Effervescent waves in a binary mixture with non-reciprocal couplings

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Non-reciprocal interactions between scalar fields that represent the concentrations of two active species are known to break the parity and time-reversal (PT) symmetries of the equilibrium state, as manifested in the emergence of travelling waves. We explore the notion of nonlinear non-reciprocity and consider a model in which the non-reciprocal interactions can depend on the local values of the scalar fields. For generic cases where such couplings exist, we observe the emergence of spatiotemporal chaos in the steady-state. We associate this chaotic behaviour with a local restoration of PT symmetry in fluctuating spatial domains, which leads to the coexistence of oscillating densities and phase-separated droplets that are spontaneously created and annihilated. We uncover that this phenomenon, which we denote as effervescence, can exist as a dynamical steady-state in large parts of the parameter space in two different incarnations, as characterized by the presence or absence of an accompanying travelling wave.

I. INTRODUCTION

Interactions between components of biological and artificial living matter are mediated in a wide variety of ways across the scales [1]: from complex behaviour patterns in humans [2], to visual perception in birds [3], hydrodynamic interactions in ensembles of cilia and flagella [4, 5], information-controlled feedback in programmable active colloids [6, 7], and chemical fields in catalytically active colloids [8–10] and enzymes [11, 12]. These microscopic interactions quite generically break action-reaction symmetry due to non-equilibrium conditions. Reciprocity breaking has already had a far reaching impact in fields like structural mechanics, in realizing metamaterials [13], and in optics, by achieving photon blockade [14]. In recent years, non-reciprocity in interactions has generated interest as an exciting new ingredient to develop minimal models for active matter systems out of thermodynamic equilibrium [12, 15–20].

Conserved active scalar field theories for two species with non-reciprocal interactions display travelling waves, moving patterns and oscillations in the steady-state [15, 16]. When activity, i.e. the strength of non-reciprocity, is strong enough to win over the hydrodynamic forces driving the system towards bulk phase separation, the system reaches novel steady-states that break the parity and time-reversal symmetry of the bulk-separated equilibrium state. The transition to travelling patterns occurs upon tuning the parameters of the model such that an exceptional point is crossed [15, 18, 21], at which the eigenvalues of the dynamical matrix determining the stability of the fully mixed state to small perturbations acquire complex values. Moreover, the pair of eigenvalues coalesce at the exceptional point and the corresponding eigenvectors become parallel [21]. This transition from a state with a spontaneously broken symme-

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FIG. 1. Emergence of effervescence in the dynamical steady-states of the nonlinear NRCH model. Panel (a) shows snapshots of the field $\phi_1$ at the times indicated, which demonstrate the effervescent waves, corresponding to $\alpha_0 = 4$ and $\alpha_1 = 5$ (see Supplemental Movie S1). The black arrows mark the progression of the travelling wave. Panel (b) shows the corresponding amplitude field $|\phi|$. Note that while (a) shows a clear striped pattern, $|\phi|$ deviates from a constant value in areas where fluctuating droplets are formed. For $\alpha_1$ sufficiently larger than $\alpha_0$, we observe an effervescent steady-state without an accompanying travelling wave. Panels (c-d) shows snapshots of $\phi_1$ and $|\phi|$ from simulations corresponding to $\alpha_0 = 2.3$ and $\alpha_1 = 4.6$ (see Movie S2 for the full time evolution).

with phase-separated droplets of matter in their wake, are generated (see Fig. 1, videos S1 and S2 in the supplement). The outline of the paper is as follows. First, we will introduce the phenomenon of effervescence and summarize our findings in Sec. II. We then introduce our theoretical model in Sec. III explaining the choice of a particular free energy that is invariant under unitary rotations in the composition plane, which makes the equations tractable to theoretical analysis. In Sec. IV we study the plane wave solutions that enable us to determine, via a stability analysis, the region of the parameter space in which these waves are unstable. The unstable region of the parameter space is then probed in Sec. V by solving the equations of motion numerically to classify the dynamical steady-states and summarize the results in a state diagram. Finally, the effect of composition is studied in Sec. VI, which is followed by concluding remarks in Sec. VII. Some details of the linear stability analysis are relegated to Appendix A.

