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Theoretical Study of a Chemical Turbulence

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Recently Kuramoto and one of the present authors have carried out a computer simulation for a chemically oscillating system and found a turbulence-like behavior similar to the hydrodynamic turbulence. The steady turbulent state of this system is theoretically studied. It is shown that there exist two characteristic regions of wavenumber k. One is a cascade region with $k\xi \ll 1$, and the other is a dissipative region with $k\xi > 1$, where ξ is a characteristic length which is much larger than the reaction mean free path l_r. Over these two regions the characteristic frequency Ω_k and the variance χ_k of phase fluctuations obey the scaling laws $\Omega_k = k^4 F(k\xi)$ and $\chi_k = k^3 G(k\xi)$, where $F(x)$ and $G(x)$ are universal functions of x. In a cascade region it is shown that $F(x) = c_1 x^{-5/2}$ and $G(x) = c_2 x^{-5}$. In a dissipative region with $k\xi \gg 1$, it is found that $G(x) = 1$ and $G(x) = c_3 \exp(-c_4 x)$. An explicit calculation of Ω_k and χ_k over the whole k region is carried out in the Kraichnan direct interaction approximation, and the results are compared with the computer simulation.

§ 1. Introduction

Recently many theoretical and experimental investigations on collective phenomena associated with chemical instabilities have been developed in various contexts.[1] Chemical reactions are usually considered on the assumption of spatial homogeneity. However, various phenomena can be understood by including a spatial inhomogeneity. For example, spiral waves and target patterns of concentrations associated with a temporal collective order cannot be understood without a spatial inhomogeneity.

Zhabotinsky[2] has reported a new, interesting phenomenon in which a spatial inhomogeneity plays a crucial role. In the Belousov-Zhabotinsky reaction, Zhabotisky has found that when α_2, the ratio of the oxidizer (BrO_3^-) concentration to the reducer ($CHBr(COOH)_2$) concentration, is smaller than 0.3, a macrooscillation synphase in space can exist under any externally stirring condition. On the other hand, when $\alpha_2 \geqq 1$, the macrooscillations depend on stirring conditions. Namely, without a stirring effect no macrooscillation has been detected, although oscillations exist locally, and then macroscopic patterns behave irregularly in space and time. When he stirred that system, the irregular motion instantly disappears and the synchronization of the oscillations revives.

Very recently, Kuramoto and one of the present authors[3] have studied the collective oscillation phenomena using the kinetic equation

$$\frac{\partial}{\partial t}w = (1+ic_0)\,w + (1+ic_1)\nabla^2 w - (1+ic_2)\,|w|^2 w\,, \tag{1·1}$$

where $w(\boldsymbol{r}, t)$ is a complex concentration of reaction molecules. A spatial inhomogeneity has been included in the diffusion term. They have shown that when $1+c_1c_2>0$, (1·1) has solutions which describe target patterns and spiral waves.[4] When $1+c_1c_2<0$, the above solutions become unstable for a long wavelength perturbation.[5),6)] In this case, (1·1) has been solved numerically,[7] leading to an irregular behavior in space and time, which is similar to the irregular motion in Zhabotinsky's experiment.

Furthermore one of the present authors and Kuramoto[8] (this is referred to as I) have numerically integrated the reduced equation of motion of (1·1),*)

$$\frac{\partial}{\partial t}\theta = (-\xi^{-2}\nabla^2 - \nabla^4)\,\theta + (\nabla\theta)^2\,, \tag{1·2}$$

where $\theta(\boldsymbol{r}, t)$ is the reduced phase of w, and the characteristic length ξ is defined by

$$\xi = \frac{l_r}{\sqrt{-(1+c_1c_2)}} \tag{1·3}$$

with l_r being a length of the order of the reaction mean free path. Equation (1·2) has been derived by eliminating the amplitude of w near $1+c_1c_2=0$. They met also an irregular motion. This irregular motion will be called 'chemical turbulence', hereafter. Carrying out a statistical treatment, they have calculated an equal-time correlation function $\overline{|\theta_k|^2}$ in a small wavenumber region such that $|k|<\xi^{-1}$, where the bar denotes a time-average in a steady turbulent state, and obtained $\overline{|\theta_k|^2}\propto k^{-2}$. In the one-dimensional case, the nonlinearity of the equation of motion for $u=\nabla\theta$ has the same form as the Burgers equation.[9] This suggests that a statistical theory of chemical turbulences can be developed from (1·2) similarly to the hydrodynamic case.[10]

It is the main purpose of the present paper to carry out this program. In § 2 the model is described and its some general properties are derived by simple and physical arguments. It is shown that there exist two characteristic regions of wavenumber which have different forms for the scaling functions from each other. In § 3 we derive self-consistent equations for the relevant quantities characteristic of the steady chemical turbulence, and results are compared with the computer simulation in I. Section 4 is devoted to summary and discussion.

