

Nonreciprocal Cahn-Hilliard Model Emerges as a Universal Amplitude Equation

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Oscillatory behavior is ubiquitous in out-of-equilibrium systems showing spatiotemporal pattern formation. Starting from a linear large-scale oscillatory instability—a conserved-Hopf instability—that naturally occurs in many active systems with two conservation laws, we derive a corresponding amplitude equation. It belongs to a hierarchy of such universal equations for the eight types of instabilities in homogeneous isotropic systems resulting from the combination of three features: large-scale vs small-scale instability, stationary vs oscillatory instability, and instability without and with conservation law(s). The derived universal equation generalizes a phenomenological model of considerable recent interest, namely, the nonreciprocal Cahn-Hilliard model, and may be of a similar relevance for the classification of pattern forming systems as the complex Ginzburg-Landau equation.

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The concept of active systems emerged as a paradigm in the description of a wide variety of biophysicochemical nonequilibrium phenomena on multiple scales ranging from the collective behavior of molecules within biological cells to the dynamics of tissues or human crowds [1]. In a narrow interpretation, active matter always involves chemomechanical coupling and shows some kind of self-sustained (collective) motion of the microscopic agents [2–5]. In a wider sense, active systems encompass open systems that are kept out of equilibrium by a throughflow of material or energy [6], and therefore may develop self-organized spatiotemporal patterns. This then includes the large spectrum of systems described by reaction-diffusion models [7–9] and systems characterized by the interplay of phase separation and chemical reactions [10].

In this context, predator-prey-type nonreciprocal interactions between constituents of active matter have recently become a particular focus as the implied breaking of Newton’s third law results in a rich spectrum of nascent self-excited dynamic behavior [11–15]. Besides various (stochastic) agent-based models of Langevin-type, continuous deterministic field theories have also been proposed [5], most notably, in the form of nonreciprocal Cahn-Hilliard models [16–18]. The latter add nonreciprocal interactions to classical Cahn-Hilliard models [19] (model B in [20]) that describe the dynamics of phase separation, e.g., in binary or ternary mixtures [21,22]. In particular, the resulting nonreciprocal Cahn-Hilliard models represent two conservation laws with nonvariational coupling. It is shown that this coupling may result in traveling and oscillating states [16–18], arrest or suppression of coarsening [18], formation of small-scale spatial (Turing) patterns as well as stationary, traveling and oscillatory localized states [23]—all features that are forbidden in standard reciprocal Cahn-Hilliard models.

However, these nonreciprocal Cahn-Hilliard models are introduced on phenomenological grounds by symmetry considerations, but no derivation of the field theory from a microscopic description or other deeper justification has been provided yet. Here, we show that the model indeed merits extensive study as it actually represents one of the universal equations of pattern formation. One may even argue that it corresponds to a “missing amplitude equation” for the basic eight types of linear instabilities in spatially extended isotropic homogeneous systems that can be described by scalar fields. An amplitude (or envelope) equation describes the universal bifurcation behavior characterizing the spatiotemporal dynamics in the vicinity of the threshold of a single instability or of several simultaneous instabilities, and can be systematically derived in a weakly nonlinear approach [24]. The mentioned eight instability types result from the combination of three features: (i) large-scale vs small-scale instability, (ii) stationary vs oscillatory instability, and (iii) instability without and with conservation law(s). The spatial and temporal character of an instability encoded in features (i) and (ii) is well captured in the classification of instabilities by Cross and Hohenberg [25], and the four corresponding amplitude equations for systems without conservation law are very well studied. One example is the complex Ginzburg-Landau equation [26] valid near the onset of a large-scale oscillatory (also known as Hopf or type III₀ [25]) instability. An overview of the basic eight instability types in our amended classification, their dispersion relations, and seven existing amplitude equations, is provided in Sec. 1 of the Supplemental Material.