II. EFFERVESCENCE

An emergent feature of the non-reciprocal interactions implemented at the linear level in a mixture of two species is the spontaneous breaking of space-translation, time-translation, time-reversal, and polar symmetries, through the formation of traveling patterns [15, 16]. When introducing nonlinear terms in the non-reciprocal interaction, in the spirit of a Landau expansion, we observe spontaneous creation and annihilation of droplets in combination with the traveling pattern, or even, in its absence, as shown in Fig. 1. We find droplets enhanced in either species 1 or 2 (described by scalar fields $\phi_1$ and $\phi_2$, respectively), as well as composites where a droplet enhanced in one species is encapsulated by another enhanced in the other species. The droplets are dynamic, both in terms of being randomly nucleated and dissolved, and their fluctuating shapes (see video S2 in the supplement). The effective non-reciprocal interaction reverses sign when the modulus of the order parameter is increased, i.e., while at low densities 1 chases 2, at higher densities 2 chases 1. The emergent imperfect PT symmetry breaking with local restoration of reciprocal interactions produces two new states, namely an effervescent wave which is a hybrid state with droplets and a traveling pattern, shown in Fig. 1(a-b), and effervescence without the traveling pattern, shown in Fig. 1(c-d).

The phenomenon of effervescence reveals a granular structure for the domains that restore reciprocal interactions, as evident in the domain size distribution shown in Fig. 2(a-b). We observe a prominent peak for a fundamental reciprocal granule and an oscillatory pattern for
larger areas [Fig. 2(a)] that originate from the composition of the domains being in the form of clusters of the reciprocal granules (see Supplemental Movies S1 and S2). For the effervescent waves, we observe a coexistence between the selected values for the modulus of the order parameter corresponding to the traveling pattern and the reciprocal granules, as observed in the distribution of the order parameter shown in Fig. 2(c). From the value of the modulus of the order parameter $|\phi|$ [see Fig. 3(a)] one can verify that the effervescent granules correspond to a local restoration of PT symmetry. Effervescence gives rise to spatiotemporal chaos, and the emergence of an effective noise from the deterministic nonlinear dynamics, due to nonlinear non-reciprocal interactions.

III. NONLINEAR NON-RECIPROCAL COUPLING

To build our theoretical framework, we can start with the dynamics of conserved fields $\phi_i$ $(i = 1, 2)$ that can be written as $\dot{\phi}_i = \Gamma_i \nabla^2 \mu_i$, in terms of the scalar chemical potentials $\mu_i$ and mobilities $\Gamma_i$. At equilibrium, $\mu_i$ can be obtained from a free energy $F$ via $\mu_i \equiv \delta F/\delta \phi_i$. The free energy is chosen as $F = \int f(\phi_i(r,t)) + \frac{K_1}{2}(\nabla \phi_1)^2 + \frac{K_2}{2}(\nabla \phi_2)^2$, where $f$ is the Helmholtz free energy (per unit volume) that describes phase separation in homogeneous systems. We now introduce non-equilibrium activity in the model by adding a non-reciprocal interaction between the two species. This can be achieved by introducing an anti-symmetric coupling between the species without any loss of generality, because the symmetric (reciprocal) part of the interaction can be absorbed in the expression for $f$. We can write the governing equations for the two fields as

$$\partial_t \phi_1 = \Gamma_1 \nabla^2 \left[ \frac{\partial f}{\partial \phi_1} + \alpha(\phi_1, \phi_2)\phi_2 \right] - \Gamma_1 K_1 \nabla^4 \phi_1, \quad (1)$$

$$\partial_t \phi_2 = \Gamma_2 \nabla^2 \left[ \frac{\partial f}{\partial \phi_2} - \alpha(\phi_1, \phi_2)\phi_1 \right] - \Gamma_2 K_2 \nabla^4 \phi_2, \quad (2)$$

where the non-reciprocal coupling $\alpha(\phi_1, \phi_2)$ is taken to be a function of the fields, as a generalization of the NRCH model introduced in Ref. [15]. We note the presence of number conservation is an important constraint that enhances the richness of the dynamics of such systems [see the examples in Figs. 3(b-c)].
Equations (1) and (2) can be written equivalently as an equation for a complex field \( \phi = \phi_1 + i\phi_2 \) with an amplitude \(|\phi| = \sqrt{\phi_1^2 + \phi_2^2}\) and a phase \(\theta = \tan^{-1} \phi_2/\phi_1\); see Fig. 3(a). Dynamical steady-states resembling chemical turbulence are observed when \(|\phi_1|,|\phi_2| \leq 0\) at equilibrium, although we find that the steady states of (1) and (2) are independent of the detailed form of \(f\), as expected. In the same spirit, we have chosen the following expression for \(\alpha\)

