Parametric resonance of rotating spiral waves under broken rotational symmetry

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Numerical simulations and analytical arguments for the resonant behavior of periodically perturbed rotating spiral waves in systems with broken rotational symmetry are presented. Resonance, i.e., the displacement of the rotation center along a straight line, appears always if the perturbed rotation does not match the symmetry given by the active medium supporting the wave. For the Z_2 case, simulations in a simple two-component reaction-diffusion model have been carried out. For the case of arbitrary rotational symmetries Z_d , a general resonance condition is derived.

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Pattern formation in nonequilibrium systems has been observed in many physical, chemical, and biological systems [1]. Among the most prominent of such patterns are rotating waves (in two dimensions: spirals), which have been reported in social amoebae [2], chicken retina [3], oocytes cytoplasm [4], and cardiac tissue [5] as well as in the Belousov-Zhabotinsky (BZ) solution [6] and in catalytic surface reactions [7]. As a common explanational framework the concept of active media is used [8], in the above cases specified by appropriate reaction-diffusion models of similar basic dynamical properties [9,10].

Up to now, such models have mostly been chosen to be isotropic or simply anisotropic, i.e., the quotient of the diffusion constants in orthogonal directions is identical for all species. While this simple anisotropy can easily be removed by rescaling the spatial coordinates (and hence is trivial), new phenomena may occur if such rescaling is impossible [11]: This complex anisotropy occurs naturally in media where the anisotropy depends on the state of the system, giving rise to a dynamical anisotropy that varies along the profile of a chemical wave. Such an effect can be found in surface reactions, where diffusion coefficients depend on adsorbate coverages and on surface structure [12], and in cardiac tissue [13]. Thus, even if only one species is diffusing, state-dependent anisotropy of diffusion may lead to qualitatively new phenomena.

As an interesting consequence for rotating spiral waves, the complex anisotropy of the medium is breaking the full rotational symmetry present in isotropic systems (ordespite the scaling of coordinates—in simply anisotropic systems). While the resulting effects on autonomous waves have been described elsewhere [14,15], we focus in this paper on the importance of rotational symmetry for the resonance behavior of perturbed spirals.

In experiment, external periodic perturbations have been applied in the BZ reaction by electric field [16], light [17-19], or stretching the gel [20], and in surface reactions by varying the temperature [21]. Both stretching the gel and applying an electric field was done along one Cartesian direction only and the electric field broke not only the rotational but also the reflectional symmetry of mere diffusion. In the other experiments, a control parameter (light or temperature—these parameters both control the rotation of a spiral in a similar way) was isotropically varied. Under these conditions, several resonances have been found which could be successfully explained with the assumption of an isotropic medium [21-23]. While this is naturally the case with homogeneous systems like the BZ reaction, the assumption of isotropy does not necessarily hold in heterogeneous catalysis, i.e., on surfaces. We will briefly summarize the theoretical results for isotropic media and then turn to anisotropicmedia. For the sake of simplicity, only isotropic perturbations are treated. As another restriction, only rigidly rotating waves are discussed and not the more complex types of rotation, known as meandering or hypermeandering [10,24], where more than one intrinsic frequency governs the spiral dynamics and the perturbation can phase-lock to the additional frequencies as shown in the light-sensitive BZ reaction [18,19].

For rotating waves, resonance corresponds to a net drift of the center of rotation along a straight line. In isotropic media, it has been shown by analytical argument and numerical simulation that in order to obtain this resonance the rotation frequency ω_0 has to be an integer multiple of the perturbation frequency ω [23]. Nearby, the spiral undergoes a net displacement along a circle with a radius inversely proportional to the difference to the full resonant frequency. Irrational ratios ω_0/ω result in nonclosing tip paths. For noninteger rational values of ω_0/ω , the contributions of the perturbation at different angles cancel themselves, resulting in closed trajectories of the spiral tip (i.e., the inner end of the spiral arm; see below). In the vicinity of the 1:1 resonance, the resulting spiral motion resembles an unperturbed meandering [8].

If the full rotational symmetry of an isotropic system is broken, additional resonances at rational ω_0/ω are possible, whenever a certain condition involving this ratio and the remaining symmetry is fulfilled. We will demonstrate this first in numerical simulations of a particular active medium for a special case of rotational symmetry, and will then analytically derive the resonance condition for systems with arbitrary rotational symmetry.

