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Fluctuations in the Complex Stochastic TDGL Equation

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Behaviors of the fluctuation near the chemical turbulence are studied using the complex stochastic TDGL equation. The fluctuation of the amplitude of the limit cycle oscillation is shown not to diverge as the system approaches the chaotic state, while the correlation length is shown to diverge. The fluctuation of phase is also studied.

Phase transitions in far-from-equilibrium states have recently been studied by many people. Two types of nonequilibrium phase transitions have been studied. The first one is a soft mode instability, beyond which a spatial pattern appears. The second one is a hard mode instability, which is related to a limit cycle. The behavior of the system near the soft mode instability is quite similar to the phase transition in equilibrium states,[1] while a new feature of the fluctuation near the hard mode instability was pointed out by Tomita and Tomita.[2]

Another type of the dissipative structure, namely, chaos, has recently attracted many physicists. The behavior of the fluctuation as the system approaches the chaotic state, however, has not yet been studied sufficiently. In this letter, we will study this problem about the chaos in a reaction diffusion system. We start with the complex stochastic TDGL equation

$$\dot{w}(\boldsymbol{r},t)=\gamma w-g|w|^2 w+D\nabla^2 w+\eta(\boldsymbol{r},t). \tag{1}$$

Here, w denotes the complex amplitude of the limit cycle oscillation. The Gaussian random force is denoted by $\eta(\boldsymbol{r},t)$ which satisfies

$$\begin{cases}\langle\eta^*(\boldsymbol{r},t)\eta(\boldsymbol{r}',t')\rangle\\ \quad=2\varepsilon\delta(\boldsymbol{r}-\boldsymbol{r}')\delta(t-t'),\\ \langle\eta(\boldsymbol{r},t)\eta(\boldsymbol{r}',t')\rangle\\ \quad=\langle\eta^*(\boldsymbol{r},t)\eta^*(\boldsymbol{r}',t')\rangle=0\,.\end{cases} \tag{2}$$

Equation (1) without the random force was obtained by Kuramoto and Tsuzuki,[3] with the usage of the reductive perturbation near the bifurcation point in the reaction diffusion system. The random force $\eta(\boldsymbol{r},t)$ is included here to take fluctuations into account. This is justified[4] by the system size expansion.[5] We note that $g=g_1+ig_2$ and $D=D_1+iD_2$ are complex variables for the hard mode instability. We assume g_1 and D_1 are positive and use the notations $C_1=D_2/D_1$ and $C_2=g_2/g_1$.

It is known that Eq. (1) has a chaotic behavior when $(1+C_1C_2)$ is negative.[6] We study the behavior of fluctuations as $1+C_1C_2\to+0$. We confine ourselves only to the region near the steady limit cycle state and treat Eq. (1) by linearizing it around the reference state.

First, we put $w=Re^{i\varphi}$ and obtain

$$\begin{cases}\dot{R}=\gamma R-g_1R^3+D_1\{\nabla^2R-R(\nabla\varphi)^2\}\\ \qquad-D_2\{R\nabla^2\varphi+\nabla R\cdot\nabla\varphi\}+\eta_R\,,\\ \dot{\varphi}=-g_2R^2+D_1\left\{\nabla^2\varphi+\dfrac{1}{R}\nabla R\cdot\nabla\varphi\right\}\\ \qquad+D_2\left\{\dfrac{1}{R}\nabla^2R-(\nabla\varphi)^2\right\}+\dfrac{1}{R}\eta_\varphi\,,\end{cases} \tag{3}$$

where η_R and η_φ satisfy

$$\begin{aligned}&\langle\eta_R(\boldsymbol{r},t)\eta_R(\boldsymbol{r}',t')\rangle=\langle\eta_\varphi(\boldsymbol{r},t)\eta_\varphi(\boldsymbol{r}',t')\rangle\\ &\qquad=\varepsilon\delta(\boldsymbol{r}-\boldsymbol{r}')\delta(t-t'),\\ &\langle\eta_R(\boldsymbol{r},t)\eta_\varphi(\boldsymbol{r}',t')\rangle=0\,.\end{aligned} \tag{4}$$

Here, R and φ are real quantities which denote the amplitude and the phase of the oscillation of the limit cycle.