\[
\alpha(\phi_1, \phi_2) = \alpha_0 - \alpha_1 |\phi|^2,
\]

which introduces a non-linear nonreciprocal coupling between the two fields. For simplicity, we set \(\Gamma_1 = \Gamma_2 = \Gamma\) and \(K_1 = K_2 = K\). With the above choices, the nonlinear NRCH model is best described in terms of the dynamics of \(\phi\), which satisfies the following equation

\[
\partial_t \phi = i\phi \nabla^2 \left[-(1 + i\alpha_0)\phi + (1 + i\alpha_1)|\phi|^2\phi - K \nabla^2 \phi\right],
\]

or equivalently, the dynamics of the amplitude and phase fields described earlier. We will now discuss the different dynamical steady-states of the non-linear NRCH model.

**IV. TRAVELLING WAVES**

We start by exploring the possibility of Eq. (5) adopting travelling wave solutions, which is a natural consequence of the number conservation constraint as shown in Fig. 3(c). Our choice of the bulk free-energy in Eq. (3) allows us to write down an exact dispersion relation for the travelling waves for a specific average composition of the system, which we choose as follows \((\phi_1) = (\phi_2) = 0\). The model is invariant under global phase rotation, i.e. the transformation \(\phi \to e^{i\theta}\phi\) leaves (5) unchanged. At this composition, the homogeneous state is linearly unstable to perturbations irrespective of the values \(\alpha_0, \alpha_1\). To capture the properties of the pattern formation process, we use a travelling wave trial solution \(\phi_q\) as parameterized by a wavenumber \(q\), namely,

\[
\phi_q(r, t) = \rho_q e^{i(q \cdot r - \omega t)},
\]

and substitute it in Eq. (5) to obtain expressions for the amplitude \(\rho_q\) and the dispersion relation \(\omega(q)\). We find

\[
\rho_q = \sqrt{1 - q^2/q_0^2}, \quad \forall q < q_0,
\]

\[
\omega(q) = \Gamma q^2 \left[-\alpha_0 + \alpha_1 \left(1 - \frac{q^2}{q_0^2}\right)\right],
\]

where \(q_0 = 1/\sqrt{\Gamma}\) [see Fig. 4(a)]. The solutions in the form of Eq. (6) exist for all values of \(\alpha_0\) and \(\alpha_1\) and for a wide spectrum of wavelengths. It is, however, imperative to examine the stability of these solutions.

To perform the stability analysis, we insert a trial solution of the form

\[
\phi_q(r, t) = [\rho_q + \delta \rho_q(r, t)] e^{i(q \cdot r - \omega t)}
\]

in Eq. (5), and derive the effective governing equation for the perturbation \(\delta \rho_q(k, t)\) in Fourier space with the wavenumber \(k\), at the linear order, as has been done for the case of metachronal waves in cilia [40]. The eigenvalues of the resulting linear dynamical equations...
in Fourier space can then be calculated (see Appendix A) and used to isolate the dominant behaviour of the system as reflected in the eigenvalue with the larger real part. Using an expansion up to quadratic order in \( k \), and a decomposition of the wavevector into the longitudinal component \( k_L = k \cdot q / q \) and the transverse component \( k_T = k \cdot (I - qq / q^2) \), we can obtain the dominant eigenvalue, which we present as

\[
\lambda(k) = iV_k L - D_L k_L^2 - D_T k_T^2,
\]

where the advection velocity \( V \), and the longitudinal and transverse diffusion coefficients \( D_L \) and \( D_T \), are found as

\[
V(q) = 2\Gamma q \left(-\alpha_0 + \alpha_1 - 2\alpha_1 q^2 / q_0^2\right),
\]

\[
D_L(q) = -\Gamma\alpha_1 (\alpha_0 - \alpha_1) + \Gamma q^2 (3q^2 - 5q_0^2) q_0 (q_0^2 - q^2),
\]

\[
D_T(q) = -\Gamma\alpha_1 (\alpha_0 - \alpha_1) + \Gamma q^2 q_0 (q_0^2 - q^2).
\]

