Spiral waves in oscillatory media with an applied electric field

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Spiral waves in oscillatory reaction-diffusion systems under the influence of a uniform, time-independent electric field are modeled by the complex Ginzburg-Landau equation extended to include a convective term with complex coefficient. Results for the spiral drift, deformation, and frequency shift due to the electric field are obtained. The coefficient of the additional convective term is derived from the original reaction-diffusion system. The equation provides a good qualitative model of experimentally seen distortion of spiral waves in the presence of an applied electric field. [S1063-651X(99)03502-3]

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Spiral waves are manifest in a broad range of experiments involving pattern formation in nonequilibrium media. Examples include the Belousov-Zhabotinsky (BZ) reaction [1], electrical excitation in cardiac tissue [2], catalytic oxidation of carbon monoxide on a platinum surface [3], spatial aggregation of social amoebae [4], and rotating waves of electrical activity on turtle cortex [5]. For BZ spiral waves, experiments have been conducted in which the reaction takes place in the presence of a uniform electric field applied in the plane of the reaction dish [6,7]. The spiral center is observed to drift primarily in the direction of the electric field, but there is a smaller component of the drift that is perpendicular to the field and whose sign depends upon the chirality of the spiral. In addition, the shape of the spiral is significantly deformed in comparison to the typical Archimedean spirals seen in the absence of the electric field (Fig. 1). These experiments were performed in the excitable regime of the BZ reaction. In the oscillatory regime, sufficiently close to the onset of oscillations, the dynamics of reaction-diffusion equations, such as those believed to govern the BZ reaction, can be modeled by the well-known complex Ginzburg-Landau equation (CGLE) [8]. In addition, it is also found that the CGLE provides a good model for qualitative behavior of real systems even though they are not near a Hopf bifurcation (including excitable regime cases) [9]. In this paper, we consider the CGLE with an additional term that we show arises from applying a weak, uniform, time-independent electric field to a reaction-diffusion system:

$$\partial A/\partial t = A - (1+i\alpha)|A|^2 A + (1+i\beta)\nabla^2 A + (\gamma + i\delta)\partial A/\partial x. \tag{1}$$

$A(\mathbf{r},t)$ is a complex scalar field that slowly modulates the fast oscillations of the physical variables, and α, β, γ, and δ are real parameters. The last term in Eq. (1) is due to the electric field which has been taken to point along the x direction. Figure 2 is a numerical solution of Eq. (1) for a pair of spirals for the case of nonzero δ. The spirals drift along the electric field direction, and they are deformed in a similar fashion to what is seen experimentally for excitable media (e.g., the experiment of Ref. [6]).

With $\gamma=\delta=0$ (i.e., zero electric field), Eq. (1) becomes the standard CGLE, which, using cylindrical coordinates (r,θ), has a two-dimensional spiral wave solution of the form [10]

$$A = A_0(r,\theta,t) = F(r)\exp\{i[-\omega_0 t + \sigma\theta + \psi(r)]\}, \tag{2}$$

where $\sigma=\pm 1$ is the ''topological charge'' of the spiral wave, and $F(r)$ and $\psi(r)$ are real functions whose large r behavior is $d\psi/dr \to k_0$ and $F \to \sqrt{1-k_0^2}$ as $r\to\infty$, where k_0 is a constant which depends on α and β. The frequency ω_0 is connected with k_0, the asymptotic plane wave number, via the dispersion relation, $\omega_0 = \alpha + (\beta-\alpha)k_0^2$.

Introducing the transformations

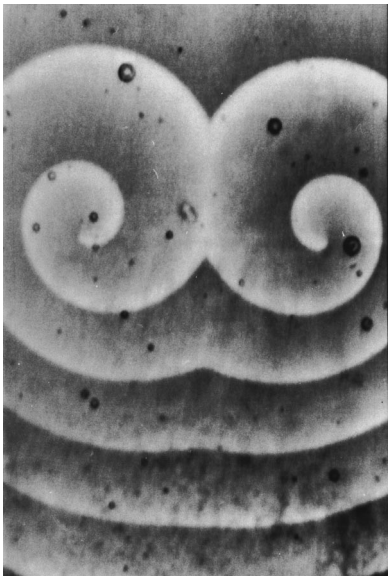

FIG. 1. Electric field induced deformation of spiral waves in the BZ reaction. From the experiment performed by Steinbock, Schütze, and Müller [6] (courtesy of S. C. Müller).

