

Hysteresis and Relaxation Phenomena of the Forced Oregonator

Yoshitaka YAMAMOTO and Kuninosuke IMAEDA

Faculty of Science, Okayama University of Science

1-1 Ridaicho, Okayama 700 Japan

(Received September 30, 1989)

Abstract

Responding to an external periodic force, the oscillation mode of the Oregonator shows a hysteresis phenomenon and a relaxation phenomenon according as the final value of the amplitude of the external periodic force is inside or outside of the bistable region. When the amplitude is changed with time while the frequency is kept a constant. In the outside of the bistable region near the transition points, we observed relaxation phenomena and obtained the critical exponent in the relation between the relaxation time and the difference of the control parameter from that of the critical point.

§1. Introduction

Hudson et al¹⁾ and Vidal et al²⁾ observed bifurcations and chaos phenomena in the temporal oscillation modes of the Br^- ion concentration in a continuous flow stirred tank reactor (CSTR) experiment of Belousov-Zhabotinskii (BZ) reaction³⁾. They observed a series of $\pi(m)$ modes consisting of m -peak periodic oscillations (m -P.O., $m=1,2,3,\dots$) and that of double periodic oscillation modes $\pi_{p,q}(m,m+1)$ consisting of p times of m -P.O. and q times of $m+1$ -P.O.. The multiplicity of the periodic oscillation depends on the flow rate of the solution into the reaction vessel i.e. the multiplicity increases with the flow rate. A double periodic mode $\pi_{p,q}(m,m+1)$ appears in a region between the two adjacent periodic modes $\pi(m)$ and $\pi(m+1)$.

In previous papers⁴⁻⁶⁾, we have reported the response of the Oregonator to an external periodic force $A\cos\omega\tau$ applied to the third component shows a single periodic mode $\pi(m)$ and a double periodic mode $\pi_{p,q}(m,m+1)$. Although there is no direct connection between the BZ reaction (CSTR) mechanism and the Oregonator response to the external periodic force, the similarity between the results of Hudson et al. and Vidal et al. and those of the forced Oregonator in the appearance of the oscillation mode $\pi(m)$ is conspicuous.

On the other hand, in our study⁵⁾ of the shift type piecewise continuous maps which approximate the attractor obtained by the forced Oregonator, we found that the piecewise continuous maps realize $\pi(m)$ and $\pi_{p,q}(m, m+1)$ modes. The order of appearance of these $\pi(m)$ and $\pi_{p,q}(m,m+1)$ does not depend essentially on the

mapping function and is quite similar to the cases of the CSTR experiments and of the forced Oregonator. Since the three different systems: the CSTR experiments, the forced Oregonator and the piecewise continuous maps which exhibit the same order of appearance of $\pi(m)$ and $\pi_{p,q}(m, m+1)$, we speculate that the characteristic feature of certain nonlinear systems under an influence of an external periodic force may be the same.

In a previous paper⁷⁾, we have observed hysteresis and relaxation phenomena in the states of the periodic oscillation modes of the forced Oregonator when the amplitude of the external periodic force is varied with time while the frequency is kept constant. A transition point of an oscillation mode differs according to whether the amplitude is increasing or is decreasing with time. Outside the bistable region and in a vicinity of the transition points, we have observed the relaxation phenomena which reveal a relation between the amplitude and the relaxation time. The relation can be expressed by a power law. We have obtained the critical exponents from the relation similar to that between the specific heat and the temperature in the critical phenomena of a matter at a transition point.

In this paper, we extended the study of the relaxation phenomena for different value of ω of the external periodic force. We report the relation between the value of critical exponent and the frequency of the external periodic force and determine the value of the critical exponent as a function of ω .

§2. Fundamental Equations and Computer Calculations

The Oregonator which simulates the BZ reaction under the influence of an external periodic force is expressed by a set of nonlinear differential equations as follows⁶⁻⁸⁾,

$$\begin{aligned} \varepsilon \frac{d\xi}{d\tau} &= \xi + \eta - q\xi^2 - \xi\eta \\ \frac{d\eta}{d\tau} &= -\eta + 2h\rho - \xi\eta \\ \frac{d\rho}{d\tau} &= (\xi - \rho) / p + A \cos \omega \tau \end{aligned} \quad (1)$$

where ξ , η , ρ are the concentrations of HBrO_2 , Br^- , Ce^{4+} components in the BZ solution, respectively, p , q , h , ε are parameters and are taken as fixed constants, 2.0, 0.006, 0.75, 0.03, respectively, and $A \cos \omega \tau$ is the external periodic force. We use the same notation convention as that used by Tyson⁹⁾. In the simulation calculation we take the amplitude A as a control parameter varying with time for different value of the angular frequency ω which is being fixed during the variation of the amplitude. We change the amplitude A after a transient oscillation which depends on initial condition has died out.

