【論 文】

Long Range Interaction of Vortices in a Chemical Active Medium

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Experimental study and computer simulations of interaction of vortices in the Belousov-Zhabotinsky reaction show that a small difference in the frequency of vortex rotation causes a marked drift of a vortex with lower frequency. The trajectory of the drift is a logarithmic spiral. Analytical estimations are in a good agreement with the experimental results.

1. Introduction

The dynamics of vortices in nonlinear excitable and oscillatory chemical systems has been attracting attention of the researchers during several decades. One of the most intriguing problem concerns an interaction of vortices. Theoretical study of the dynamics in generic models of active media: analytical study of Ginzburg-Landau equations [1] and numerical simulations of FitzHugh-Nagumo type models [2,3] show the possibility of both attraction and repulsion of the vortices. Ordinarily the interaction becomes important if the distance between the centers of rotation of vortices is small (less than a wavelength).

A possibility of interaction of vortices spaced by a large distance has been unclear until now. The fail in the experimental observation of such an interaction is conventionally explained by the fact that the waves emitted by the vortices annihilate after collision, that is why vortices cannot "feel" each other at distance larger than a wavelength.

However, the above said is valid for wave sources with identical frequencies. Experimental study [4] of stimulation of a vortex by planar waves showed that in the case the frequency of planar waves is higher than that of a vortex, a drift of the vortex occurred. It was also shown a possibility of similar drift induced by a high frequency wavetrain emitted by another vortex. Analytical estimations [4] explain the possibility of such a drift along a straight line in a direction parallel to the wavevector of high frequency planar waves.

In the present paper we study the interaction of vortices having close, but not the same frequencies. To maintain such vortices we used a light sensitive Belousov-Zhabotinsky reaction. We observed the vortex with a lower frequency drifted away of the vortex with high frequency. The trajectory of such a drift was not a straight line, but a part of a logarithmic spiral. Computer simulations and analytical study reveal the mechanism of such a dynamics.

2. Experimental method and numerical procedure

We carried out the experiments using the light sensitive BZ reaction of the following composition: $NaBrO_3$- 0.25 M, $CH_2(COOH)_2$ - 0.2 M, H_2SO_4 - 0.2 M, $Ru(bpy)_3Cl_2$- 2 mM. The solution was carefully stirred and poured into a 5.9 cmdiameter Petri dish, which was then covered with a glass lid.

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After a pair of vortices was initiated, one of them was subjected to irradiation by visible light. The irradiation resulted in a decrease of the velocity of propagating waves and in a decrease of frequency of vortex rotation by approximately 10 %. We used light from a 50 mW argon ion laser, wavelength 488 and 514.5 nm, to irradiate the medium.

The dynamics was followed with a video-recorder connected to an SGI Indy computer.

For computer simulations we used a two-variables model of the BZ reaction developed by Rovinsky and Zhabotinsky[5] with rate constants estimated in ref.[6]. According to the model, the dynamics in a perfectly stirred vessel is described by the differential equations for bromous acid (x) and ferriin (z)[5,6]:

$$\frac{dx}{d\tau} = \frac{1}{\varepsilon}\left[x(1-x) - \left(2q\alpha\frac{z}{1-z} + \beta+\nu\right)\frac{x-\mu}{x+\mu}\right]$$

$$\frac{dz}{d\tau} = x - \alpha\frac{z}{1-z}. \qquad (1)$$

To study spatio-temporal effects, we added diffusion terms to

$$\frac{dx}{d\tau} = F(x,z) + \nabla_\rho^2 x$$

$$\frac{dz}{d\tau} = G(x,z) + \delta\nabla_\rho^2 z \qquad (2)$$

where $F(x,z)$ and $G(x,z)$ are the right-hand sides of eqs.(1); ρ denotes scaled spatial coordinates; ∇_ρ^2 is the Laplacian operator with respect to the coordinates ρ; $\delta = D_z/D_x$ is the ratio of the diffusion coefficients and assumed to be unity here.

