A model for jumping and bubble waves in the Belousov–Zhabotinsky-aerosol OT system

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We develop a four-variable model, based on the classic Field–Körös–Noyes mechanism for the oscillatory Belousov-Zhabotinsky (BZ) reaction, that describes recently discovered jumping waves and bubble waves in the BZ reaction in aerosol OT microemulsion and provides insight into their origins. Contrary to suggestions based on previous models, it appears that these phenomena can arise from interaction between a Turing instability and either excitability or a Hopf instability of the steady state, rather than requiring a wave instability. The model should be useful for studying other patterns in BZ microemulsions as well as the behavior of systems of BZ microdroplets coupled through bromine diffusion. © 2009 American Institute of Physics. [doi:10.1063/1.3231488]

I. INTRODUCTION

The well-known Belousov-Zhabotinsky (BZ) reaction, in which malonic acid is catalytically oxidized by bromate in acidic medium\textsuperscript{1,2} has been described by a number of models, many of which are based on the detailed Field–Körös–Noyes (FKN) mechanism\textsuperscript{3} or simplifications of it, e.g., the Oregonator model.\textsuperscript{4} We have recently proposed further modifications of the Oregonator model in order to describe patterns in the BZ-AOT water-in-oil microemulsion system (AOT is the surfactant, aerosol OT).\textsuperscript{5–7} The recent discovery of novel jumping waves (JWs) and bubble waves (BWs) in the BZ-AOT system,\textsuperscript{8} as well as the development of a new experimental system, water microdroplets containing the BZ reactants coupled via diffusion of oil-soluble intermediates of the BZ reaction, Br\textsubscript{2} and BrO\textsubscript{2}, through oil gaps between droplets,\textsuperscript{9} suggest the need for a new model for the BZ reaction. Earlier detailed models (such as FKN) are too cumbersome to describe systems containing large numbers of drops; others (such as the Oregonator) are too simple to account for phenomena like JW. We seek to develop a model of intermediate complexity that is simple enough to allow some degree of analysis, yet retains enough detail to describe the new phenomena (JW and BW) found in experiments.

Our approach is as follows. We first derive a suitable model for the BZ reaction starting from the full FKN model augmented by terms that describe the interaction of the catalyst with light. We simplify the model by a series of approximations in which some concentrations are taken as constant, while others are eliminated through quasiequilibrium or steady state (SS) approximations. In this fashion, we are able to reduce the model to four variables, including one that will be used at the next stage to characterize interactions between droplets in the microemulsion system. After some analysis of the steady state of this model and its stability properties, we add two new variables to describe the messenger species bromine in the surfactant and oil phases of the microemulsion. A further reduction process enables us to obtain a four-variable model for the BZ-AOT system with renormalized diffusion and rate constants to take into account the effects of the microemulsion. We then carry out numerical simulations of JW and BW and analyze our results to gain insight into the origin of these phenomena.

II. MODELING THE BZ REACTION

We start from the FKN model\textsuperscript{3}

\begin{align*}
X + Y + H^+ & \rightarrow 2P, \quad k_1 = 2 \times 10^6 \text{ M}^{-1} \text{s}^{-1}, \\
Y + A + 2H^+ & \rightarrow X + P, \quad k_2 = 2h^2a \text{ (s}^{-1}), \\
X + X & \rightarrow P, \quad k_3 = 3000 \text{ (M}^{-1} \text{s}^{-1}), \\
X + A + H^+ & \rightarrow 2W, \quad k_4 = 42h \text{ (s}^{-1}), \\
Y + P + H^+ & \rightarrow U, \quad k_5 = 5 \times 10^9 \text{ (M}^{-1} \text{s}^{-1}), \\
U & \rightarrow Y + P + H^+, \quad k_6 = 10 \text{ (s}^{-1}), \\
U + MA & \rightarrow Y + BrMA, \quad k_7 = 29m \text{ (s}^{-1}), \\
P + MA & \rightarrow BrMA, \quad k_8 = 9.3m \text{ (s}^{-1}), \\
H_2O + W + W & \rightarrow X + A + H^+, \quad k_9 = 2 \times 10^5 \text{ (M}^{-1} \text{s}^{-1}), \\
W + C & \rightarrow X + Z, \quad k_{red} = 5 \times 10^6 \text{ (M}^{-1} \text{s}^{-1}), \\
Z + BrMA & \rightarrow Y + C, \quad k_9 = k_9b \text{ (s}^{-1}), \\
Z + MA & \rightarrow C, \quad k_{10} = 0.05m \text{ (s}^{-1}),
\end{align*}