However, the consequences of conservation laws in the full range of pattern-forming systems are less well studied: Small-scale stationary and oscillatory cases with a conservation law are considered in [27] and [28], respectively,

with applications to pattern formation in the actin cortex of motile cells [29,30], in crystallization [31], and in magnetoconvection [32]. However, only recently it was shown that the standard single-species Cahn-Hilliard equation does not only describe phase separation in a binary mixture [19,33] but furthermore can be derived as an amplitude equation valid in the vicinity of a large-scale stationary instability in a system with a single conservation law [34]. In consequence, close to onset, a reaction-diffusion system with one conservation law as, e.g., discussed in [29,35–43], can be quantitatively mapped onto a Cahn-Hilliard equation. Similarly, the equation captures core features of certain collective behavior in chemotactic systems [44] and of cell polarization in eukaryotic cells [45].

This leaves only one of the eight cases unaccounted for, namely, the large-scale oscillatory instability with conservation laws, that we call conserved-Hopf instability. In the following, we consider active systems with two conservation laws and show that the general nonreciprocal Cahn-Hilliard model emerges as a corresponding universal amplitude equation. Thereby, all the particular phenomenological models studied in [16–18,23] are recovered as special cases. This also applies to the complex Cahn-Hilliard equation appearing as a mass-conserving limiting case in Ref. [46].

Before we embark on a general derivation of the amplitude equation we emphasize its applicability to the wide spectrum of systems where the conserved-Hopf instability and related intricate nonlinear oscillatory behavior can occur: A prominent example is the spatiotemporal pattern formation of proteins vital for cellular processes. Although chemical reactions cause conformation changes of proteins, their overall number is conserved on the relevant time scale, e.g., MinE and MinD in ATP-driven cellular Min oscillations [41]. Such an instability can also be expected in other reaction-diffusion systems with more than one conservation law, e.g., the full cell polarity model in Ref. [36]. Relevant examples beyond reaction-diffusion systems include oscillations in two-species chemotactic systems [47]; an active poroelastic model for mechanochemical waves in cytoskeleton and cytosol [48]; thin liquid layers covered by self-propelled surfactant particles [49]; oscillatory coupled lipid and protein dynamics in cell membranes [50]; multicomponent phase-separating reactive or surface-active systems [51,52]; and two-layer liquid films or drops on a liquid layer with mass transfer [53] or heating [54] where the two interfaces may show intricate spatiotemporal oscillation patterns [53,55,56].

In most cases, the two conserved quantities correspond to concentration fields, film or drop thickness profiles, particle number densities, and the conserved-Hopf instability occurs as a primary instability. However, another class of examples exists where it appears as a secondary instability. For example, in Marangoni convection the interaction between a large-scale deformational and a

small-scale convective instability is described by coupled kinetic equations for the film height and a complex amplitude [57]. There, the liquid layer profile and the phase of the complex amplitude represent the two conserved quantities and the occurring conserved-Hopf instability corresponds to an oscillatory sideband instability.

Systems like the given examples that feature two conservation laws and exist in a sustained out-of-equilibrium setting can become unstable through a conserved-Hopf instability, i.e., the linear marginal mode [growth rate $\Delta(k_c) = 0$] occurs at zero wave number ($k_c = 0$) and zero frequency [$\Omega(k_c) = \Omega_c = 0$]. This is determined via a linear stability analysis of the trivial uniform steady state yielding the dispersion relations $\lambda_{\pm}(k)$ of the dominant pair of complex conjugate modes where $\Delta = \text{Re}\lambda_{\pm}$ and $\pm\Omega = \text{Im}\lambda_{\pm}$. Although $\lambda_{\pm}(k=0) = 0$ always holds, as the two conservation laws imply the existence of two neutral modes, the conserved-Hopf mode is oscillatory at arbitrarily small wave numbers. In other words, directly beyond instability onset the system undergoes large-scale small-frequency oscillations, i.e., the conservation laws imply that the first excited mode has the smallest wave number compatible with the domain boundaries and oscillates on a correspondingly large time scale as $\Omega \rightarrow 0$ for $k \rightarrow 0$. In consequence, the weakly nonlinear behavior is not covered by any of the seven amplitude equations summarized in Sec. 1 of the Supplemental Material.