§ 2. One-dimensional model and its general properties

In terms of the Fourier transform of $\theta(r, t)$,

*) By putting θ, $\boldsymbol{r}$ and t by $\xi^{-2}\theta$, $\xi\boldsymbol{r}$ and $\xi^4 t$, respectively, (1·2) becomes[8] $(\partial/\partial t)\theta = (-\nabla^2-\nabla^4)\theta + (\nabla\theta)^2$. This equation has been solved in I instead of (1·2) itself.

$$\theta_k(t)=\frac{1}{\sqrt{\Omega}}\int e^{-ikr}\theta(r,t)\,dr,\quad \theta(r,t)=\frac{1}{\sqrt{\Omega}}\sum_k e^{ikr}\theta_k(t), \tag{2·1}$$

Ω being the size of the system, (1·2) takes the form

$$\frac{\partial}{\partial t}\theta_k(t)=-\Gamma_k{}^0\theta_k(t)+v_k(\theta(t)), \tag{2·2}$$

$$\Gamma_k{}^0\equiv-\xi^{-2}k^2+k^4,\quad v_k(\theta)\equiv-\frac{1}{\sqrt{\Omega}}\sum_{k'}k'(k-k')\theta_{k'}\theta_{k-k'}. \tag{2·3}$$

In the absence of the nonlinear coupling, a small perturbation θ_k grows or damps according as $|k|<\xi^{-1}$ or $|k|>\xi^{-1}$, respectively, and the maximum growth rate occurs at $k_m=2^{-1/2}\xi^{-1}$. The nonlinear term can easily be shown to satisfy an 'energy' conservation law

$$\sum_k k^2\theta_k(\dot{\theta}_{-k})_{\rm nl}=\sum_k k^2\theta_k v_{-k}(\theta)=0, \tag{2·4}$$

which plays an important role in the following analysis: The above property is similar to the hydrodynamic case. As stated in § 1, the numerical integrations of (2·2) with deterministic initial values show a turbulence-like irregular motion.[8]

This irregular motion originates from an instability of modes and the cascade process through nonlinear terms. The collisions (or the cascade process) among the unstable modes rapidly wash out the initial memory. Therefore, it is appropriate to adopt a statistical description, in which the time-average is replaced by an ensemble average. The same situation arises in the hydrodynamic turbulence.

Thus the probability distribution function $P(\theta,t)$ is given by an ensemble average of

$$\delta(\theta(t)-\theta)\equiv\prod_k\delta(\theta_k(t)-\theta_k), \tag{2·5}$$

where δ is the Dirac delta function. Equation (2·2) leads to

$$\frac{\partial}{\partial t}P(\theta,t)=-\mathcal{H}(\theta)P(\theta,t), \tag{2·6}$$

where

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}',$$
$$\mathcal{H}_0=\sum_k\tilde{\theta}_k\Gamma_k{}^0\theta_k,\quad \mathcal{H}'=-\sum_k\tilde{\theta}_k v_k(\theta), \tag{2·7}$$

with $\tilde{\theta}_k\equiv-\partial/\partial\theta_k$. The steady turbulent state is characterized by the steady distribution function $P_s(\theta)$ which satisfies

$$\mathcal{H}(\theta)P_s(\theta)=0. \tag{2·8}$$

Furthermore the energy conservation law (2·4) is reduced to

$$\sum_k\Gamma_k{}^0k^2\langle|\theta_k|^2\rangle=0, \tag{2·9}$$

where $\langle\cdots\rangle$ is the average over $P_s(\theta)$. This can be rewritten as

$$\sum_k^{<}|\Gamma_k{}^0|k^2\langle|\theta_k|^2\rangle=\sum_k^{>}|\Gamma_k{}^0|k^2\langle|\theta_k|^2\rangle \tag{2·10}$$

with $\sum_k^{<}$ and $\sum_k^{>}$ defining the sums over the regions $|k|<\xi^{-1}$ and $|k|>\xi^{-1}$, respectively. Equation (2·10) shows that growing irregularities in the small wavenumber region with $|k|<\xi^{-1}$ flow out toward the large wavenumber region (dissipative region) with $|k|>\xi^{-1}$, in which intensities die out. Thus we can describe the present turbulence as an 'energy' cascade process. In terminology of a hydrodynamic turbulence,[11] the energy-input to the system occurs in the region with $|k|<\xi^{-1}$, and the 'energy' flows toward the region with $|k|>\xi^{-1}$ through nonlinear couplings. It should be noted that in a hydrodynamic turbulence many authors[12] have introduced an external stirring force which maintains a steady turbulence, and which is represented by an additional diffusion term to (2·6), whereas in the present model the mechanism of 'energy'-input is automatically included in the wavenumber region with a negative $\Gamma_k{}^0$.