Numerical studies of anisotropic resonance behavior were performed with a reaction-diffusion model that was developed for a catalytic surface reaction, namely the CO oxidation on Pt(110) single-crystal surfaces [23]:

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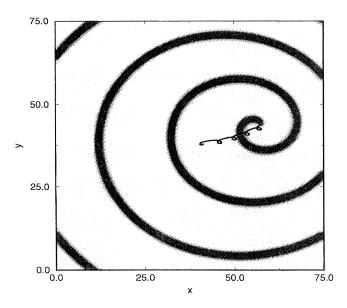


FIG. 1. Snapshot of a drifting spiral in model (1) for equal perturbation and rotation frequencies. The unperturbed spiral was located in the center of the medium. We monitor the value of u after 4.6 periods of the perturbation (with gray denoting higher values) and the trajectory of the spiral tip under perturbation (black line). The parameters are a=0.84, b=0.19, $\varepsilon=0.025$; modulation amplitude 5% in b, period $2\pi/\omega=7.0$ time units; semi-implicit integration (see [25] with dt=0.0073, grid size 384×384 corresponding to 75×75 spatial units.

$$\partial_{t}u = -\varepsilon^{-1}u(u-1)[u-(v+b)/a] + \nabla[D(v)\nabla u],$$

$$\partial_{t}v = f(u) - v \text{ with } f(u) = \begin{cases} 0, & u < 1/3 \\ 1 - 6.75u(u-1)^{2}, & \text{else} \\ 1, & u > 1. \end{cases}$$
(1)

These equations are similar to a model proposed by Barkley [25], allowing oscillatory, excitable or bistable local kinetics depending on the number of stable fixed points [23]. In the

above equations, the fast activator variable u denotes the adsorbate coverage of the surface (changing rapidly due to adsorption and reaction processes), while the slow inhibitor variable v stands for the degree of surface reconstruction, influencing the reactivity of the surface and the diffusion of u. We chose the diffusion to be

$$D_x(v) = 1 + v/2, \quad D_v(v) = 1 - v/2.$$
 (2)

This nonstandard diffusion term leads to complex anisotropy of the medium, which is no longer identical under every rotation but only under turns of 180° and cannot be scaled out.

Perturbations were applied by varying the parameter b, which determines the excitability threshold in Eqs. (1) and is related to the temperature of the catalyst, but the other parameters a and ε would be equally well. We restrict ourselves to the excitable region, but spirals in the oscillatory or bistable region are expected to show identical resonant behavior, since in all regions the parameters influence the rotation period monotonously [23]. Consequently the occurrence of resonances depends on the symmetry only. In Fig. 1, a spiral is shown after several rotations, together with the trajectory of the spiral tip, for a perturbation frequency equal to its rotation frequency. The spiral arm is distorted by the Doppler effect of the spiral's net displacement (drift) and by the anisotropic wave propagation in the medium. For the resonances, we further concentrate on the tip trajectories only. The tip is the inner end of the spiral arm, its location being defined as the intersection of isoconcentration lines of u and v [for Eqs. (1), u = 0.5 and v = a/2 - b were used]. The loopy trajectory of the tip consists of rotation and translation of the wave. Resonances appear as a net drift of the rotating wave along a straight line, i.e., the tip trajectory exhibits a loopy line with a straight net translation (cf. Fig. 1).

The application of different perturbation frequencies is shown in Fig. 2. Resonances occur for every odd integer m, when $\omega_0/\omega = n/m$, and arbitrary integer n, while even m result in closed paths of the spiral tip. (Without loss of generality it can be assumed that n,m are coprime natural numbers.)

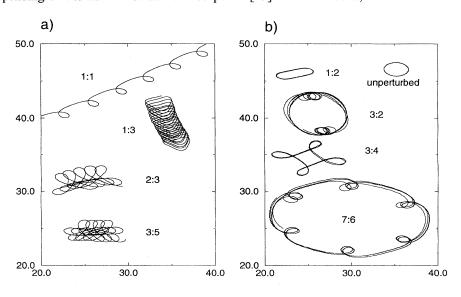


FIG. 2. Tip trajectories for different ratios $\omega_0/\omega = n : m$ of rotation frequency ω_0 and perturbation frequency ω in the model, which has 180°-rotational symmetry. Therefore (a) resonance occurs for all m odd, and (b) no resonance is found for m even. Other parameters as Fig. 1.

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Why not resonance for all rational ratios? It seems that the even numbers of m do not cause a resonance because they match the rotational symmetry of 180° . We will now generalize this intuitive argument and give a simple proof of the relationship between the rotational symmetry and the occurrence of resonance.