§3. Results on Hysteresis and Relaxation Phenomena

In a calculation the frequency ω of the external periodic force is kept at a constant, $\omega = 3.14159$. Then the amplitude A is kept constant $A = 40$ for $\tau < 100$ and after a transient oscillation has died out, we decrease the amplitude A linearly in time from 40 to 20, namely, $A = 40 - 0.2(\tau - 100)$ for an interval of time $100 \leq \tau \leq 200$. The oscillation mode of the system changes from $\pi(2)$ to $\pi(3)$ at $A = 31.6$ and the time variation of the oscillation of the η is shown in Fig.1. On the contrary, when we increase A from 20 to 40, $\pi(3)$ changes to $\pi(2)$ at a transition point $A = 32.7$. The transition points differ whether A is decreasing or increasing. This difference of the value of the transition point A is shown in Fig.2, where the solid lines (or the dotted lines) represent the minimum values of η when A is decreased (or increased) in time

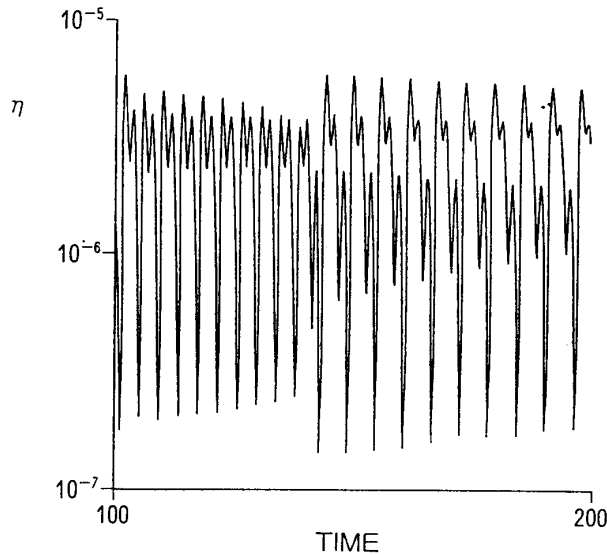


Fig.1. The change of the oscillation mode of the Oregonator in the component η from $\pi(2)$ to $\pi(3)$. For $\tau < 100$, the amplitude of the external periodic force $A \cos \omega \tau$ is 40. After a transient oscillation depending on the initial condition has died out, the amplitude A is decreased as $A = 40 - 0.2(\tau - 100)$ for $100 \leq \tau \leq 200$ while the frequency ω being fixed at 3.14159.

while ω is being fixed at 3.14159. The rate of change of A with time $dA/d\tau$ is taken as 0.04. In the region (I) $A > 32.7$, the oscillation mode is $\pi(2)$ and in the region (II) $A < 31.6$, it is $\pi(3)$. In the two regions (I) and (II), the modes are independent of whether A is increased or decreased (the values of η show a small difference in the lowest branch of the mode). But in the region (III) $31.6 < A < 32.7$ (we call the region a bistable region) when A is increasing, the mode is $\pi(3)$ while the mode is $\pi(2)$ when A is decreasing. Therefore, in the region (III) for the same value of A , the mode is different depending on whether A is increasing or decreasing. On the other