where the concentrations of protons $h=[H^+]$, bromate $a=[A]$, malonic acid $m=[MA]$, and bromomalonic acid $b=[BrMA]$ are taken as constants, and $x=[X]=[HBrO_2]$, $y=[Y]=[Br^-]$, $p=[P]=[HOBr]$, $w=[W]=[BrO_2]$.
waves, which require coupling through an activator, were not
limited by the depletion of the reduced form of the cata-
lyst. This constant can be determined from the condition
\[ k_\text{red} w_{\text{max}} = 2k_r w_{\text{max}}, \] (26)
where the maximum concentration of the radical \( w_{\text{max}} \) can be
found with the aid of the following equation [cf. Eq. (18) when
\( k(I)=0 \)]:
\[ k_\text{red} w_{\text{max}} c_{\text{min}} = k_c c_0 + k_{10} c_{\text{min}}. \] (27)
Equations (26) and (27) give
\[ c_{\text{min}}^2 = 2k_r(k_c + k_{10})c_0/k_{\text{red}}. \] (28)

Our next simplification is to eliminate the fast variable \( p \).
We set \( dp/dt=0 \) in Eq. (19) and find the quasistationary p.
We note that \( k_3 y_p \approx k_9 p \) at nearly all times, since \( k_3 y_p \gg k_9 \),
where \( y_p = k_5/k_1 \) (\( y_p \) is the critical concentration of the
inhibitor bromide below which autocatalytic reactions 4 and 9
start). If so, we can drop the term \( k_9 p \) from Eq. (19) to give
\( k_3 y_p = -2k_1 x y - k_2 y - k_3 x^2 - k_4 p \). Substituting this result in
Eqs. (17) and (20) and taking into account the previous sim-
plications, we finally have our four-variable model

(A) \[ dx/dt = -k_1 x y + k_2 y - 2k_3 x^2 + k_4 x / (c_0 - z + c_{\text{min}}), \] (29)
\[ dy/dt = -k_1 x y - k_2 y - k_3 y p + k_d u + k_9 z + k(I)c_b / (b_c + b), \] (30)
\[ dz/dt = k_3 y p - k_d u - k_7 u, \] (31)
\[ dw/dt = 2k_4 x - 2k_5 x^2 - k_\text{red} w_c, \] (32)
\[ dc/dt = -k_\text{red} w_c + k_2 z - k_10 z - k(I)c_b / (b_c + b), \] (33)

where \( b_c = k_d / k_c = 0.05M. \) (10,12)

Experiments on patterns in a one-dimensional (1D) array
of coupled BZ droplets\(^9\) imply that inhibitory coupling of the
BZ microdroplets through soluble Br2 dominates, since
waves, which require coupling through an activator, were not
found. This result allows us to eliminate \( \text{Br}_2\) (w) from the
model by making the SS approximation, i.e., setting
\( dw/dt = 0 \). From Eq. (21) we have \( k_\text{red} w_c = 2k_3 x - 2k_5 x^2 \).
Substituting this expression into Eqs. (16), (18), and (22)
and ignoring \( k_5 w_c \) (since \( k_\text{red} w_c \approx 2k_5 x^2 \) at all times during the reaction), we have

\[ dx/dt = -k_1 x y + k_2 y - 2k_3 x^2 + k_4 x, \] (23)
\[ dz/dt = 2k_4 x - k_9 z - k_10 z + k(I)c_b / (b_c + b + 1), \] (24)
\[ dc/dt = -2k_4 x + k_9 z + k_10 z - k(I)c_b / (b_c + b + 1). \] (25)