Dispersion relations below, at, and above the threshold of a conserved-Hopf instability are sketched in Fig. 1 and are at small k given by

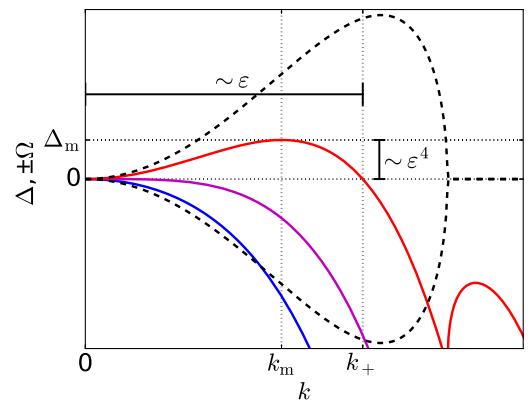


FIG. 1. Linear growth rates $\Delta(k) = \text{Re}\lambda_{\pm}(k)$ in dependence of the wave number k below (solid blue line, $\delta < 0$), at (solid purple line, $\delta = 0$) and above (solid red line, $\delta > 0$) the threshold of a conserved-Hopf instability as described by the dispersion relation $\lambda_{\pm}(k)$ given by the series expansion Eq. (1). The black dashed lines give the frequencies $\pm\Omega(k) = \text{Im}\lambda_{\pm}(k)$ that are identical in all three cases. Labeled thin dotted lines and solid bars indicate typical quantities and scalings above onset as described in the main text.

$$\begin{aligned}
 \lambda_{\pm}(k) &= \Delta(k) \pm i\Omega(k) \\
 \text{with } \Delta(k) &= \delta k^2 - \tilde{\delta} k^4 + \mathcal{O}(k^6) \\
 \text{and } \Omega(k) &= \omega k^2 + \tilde{\omega} k^4 + \mathcal{O}(k^6). \quad (1)
 \end{aligned}$$

The onset occurs when δ becomes positive while $\tilde{\delta} > 0$. Above onset, Eq. (1) indicates modes within a band of wave numbers $0 < k < k_+ = \sqrt{\delta/\tilde{\delta}}$ that exponentially grow. The fastest mode is at $k_m = k_+/\sqrt{2}$ and has the growth rate $\Delta_m = \delta^2/(4\tilde{\delta})$.

To determine an amplitude equation that captures the bifurcation structure characterizing the spatiotemporal pattern formation in the vicinity of the onset of a conserved-Hopf bifurcation with a dispersion relation as depicted in Fig. 1, we apply a weakly nonlinear approach [24]. First, we introduce a smallness parameter ε with $|\varepsilon| \ll 1$ and consider the system close to onset where $\delta = \delta_2 \varepsilon^2$. From hereon subscript numerals indicate the order in ε of the corresponding term. Then, the width of the band of growing wave numbers and the maximal growth rate scale as ε and ε^4 , respectively. This determines the additional large spatial scale $\vec{X} = \varepsilon \vec{x}$ and slow timescale $T = \varepsilon^4 t$ relevant for the dynamics. Additionally, Eq. (1) indicates that the leading order oscillation frequency scales like $\Omega \approx \omega k^2 \sim \varepsilon^2$. This implies that a second slow timescale $\tau = \varepsilon^2 t$ has to be taken into account.

Specifically, we now consider a general homogeneous isotropic multicomponent system with two conservation laws,

$$\begin{aligned}
 \partial_t \rho &= -\vec{\nabla} \cdot \left(Q(\mathbf{u}) \vec{\nabla} \eta(\mathbf{u}, \vec{\nabla}) \right) \\
 \partial_t \sigma &= -\vec{\nabla} \cdot \left(R(\mathbf{u}) \vec{\nabla} \mu(\mathbf{u}, \vec{\nabla}) \right) \\
 \partial_t \mathbf{n} &= \mathbf{F}(\mathbf{u}, \vec{\nabla}), \quad (2)
 \end{aligned}$$