The model has been verified experimentally to adequately simulate spatio-temporal phenomena in the BZ reaction.[6-9]

An additional bromide ion production during the irradiation of the reaction by visible light[10-13] was accounted for by the term ν in eq.1. The value of this term was $\nu = 0.002$. The meaning and the values of the other model parameters is described in detail in ref.[6].

The equations 1 and 2 were integrated in a 2D array of 400 by 400 elements using the Euller's method of integration with Neumann's boundary conditions. The spatial and temporal steps where chosen small enough so that further decrease in their values would improve the accuracy of measured values, particularly wavelength and velocity, by few percent only.

3. Results

Initially, the two vortices form a symmetrical wavepattern (**Fig.1**(a)). After switching on the light, the frequency of rotation of the irradiated vortex S_1 (upper one on each frame of **Fig.1**) was reduced by 10 % with respect to the vortex S_2 which was not subjected to irradiation. This resulted in the symmetry break (**Fig.1**(b)). However, there was no observed a drift of the vortex until the waves from S_2 reach the core of S_1 (**Fig.1**(c)). Since that moment the vortex S_1 was drifting away of S_2 (**Fig.1**(c)-(f)).

In **Fig.2** it is plotted the trajectory of the drift. Surprisingly, the drift was not directed along the **Y**-axis only; it had a large **X**-component.

To comprehend the long term dynamics of the

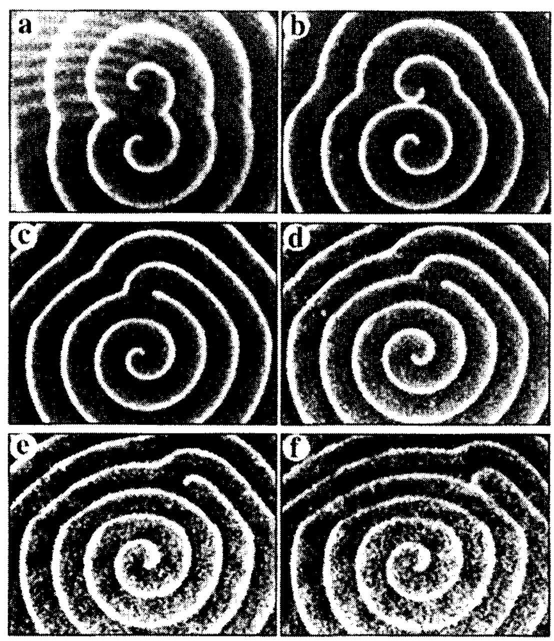

Fig.1 Interaction of vortices in the BZ reaction. The vortex with lower frequency (upper one in each frame) drifts away from the vortex with higher frequency. The difference in frequencies is maintained because upper half of the medium on each frame was subjected to light irradiation. The period of rotation was T_1 = 20.0 s for the lower vortex and T_2 = 22.2 s for the upper vortex. Interval between snapshots is 4 min.; size of each frame is 11.4 by 8.8 mm.

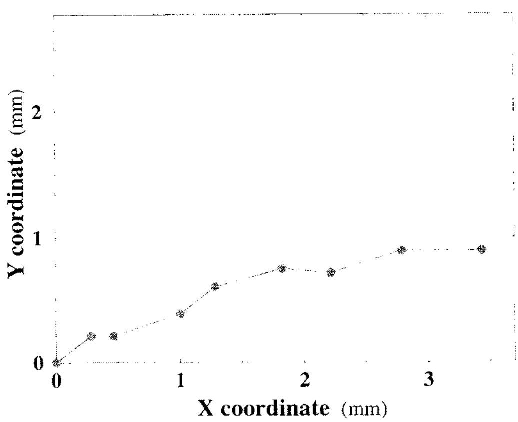

Fig.2 Experimentally measured trajectory of the drift of a vortex with lower frequency. **Y**-axis is along the line that connected tips of vortices soon after initiation (see **Fig.1**(a)), **X**-axis is perpendicular to the **Y**-axis. Origin is on the initial position of a drifting spiral.