Consider the general case of a system with Z_d rotational symmetry, $360^\circ/d$ being the smallest angle of symmetrical rotation. The appropriate symmetry group for the isotropic case is E_2 , the set of all reflections as well as infinitesimal translation and rotation on a plane [26]. When the rotational symmetry is broken into finite rotations, the translational symmetry remains intact. The possible axes of symmetrical reflections are determined by the rotational symmetry. Keeping this in mind, it is sufficient to talk about the rotational symmetries. The numerical results for the d=2 case suggest the following generalization of the resonance condition.

Generalization. Given a rigidly rotating wave in a twodimensional medium with Z_d rotational symmetry. An isotropic perturbation of frequency ω which has a rational relation to the autonomous frequency ω_0 , i.e., $\omega_0/\omega = n/m$ (with n,m coprime integers), gives rise to resonance, if and only if m and d are coprime. For all other rational values of n/m, closed trajectories of the wave tip are obtained.

In order to prove this statement, we first consider the symmetry of the unperturbed rotating wave. In a polar coordinate frame let $R_0(t)$ and $\phi_0(t)$ denote the time-dependent radius and angle coordinates respectively of the unperturbed tip. Obviously, the Z_d symmetry of the underlying medium imposes the following restrictions on the motion of the tip:

$$R_0(t+T_0/d) = R_0(t),$$
 (3a)

$$\phi_0(t+T_0/d) = \phi_0(t) + 2\pi/d,$$
 (3b)

where $T_0 = 2\pi/\omega_0$. Let $c_0(t)$ denote the time-dependent complex velocity of the unperturbed tip. Then we see from Eqs. (3a) and (3b) that $c_0(t)$ is a T_0 -periodic function satisfying the following symmetry requirement:

$$c_0(t+T_0/d) = e^{i2\pi/d}c_0(t).$$
 (3c)

Being T_0 -periodic, $c_0(t)$ can be expanded as a Fourier series with condition (3c) eliminating certain coefficients, so that one obtains

$$c_0(t) = \sum_{r = -\infty}^{\infty} c_r e^{i(rd + 1)\omega_0 t}.$$
 (4)

Now let us assume that an isotropic periodic perturbation of the medium with frequency $\omega = m/n\omega_0$ modulates the tip velocity in the following way:

$$c(t) = (1+\delta)c_0(t), \tag{5}$$

where c(t) is the tip velocity under perturbation and δ is an ω -periodic function that hence allows the following Fourier expansion:

$$\delta = \sum_{s=-\infty}^{\infty} b_s e^{-is\omega t}.$$
 (6)

Combining Eqs. (4)-(6) results in

$$c(t) = c_0(t) + \sum_{r = -\infty}^{\infty} \sum_{s = -\infty}^{\infty} c_r b_s e^{i(rd + 1 - sm/n)\omega_0 t}.$$
 (7)

The net displacement ΔR of the tip after time nT_0 is given by

$$\Delta R = \int_0^{nT_0} c(t)dt = n \int_0^{T_0} [c(nt) - c_0(nt)]dt, \qquad (8)$$

where the latter expression is obtained by linear substitution in the integral and the fact that the T_0 -periodic unperturbed motion does not contribute to ΔR . Combining Eqs. (7) and (8) we obtain

$$\Delta R = \sum_{r=-\infty}^{\infty} \sum_{s=-\infty}^{\infty} c_r b_s \int_0^{T_0} e^{i[(rd+1)n-sm]\omega_0 t} dt.$$
 (9)

The integrals in Eq. (9) provide nonvanishing contributions to ΔR only if the following condition holds:

$$\exists r,s \in \mathbb{Z}: sm = (rd+1)n, \tag{10}$$

where \mathbb{Z} denotes the set of integer numbers. It is immediately clear that for m=1 condition (10) can always be fulfilled by fixing r and n and choosing s=(rd+1)n. In general, in order to fulfill Eq. (1) with m and n having no common factor, s must be a multiple of n. Hence, fulfilling Eq. (10) amounts to finding a linear combination of m and d in \mathbb{Z} equal to one (sm-rd=1), which can be solved—as established in basic number theoretical literature [27]—if and only if the greatest common factor of m and d is 1. Thus one gets nonzero contributions to ΔR only if m and d are coprime natural numbers, and closed tip trajectories otherwise (end of proof).