The travelling wave solutions in Eq. (6) are unstable in the part of the phase space where \( D_L < 0 \). First, note that for \( \alpha_1 = 0 \), \( D_L \) reduces to the expression that holds for the conserved real Landau-Ginzburg dynamics, i.e. \( \alpha_0 \) alone does not create turbulence. For \( q \to 0 \), \( D_L \approx -\alpha_1 (\alpha_0 - \alpha_1) \), indicating that an interplay of \( \alpha_0 \) and \( \alpha_1 \) is necessary for destabilizing plane waves. However, \( \alpha_1 \) alone can be used to tune \( D_L \) to negative values at sufficiently large values of \( q \).

The stability diagram in the \((\alpha_0, \alpha_1)\) plane is shown in Fig. 4(b), with the unstable regions corresponding to \( D_L < 0 \) being shaded (and the colours correspond to the wave-numbers indicated in the legend). For wavenumbers lower than a threshold value of \( q_0 / \sqrt{3} \), the unstable region consists of two unconnected pieces in the quadrants \( \alpha_0, \alpha_1 > 0 \) and \( \alpha_0, \alpha_1 < 0 \). Above the threshold, the two regions connect to form a single connected unstable region enclosing the origin. The Eckhaus stability criterion at equilibrium, which states that all wavelengths greater than \( q_0 / \sqrt{3} \) are unstable, thus determines the topology of the stability diagram.

The result (11) is checked using numerical simulations with slightly perturbed travelling waves of a chosen wavelength as the initial condition and allowing the system to evolve for a sufficiently long time. The difference between the space averaged amplitude and the amplitude of the input wave defined as

\[
\Delta(q) = \frac{1}{A} \int d^2 r |\phi(r, t)| - \rho q,
\]

is calculated in the \((\alpha_0, \alpha_1)\) plane to determine the stability of the travelling waves; see Fig. 4(c). Here \( A = 4L^2 \) is the area of the system. The wavelength of the sinusoidal wave, \( q \), and the time periodicity, \( \omega(q) \), of the wave at a fixed position in space, are determined using Fourier transforms.
\[ \tau = \frac{1}{T} \int_0^T dt \, \delta \phi_i(k,t) \phi_i(-k,t), \]

where summation over repeated index \( i \) is implied, and the power spectrum \( S(\omega) \), defined as

\[ S(\omega) = \frac{1}{A} \int \mathcal{D}^2 r \, \phi_i(r,-\omega) \bar{\phi}_i(r,\omega). \]

We observe that \( S(k) \) is isotropic in the effervescent state [Fig. 5(b)] and shows distinct peaks corresponding to the wavelength of the travelling wave in the effervescent waves [Fig. 5(c)]. Moreover, \( S(\omega) \) exhibits a nearly constant plateau and is indistinguishable from white noise in the effervescent state, while a pronounced peak appears in addition to the nearly constant background for effervescent waves [Fig. 5(d)]. We can also probe the heat-map of \( S(\omega) \) as a function of \( \omega \) and \( \alpha_1 \) for fixed \( \alpha_0 \) [Fig. 5(e)]. We observe that the peak for travelling waves disappears in the effervescence case and reappears for effervescent waves.

\[ \mathcal{D} \mathcal{D}^* \mathcal{D}^\dagger = \Gamma \mathcal{G}^2 \left( 1 + i \alpha_0 - 2(1 + i \alpha_1)|\phi_0|^2 \right). \]
up to order $q^2$. The eigenvalues $\lambda_{\pm}$ of the non-Hermitian stability matrix $D$ in Eq. (15) can be complex. For the stability of the homogeneous state, real parts of $\lambda_{\pm} = \text{tr}(D)/2 \pm \frac{1}{2} \sqrt{\text{tr}(D)^2 - 4\text{det}(D)}$ should be negative. Complex values for $\lambda_{\pm}$ imply that a slightly perturbed mixed state develops an oscillatory instability and the system generally evolves into a steady-state that carries signatures of these oscillations like the travelling wave or those summarized in Fig. 1. The determinant and the trace of the $D$, given as