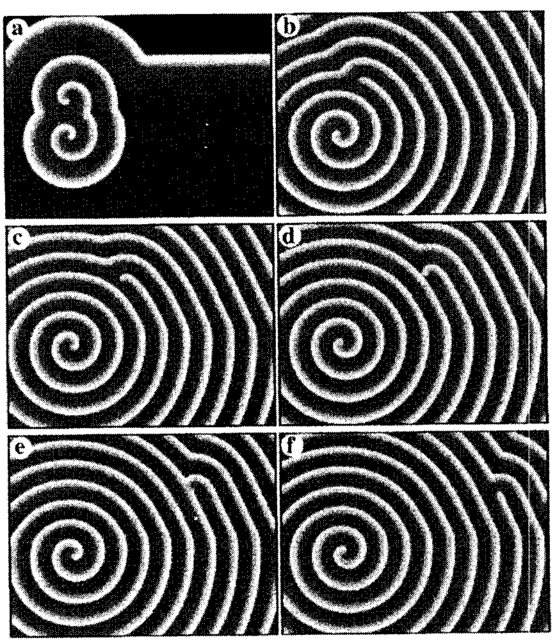

Fig.3 Computer simulation of interaction of vortices in the BZ reaction. The vortex with lower frequency (upper one in each frame) drifts away from the vortex with higher frequency. Interval between snapshots is 7.2 min.; size of each frame is 24 by 18 mm.

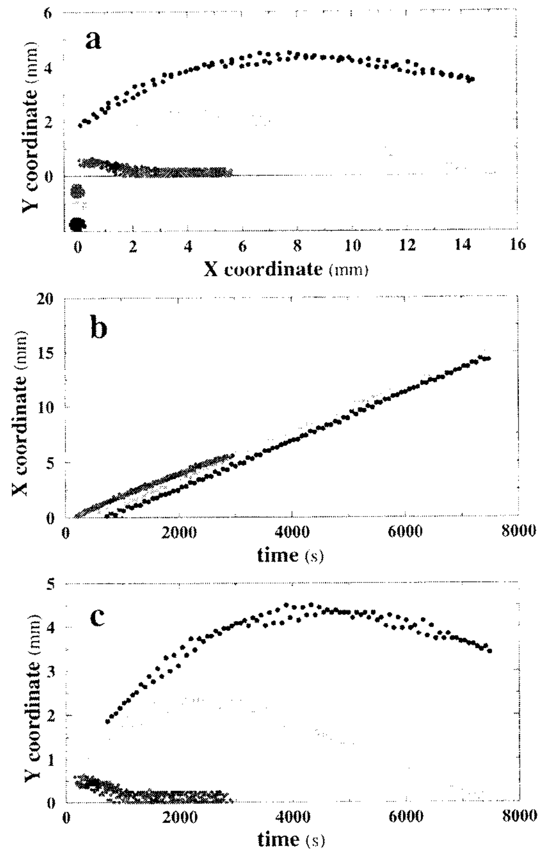

Fig.4 Trajectory of the vortex drift in computer simulations (a). Different trajectories correspond to three different computer simulations. The circles mark the position of the tip of the vortex. In each simulation the vortices with lower frequency (upper ones) drift, while the vortices with higher frequency (lower ones) rotate stationary. Initially the vortices were situated symmetrically with respect to the boundary of inhomogeneity (the line **Y**= 0).
Change of **X** and **Y** coordinates with time (b, c). Note that the **X** coordinate changes about linearly while the change of the **Y** coordinate is nonlinear and nonmonotonic.

vortex drift we carried out computer simulations. Simulating the experimental conditions, we observed similar wavepatterns (**Fig.3**). As in the experiments, the vortex S_2 having high frequency rotated stationary (see **Fig.4**(a), below the line **Y**=0). However, the long term simulations show that vortex S_1 drifts along a more complicated trajectory (**Fig.4**(a), over the line **Y**=0) than it was observed experimentally (**Fig.2**).

The trajectory typically had several distinct parts and **X** and **Y** coordinates changed in a different manner (**Fig.4**(b), (c)). Along the **X**-axis it was observed a drift with a nearly constant velocity $v = 1.7 \times 10^{-3}$ mm s^{-1}. Along the **Y**-axis after some increase of this coordinate, there followed its decrease until the trajectory reached the boundary of the inhomogeneity (**Y**=0). From this point the drift was strictly along the boundary.