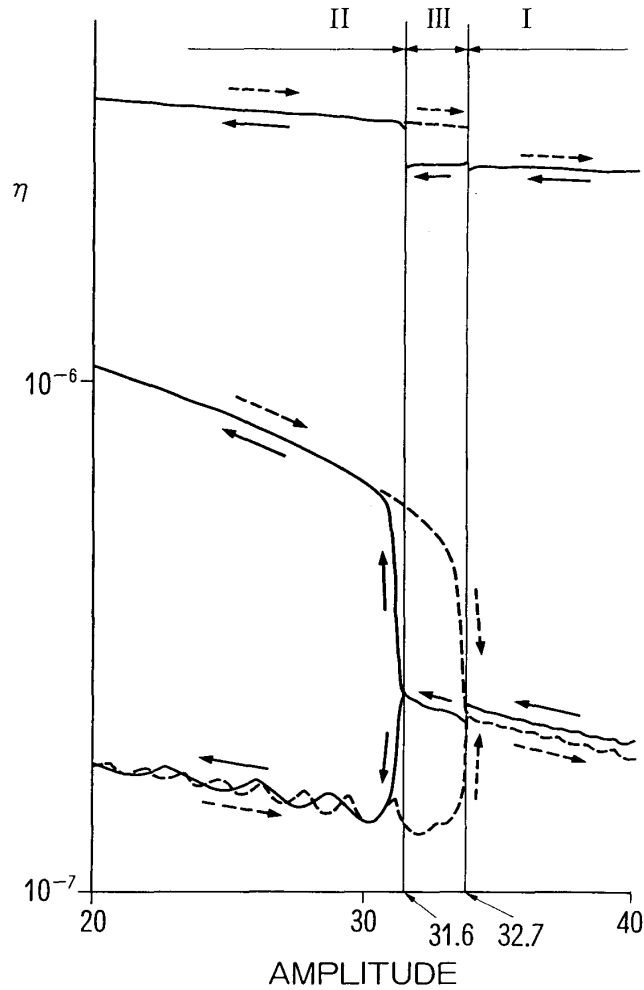


Fig.2. The trajectories of the minimum points of the temporal oscillation of the η -component when the amplitude of the external periodic force is increased from $A=20$ to 40 linearly in time (indicated by the dotted lines) and then decreased to $A=20$ (indicated by the solid lines) while the frequency is being fixed at 3.14159 . The rate of change of A with time, $dA/d\tau$ is 0.04 . In the region (I I I) $32.7 > A > 31.6$, the trajectories show a typical hysteresis phenomenon.

hand, when the value of A is changed from that less than 31.6 to a value A_f which is larger than 32.7 , the mode changes from $\pi(3)$ to $\pi(2)$, but it takes a time to jump to the final state $\pi(2)$. The $\pi(3)$ mode is a metastable state and the relaxation time depends on the difference $A_f - 32.7 = \Delta A$. We observed that as $\Delta A \rightarrow 0$, the relaxation time tends to infinity. Thus, we have confirmed a hysteresis phenomenon and a relaxation phenomenon in the response of the forced Oregonator.