The limit cycle appears for $\gamma>0$ and is represented by

$$\begin{cases} R_0=\sqrt{\dfrac{\gamma}{g_1}}, \\ \varphi_0=-\dfrac{g_2}{g_1}\gamma t+\text{constant}. \end{cases} \qquad (5)$$

By linearizing Eqs. (3) around the reference state (5), we obtain the following equations:

$$\delta\dot{R}_{\boldsymbol{k}}=-(2\gamma+D_1k^2)\delta R_{\boldsymbol{k}} + D_2k^2\delta\Phi_{\boldsymbol{k}}+\eta_{\boldsymbol{k}}^R, \qquad (6)$$

$$\delta\dot{\Phi}_{\boldsymbol{k}}=-(2C_2\gamma+D_2k^2)\delta R_{\boldsymbol{k}} - D_1k^2\delta\Phi_{\boldsymbol{k}}+\eta_{\boldsymbol{k}}^{\varphi} \qquad (7)$$

after making the Fourier transformations

$$\delta R_{\boldsymbol{k}}=\int(R-R_0)e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}d\boldsymbol{r} = \int\delta Re^{-i\boldsymbol{k}\cdot\boldsymbol{r}}d\boldsymbol{r} \qquad (8)$$

and

$$\delta\Phi_{\boldsymbol{k}}=R_0\int(\varphi-\varphi_0)e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}d\boldsymbol{r} = \int\delta\Phi e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}d\boldsymbol{r}. \qquad (9)$$

We can study the fluctuations around the limit cycle state by diagonalizing the matrix

$$\begin{pmatrix} 2\gamma+D_1k^2 & -D_2k^2 \\ 2C_2\gamma+D_2k^2 & D_1k^2 \end{pmatrix}. \qquad (10)$$

The eigenvalues of this matrix are given by

$$\lambda_{\pm}=\gamma+D_1k^2 \pm(\gamma^2-2\gamma D_2C_2k^2-D_2{}^2k^4)^{1/2}. \qquad (11)$$

If the wave number k is small, we obtain

$$\lambda_+=2\gamma+D_1(1-C_1C_2)k^2$$

and $\lambda_-=D_1(1+C_1C_2)k^2$.

Thus, the correlation length corresponding to λ_+ is given by $\xi_+=[D_1(1-C_1C_2)/2\gamma]^{1/2}$. On the other hand, due to λ_-, which goes to 0 as k goes to 0, $\langle(\delta\Phi)^2\rangle$ shows the 'ensemble dephasing'. These expressions were studied previously by Mashiyama et al.[7] However, their results are not satisfactory, since the correlation length for the region $1-C_1C_2<0$ becomes imaginary within the above approximation and the inclusion of k^{2n} ($n>1$) terms change the behavior of fluctuations near the chaotic region also, as will be seen later. Therefore, we study Eqs. (6) and (7) without making a linearization about k^2.

After a straightforward calculation, we obtain

$$\langle\delta R_{\boldsymbol{k}}(t)\delta R_{\boldsymbol{k}'}(t)\rangle=\frac{\varepsilon\delta(\boldsymbol{k}+\boldsymbol{k}')}{B(k)} \times\left\{\frac{\gamma}{4\lambda_+}(\gamma-D_2C_2k^2+\sqrt{B})(1-e^{-2\lambda_+t}) -\frac{D_2k^2}{\lambda_++\lambda_-}(\gamma C_2+D_2k^2)(1-e^{-(\lambda_++\lambda_-)t}) +\frac{\gamma}{4\lambda_-}(\gamma-D_2C_2k^2-\sqrt{B})(1-e^{-2\lambda_-t})\right\},$$

$$\langle\delta\Phi_{\boldsymbol{k}}(t)\delta\Phi_{\boldsymbol{k}'}(t')\rangle=\frac{\varepsilon\delta(\boldsymbol{k}+\boldsymbol{k}')}{B(k)} \times\left\{\frac{\gamma(\gamma-D_2C_2k^2+\sqrt{B})(\gamma^2+B-2\gamma\sqrt{B})}{4\lambda_+D_2{}^2k^4}\times(1-e^{-2\lambda_+t}) -\frac{(\gamma C_2+D_2k^2)(2\gamma C_2+D_2k_2)}{\lambda_++\lambda_-}\times(1-e^{-(\lambda_++\lambda_-)t}) +\frac{\gamma(\gamma-D_2C_2k^2-\sqrt{B})(\gamma^2+B+2\gamma\sqrt{B})}{4\lambda_-\cdot D_2{}^2k^4}\times(1-e^{-2\lambda_-t})\right\} \qquad (12)$$