It is difficult to solve (2·6) and (2·8) exactly. Some general properties, however, can be derived by simple and physical arguments. First the reductive perturbation method[5] ensures the forms of the characteristic frequency Ω_k and the variance $\chi_k(\equiv\langle|\theta_k|^2\rangle)$ as follows: $\Omega_k=\xi^{-4}f(k\xi, \xi/l_r)$ and $\chi_k=\xi^{-3}g(k\xi, \xi/l_r)$, where $f(x, y)$ and $g(x, y)$ are universal functions of x and y. The cutoff wavenumber k_c is assumed such that $\xi>k_c{}^{-1}\gg l_r$. Thus in the limit $\xi/l_r\to\infty$, $(k_c l_r\to 0)$, one finds

$$\Omega_k=\xi^{-4}f(k\xi), \tag{2·11}$$

$$\chi_k=\xi^{-3}g(k\xi) \tag{2·12}$$

with $f(x)\equiv f(x, \infty)$ and $g(x)\equiv g(x, \infty)$. In analogy with a thermodynamic critical phenomena,[13] these relations will be called scaling laws, hereafter.

Since the energy cascade is realized through nonlinear couplings, we may assume the existence of a cascade region characterized by $|k|\ll\xi^{-1}$ where $v_k(\theta)$ dominates over the other terms. In this region the steady distribution $P_s(\theta)$ must satisfy

$$\mathcal{H}'(\theta)P_s(\theta)=0. \tag{2·13}$$

The solution of (2·13) consistent to the above assumption is found to be a Gaussian distribution

$$P_s(\theta)\propto\exp\left[-\frac{1}{2}\sum_k{}'\frac{\theta_k\theta_{-k}}{\chi_k}\right]. \tag{2·14}$$

Here the prime denotes the sum over the cascade region, and the variance is given by

$$\chi_k=\text{const}\ \xi^{-5}|k|^{-2}, \tag{2·15}$$

where we have used the scaling law (2·12). Hence we obtain $g(x)\propto x^{-2}$ for $x\ll 1$. The numerical constant factor in (2·15) can be determined by including the statistics of large wavenumber modes. Equation (2·15) is equivalent to the scaling $\theta_k \to L\theta_k$ with

$$k\to k/L\,,\quad \xi\to\xi\,,\quad l_r\to l_r\,,\quad (L\gg 1) \tag{2·16}$$

for $|k|\xi\ll 1$. Hence Mori's scaling method[14),15)] leads to the scaling $v_k(0)\to L^{-1/2}v_k(0)$. Since this is larger than $\Gamma_k{}^0$ whose scaling is $\Gamma_k{}^0\to L^{-2}\Gamma_k{}^0$ for $L\gg 1$, the characteristic frequency Ω_k is determined by $v_k(\theta)$ and is scaled similarly to $v_k(0)/\theta_k$. This leads to the frequency spectrum

$$\Omega_k = \text{const}\ \xi^{-5/2}|k|^{3/2}\,, \tag{2·17}$$

where we have used (2·11). Hence one finds $f(x)\propto x^{3/2}$ for $x\ll 1$. Furthermore (2·15) is equivalent to $\langle|u_k|^2\rangle=\text{const}$ with $u=\nabla\theta$. This spectrum and (2·17) seems to be a feature characteristic of the one-dimensional system with a mode-coupling of the inertial-term type, and indeed the same spectra as these have been derived by Mori about hydrodynamic fluctuations.[14),15)]

The dissipative region with large wavenumbers has a quite different beahvior. In this region the linear term as well as the nonlinear coupling plays a crucial role. With the characteristic length ξ the dissipative region is specified by $|k|\xi>1$. Mori's scaling method leads to the scaling

$$k\to k/L\,,\quad \xi\to L\xi\,,\quad l_r\to l_r\,,\quad (L\gg 1). \tag{2·18}$$

Furthermore since the magnitude of the irregularity is smaller in this region than in the cascade region, it is expected that the renormalization of $\Gamma_k{}^0$ by nonlinear mode-couplings can be neglected. Thus one can put $\Gamma_k=k^4$. This leads to $f(x)=x^4$ for $x\gg 1$ from (2·11). In this stage, however, we cannot determine the asymptotic behavior of $g(x)$ for $x\gg 1$, and further investigations are necessary. A detailed analysis of the whole k region will be given in the next section.

