

The Benjamin-Feir instability leads to defect mediated turbulence [14]. The presence of the defects, where the oscillation vanishes, reduces the mean square value of the fluctuations. This accounts for the parameter shift between the nucleation transition and the homoclinic bifurcation. A precise numerical computation of this mean square value allows us to get a good quantitative estimate of the nucleation threshold.

The domains of the state  $C$  (the white triangle in Fig. 2), once nucleated, retract almost uniformly in time with the velocity  $c \approx 0.210$ , for  $\mu \approx 0.2$ . The existence of an apparent constant velocity of a domain wall that separates the homogeneous stationary solution  $C$  and the chaotically oscillating state needs some explanations. The velocity of the front that connects the turbulent state and  $C$  may be computed by performing the bifurcation analysis of the weakly unstable front which connects  $A$  to  $C$ , near the value of  $a$  where  $V_A = V_C$  and  $\mu \approx 0$ . When  $V_A = V_C$ , the front that connects  $A$  to  $C$  is stationary and reads

$$U_{A-C} = -\frac{\sqrt{2}}{2} \left[ 1 + \tanh\left(\frac{x-p}{2}\right) \right], \quad (5)$$

where  $p$  represents the arbitrary position of the domain wall. For slightly positive  $\mu$ , this solution is unstable because  $A$  itself is unstable. As usual [16], this instability couples to the translation degree of freedom of the domain wall, and leads to its propagation. Close to the Maxwell condition for  $V$ , and for  $\mu \approx 0$ , the velocity of the perturbed front is found, as a solvability condition, by looking for solutions of Eq. (2) in the form  $U(x, t) = U_{A-C}(x-p) + W$ . One gets

$$\frac{\partial p}{\partial t} = C_{eq} + C_{neq}, \quad (6)$$

where

$$\begin{cases} C_{eq} = \frac{5\sqrt{2}}{2}(a_M - a) \\ C_{neq} = \frac{15}{4} \int_{-\infty}^{\infty} |\chi|^2 |A|^2 \left( \frac{\partial U_{A-C}}{\partial z} \right) \frac{\partial^3 V}{\partial U^3} \Big|_{U=U_{A-C}} dz \end{cases} \quad (7)$$

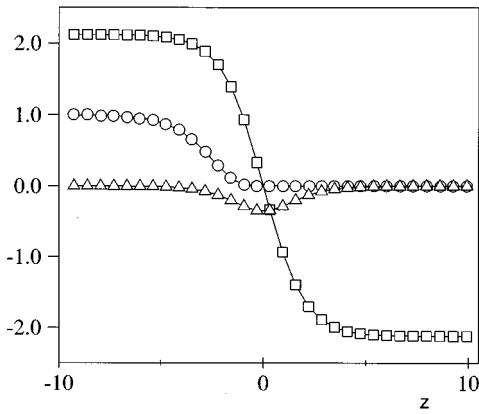


FIG. 4. Plot of the functions ( $\Delta$ )  $\partial U_{A-C}/\partial z$ , ( $\circ$ )  $|\chi|^2$ , ( $\square$ )  $\frac{1}{2}\partial^3 V/\partial U^3$ .

and  $W = A\chi e^{it} + c.c. + (\text{higher-order terms})$  (see Fig. 4). Here  $\chi$  represents the most unstable eigenfunction of the front solution. It obeys the equation  $L(x)\chi = 0$ , where

$$L = \frac{\partial^2}{\partial x^2} - \frac{(1-i)}{2} U_{A-C} [2a + U_{A-C}(3+i)] \quad (8)$$

and  $\partial\chi/\partial x = 0$  at the boundaries.  $A$  represents the complex envelop of the oscillations whose dynamics, determined at next order, is described by a generalization of the complex Ginzburg Landau equation.

The first term  $C_{eq}$  represents the shift in velocity observed for the domain wall induced by the relative change of the stability of the states  $A$  and  $C$  associated with the ‘‘potential’’  $V$ . The second term  $C_{neq}$ , is associated with the supercritical instability of the  $A$  state. Owe to the Benjamin-Feir instability [13],  $A$  is fluctuating. This term can be split into two parts: a constant part defined by its temporal mean value  $\langle C_{neq} \rangle$ , and a zero mean value fluctuating part.

The constant shift leads to a small correction to the Maxwell condition, obviously related to the asymmetry of the oscillations (see Fig. 5). Its behavior as a function of  $A$  is consistent with the symmetry  $A \rightarrow Ae^{i\phi}$  and  $t \rightarrow t + \phi$  of the reduced equations.

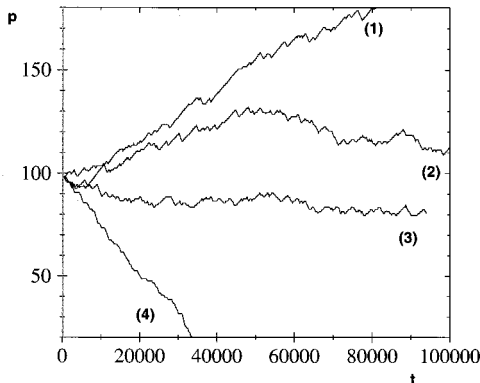


FIG. 5. Position of the interface as a function of the time for different values of  $a$  with  $\mu = 0.05$ . (1)  $a = 2.1205$ , (2)  $a = 2.1206$ , (3)  $a = 2.1207$ , (4)  $a = a_M = 2.1213$ .