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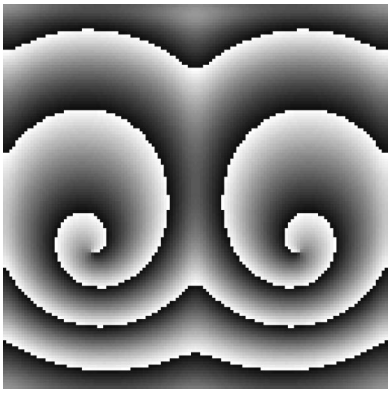

FIG. 2. Deformation of spiral waves. Plotted is the phase of A for a numerical solution of Eq. (1) with the electric field taken along the vertical direction and $\alpha=0.25$, $\beta=-1$, $\gamma=0$, $\delta=0.3$. The spiral cores have drifted down from their initial location halfway up the box.

$$\boldsymbol{r}'=\lambda^{1/2}(x-vt)\boldsymbol{x}_0+\lambda^{1/2}y\boldsymbol{y}_0\equiv x'\boldsymbol{x}_0+y'\boldsymbol{y}_0,$$

$$t'=\lambda t,\quad A'(x',y',t')=\lambda^{-1/2}A(x,y,t)\times\exp\{i[\nu t-\kappa(x-vt)]\},$$

Eq. (1) becomes

$$\partial A'/\partial t'=\lambda^{-1}[1+i\nu-\kappa^2(1+i\beta)+i\kappa(v+\gamma+i\delta)]A'+(1+i\alpha)|A'|^2A'+(1+i\beta)\nabla'^2A'+\lambda^{-1/2}[\gamma+i\delta+v+2i\kappa(1+i\beta)]\partial A'/\partial x',\quad(3)$$

where $\nabla'^2=\partial^2/\partial x'^2+\partial^2/\partial y'^2$. The complex coefficient of $\partial A'/\partial x'$ can be made zero by the following choice of the two real transformation parameters v and κ:

$$v=-(\gamma+\beta\delta),\quad(4a)$$

$$\kappa=-\delta/2.\quad(4b)$$

Inserting Eqs. (4a) and (4b) in Eq. (3), we can now make the complex coefficient of A' one by the following choice of the remaining two parameters ν and λ:

$$\nu=-\beta(\delta/2)^2,\quad(4c)$$

$$\lambda=1+(\delta/2)^2.\quad(4d)$$

Thus we have transformed Eq. (1) to the conventional CGLE [i.e., Eq. (1) with $\gamma=\delta=0$]. Hence Eq. (1) with $\gamma\neq0$, $\delta\neq0$ has the following spiral wave solution:

$$A(x,y,t)=\lambda^{1/2}F(\lambda^{1/2}\tilde{r})\exp\{i[-(\lambda\omega_0+\nu)t+i\kappa(x-vt)+\sigma\tilde{\theta}+\psi(\lambda^{1/2}\tilde{r})]\},\quad(5)$$

where F and ψ are the amplitude and phase functions for the conventional ($\gamma=\delta=0$) CGLE and $(\tilde{r},\tilde{\theta})$ are cylindrical coordinates in the frame comoving with the spiral, $\tilde{r}^2=(x-vt)^2+y^2$. Comparing Eqs. (2) and (5), we see that application of an electric field has the following effects. (i) The large $\tilde{r}$ amplitude is shifted by the factor $\lambda^{1/2}$. (ii) The scale of spatial variation is also shifted by the factor $\lambda^{1/2}$. (iii) The oscillation frequency is changed by an amount $\Delta\omega=(\omega_0-\beta)(\delta/2)^2$. (iv) The large $\tilde{r}$ asymptotic wave number now becomes $\tilde{\theta}$ dependent,

$$\boldsymbol{k}(\tilde{\theta})=(\lambda^{1/2}k_0\cos\tilde{\theta}+\kappa)\boldsymbol{x}_0+\lambda^{1/2}k_0\sin\tilde{\theta}\boldsymbol{y}_0.\quad(6)$$

The directional dependence of $|\boldsymbol{k}|$ is responsible for the deformation of the spiral shown in Fig. 2 and found in experiments, and $\kappa=-\delta/2$ quantifies the extent of the deformation. The deformation and the frequency shift depend upon δ and not γ, which only contributes to the uniform translation. As shown below, γ and δ are proportional to the electric field and so results (4a) and (4b) imply that the drift and deformation velocity are proportional to the electric field. Experiments done on the BZ reaction likewise find a drift velocity that is roughly proportional to the electric field [6,7]. The perpendicular component of drift seen in these experiments does not appear in our results, indicating that this aspect of the observations is not captured by the perturbation expansion yielding Eq. (1); e.g., the experiments were performed in the excitable regime of the BZ reaction whereas the perturbation expansion applies for oscillatory media. Perhaps for the same reason, the above solution corresponds to a uniformly translating deformed spiral without the periodic relaxations to an undeformed shape as reported in Ref. [7].