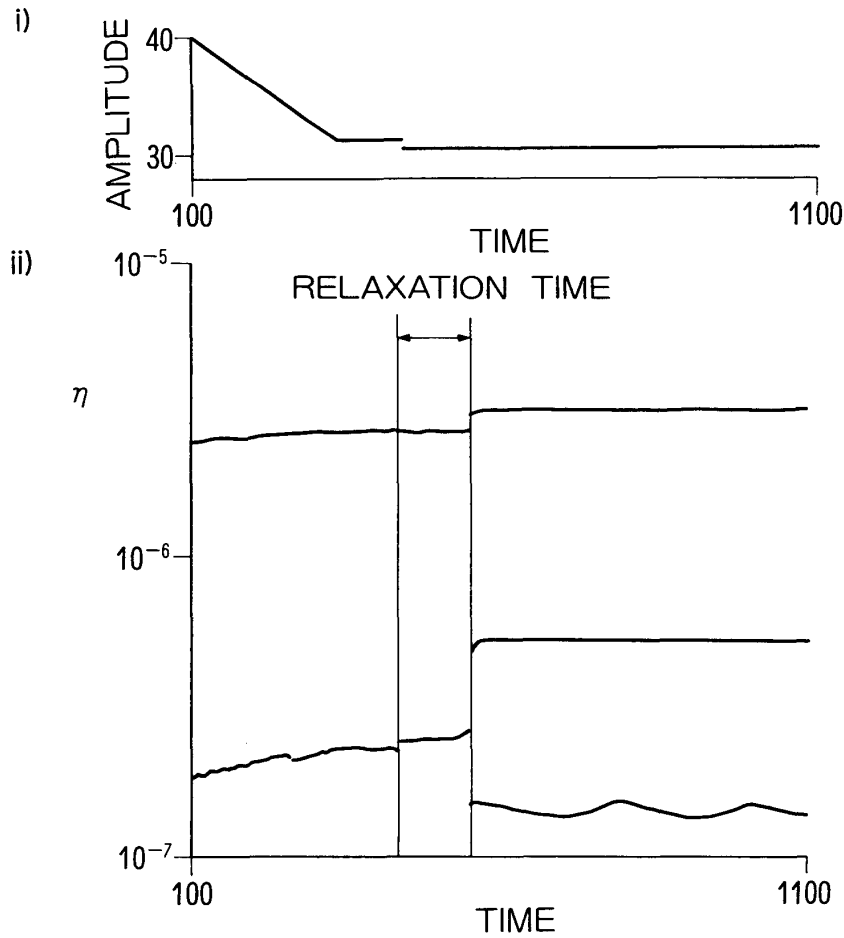


Fig.3. i) Figure shows the time change of A . For $\tau < 100$ the value of A is 40 and for $\tau \geq 100$, A is decreased as $A = 40 - 0.04(\tau - 100)$. When the value of A becomes 31, the value of A is maintained at 31 for a time 100. Then the value of A is decreased to $A_f = 30.335$ and then maintained for the rest of time while ω is fixed at 3.1.

ii) The mode is $\pi(2)$ for $A \geq 31.0$ and then the value of A is changed to 30.335. The mode still remains in $\pi(2)$ for a time 120 (relaxation time) After a relaxation time of 120 the mode suddenly changes to $\pi(3)$.

Figure 3 shows the relaxation time in the time oscillation of the η component when ω is fixed at 3.1. The value of A is decreased from 40 to 31 linearly (where $A = 31$ is the bistable state) and then it is maintained at 31 for a while. The mode stays in $\pi(2)$. Then the value of A is changed to $A_f = 30.335$ and this value is maintained after that. The mode remains in $\pi(2)$. After a time of 120, the mode changes suddenly to $\pi(3)$.

The relaxation time becomes longer as the final value A_f gets nearer to the transition point A_c , where the value of A_c is found to be 30.3399. The relaxation

between the value of $A_c - A_f$ and the relaxation time T is shown in Fig.4. The value of A is decreased linearly from 40 to 31, and is maintained at 31 for a time of 100. Then the value is jumped to A_f while ω is fixed at 3.1. For $A_f = 30.3399$, the relaxation time becomes infinity. We consider that the value of 30.3399 is the transition point from $\pi(2)$ to $\pi(3)$. The relation between the value of $A_c - A_f$ and the relaxation time T fits the familiar equation

$$T = B (A_c - A_f)^{-\lambda} \quad (2)$$

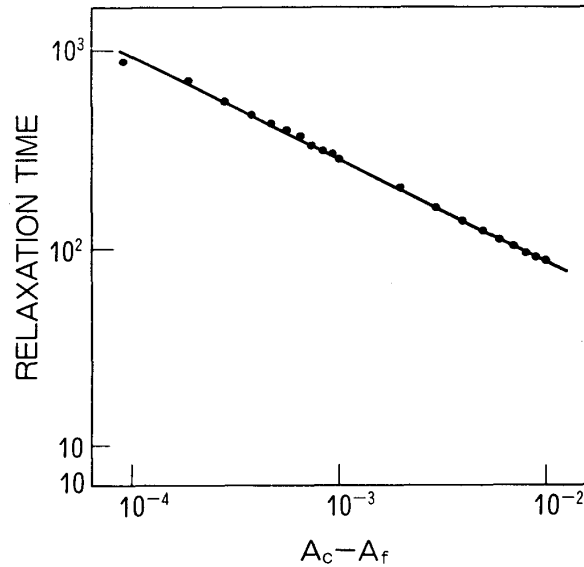


Fig.4. The relationship between $A_c - A_f$ and the relaxation time T . The value of A is decreased as in the case of Fig.9 i). ω is fixed at 3.1. The relationship between $A_c - A_f$ and the relaxation time T obtained is given by the expression $T = B(A_c - A_f)^{-\lambda}$, where we find $B = 7.92$, the transition point is $A_c = 30.3399$, and the critical exponent is $\lambda = 0.5173 \pm 0.0097$.