$$\text{det}(D) = \Gamma^2 q^4 \left[ (1 - 2|\phi_0|^2)^2 + (\alpha_0 - 2\alpha_1|\phi_0|^2) \right],$$

$$\text{tr}(D) = \Gamma q^2 \left[ 2 - 4|\phi_0|^2 \right],$$

are functions of $|\phi_0|$ only. Since $\lambda_{\pm}$ depend on the trace and determinant of $D$, the stability is thus determined by $|\phi_0|$ alone. For complex values of $\lambda_{\pm}$, the real and imaginary parts are

$$\text{Re}(\lambda) = \Gamma q^2 \left[ 1 - 2|\phi_0|^2 \right],$$

$$\text{Im}(\lambda) = \Gamma q^2 \left[ (\alpha_0 - 2\alpha_1|\phi_0|^2)^2 - |\phi_0|^4(1 + \alpha_1^2) \right].$$

(17)

For $|\phi_0| = 0$, we observe that $\text{Re}(\lambda) > 0$ and $\text{Im}(\lambda) \neq 0$ independently of $\alpha_{0,1}$. Therefore, in the middle of the unstable region the homogeneous state develops oscillatory instabilities in response to small perturbations. The real and imaginary parts of the eigenvalues are plotted as functions of $|\phi_0|$. Two types of bifurcation points arise as $|\phi_0|$ is changed while keeping other parameters constant (as shown in Fig. 6): Hopf bifurcation where the real parts of a pair of complex eigenvalues change sign, and exceptional point where the two $\lambda_\pm$ coalesce while the corresponding eigenvectors are parallel. Upon crossing an exceptional point, the eigenvalues develop imaginary parts. The nonlinearity of the non-reciprocal parameter being considered enhances the richness of the stability diagram. For $|\phi_0| < 1/\sqrt{2}$, two exceptional points appear at the following values of $|\phi_0|$:

$$|\phi_0|^2 = \frac{2\alpha_0\alpha_1}{3\alpha_1^2 - 1} \pm \frac{\alpha_0\sqrt{\alpha_1^2 + 1}}{3\alpha_1^2 - 1}. \quad (19)$$

A third possibility occurs for $|\phi_0| = 1/\sqrt{2}$ and $\alpha_0 = \alpha_1 \pm \frac{1}{2} \sqrt{1 + \alpha_1^2}$ where $\text{Re}(\lambda) = \text{Im}(\lambda) = 0$. Finally, a pair of real eigenvalues could both change sign signalling an instability where perturbations grow and lead to the formation of a bulk separated state.

The results of the stability analysis are verified numerically by running $81 \times 81$ simulations keeping $\alpha_{0,1}$ fixed and by varying $\phi_{1,2}$ (see Fig. 7). For $\alpha_0 = 5$ and $\alpha_1 = 2$, the states develop an oscillatory instability where perturbations grow and lead to the formation of a bulk separated state. To identify the oscillatory steady state we calculate the area enclosed in $(\phi_1, \phi_2)$ space by the
boundary enclosing the trajectory \((\phi_1(r, t), \phi_2(r, t))\) at a constant \(r\), namely, \(\sigma(r) = \frac{1}{2} \int \phi_1 d\phi_2 - \phi_1 d\phi_2\). We observe that \(\delta \phi_1 > 0\) where \(\text{Re}(\lambda_k) > 0\), while we obtain nonzero values of \(\sigma\) where \(\text{Im}(\lambda_k) > 0\), indicating oscillatory steady-states.

Moving away radially from the centre |\(\phi_0| = 0\) in the composition space \((\phi_1, \phi_2)\), the currents driving the phase separation appear to dominate over the non-reciprocal interactions. For \(\alpha_0 = \alpha_1 = 4\), the effervescent waves change into a predominantly phase separated state with domains spanning the system size and with fluctuating interfaces (see Supplemental Movies S3 and S4). For \(\alpha_0 = 5\) and \(\alpha_1 = 2\), the effervescent waves persist until the very edge of the region beyond which the homogeneous state is stable.