The transformation of Eq. (1) to the case with $\gamma=\delta=0$ with the values of α and β preserved implies that spiral wave solutions to Eq. (1) will have the same stability properties as CGLE spiral waves for a given α and β. The stability properties of CGLE spiral waves have been investigated with respect to the stability of plane waves [11], core acceleration [12], and phase twist along the vortex filament in three dimensions [13]. In addition, the transformation does not assume that the solution is a spiral wave. This implies that the borderlines in α-β parameter space which demarcate qualitatively different regimes of behavior, such as the so-called ''frozen'' state of relatively stationary defects and the defect-mediated turbulence state [14], will be the same for the CGLE with the added $\partial A/\partial x$ term as for the original CGLE.

We now explain the origin of the $\partial A/\partial x$ term in Eq. (1). In the presence of a uniform electric field in the x direction, the equation modeling a system of reacting and diffusing chemicals is of the form [15]

$$\partial\boldsymbol{c}/\partial t=\boldsymbol{f}(\boldsymbol{c},\mu)+\boldsymbol{D}\nabla^2\boldsymbol{c}+E\boldsymbol{M}\partial\boldsymbol{c}/\partial x.\quad(7)$$

The components c_i of the vector field $\boldsymbol{c}(\boldsymbol{r},t)$ represent the concentrations of the chemical species of concern in the experiment. The nonlinear function $\boldsymbol{f}$ describes the reaction kinetics of the chemicals, and μ is a dimensionless parameter. $\boldsymbol{D}$ is a diagonal matrix with D_{ii} the diffusion coefficient of the ith species. The diagonal matrix $\boldsymbol{M}$ contains the ionic mobilities which couple the chemical species to the electric field of magnitude E.

The last term in Eq. (7) is the origin of the convective term in Eq. (1). The parameter β in Eq. (1) will be nonzero if the diffusion coefficients in $\boldsymbol{D}$ are not all equal [8]. Similarly the parameter δ is related to the inequality of the ionic mobilities in $\boldsymbol{M}$. To demonstrate this, it suffices to derive

only the terms in Eq. (1) that are linear in A starting with the reaction-diffusion equation (7) (for a full derivation of the CGLE see Ref. [8]). It is assumed that as the parameter μ in the rate kinetics function $\boldsymbol{f}$ passes through zero from negative to positive, a supercritical Hopf bifurcation from an equilibrium to a limit cycle takes place. The equilibrium solution $\boldsymbol{c}_0(\mu)$ of the spatially homogeneous system is given by $\boldsymbol{f}(\boldsymbol{c}_0,\mu)=0$, and is stable for $\mu<0$. Given a small parameter, $\epsilon \ll 1$, and a small deviation from the equilibrium solution, $\boldsymbol{u}(\boldsymbol{r},t)=\boldsymbol{c}(\boldsymbol{r},t)-\boldsymbol{c}_0(\mu)$, with $\boldsymbol{u}\sim\epsilon$, linearization of Eq. (7) yields

$$\partial \boldsymbol{u}/\partial t = \boldsymbol{L}\boldsymbol{u} + \boldsymbol{D}\nabla^2\boldsymbol{u} + E\boldsymbol{M}\partial \boldsymbol{u}/\partial x, \tag{8}$$

where $\boldsymbol{L}$ is the Jacobian matrix, $L_{mn}=(\partial f_m/\partial c_n)_{c_0(\mu)}$. Considering only values of $\mu \ll 1$ limits our analysis to the immediate vicinity of the Hopf bifurcation at $\mu=0$, and yields the approximations $\boldsymbol{c}_0(\mu)\approx \boldsymbol{c}_0(0)+\mu(\partial \boldsymbol{c}_0/\partial\mu)_{\mu=0}$ and $\boldsymbol{L}\approx \boldsymbol{L}_0+\mu\boldsymbol{L}_1$, with $(L_0)_{mn}=(\partial f_m/\partial c_n)_{c_0(0)}$ and $(L_1)_{mn}=\Sigma_k(\partial^2 f_m/\partial c_k \partial c_n)_{c_0(0)}(\partial c_{0k}/\partial\mu)_{\mu=0}$. Accordingly we may approximate Eq. (8) as

$$\partial \boldsymbol{u}/\partial t = \boldsymbol{L}_0\boldsymbol{u} + \mu\boldsymbol{L}_1\boldsymbol{u} + \boldsymbol{D}\nabla^2\boldsymbol{u} + E\boldsymbol{M}\partial \boldsymbol{u}/\partial x. \tag{9}$$