and

$$\langle\delta R_{\boldsymbol{k}}(t)\delta\Phi_{\boldsymbol{k}'}(t)\rangle=\frac{\varepsilon\delta(\boldsymbol{k}+\boldsymbol{k}')}{B(k)} \times\left\{\frac{\gamma(\gamma-D_2C_2k^2+\sqrt{B})(\gamma-\sqrt{B})}{4\lambda_+\cdot D_2k^2}\times(1-e^{-2\lambda_+t}) -\frac{\gamma(\gamma C_2+D_2k^2)}{\lambda_++\lambda_-}(1-e^{-(\lambda_++\lambda_-)t}) +\frac{\gamma(\gamma-D_2C_2k^2-\sqrt{B})(\gamma+\sqrt{B})}{4\lambda_-\cdot D_2k^2}\times(1-e^{-2\lambda_-t})\right\},$$

where B is defined by

$$B \equiv (\gamma^2 - 2\gamma D_2 C_2 k^2 - D_2^2 k^4).$$

From these expressions, we can calculate the correlation length, the strength of fluctuations $\langle \delta R(r)^2 \rangle$ etc. First, we note that there are three poles corresponding to $\lambda_+(k)=0$, $\lambda_-(k)=0$, $\lambda_+(k)+\lambda_-(k)=0$. (The pole $B(k')=0$ is spurious, since the numerator goes to 0 as $k\to k'$.) We note that $\lambda_-(0)=0$ holds. The pole at $k=0$ corresponds to the ensemble dephasing. This pole appears only for $\langle \delta\Phi_k \delta\Phi_{k'} \rangle$, since for the other quantities, the numerator is proportional to k^2 as $k\to 0$. Thus $\langle \delta\Phi(\boldsymbol{r})\delta\Phi(0)\rangle$ has a divergent behavior as t goes to ∞ for one or two dimensional systems as

$$\langle \delta\Phi(r,t)\delta\Phi(0,t)\rangle \sim \frac{(1+C_2^2)\varepsilon}{\sqrt{D_1}(1+C_1C_2)} \times \sqrt{t}\,, \quad (d=1)$$

$$\langle \delta\Phi(\boldsymbol{r},t)\delta\Phi(0,t)\rangle \sim \frac{(1+C_2^2)\varepsilon}{D_1(1+C_1C_2)} \log\left(\frac{D_1(1+C_1C_2)t}{2\gamma^2}\right). \quad (d=2)$$

For $d=3$, we have $\langle \delta\Phi(\boldsymbol{r})\delta\Phi(0)\rangle \sim (1+C_2^2)\varepsilon / \{D_1(1+C_1C_2)r\}$ as $t\to\infty$. This behavior was noted in a simpler model by Schnakenberg.[8] We note that the ensemble dephasing shows divergent behavior as the system approaches the instability point $1+C_1C_2=0$. This clearly shows the instability of the phase mode.

Next, we study the behavior of $\langle \delta R(\boldsymbol{r})\delta R(0)\rangle$ in the steady state ($t\to\infty$). By making the inverse Fourier transformation, we obtain

$$\langle \delta R(\boldsymbol{r})\delta R(0)\rangle \propto \frac{\varepsilon}{\sqrt{\gamma D_1}}\left(\frac{\gamma}{D_1}\right)^{(d-1)/4} \frac{1}{r^{(d-1)/2}} \frac{1}{(1+2C_1C_2-C_1^2)} \times\left\{(C_1C_2-C_1^2)e^{-r/\xi_1} + \left(\frac{1+C_1^2}{2}\right)^{(3-d)/4}(1+C_1C_2)^{(1+d)/4}e^{-r/\xi}\right\}, \quad (13)$$

where $r=|\boldsymbol{r}|$ and

$$\xi_1 = (D_1/\gamma)^{1/2}$$

and

$$\xi = \{D_1(1+C_1^2)/(2\gamma(1+C_1C_2))\}^{1/2}. \quad (14)$$

The result is valid for $d=1$ and 3, for arbitrary r and for $d=2$, Eq. (13) is valid only for large r. The behavior of correlation length is shown in Fig. 1. The correlation length ξ