Also, \( z + c = c_0 \), where \( c_0 \) is the total concentration of the cata-
lyst, so we can replace \( c \) by \( c_0 - z \). The resulting equations
have the defect that the autocatalytic term, \( k_4 x \) in Eq. (23), is
not limited by the depletion of the reduced form of the cata-
lyst. The classic Oregonator model\(^4\) suffers from the same
flaw. To resolve this problem, some authors\(^6,13\) modify the
autocatalytic term as \( k_4 x c / (c + c_{\text{min}}) \), where \( c_{\text{min}} \) is a small
constant. This constant can be determined from the condition
that the rates of reactions 9 and 10 when \( c = c_{\text{min}} \) (when the
rate of autocatalysis is a maximum) are approximately equal
(note that in most other phases of the reaction \( k_\text{red} c \approx 2k_r w \)) so that

\[ k_\text{red} c_{\text{min}} = 2k_r w_{\text{max}}. \] (26)
A. Steady state

The steady state(s) (SS) of system (29)–(32) (model A) at \( \phi(I)=0 \) can be found by solving numerically the following equations:

\[
x_{SS} = \alpha z_{SS}(1 - z_{SS}/c_0 + \epsilon)/(1 - z_{SS}/c_0),
\]

\[
x_{SS} = \mu[k_9 + F(z_{SS})]/[k_9 - F(z_{SS})],
\]

where \( F(z_{SS}) = ak_4 - 2k_5a_2z_{SS}(1 - z_{SS}/c_0 + \epsilon)^2/(1 - z_{SS}/c_0)^2 \), \( \mu = k_2/k_1 \), and \( \alpha = (k_9 + k_{10})/(2k_2) \). Then \( y_{SS} = k_0z_{SS}/(k_3z_{SS} + k_2) \), \( u_{SS} = (2k_1z_{SS})/z_{SS} + k_2y_{SS} + k_2z_{SS}^2/k_7 \). Model A has a single SS, since the right hand side of Eq. (33) is monotonically increasing and that of Eq. (34) is monotonically decreasing for allowable values of \( z_{SS} \), if \( k_9 > k_{10} \), which is true. Note that \( k_9 > k_{10} \) implies that the stoichiometric factor in Eq. (30'), \( g > 0.5 \).

When \( \phi(I) \neq 0 \), the solution of model (A) can be found numerically by solving

\[
f_2(z)(k_2 - k_1x_{SS})/(k_1x_{SS} + k_2) - 2k_2x_{SS}^2 + x_{SS}f_1(z) = 0,
\]

where

\[
x_{SS} = f_1(z)/2f_1(z),
\]

\[
f_1(z) = k_4(c_0 - z)/(c_0 - z + c_{min}), \quad f_2(z) = k_5 + k_2/(c_0 - z)/(b_c + b_1), \quad f_3(z) = k_6 + k_2/(c_0 - z)/(b_c + b_1).
\]

After finding \( z_{SS} \) and \( x_{SS} \), we find first \( y_{SS} \) and then \( u_{SS} = f_2(z_{SS})/(k_1x_{SS} + k_2) \) and \( u_{SS} = (2k_1x_{SS} + k_2x_{SS}^2 + k_2z_{SS}^2)/k_7 \).