i.e., coupled kinetic equations for two conserved (ρ and σ) and N nonconserved [$\mathbf{n} = (n_1, \dots, n_N)$] scalar field variables. Note that Eq. (2) can represent any of the examples mentioned above. From here onwards, $\mathbf{u} = (\rho, \sigma, \mathbf{n})$ is used as an abbreviation where convenient. The dynamics of the two conserved quantities is given by the divergence of corresponding fluxes that consist of the product of a mobility (Q or R) and the gradient of a nonequilibrium (chemical) potential (η or μ) that, in general, still depends on spatial derivatives $\vec{\nabla}$. The dynamics of the nonconserved quantities is given by the vector \mathbf{F} of general functions of fields and their spatial derivatives. In the simplest case, the system may represent a reaction-diffusion system with $N + 2$ species that has been rearranged (similar to [42]) to explicitly show the two conservation laws [58]. More complicated examples include multifield thin-film descriptions where two components are conserved [56] and

multispecies membrane models showing phase separation and chemical reactions [50]. For an active system the potentials cannot be obtained as variational derivatives of a single underlying energy functional. Here, we sketch the derivation of an amplitude equation for the conserved-Hopf instability (Fig. 1) of a homogeneous steady state of a general system (2) while Sec. 2 of the Supplemental Material presents the complete algebra.

To perform the weakly nonlinear analysis valid in the vicinity of instability onset, we expand all fields in ε , i.e., $\mathbf{u}(\vec{X}, \tau, T) = \mathbf{u}_0 + \varepsilon \mathbf{u}_1(\vec{X}, \tau, T) + \varepsilon^2 \mathbf{u}_2(\vec{X}, \tau, T) + \dots$, where \mathbf{u}_0 is the steady uniform state with $\mathbf{F}(\mathbf{u}_0) = 0$ and $\mathbf{u}_i(\vec{X}, \tau, T)$, $i = 1, 2, \dots$ are the deviations that describe the (weakly) nonlinear behavior. We take the above discussed scaling of space and time implied by the dispersion relation into account by writing $\vec{\nabla}_{\vec{x}} = \varepsilon \vec{\nabla}_{\vec{X}}$ and $\partial_t = \varepsilon^2 \partial_{\tau} + \varepsilon^4 \partial_T$, respectively. With this we then consider Eqs. (2) order by order. The scaling implies that we need to successively consider all orders up to $\mathcal{O}(\varepsilon^5)$ to discover evolution equations that capture dynamic effects on the slow timescale T .

In principle, at each order we first determine the non-conserved fields as (nonlinear) functions of the conserved fields, reflecting that the dynamics of the former is slaved to the latter. Second, we obtain the continuity equations to the corresponding order by inserting the obtained expressions into the appropriate mobilities and potentials similar to Taylor-expanding them. In particular, at order ε , the contributions of the two continuity equations vanish and the remaining N equations become a homogeneous linear algebraic system for the slaved quantities, solved by $\mathbf{n}_1(\vec{X}, \tau, T) = \mathbf{n}_{\rho} \rho_1(\vec{X}, \tau, T) + \mathbf{n}_{\sigma} \sigma_1(\vec{X}, \tau, T)$ where \mathbf{n}_{ρ} and \mathbf{n}_{σ} correspond to the zero eigenmodes $(1, 0, \mathbf{n}_{\rho})$ and $(0, 1, \mathbf{n}_{\sigma})$ of the dominant eigenspace at $k = 0$ [59].

At order ε^2 , again the continuity equations are again trivially fulfilled, and the remaining equations form an inhomogeneous linear algebraic system for \mathbf{n}_2 . Thereby, the inhomogeneity is nonlinear in lower order quantities. At order ε^3 , the first nonvanishing contributions from the continuity equations appear, that, after eliminating \mathbf{n}_1 , correspond to linear equations in $\vec{\nabla}^2 \rho_1$ and $\vec{\nabla}^2 \sigma_1$. They provide the conditions for the instability onset at $\delta = 0$ in Eq. (1). They also capture the leading order oscillations with frequency ω on the time scale τ by an antisymmetric dynamic coupling that represents a nonreciprocal coupling of lowest order (a structure equivalent to the Schrödinger equation for a free particle). Also for \mathbf{n}_3 an inhomogeneous linear algebraic system emerges. At the subsequent order ε^4 , further contributions to the evolution on the time scale τ are obtained from the continuity equations. Finally, at order ε^5 we obtain expressions for $\partial_T \rho_1$ and $\partial_T \sigma_1$. Using the earlier obtained results for \mathbf{n}_1 , \mathbf{n}_2 , and \mathbf{n}_3 , the complete continuity equations at this order can be written as nonlinear functions of the ρ_i and σ_i . This provides the weakly