§ 3. Self-consistent equations

In order to simplify the discussions we will start with the scaled quantities $\hat{k}$, $\hat{r}$, $\hat{t}$ and $\hat{\theta}(\hat{r},\hat{t})$ defined by $\hat{k}\equiv k\xi$, $\hat{r}\equiv r/\xi$, $\hat{t}\equiv\xi^{-4}t$ and $\hat{\theta}(\hat{r},\hat{t})\equiv\xi^2\theta(r/\xi,\xi^{-4}t)$.[8)] Furthermore, the symbol $\hat{A}$ will be neglected, hereafter, as long as stated otherwise. These scaled equations are the same as (2·2) and (2·3) with $\xi=1$. After all the calculations are carried out, we will return to the original units of the system by replacing k and r *by* $k\xi$ and r/ξ, respectively.

(A) *Diagrammatic method*[16),17)]

Let us consider the steady turbulent state and introduce two propagators by

$$\langle(\theta_{k_1}(t_1)\theta_{k_2}(t_2))_+\rangle=\delta_{k_1-k_2}F_{k_1}(t_1-t_2)k_1^{-2}\,, \tag{3·1}$$

$$\langle(\theta_{k_1}(t_1)\tilde{\theta}_{k_2}(t_2))_+\rangle=\delta_{k_1-k_2}G_{k_1}(t_1-t_2) \tag{3·2}$$

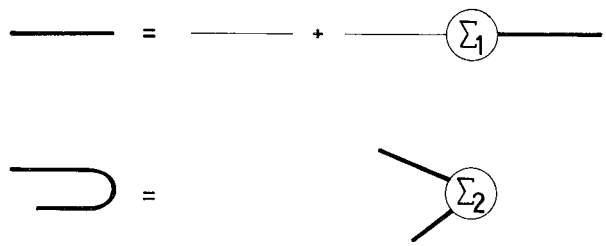

Fig. 1. Dyson equations for the propagators G and F. The thin straight line, the thick straight line and the thick curved line represent the propagators $G_k^0(t)\equiv\eta(t)\exp(-\Gamma_k^0 t)$, $G_k(t)$ and $F_k(t)$, respectively, where $\eta(t)$ is a step function; $\eta(t)=1$ for $t>0$ and 0 otherwise.

with the definitions of $\theta_k(t)$ and $\tilde{\theta}_k(t)$,

$$\begin{pmatrix}\theta_k(t)\\ \tilde{\theta}_k(t)\end{pmatrix}=\exp[t\mathcal{H}(\theta)]\begin{pmatrix}\theta_k\\ \tilde{\theta}_k\end{pmatrix}\exp[-t\mathcal{H}(\theta)], \tag{3·3}$$

where δ_k is unity if $k=0$, and zero otherwise. Here $(\cdots)_+$ denotes a time ordering operator such that operators are ordered from the right to the left as time increases and the average is given by $\langle(\cdots)\rangle\equiv\int\prod_k d\theta_k(\cdots)P_s(\theta)$, $P_s(\theta)$ being the steady distribution function given by (2·8) with $\Gamma_k{}^0=-k^2+k^4$.

The Dyson equations for G and F are shown in Fig. 1. These yield

$$\frac{\partial}{\partial t}G_k(t)=\delta(t)-\Gamma_k{}^0G_k(t)+\int_{-\infty}^{\infty}ds\Sigma_{1k}(t-s)G_k(s), \tag{3·4}$$

$$F_k(t-t')=\int_{-\infty}^{\infty}ds_1\int_{-\infty}^{\infty}ds_2G_k(t-s_1)G_{-k}(t-s_2)\Sigma_{2k}(s_1-s_2). \tag{3·5}$$