B. Linear stability analysis of model (A)

Knowing the SS of model (A), we can proceed to analyze its stability. With \( \phi(I)=0 \), we varied the key parameters of the model, \( h, a, m, \), and \( k_9'' \), where \( k_9'' = k_{10}' \) (\( k_9'' \) depends on the initial concentration of \( Br^- \) and consequently on the initial \( b \)), over broad ranges, though in experiments \( m \) is usually confined to values between 0.03 and 1, \( h \) between 0.05 and 1, and \( a \) between 0.05 and 0.5. The phase diagram is shown in Fig. 2. The onset of Hopf instability (lines 1–6 in Fig. 2) occurs from both sides of the oscillatory region via a canard phenomenon.14 The boundary between the oscillatory state and the reduced SS (curves 1–3) shifts toward the SS as \( k_9'' \) decreases or \( h \) increases. The boundary between the oscillatory state and the oxidized SS (curves 4–6) moves toward the SS as \( k_9'' \) increases or \( h \) decreases.

III. MODELING THE BZ-AOT SYSTEM

To describe the BZ-AOT system, we add two new variables to model (A), \( Br_2 \) in the oil phase, \( u_{Ob} \), and \( Br_2 \) in the surfactant pseudophase, \( u_{St} \), and we rename the variable \( u \) in the aqueous phase as \( u_W \). We normalize the concentrations in the three phases (the aqueous phase with volume fraction \( \varphi_W \), the oil phase with volume fraction \( \varphi_{oil} \), and the surfactant phase with volume fraction \( \varphi_{oil} + \varphi_{oil} + \varphi_{oil} = 1 \) to the bulk concentrations (subscript b) as \( \varphi_{Wb} = \varphi_{Wb}, \varphi_{Ob} = \varphi_{Ob}, \varphi_{Ob} = \varphi_{Ob} \). We also add diffusion coefficients for all variables, since we
where $k_f$ of $\text{Br}_2$ from water to the AOT shell is $k_b$ as $u_O$. The effective diffusion coefficient $D_u$ is the diffusion coefficient of $\text{Br}_2$ in the surfactant droplet, is $10^{-6} - 10^{-7} \text{cm}^2/\text{s}$, while $D_d$ is above and to the left of the curves, while for curves 4–6, the SS is oxidized is below and to the right.

wish now to model the reaction-diffusion system in order to explain the reaction-diffusion. The effective diffusion coefficient $D_u$ is determined from the equation

$$D_u \nabla^2 u_W + D_s \nabla^2 u_S + D_o \nabla^2 u_O = D'_u \nabla^2 u_W$$

$D_u' = [D_u + D_k k_{f_2} k_{f_2} + D_d k_{f_2} k_{f_2} k_{f_1} k_{f_1}]/[1 + k_{f_2} k_{f_2} + k_{f_2} k_{f_2} k_{f_1} k_{f_1}].$

Equations (41) and (43) are obtained by assuming that all rate constants $k_f$, $k_{f_2}$, and $k_{f_1}$ are very large, since the diffusion of $\text{Br}_2$ between phases is rapid at the nanometer distances involved here, and that the following equilibria hold

$$k_{f_2} u_S = k_{f_2} u_O$$

$$k_{f_1} u_W = k_{f_1} u_S.$$  

Returning in Eq. (40) to the concentrations in the aqueous phase and using the equality $u_W = u_s[1 + k_{f_1} k_{f_1} k_{f_2} k_{f_2} (k_{f_1} k_{f_1})]$ in Eq. (30) (where $u_t = u_w + u_s + u_O$) to replace $k_{f_1} u_W$ with $k_f u_t$, we finally have the model (B) for the BZ-AOT system at $k(t) = 0$ (where we omit for simplicity subscripts $W$ for $x$, $y$, and $z$, and subscript $t$ for $u$)

(B) $\frac{dx}{dt} = -k_{f_1} x y + k_{f_2} y - 2 k_{f_1} x^2 + k_{f_2} x (c_0 - z)/(c_0 - z + c_{\min}) + D_d \nabla^2 x,$

$\frac{dy}{dt} = -3 k_{f_1} x y - 2 k_{f_2} y - k_{f_2} x^2 + k_{f_2} x + k_{f_2} + D_d \nabla^2 y,$