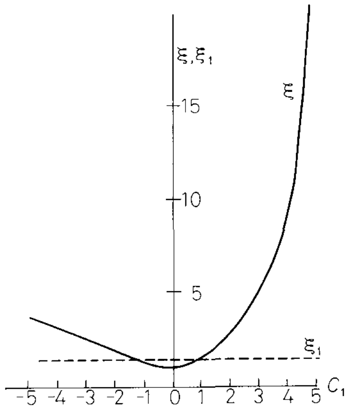

Fig. 1. The correlation lengths ξ and ξ_1 defined by Eq. (14) as functions of C_1. The ordinate is scaled by $\sqrt{D_1/(2\gamma)}$. We take $C_2=-0.2$.

diverges as $(1+C_1C_2)\to 0$. This is a common feature of fluctuations as the system approaches an instability point. In this case, however, the strength of a fluctuation does not diverge, even for $d=1$. The prefactor for $e^{-r/\xi}$ goes to 0 when $1+C_1C_2\to 0$ by the exponent $(1+d)/4$. Thus the contribution of the mode corresponding to ξ vanishes as $(1+C_1C_2)\to 0$. This is unfamiliar for usual phase transitions. For example, as $\gamma\to 0$, the correlation length $\sqrt{D_1/\gamma}$ diverges and the corresponding prefactor $\gamma^{(d-3)/4}$ does not vanish for $d\leq 3$. The point $\gamma=0$ corresponds to the bifurcation point to a limit cycle, and this transition point has a common feature to the phase transition in equilibrium, in the above sense. However, the instability point beyond which the system has a chaotic behavior, differs from usual transitions, in the sense that this instability is too weak about the fluctuation of the amplitude. The behavior of $\langle \delta R^2 \rangle$ is shown in Fig. 2 for $d=1$. In the same way, we can calculate the correlation $\langle \delta \Phi(\boldsymbol{r}) \delta R(0) \rangle$ and have

$$\langle \delta \Phi(\boldsymbol{r}) \delta R(0) \rangle \propto \frac{\varepsilon}{\sqrt{\gamma D_1}} \left(\frac{D_1}{\gamma} \right)^{(d-1)/4} \times \frac{1}{\gamma^{(d-1)/2}} \frac{C_1 - C_2}{(1+2C_1C_2 - C_1^2)} \times \left\{ e^{-r/\xi_1} - \left(\frac{1+C_1^2}{2(1+C_1C_2)} \right)^{(3-d)/4} e^{-r/\xi} \right\}. \quad (15)$$

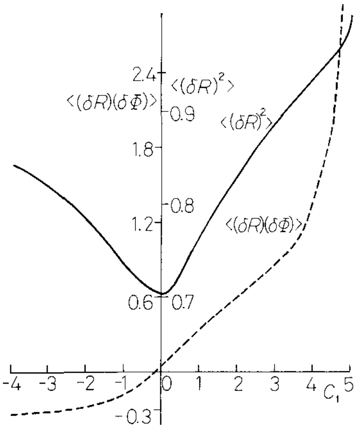

Fig. 2. The fluctuations $\langle (\delta R)^2 \rangle$ and $\langle (\delta R)(\delta \Phi) \rangle$ as functions of C_1, for the same value of C_2 as in Fig. 1. The ordinate is scaled by $\varepsilon / (8\pi\sqrt{\gamma D_1})$.

Here the result for $d=2$ is valid only for large r. Thus, $\langle \delta \Phi(\boldsymbol{r}) \delta R(0) \rangle$ diverges as $(1+C_1C_2)$ goes to 0 with the singularity $(1+C_1C_2)^{(3-d)/4}$ for $d=1$ or 2. The behavior of $\langle \delta \Phi \delta R \rangle$ for $d=1$ is shown in Fig. 2. Therefore, the effect of the instability of the system appears more strongly in the fluctuation of the phase.

Our results are confined within the Gaussian approximation. (See Eqs. (6) and (7).) In the very vicinity of the critical point $\gamma \to 0$ or $1+C_1C_2 \to 0$, we have to take account of the terms neglected in Eqs. (6) and (7). Although it will be rather difficult, the renormalization group[1] may be a useful method in this case.

We can control the parameters C_1 and C_2 by changing the diffusion constants and the concentrations of the two reactants. Experimental evidence of the existence of a chemical turbulence has recently been reported,[9),10)] and it is expected that as the system approaches a chaotic condition the predicted behavior of fluctuations will be observed through some experiments with spectroscopical methods.

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