Now the Fourier components are defined by*)

$$X(k,\omega)=\int_{-\infty}^{\infty}dt\,X_k(t)\exp(i\omega t), \tag{3·6}$$

where a function X denotes one of G, F, Σ_1 and Σ_2. The equations of motion for G and F become

$$-i\omega G(k,\omega)=1-[\Gamma_k{}^0-\Sigma_1(k,\omega)]G(k,\omega), \tag{3·7}$$

$$-i\omega F(k,\omega)=-[\Gamma_k{}^0-\Sigma_1(k,\omega)]F(k,\omega)+G(-k,-\omega)\Sigma_2(k,\omega), \tag{3·8}$$

respectively. The self-energy Σ_1 represents a renormalization of the bare growth or damping rate Γ^0 by the nonlinear coupling, and the last term in the r.h.s. of (3·8) represents the nonlinear source term by which the system is maintained to be steady in time. Integrating the both sides of (3·8) over ω, we obtain

$$0=\frac{1}{2}\dot{I}_k=-\Gamma_k{}^0I_k+\frac{1}{2\pi}\int_{-\infty}^{\infty}[\Sigma_1(k,\omega)F(k,\omega)+\Sigma_2(k,\omega)G(-k,-\omega)]d\omega \tag{3·9}$$

*) We here assume the stability of the steady state or the existence of the Fourier components, $G(k,\omega)$ and $F(k,\omega)$, etc.

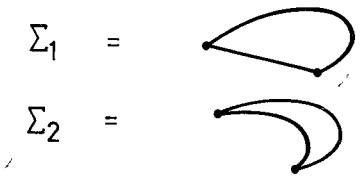

Fig. 2. Lowest order diagrams for the self-energy parts of G and F.

with $\Gamma_k^0 = -k^2 + k^4$, where

$$I_k = \frac{1}{2\pi}\int_{-\infty}^{\infty} F(k, \omega)\, d\omega = k^2 \chi_k \,. \tag{3·10}$$

The self-energy parts Σ_1 and Σ_2 in the lowest order approximation are shown in Fig. 2. On the assumption of the following equations

$$G(k, \omega) = (-i\omega + \Gamma_k)^{-1}, \tag{3·11}$$

$$F(k, \omega) = [G(k, \omega) + G(-k, -\omega)]\, I_k, \tag{3·12}$$

the contributions from these diagrams can be evaluated as

$$\begin{aligned}\Sigma_1(k, \omega) &= -\int_{k'}\int_{\omega'} G(k', \omega') F(k-k', \omega-\omega')\, 4kk' \,, \\ &= -\int_{k'} \frac{4kk' I_{k-k'}}{\Gamma_{k'} + \Gamma_{k-k'} - i\omega} \,, \end{aligned} \tag{3·13}$$

$$\begin{aligned}\Sigma_2(k, \omega) &= \frac{k^2}{2}\int_{k'}\int_{\omega'} 4F(k', \omega') F(k-k', \omega-\omega') \,, \\ &= \frac{k^2}{2}\int_{k'} 4I_{k'} I_{k-k'} \left(\frac{1}{\Gamma_{k'} + \Gamma_{k-k'} - i\omega} + \frac{1}{\Gamma_{k'} + \Gamma_{k-k'} + i\omega} \right), \end{aligned} \tag{3·14}$$

where $\int_{k'} \equiv (1/2\pi)\int_{-\infty}^{\infty} dk'$ and $\int_{\omega'} \equiv (1/2\pi)\int_{-\infty}^{\infty} d\omega'$. Equations (3·11) and (3·12) exactly hold on a Markoffian assumption. Finally, substitution of these expressions into (3·11) and (3·12) yields the coupled equations for Γ_k and I_k

$$\Gamma_k = \Gamma_k^0 - \Sigma_1(k, 0) = \Gamma_k^0 + \frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{4kk' I_{k-k'}}{\Gamma_{k'} + \Gamma_{k-k'}} dk' \,, \tag{3·15}$$

$$\begin{aligned} 0 = &-\Gamma_k^0 I_k - \frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{4kk' I_{k-k'}}{\Gamma_{k'} + \Gamma_{k-k'} + \Gamma_k} I_k dk' \\ &+ \frac{k^2}{2}\frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{4 I_{k'} I_{k-k'}}{\Gamma_{k'} + \Gamma_{k-k'} + \Gamma_k} dk' \,. \end{aligned} \tag{3·16}$$

The set of (3·15) and (3·16) is equivalent to the one derived by Kraichnan's direct interaction approximation[18] in the theory of the hydrodynamic turbulence when a Markoffian approximation is adopted in the latter. It is worth while to note that in (3·16) the sum-rule $\sum \Gamma_k^0 I_k = 0$ is satisfied. Asymptotic solutions

of (3·15) and (3·16) in the small and large wavenumber regions can be derived in the following.