$\frac{dz}{dt} = 2 k_{f_2} x (c_0 - z)/(c_0 - z + c_{\min}) - k_{f_2} - k_{f_2} + D_d \nabla^2 z.$

We can estimate the constants $k_f$ and $D_u'$, where $P_{B_1}$ and $P_{B_2}$ are the partition coefficients for $\text{Br}_2$ between AOT and water and between oil and AOT, respectively, and $r_{v_1} = V_o/V_w = \varphi_o/\varphi_W$, $r_{v_2} = V_o/V_o = \varphi_o/\varphi_o$, the volume ratios of the corresponding phases. For $\varphi_o = \varphi_w + \varphi_s = 0.4$ (i.e., below the percolation threshold), using the relation $\varphi_s = 2.5 \varphi_W$ [for $\omega = 15$ (Ref. 15)], which gives $r_{v_1} \equiv 1.5$ and $r_{v_2} \equiv 2.5$, and assuming that $P_{B_1} = 2.0$ (Ref. 16) and $P_{B_2} = 100$ (since $P_{B_1} = P_{B_2} = P_{B_3}$, where $P_{B_3} \equiv 20$, the partition coefficient between water and octane), we obtain $k_{f_1} \equiv k_f/73$ and $D'_{d} = D_{d}/14$ for $D_{d} = D_{d}/0.01 D_{d}$ and $D'_{d} = D_{d}$ for $D_{d} = D_{d}/100 D_{d}$. The coefficient $D_{u}'$ should in general be smaller than $D_u$ due to the ability of the AOT-shell (large aggregate of surfactant molecules) to transiently capture $\text{Br}_2$ and thereby slow its diffusion.

In the dimensionless Eqs. (29’)-(32’), $k_f$ should be replaced by $k_f: u = \bar{u} k_f k_s k_2^2$, and $e_s = (k_{f_1} + k_{f_2})k_f k_s^2$. Note that $e_s$ becomes almost 100 times larger and the variable $\bar{u}$ is no longer a fast variable. The other constants in the model also require adjustment to take into account the fact that the environment in the water droplets of the BZ-AOT system differs from that in the pure aqueous system. For example, the concentration of protons is much smaller due to protonation of the $\text{SO}_3^-$ groups of the AOT molecules.

A. Linear stability analysis of model (B)

Linear stability analysis of system (B) reveals a broad range of parameters in which Turing instability occurs. A typical dispersion curve for Turing instability is shown in Fig. 2. Phase diagram in the $a$-$m$ plane for models (29)-(32). All curves = (curves 1 and 3) 0.16, (2, 6) 0.2, (4 and 5) 0.4, $k_d = (1) 0.15$, (2, 3) 0.2, (4) 0.06, (5, 6) 0.1. “Osc.” denotes the oscillatory region. Curves 1–3 are well fitted by lines $m = C_1 \times a$, where $C_1 = (i=1) 10$, (2) 3, (3) 1.92, while curves 4 and 5 are well fitted by lines $m = C_2 \times a^2$, where $n = 2.1$–2.3 and $C_2 = (i=4) 0.306$, (5) 0.064, and (6) 0.0207. For curves 1–3, the SS (reduced) is above and to the left of the curves, while for curves 4–6, the SS (oxidized) is below and to the right.
the behavior of the model, which can lead to inwardly propagating waves. We also find wave instability, in some cases with negative dispersion.