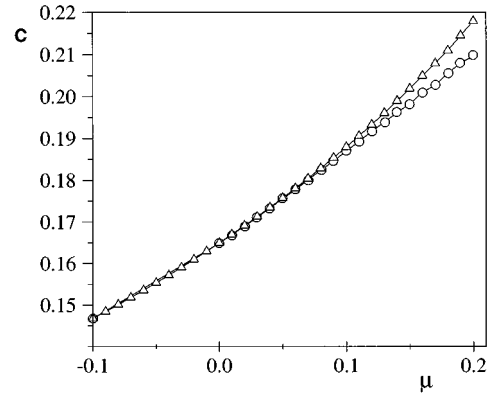


FIG. 6. Velocity of the fronts as functions of  $\mu$  for  $a = 2.08$ . ( $\Delta$ ) Velocity of the unstable front that connects  $C$  to  $A$ , ( $\circ$ ) averaged velocity of turbulent front.

Although, this formula does not apply for parameter values where the nucleation takes place, it gives a correct qualitative interpretation of the front motion. The front that connects  $C$  to the chaotic oscillation can be approximated, close to the core of the interface, as a perturbation of the unstable front  $U_{A-C}$  which connects  $A$  to  $C$ . This front and its velocity have been computed numerically by a shooting method allowing to find a heteroclinic solution of the equation

$$cU_{\xi\xi\xi} + (c^2 - 1)U_{\xi\xi} - cv(U)U_{\xi} + \frac{\partial V}{\partial U} = 0. \quad (9)$$

Equation (9) is obtained by looking solutions of Eq. (2) in the form  $U(\xi)$ , where  $\xi = x - ct$ .

The velocity of the actual interface can be obtained by a formula analogous to Eq. (6), where  $C_{eq}$  becomes the velocity of the unstable front  $U_{A-C}$ . Figure 6 shows the difference between the velocity of the interface  $U_{A-C}$  and the actual front. Close to  $\mu \geq 0$ , for a finite range of parameter, the instability is convective [17]. Consequently, there is no difference between the two velocities. When the instability becomes absolute, the contribution of the instability to the front motion becomes evident.

The behavior of the solutions when  $0.12 < \mu < 0.28$  in which the chaotic nucleation of the metastable phase  $C$  is observed presents many of the characteristics of statistical properties of spatiotemporal intermittency [8] (see Fig. 2). This will be detailed in a forthcoming work. In this regime, the fraction of ‘‘turbulent’’ state  $R$  decreases from 1.0 to 0.0, where the metastable phase ‘‘percolates’’ (see Fig. 7).

As usual, robust phenomena, which can be observed close to a degenerate bifurcation, are also likely to be observed for fully nonlinear systems [19]. It is in particular the case for the cascade of period doubling bifurcation that can be captured in the unfolding of a degenerate bifurcation corresponding to a triple zero eigenvalue.

We have described in this paper a spatiotemporal complex state that consists of the nucleation of metastable domains. The mechanism underlying this deterministic ‘‘cavitation process’’ is intimately related to the modulational instability of the oscillation induced by the bistability. Close to the ‘‘Maxwell point  $a_M$ ,’’ ‘‘robust localized states,’’ whose na-

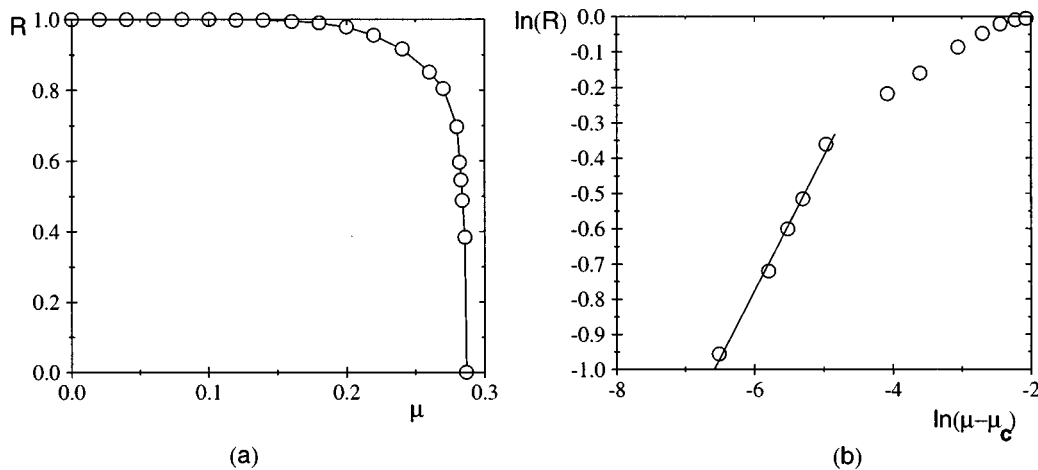


FIG. 7. Transition from turbulent state to stationary state. (a) Plot of the mean turbulent fraction  $R$  for  $a=2.08$ . (b) Log-log plot of  $R$  that emphasizes a critical exponent  $(\mu - \mu_c)^\beta$ , with  $\beta=0.38 \pm 0.02$  and  $\mu_c=0.287 \pm 0.01$ . The box's width is 100 000, and the interval of time is over 100.000 units of time.

ture seems to be different from those related to a subcritical Hopf bifurcation [18], are observed. Close to the spinodal point where the  $B$  and  $C$  disappear through a saddle node bifurcation, excitable waves are observed. The parameter range of their stabilities is particularly interesting. On one side, it is bounded by a transition from excitation to oscillation [20] and on the other side, by an instability of the excitable waves that leads to back emission of propagating

pulses [21]. Owing to the existence of an analog of surface tension, in two space dimensions, the nucleated domains, take the form of circular bubbles that eventually retract self-similarly in time.

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