(a) *Small wavenumber (cascade) region* ($k \ll 1$)

The dominant term in this region is the nonlinear mode-coupling term, and the asymptotic solution can be obtained in the limit, $\Gamma_k^0 \to 0$. By setting $\Gamma_k = \Gamma_1 k^\phi$ and $I_k = I_1 k^\alpha$ in (3·15) and (3·16), k being $|k|$, a self-consistent calculation yields $\phi = 3/2$ and $\alpha = 0$, and thus $\chi_k = I_1 k^{-2}$. These results are consistent with the assumption $\Gamma_k \gg |\Gamma_k^0|$, and agree with (2·15) and (2·17) with $\Omega_k = \Gamma_k$. From (3·15) constant factors I_1 and Γ_1 are related each other by

$$\frac{\Gamma_1^2}{I_1} = \frac{4}{2\pi} \int_{-\infty}^{\infty} dx \frac{x}{|x|^{3/2} + |1-x|^{3/2}} \simeq 4 \times 0.311 \simeq 1.24 . \tag{3·17}$$

The I_1 and Γ_1 cannot be determined, since in the limit $\Gamma_k^0 \to 0$, (3·16) is always satisfied for any I_1 and Γ_1. Thus in the small wavenumber region, nonlinear couplings give two important effects; one is the renormalization of damping rates and the other the self consistent formation of the nonlinear source.

Let us introduce an equal-time average difference of phases $\theta(r_1)$ and $\theta(r_2)$ at points r_1 and r_2. Defining this by $\Delta\theta(r) \equiv \langle [\theta(r_1) - \theta(r_2)]^2 \rangle^{1/2}$ with $r \equiv |r_1 - r_2|$ ($\gg 1$), one finds

$$\Delta\theta(r) \propto r^{1/2} . \tag{3·18}$$

Let us summarize above results. Returning to the original units of the system, from (2·11) and (2·12) one finds that the characteristic frequency $\Omega_k (= \Gamma_k)$, the variance χ_k and the phase difference $\Delta\theta(r)$ behave as $\xi^{-5/2} k^{3/2}$, $\xi^{-5} k^{-2}$ and $\xi^{-5/2} r^{1/2}$, respectively, for $k \ll \xi^{-1}$, ($r \gg \xi$), and that universal functions f and g take the forms $c_1 x^{3/2}$ and $c_2 x^{-2}$, respectively, where c_1 and c_2 are certain numerical factors.

(b) *Large wavenumber (dissipative) region* ($k \gg 1$)

In the dissipative region with $k \gg 1$, the irregularity is expected to be small, compared with the one in the small wavenumber region. Thus the renormalization of damping rates can be dropped out, and one can put $\Gamma_k \cong \Gamma_k^0 \cong k^4$. However, the nonlinear source term must be retained. Equation (3·16) becomes

$$k^4 I_k = \frac{k^2}{2} \int_{-\infty}^{\infty} \frac{dk'}{2\pi} \frac{4 I_{k'} I_{k-k'}}{|k'|^4 + |k-k'|^4 + |k|^4} . \tag{3·19}$$

By assuming the following form of the solution in (3·19)

$$I_k = B k^\alpha \exp(-A k^\beta), \tag{3·20}$$

the main contribution to the right-hand side of (3·19) comes from the region $k' \sim k/2$. Thus (3·19) can be approximated as

$$k^6 I_k \sim C \int_{a|k|}^{b|k|} dk' I_{k'} I_{k-k'} , \tag{3·21}$$

where a, b and C are certain numerical constants such that $0 \leqq a < b \leqq 1$ and $C > 0$.

Inserting (3·20) into (3·21), we obtain

$$k^{\beta+\alpha}\exp(-Ak^{\beta})\sim BCk^{2\alpha+1}\int_a^b dx\, x^{\alpha}(1-x)^{\alpha} \times\exp[-Ak^{\beta}\{x^{\beta}+(1-x)^{\beta}\}]. \tag{3·22}$$

This equation can be solved by putting

$$\alpha=5\,,\quad \beta=1\,. \tag{3·23}$$

Thus we find the intensity in the dissipative region has the form

$$I_k\cong Bk^5\exp(-Ak),\quad \chi_k\cong Bk^3\exp(-Ak), \tag{3·24}$$

where $B=[C\int_a^b dx\, x^5(1-x)^5]^{-1}$, and A is determined by the statistics over a whole region.