Simulations were performed both in 1D and two dimensions. Figures 3 and 4 show dispersion curves for model (B). (a) Turing instability. (b) Wave instability. Curves 1 and 2 are, respectively, Re(A) and Im(A), where $A$ is the eigenvalue of the linearized model (B) with the largest real part. Parameters: (a) $h=0.08$, $a=0.1$, $m=0.5$, $c_0=0.003$, $k_1=2 \times 10^6$, $k_2=2h$, $k_3=3000$, $k_4=2n$, $k_5=29m/150$, $k_6=0.12m$, $k_7=0.05m$, $c_{\text{min}}$ is calculated from Eq. (28) at $k_z=2 \times 10^8$ and $k_{\text{opt}}=5 \times 10^6$. $D_u=0.05$, $D_u=1$. (b) $h=0.21$, $a=0.29$, $m=0.024$, $c_0=0.003$, $k_1=4.2 \times 10^5$, $k_2=0.025578$, $k_3=3000$, $k_4=2.5758$, $k_5=6.96$, $k_6=0.051m$, $k_7=0.055m$, $c_{\text{min}}=1.07867 \times 10^{-3}$, $D_j=0.01$, $D_j=1$.

Fig. 3(a). However, as we will see in our numerical simulations, the presence of Turing instability does not guarantee the emergence of Turing patterns in model (B). This feature probably arises from the large excitability of the SS in this system.

If the SS is close to the fully oxidized state ($z_{\text{SS}}=c_0$), we also find wave instability, in some cases with negative dispersion, which can lead to inwardly propagating (anti)waves. An example is shown in Fig. 3(b). Typically, models that produce a wave instability contain a second fast-diffusing activator (in addition to our activator $x$ and inhibitor $y$). Here, we have no additional fast-diffusing activator. Instead, the same fast-diffusing inhibitor $u$ that gives rise to Turing instability produces our wave instability, because the terms $2k_{1,xy}$ and $k_{3,x}$ that involve activator $x$ are responsible for the production of $u$, thus coupling $u$ with $x$.

B. Computer simulation of model (B)

To integrate the partial differential equations, we employ the commercially available software package FLEXPDE, with a typical error (ERRLIM) of $1.0 \times 10^{-7}$ for each variable in each spatial cell. Simulations were performed both in 1D and two dimensions (2D).

A typical set of JW in 1D is shown in Fig. 4. Figure 4(a) is a space-time plot, in which the saltatory manner of wave propagation is clearly visible. Since the range of parameters for JW is surprisingly broad, we can easily control the wave speed and the length of a single jump by changing our parameters, in particular $m$, $a$, $h$, and $k_0$. The last constant depends on the concentration of BrMA and strongly affects the behavior of model (B). For the parameters we used, model (B) exhibits excitability in zero dimensions, i.e., without diffusion terms, and Turing instability when diffusion terms are included.

To understand the origin of this jumping behavior, we followed the values of all the model variables. Figure 4(b) shows the spatial distribution of variables $z$ (oxidized catalyst) and $y$ (inhibitor) at several times in the vicinity of the third jump shown in Fig. 4(a). The first panel of Fig. 4(b) is a coarse scaling, while the next three panels show a finer resolution. We see that when a peak of $z$ forms, two peaks of $y$ begin to grow at its edges, thus preventing the $z$-peak from spreading. These $y$-peaks, and a third peak that emerges between them ($t=1128$), result from the fast diffusion of bromine ($u$). The $z$-peak is soon destroyed by the central $y$-peak, which grows in its place [two last panels of Fig. 4(b)]. The next $z$-peak arises in a distant region, where $y$ and $u$ are relatively small and $x$ is relatively large (here, at position 135 a.u. and time 1428 s).

In Fig. 4(a), we see that after the first JW, a second wave emerges and then a third, despite the fact that the system is excitable (stable to small perturbations), not oscillatory. This behavior results from the proximity of the system to the onset of Hopf bifurcation: the initial perturbation decays via damped oscillations, which are large enough to induce a new cycle of excitability that starts at the left end of the segment. Therefore the frequency of JW, which is determined by that of the slowly decaying perturbation, is relatively small and there are no bulk oscillations in the system. We can change the parameters slightly, for example, by increasing $h$, and push the system into the oscillatory domain. An example of JW in this case (Hopf + Turing) is shown in Fig. 5. As in Fig. 4(a), we see JW as cascading white dashes. In addition, the horizontal lines at the right represent bulk oscillations. The periods $T_{JW}$ of JW and $T_b$ of bulk oscillations are 1103 and 1317 s, respectively. The point at which the JW and bulk oscillation collide is a shock point. The velocity of this shock point (marked by the white dashed line) can be calculated from $T_{JW}$, $T_b$, $L_f$ (the length of a single jump) $\approx 35$ a.u. (cf. the Turing wavelength found from the dispersion curve as $2\pi/k_{\text{opt}}=15$ a.u.), and the time interval between two consecutive jumps $T_J=191$ s as $(T_b-T_{JW})L_f/(T_JT_{JW})$. 