Finally, it should be mentioned that for intrinsic anisotropy in simple reaction-diffusion systems the invariance of the diffusion operator under inversion of coordinates requires d to be an even number. However, an odd number of d could be obtained by introducing anisotropy in a simple isotropic system through the application of external forces, e.g., electrostatic fields, or in more complex systems with spatial coupling different from mere diffusion.

To summarize, we presented numerical evidence and analytical arguments for a resonance (drift) condition in the case of isotropically perturbed rotating waves in an anisotropic medium. Simply speaking, more and more resonances occur when lowering the rotational symmetry of the medium, i.e., in the absence of any rotation axes (Z_1) all rational values of ω_0/ω cause resonance, while for full rotational symmetry (Z_∞) only the resonances with natural ω_0/ω persist. In principle, the arguments given can be extended to rotating waves in other media by taking the respective symmetry into account.

- M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. 65, 851 (1993).
- [2] G. Gerisch, Naturwissenschaften 58, 430 (1971).
- [3] G. Gorelova and J. Bures, J. Neurobiol. 14, 353 (1983).
- [4] J. Lechleiter, S. Girard, E. Peralta, and J. Clapham, Science **252**, 123 (1991).
- [5] J. M. Davidenko, A. V. Pertsov, R. Salomonsz, W. Baxter, and J. Jalife, Nature (London) 355, 349 (1992).
- [6] A. T. Winfree, Science 175, 634 (1972).
- [7] G. Ertl, Science 254, 1750 (1991).
- [8] A. S. Mikhailov, Foundations of Synergetics I (Springer, Berlin, 1990).
- [9] V. S. Zykov, Simulation of Wave Processes in Excitable Media (Manchester University Press, Manchester, 1987).
- [10] A. T. Winfree, CHAOS 1, 303 (1991).
- [11] A. S. Mikhailov, Phys. Rev. E 49, 5875 (1994).
- [12] F. Mertens and R. Imbihl, Nature (London) 370, 124 (1994).
- [13] A. T. Winfree, When Time Breaks Down (Princeton University Press, Princeton, 1987).
- [14] N. Gottschalk, F. Mertens, M. Bär, M. Eiswirth, and R. Imbihl, Phys. Rev. Lett. 73, 3483 (1994).
- [15] F. Mertens, N. Gottschalk, M. Bär, M. Eiswirth, R. Imbihl, and

- A. Mikhailov, Phys. Rev. E 51, R5193 (1995).
- [16] A. P. Muñuzuri, M. Gómez-Gesteira, P. Pérez-Muñuzuri, V. I. Krinsky, and V. Pérez-Villar, Phys. Rev. E 50, 4258 (1994).
- [17] K. I. Agladze, V. A. Davydov, and A. S. Mikhailov, Pis'ma Zh. Éksp. Teor. Fiz. 45, 601 (1987) [JETP Lett. 45, 767 (1987)].
- [18] M. Braune, A. Schrader, and H. Engel, Chem. Phys. Lett. 222, 358 (1994).
- [19] O. Steinbock, V. Zykov, and S. Müller, Nature (London) 366, 322 (1994).
- [20] A. P. Muñuzuri, M. Gómez-Gesteira, P. Pérez-Muñuzuri, V. I. Krinsky, and V. Pérez-Villar, Phys. Rev. E 50, R667 (1994).
- [21] S. Nettesheim, A. V. Oertzen, H. H. Rotermund, and G. Ertl, J. Chem. Phys. 98, 9977 (1993).
- [22] A. S. Mikhailov, V. A. Davydov, and V. S. Zykov, Physica D 70, 1 (1994).
- [23] M. Bär, N. Gottschalk, M. Eiswirth, and G. Ertl, J. Chem. Phys. 100, 1202 (1994).
- [24] D. Barkley, Phys. Rev. Lett. 68, 2090 (1992).
- [25] D. Barkley, Physica D 49, 61 (1991).
- [26] D. Barkley, Phys. Rev. Lett. 72, 164 (1994).
- [27] See for example G. E. Andrews, *Number Theory* (W. B. Saunders, Philadelphia, 1971).

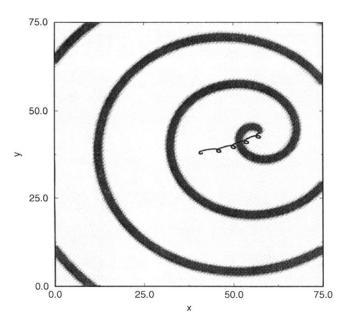


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