VII. CONCLUDING REMARKS

We have introduced a model with nonlinear non-reciprocal interactions between two species, and studied the phase separation dynamics of the system and its dynamical steady states. We have observed a new type of chemical spatiotemporal chaos that arises due to imperfect breaking of PT symmetry, involving fluctuating domains in space where the symmetry is temporarily restored. This effect produces the startling phenomena of effervescence and effervescent traveling waves. We also observe that our model exhibits fluctuations that can act as a background effective white noise due to the non-reciprocal nonlinearities, similarly to the case of Kuramoto-Sivashinsky equation.

The non-reciprocal coupling \(\alpha\) and the free energy \(f\) were chosen such that we can obtain an analytical form for the travelling waves first reported in [15, 16], thereby establishing the stability of the waves. The emergence of spatiotemporal chaos is attributed to the non-reciprocal coupling that changes sign as a function of the amplitude of scalar fields. A linear stability analysis enables us to highlight the interplay between the two non-reciprocal coefficients, which destabilizes the traveling waves. We have verified that the results presented here hold quite generally, independently of the choice of the bulk free energy \(f\), and also for two alternative forms of \(\alpha\).

The effect of non-reciprocal interactions in the presence of number conservation constraint, which is characteristic of many active matter systems, leads to the emergence of novel dynamical states. We have also highlighted the role of composition in tuning the pattern-forming behaviour of the system, which enhances the connection of the model to bulk phase separating systems.

Our work sheds light onto the rich and complex behaviour that can arise in minimal models of active matter system with non-reciprocal interactions. We hope that our work will pave the way for new studies of the role of non-reciprocity in colloidal systems with tunable interactions [6] or in the field of swarm robotics [41].

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Appendix A: Linear Stability Analysis

To investigate the stability of the plane wave solution (6) to the nonlinear NRCH equation (5), we insert the form (9) into Eq. (5), and expand the equation up to the first order in \(\delta \rho_q\) in Fourier space, taking into account the definitions given in Eq. (8). We obtain

\[
\begin{bmatrix}
\delta \rho_q(k)
\end{bmatrix}
=
\begin{bmatrix}
M_{11} & M_{12} \\
M_{21} & M_{22}
\end{bmatrix}
\begin{bmatrix}
\delta \rho_q(k)
\end{bmatrix},
\]

where

\[
M_{11}(q, k) = i \Gamma q^2 \left[ -\alpha_0 + \alpha_1 \left( 1 - \frac{q^2}{q_0^2} \right) \right] - \Gamma K (k - q)^4
+ \Gamma \left[ (1 + i \alpha_0) - 2(1 + i \alpha_1) \left( 1 - \frac{q^2}{q_0^2} \right) \right] (k - q)^2,
\]

\[
M_{12}(q, k) = -\Gamma (1 + i \alpha_1) \left( 1 - \frac{q^2}{q_0^2} \right) (k - q)^2,
\]

\[
M_{21}(q, k) = -\Gamma (1 - i \alpha_1) \left( 1 - \frac{q^2}{q_0^2} \right) (k + q)^2,
\]

\[
M_{22}(q, k) = -i \Gamma q^2 \left[ -\alpha_0 + \alpha_1 \left( 1 - \frac{q^2}{q_0^2} \right) \right] - \Gamma K (k + q)^4
+ \Gamma \left[ (1 - i \alpha_0) - 2(1 - i \alpha_1) \left( 1 - \frac{q^2}{q_0^2} \right) \right] (k + q)^2.
\]

The stability of the plane wave solution (6) is determined by the eigenvalues of the \(M\), which are given as \(\lambda_\pm \equiv \text{tr} M/2 \mp \frac{1}{2} \sqrt{\text{tr} M^2 - 4 \text{det} M}\). To the zeroth order in \(k\), the eigenvalues are \(\lambda_+ = -2q^2 \left( 1 - \frac{q^2}{q_0^2} \right) \) and \(\lambda_- = 0\). For small wavelength perturbations \(k \ll q\), the branch of eigenvalues \(\lambda_-\) remains negative, and thus stabilizing, and the stability of the travelling waves is determined by \(\lambda_+\) alone. We calculate \(\lambda_+\) up to quadratic order in \(k^2\) to probe the advection and diffusion effects. To \(O(k^2)\), we obtain the results presented in Eqs. (10) and (11).

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