By making use of the result (3·24), an average equal-time phase difference of two points separated by a distance $r(\ll 1)$ is obtained as

$$\Delta\theta(r)\propto r\,, \tag{3·25}$$

which agrees with the result in the dissipative region in a hydrodynamic turbulence.[19]

A summary of the above is as follows: Returning to the original units, from (2·11) and (2·12) one finds that Ω_k, χ_k and $\Delta\theta(r)$ behave as k^4, $k^3\exp(-Ak\bar{\xi})$ and $\bar{\xi}^{-3}r$, respectively, for $k\gg\bar{\xi}^{-1}$, $(r\ll\bar{\xi})$, and that universal functions become $f(x)=x^4$ and $g(x)=c_3x^3\exp(-c_4x)$, where c_3 and c_4 are certain numerical constants.

(B) *Vertex corrections*

We here evaluate the lowest order vertex corrections. The dominant corrections to the renormalized damping rate and the kinetic equation (3·9) come from those diagrams whose intermediate states consist of the small wavenumber modes only. Since the distribution function for these modes is of the Gaussian, one can neglect the vertex corrections to the kinetic equation (3·9), and the vertex corrections become important for the renormalized damping rates. The simplest vertex correction $\Delta\Sigma_1$ of Σ_1 is given by Fig. 3, which gives

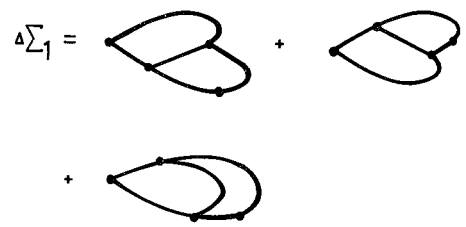

Fig. 3. Diagrams representing the simplest vertex correction of the self-energy part of G.

$$\Delta\Sigma_1(k,0)=16\int_{k'}\int_{k''}\frac{I_{k'}}{\Gamma_{k'}+\Gamma_{k-k'}}\,\frac{I_{k'-k''}}{\Gamma_{k''}+\Gamma_{k-k''}} \times\frac{I_{k-k''}}{\Gamma_{k'}+\Gamma_{k'-k''}+\Gamma_{k-k''}}\,\frac{k^2k''(k-k')}{I_k}\,, \tag{3·26}$$

where we have used the relation for the cascade region,

$$\frac{k_1}{I_{k_1}}+\frac{k_2}{I_{k_2}}+\frac{k_3}{I_{k_3}}=0$$

with $k_1+k_2+k_3=0$. This type of relation is characteristic of the Gaussian distribution.[20] Inserting the asymptotic solutions $\Gamma_k=\Gamma_1|k|^{3/2}$ and $I_k=I_1$ into (3·26), one finds $\Delta\Sigma_1(k,0)=(I_1^2/\Gamma_1^3)\times16\times0.0233k^{3/2}$, and thus $[\Delta\Sigma_1(k,0)/\Sigma_1(k,0)]\cong0.24$. This correction decreases damping rates. It is worth while to note that to all orders the vertex corrections have the same power spectra $k^{3/2}$ and hence these contribute to Γ_k only through the modulation of the constant factor Γ_1.

(C) *Numerical results*

Numerical solutions of Eqs. (3·15) and (3·16) in a whole region in k-space are shown in Figs. 4∼5. The variance χ_k and renormalized damping rate Γ_k are shown in Figs. 4(a) and (b), respectively. The solid line in Fig. 4 represents solutions of (3·15) and (3·16). Our results should be compared with those of the computer simulation in I. The qualitative features in the small and large wavenumber regions are good. However, the I_1 is larger by about decuple than the one in the simulation. Furthermore the most distinct difference is the non-existence of a hump in the middle region of χ_k, which appears in the simulation. Noting that vertex correction have the same power spectra $k^{3/2}$, and are dominant only in the small wavenumber region, we can parametrically take into account these effects by replacing Σ_1 by $c\Sigma_1$ in (3·15), where c is a certain positive constant. By the use of the fact that the lowest order vertex correction decreases the damping rate, it may be natural to select $0<c<1$. For $c=0.5$, the numerical