The location of turn-points of the trajectory was dependent on the initial distance between vortices: the larger initial distance was, the far in space (**Fig.4**(a)) and later in time (**Fig.4**(c)) the turn-points were situated.

The observed dynamics cannot be explained in terms of the early developed theory.[4] Recently we developed an analytical approach which provides a finer description of the interaction of vortices. This approach is based on the kinematic consideration of the vortex dynamics and is described in detail in ref.[14]. Here we reproduce the main statements of this theory.

One of the main conclusions is that in the case of a stimulation by planar waves of high frequency the vortex drifts with the both **X** and **Y** components nonvanished. The angle of the drift, which is determined by the ratio of drift velocities along the two axes:

$$\frac{\dot{\mathbf{Y}}}{\dot{\mathbf{X}}} = \tan\theta \qquad (3)$$

is determined by the formula:

$$\theta = \left[\frac{3}{2}\pi\frac{T_1 - T_2}{T_1}\right]^{1/3} \qquad (4)$$

where T_1 is the period of vortex rotation, T_2 is the period of stimulating waves.

The velocities of the drift in this case are:

$$\dot{\mathbf{X}} \propto \frac{T - T_1}{T} \qquad (5)$$

$$\dot{\mathbf{Y}} \propto \left[\frac{T - T_1}{T}\right]^{2/3}. \qquad (6)$$

It is important to note that according the formulas (4)-(6) there is a drift along the both **X** and **Y** axes.

If to consider a stimulation of a vortex by circular waves, it is easy to understand that in this case the direction **Y** is along the radius from the source of circular waves. Along with the drift of the vortex, this direction varies in time so that the trajectory of the drift is a logarithmic spiral:

$$r = r_0 \exp(a\phi) \qquad (7)$$

where r and ϕ are the radius and the azimuthal angle in a polar coordinate system with the pole coinciding with the source of circular waves. The coefficient a is expressed as:

$$a = 1/\tan\left[\left[\frac{3}{2}\pi\frac{T_1 - T_2}{T_1}\right]^{1/3}\right]. \qquad (8)$$

To apply the above formula for the case under study, one should recall that the vortex having the form of Archimedian spiral is a source of waves which at large distance from the core become indistinguishable from circular waves. Thus, the formula (7) describes the trajectory of the drift of a vortex at large distances between vortices.

4. Discussion

In this paper we present the results of an experimental study of interaction of vortices at large distances in the case of unequal frequencies of rotation. The performed computer simulations fit experimental results well and allow us to describe the dynamics of the drift in more detail. Particularly, due to a limited life time of the reaction in closed reactors, only a part of the trajectory (**Fig.2**) was traced experimentally. To observe the full scale trajectory, like the presented in **Fig.4** it is required to use a more complicated experimental setup. Particularly continuously-fed open reactors will do for this purpose.

The analytical approach used to describe the phenomena yields simple and clear results for the two case: when the stimulating waves are planar or circular ones. However, in practice we deal with stimulating waves irradiated by a vortex having the form of Archimedian spiral, besides the waves undergo the refraction near the boundary of inhomogeneity. Despite this, the theory explain the main features of the behavior observed, particularly, it explains the occurrence of the **X** component of the drift. See ref.[14] for more discussion of this theory.

We studied interaction of the two vortices in an inhomogeneous medium. However, the major statements are based only on the fact of the non-identity of frequencies of the two vortices. Such vortices can be found in a homogeneous medium as well. An illustration to this is recent observations presented in ref.[3] where the authors have introduced an additional slow variable to the FHN model. This resulted in a shift of frequencies of interacting vortices followed by a drift of a vortex, which we believe has the same nature as described in the current article.

It should be noted that the described dynamics is characteristic of an open system far from the thermodynamic equilibrium. We believe that the dynamics is valid for a wide class of non-linear systems and can be useful to testify the properties of new materials in the non-equilibrium conditions.

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