![Fig. 3](image1.png)

![Fig. 4](image2.png)
Analogous shock points were observed in the experiment. Since the range of parameters for JW is very broad and JW can arise when the system exhibits Turing instability, excitability and/or Hopf instability, it is important to determine whether such behaviors as Turing patterns and conventional continuously propagating waves can also occur in this model as they do in the actual BZ-AOT system. Turing patterns are found at very small diffusion coefficient for species $x, y, z$. Note that the distance between the outer $z$-peaks is about $5.2 \lambda_T$, where $\lambda_T=5.24$ is the Turing wavelength obtained from the dispersion curves by linear stability analysis, while the distance between the two central $z$-peaks is $\lambda_T/2$. These two central peaks emerge first due to a small, narrow initial perturbation at the middle of the segment, while the other peaks arise later as the perturbation spreads out from the central peaks. The problem of stabilization of large-amplitude and sharp Turing patterns is still an open question. We cannot predict their wavelengths. Only for small-amplitude Turing patterns for which the concentrations are close to their SS values, can we guarantee that the observed Turing wavelength will be near $\lambda_T$. Traveling waves occur when species $x, y, z$ diffuse more rapidly. In these simulations, we used slightly different coefficients $D_X, D_Y, D_Z$ instead of a single $D_d$ for all species. The JW found in 1D can assume various shapes in 2D. In Fig. 7, we show circular JW found in 2D. In Fig. 8, we show bubble waves found at another set of parameters. In
In 1D, the waves found with these parameters resemble ordinary JW. In 2D, however, each emerging ring splits into many small circular waves that look like small bubbles. When bubbles collide, they annihilate. This splitting is due to a Turing instability of the smooth circular ring. Bubbles can move independently of one another. As a result, some bubbles propagate (by jumping) slightly faster or have slightly larger jump lengths ($L_j$) than others (upper left quadrants of snapshots in Fig. 8, where bubbles emerge at $t = 700–750$, while in other areas, bubbles emerge at $t = 600$). In these simulations we made $D_Z$ slightly smaller than $D_X = D_Y$, though this condition is not essential for BW. The 2D simulations are quite time-consuming, and we did not investigate a broad range of parameters as we did in 1D.

IV. DISCUSSION

We have developed a new model for the BZ and BZ-AOT systems and found that this model is capable of simulating the JW and BW found in recent experiments. The results suggest that JW and BW arise from a combination of Turing instability and excitability or Turing + Hopf, rather than from a wave instability as suggested previously. That this model is derived from the detailed FKN mechanism and is able to explain BW, which could not be obtained with earlier models containing a wave instability, is encouraging. The existence of two different mechanisms that show JW emphasizes the fact that any single pattern in a reaction-diffusion system may have multiple explanations. To evaluate a proposed mechanism, one needs to see if it reproduces other patterns close in the parameter space to the pattern of interest. Further tests of model (B) might, for example, use it to simulate segmented waves. Note that we have used in our present simulations the same constants employed in the FKN mechanism for the BZ reaction in the aqueous phase.

We anticipate that models (A) and (B) will find many applications, for example, in simulations of diffusively coupled BZ-droplets or in investigating additional patterns in the BZ-AOT system, where bromine is important as a messenger molecule.

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