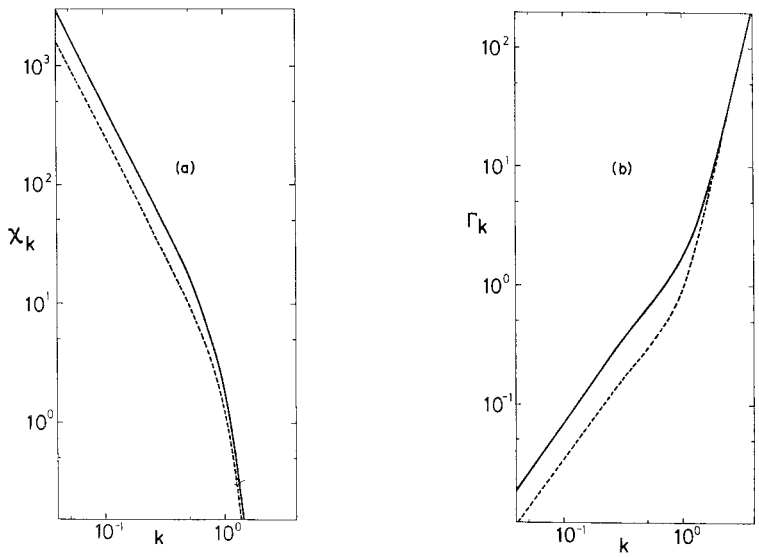

Fig. 4. Solid lines correspond to numerical results for $c=1$, and dotted lines for $c=0.5$. In the small wavenumber region, $\chi_k(\equiv\langle|\theta_k|^2\rangle)$ and Γ_k behave as k^{-2} and $k^{3/2}$, respectively.

results are shown by the dotted lines in Figs. 4(a), (b). The curve in the small wavenumber region has a tendency to agree with the simulation, but a quantitative agreement is not still good. Moreover, even under the above improvement, there appears no hump. As will be discussed in § 4, the hump seems to be caused by the memory effect[21] which becomes crucial in one-dimensional case.

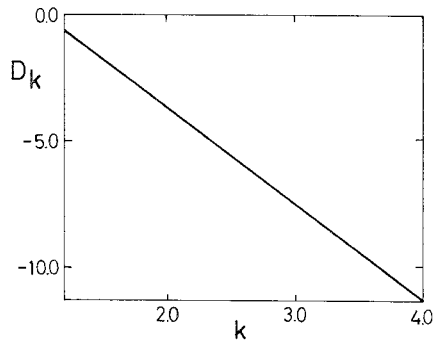

Fig. 5. Numerical result of $D_k \equiv \log_{10}(k^{-3}\chi_k)$ vs k in the dissipative region.

Figure 5 shows the numerical result of $k^{-3}\chi_k$ vs k in the large wavenumber region. The qualitative agreement with the theoretical prediction (3·24) is clear. The unknown constant A is about 8.7. We have analyzed the simulation in the large wavenumber region, and have found that for $k \gtrsim 1$ the form (3·24) is more favorable than a simple power-law spectrum.

§ 4. Summary and discussion

It has been shown that the chemical turbulence which was numerically found by Kuramoto and one of the present authors[9] can be theoretically investigated with the methods similar to the hydrodynamic case. We have derived self-consistent equations for the variance $\chi_k (\equiv \langle |\theta_k|^2 \rangle)$ and the renormalized damping rate Γ_k, which have the same form as Kraichnan's direct interaction approximation,[18] if a Markoffian approximation is adopted in the latter.

Qualitative agreements of the present results with the computer simulation have been also shown in the two limits; the cascade region with $k \ll \xi^{-1}$ and the dissipative region with $k > \xi^{-1}$. In the whole k region χ_k and Γ_k have the scaling formulae $\Gamma_k = \xi^{-4} f(k\xi)$ and $\chi_k = \xi^{-3} g(k\xi)$, $f(x)$ and $g(x)$ being universal functions of x. The functions f and g behave as $x^{3/2}[x^4]$ and $x^{-2}[x^3 \exp(-Ax)]$, respectively, in the cascade [dissipative] region. It should be emphasized that these two limiting regions obey the different scaling (2·16) and (2·18). In the lowest order approximation quantitative agreements in the cascade region is not so good. However, by including the vertex corrections of Σ_1 this disagreement tends to be improved.

Qualitative features in the middle wavenumber region ($k \approx \xi^{-1}$) cannot be comprehensible even by including the vertex corrections. This difficulty may be attributed to the Markoffian assumption. Expanding (3·13) as

$$\Sigma_1(k, \omega) = \Sigma_1(k, 0) + i\omega \Sigma_1'(k, 0) + \cdots , \qquad (4\cdot1)$$

we can evaluate the memory effect, obtaining $[i\omega \Sigma_1'(k, 0)/\Sigma_1(k, 0)] \cong 0.59$. This correction is rather large, and the non-Markoffian corrections seem to have impor-

tant effects on the decay processes. In fact, in one-dimensional case a long-time tail appears and plays a crucial role in various phenomena.[21]

Therefore, in order to study the present model in a more precise way, the original equations (3·7) and (3·8) with (3·13) and (3·14) are hoped to be solved.

The extension of the present formalism to higher dimension can be made straightforward. This problem is also hoped to be studied elsewhere.

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