In the neighborhood of the bifurcation, the dynamics are determined by the most unstable modes of the homogeneous system, $d\boldsymbol{c}/dt=\boldsymbol{f}(\boldsymbol{c},\mu)$. At the threshold value $\mu=0$, these critical modes are eigenvectors of the matrix $\boldsymbol{L}_0$, denoted by $\boldsymbol{v}_+$ and $\boldsymbol{v}_-$, and have eigenvalues $+i\Omega_0$ and $-i\Omega_0$, respectively, where Ω_0 is the limit cycle frequency. The following ansatz can now be made: $\boldsymbol{u}(\boldsymbol{r},t)=\epsilon A(\boldsymbol{r},t)e^{i\Omega_0 t}\boldsymbol{v}_+ + \epsilon\overline{A}(\boldsymbol{r},t)e^{-i\Omega_0 t}\boldsymbol{v}_-$, where $\overline{A}$ is the complex conjugate of A. Since $\boldsymbol{L}_0$ is real, $\boldsymbol{v}_-=\overline{\boldsymbol{v}}_+$ and denoting the dual vectors to $\boldsymbol{v}_+$ and $\boldsymbol{v}_-$ by $\boldsymbol{v}^+$ and $\boldsymbol{v}^-$, we have $\boldsymbol{v}^+\boldsymbol{v}_-=\boldsymbol{v}^-\boldsymbol{v}_+=0$. Furthermore, we impose the normalization, $\boldsymbol{v}^+\boldsymbol{v}_+=\boldsymbol{v}^-\boldsymbol{v}_-=1$. Inserting this ansatz into Eq. (9) and left multiplying by $e^{-i\Omega_0 t}\boldsymbol{v}_+$ gives

$$\partial A/\partial t = \mu b_1 A + b_2\nabla^2 A + E b_3 \partial A/\partial x, \tag{10}$$

where we have neglected rapidly oscillating terms varying as $\exp(-2i\Omega_0 t)$ by taking $\partial A/\partial t \sim \mu$. In addition, the spatial variation of A is taken to be slow and the electric field is assumed to be weak, so that $\nabla A\sim\mu^{1/2}$ and $E\sim\mu^{1/2}$. The constants $b_{1,2,3}$ are given by $b_1=\boldsymbol{v}^+\boldsymbol{L}_1\boldsymbol{v}_+$, $b_2=\boldsymbol{v}^+\boldsymbol{D}\boldsymbol{v}_+$, and $b_3=\boldsymbol{v}^+\boldsymbol{M}\boldsymbol{v}_+$. This is Eq. (1) (up to rescaling and the frequency shift noted below) except for the cubic nonlinear term which results from setting $\epsilon=\mu^{1/2}$, necessitating the inclusion of second- and third-order terms in the expansion of $\boldsymbol{f}(\boldsymbol{c}_0+\boldsymbol{u})$. The parameters γ and δ in Eq. (1) are seen to be proportional to the electric field E. If all the chemical species have the same ionic mobility M, then $\boldsymbol{M}$ is proportional to the identity matrix and b_3 will be real, i.e., $b_3=M$. On the other hand, if the ionic mobilities are not all equal, then b_3 will have a nonzero imaginary part. Thus the parameter δ which is proportional to Im b_3 will be nonzero as claimed above. Similarly, β will be nonzero if the diffusion coefficients differ. The coefficient of the linear growth term in Eq. (1) is real because any imaginary part of b_1 can be absorbed as a frequency shift to A, $A\rightarrow Ae^{i(\mathrm{Im}\, b_1)t}$. As shown in Ref. [16], the components of the eigenvector $\boldsymbol{v}_+$ and its dual $\boldsymbol{v}_-$ can be determined experimentally for chemical reaction-diffusion systems, and therefore it is possible for the parameters γ and δ to be calculated for an actual experimental system [as can the other parameters appearing in Eq. (1)].

In summary, we have shown that the presence of an electric field modifies the CGLE by inclusion of a convective term with complex coefficient. This leads to spiral wave distortion (see Fig. 2) and drift similar to that seen in experiments [17].

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[16] F. Hynne and P. G. Sørensen, Phys. Rev. E **48**, 4106 (1993).
[17] The transformation used for the electric field problem can also be used to find the velocity of a slightly curved vortex filament in the three-dimensional CGLE. This problem has been solved

previously by Gabbay *et al.* using perturbation theory where the filament curvature is assumed to effect a small correction to the two-dimensional spiral wave solution [M. Gabbay, E. Ott, and P. N. Guzdar, Phys. Rev. Lett. **78**, 2012 (1997); Physica D **118**, 371 (1998)]. At a given point on a filament of small curvature, $K \ll 1$ (and no phase twist), the CGLE can be written in the Frenet frame moving with the filament as $\partial A/\partial t = A-(1+i\alpha)|A|^2A+(1+i\beta)\nabla^2A+[v-(1+i\beta)K]\partial A/\partial x$, where ∇^2 is the two-dimensional Laplacian in the plane perpendicular to the filament, the x direction points toward the center of curvature, and v is the filament velocity. This is formally the same as the situation in Eq. (1). Using Eqs. (4a) and (4b) yields $v=(1+\beta^2)K$ for the filament velocity and $\kappa = \beta K/2$ for the first-order wave-number shift, in agreement with Gabbay *et al.*