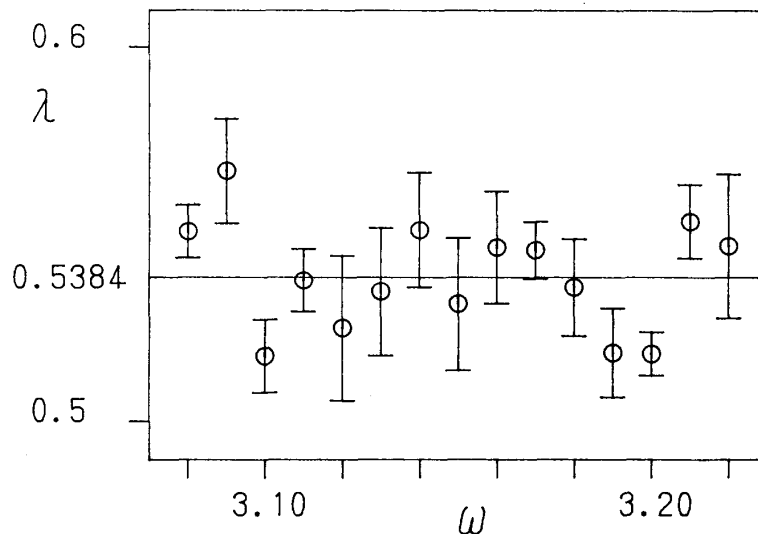


Fig.5. The critical exponent λ for various values of the frequency ω . The horizontal line is the average value of $\lambda = 0.5384$.

Figure 5 shows the critical exponents λ for the different values of $\omega = 3.08 \sim 3.22$. The Fig.5 shows that the value of λ does not depend on ω sensibly. The average value of the critical exponent is 0.5384. In Fig.5, nine critical exponents agree within errors and is approximately equal to 0.54. The values of all critical exponents obtained are between 0.51 and 0.57. Therefore, we think that the critical exponent does not depend sensitively on the value of ω and is a constant at about 0.54.

§4. Discussion and Conclusions

We have some reports on the relaxation phenomena in the response of nonlinear systems. Y. Morimoto⁹) observed the relaxation phenomena near the critical point in a forced Van der Pol oscillator equation. He obtained that the relation between the relaxation time and the distance of the amplitude A_f from the critical point A_c can be expressed by the same power law given by eq. (2).

For a two dimensional map, C. Grebogi et al.¹⁰) studied chaotic transients. In the chaotic transient, the average lifetime depends upon the system parameter p namely, as $T \sim |p - p_c|^{-\gamma}$, where p_c and γ are the values of p at the crisis and the critical exponent, respectively. The relation between the average lifetime and the system parameter is the same form as eq (2). But whether the mechanism which gives rise to the transient phenomena for the chaotic transient phenomena and for our case of the relaxation phenomena are the same or not is not yet clear to us.

In this study we report the critical exponent when the mode transits from $\pi(2)$ to $\pi(3)$. It is interesting to study the critical phenomena as the mode transits from $\pi(3)$ to $\pi(2)$, i. e., the value of A is increased to the bistable region and then changed to a value near the transition point so that the mode changes from $\pi(3)$ to $\pi(2)$ (but out side of bistable region).

In the critical phenomena, even though the hysteresis and relaxation phenomena are caused by different mechanisms, it often happens that the critical exponents obtained for these phenomena by the power law given by eq (2) are the same. The reason for this is similar in the case of phase transition in the property of matter. However we do not know whether the critical exponent can appear in different kinds of transitions in the systems expressed by a set of nonlinear differential equations or not.

Acknowledgments

One of authors (Y.Y.) would like to express his sincere thanks to Professor Takeo MATSUBARA for his advice in this research. We are grateful to Mr. Takahiro YAMAGUCHI for his continued help in this work.

References

- 1) J. L. Hudson, M. Hart and D. Marinko: J. Chem. Phys. **71** (1979) 1601.
- 2) C. Vidal, J. C. Roux, S. Bachelart and A. Rossi: C. R. Acad Sci. Paris **289** (1979) 17
- 3) P. De Kepper and J. Boissonade: J. Chem. Phys. **75** (1981) 189.

- 4) K. Imaeda: and T. Tei: J. Phys. Soc. Jpn. **55** (1986) 743.
- 5) K. Imaeda, Y. Yamamoto and T. Yamaguchi: J. Phys. Soc. Jpn. **56** (1987) 3832.
- 6) R. J. Field, E. Koros and: R. M. Noyes: J. Am. Chem. Soc. **94** (1972) 8649.
- 7) R. J. Field and E. Koros: J. Chem. Phys. **60** (1974) 1877.
- 8) J. J. Tyson: Belousov—Zhabotinskii Reaction, Lecture Notes in Biomathematics. Vol. 10, ed. S. Leven) Springer—Verlag, Berlin, Heidelberg, New York, 1976).
- 9) Y. Morimoto: J. Phys. Soc. Jpn. **55** (1986) 2937.
- 10) C. Grebogi, E. Ott, and J. A. Yorke: Phys. Rev. Lett. **57** (1986) 1284.