nonlinear expression for the leading order time evolution on the timescale T . Next, the expressions found at the different orders are recombined, in passing “inverting” the scalings and expansions of time, coordinates, and fields ρ and σ . The resulting amplitude equation corresponds to a generalized nonreciprocal Cahn-Hilliard model (i.e., two nonreciprocally coupled Cahn-Hilliard equations) and is given in Sec. 2 of the Supplemental Material. In the common case of constant mobilities ($Q = Q_0$ and $R = R_0$) in Eq. (2), cross-couplings in the highest-order derivatives may be removed by a principal axis transformation, resulting in

$$\begin{aligned}\partial_t A &= \vec{\nabla}^2 \left[\alpha_1 A + \alpha_2 B + N_A(A, B) - D_A \vec{\nabla}^2 A \right] \\ \partial_t B &= \vec{\nabla}^2 \left[\beta_1 A + \beta_2 B + N_B(A, B) - D_B \vec{\nabla}^2 B \right].\end{aligned}\quad (3)$$

Here, the spatially slowly varying real amplitudes A and B are linear combinations of the deviations of the conserved fields from their mean values, D_A and D_B are effective interface rigidities, and N_A and N_B are general cubic polynomials in A and B , e.g., $N_A = \alpha_3 A^2 + \alpha_4 AB + \alpha_5 B^2 + \alpha_6 A^3 + \alpha_7 A^2 B + \alpha_8 AB^2 + \alpha_9 B^3$. All parameters are real [60].

The derived general nonreciprocal two-component Cahn-Hilliard model describes the universal bifurcation behavior in the vicinity of any conserved-Hopf instability independently of the particular system studied—all such systems and most of their parameters at instability onset are encoded in the rich parameter set of the derived equations. It should further be noted that the derived general model encompasses further primary bifurcations as it actually corresponds to the amplitude equation for an instability of higher codimension. This is shown in Sec. 3 of the Supplemental Material employing the example of a Cahn-Hilliard instability of codimension two. In other words, the derived amplitude equation may be considered as belonging to a higher level of a hierarchy of such equations. It captures several qualitatively different linear instability scenarios. Such hierarchies are useful to understand the qualitative differences and transitions between instability types. Amplitude equations on a higher hierarchy level describe the bifurcation behavior close to higher codimension points, i.e., the behavior in the vicinity of several different instabilities. In the limiting case where only one of the contained instabilities is close to its onset, the higher level equation can often be reduced to a simpler lower level equation [61]. However, such a further reduction of the derived nonreciprocal Cahn-Hilliard equation remains a task for the future.

Note that the presence of additional subdominant neutral modes (e.g., resulting from additional conservation laws) or the simultaneous onset of several distinct instabilities would (possibly in extension of the present work) also

result in amplitude equations on a higher level of the “codimension hierarchy” [30,62].

It is an interesting observation that the various *ad hoc* nonreciprocal Cahn-Hilliard models studied in [16–18] emerge as special cases of the equation derived here [63]. Table 3 in Sec. 2 of the Supplemental Material provides the corresponding parameter choices in Eq. (3). Two other limiting cases are also included: (i) If certain symmetries between coefficients hold, one may introduce a complex amplitude $C = A + iB$ and present Eq. (3) as a complex Cahn-Hilliard equation $\partial_t C = -G \vec{\nabla}^2 \left[\varepsilon + (1 + ib) \vec{\nabla}^2 - (1 + ic) |C|^2 \right] C$, i.e., as a complex Ginzburg-Landau equation with an additional outer Laplace operator reflecting the conservation property, as briefly considered in Ref. [46]. This, in passing clarifies that Eq. (3) is more than just a “conserved complex Ginzburg-Landau equation” because it does not show its phase-shift invariance. (ii) Imposing another symmetry between coefficients renders the coupled equations variational. Then they represent a generic model for the dynamics of phase separation in a ternary system [22,71].

To conclude, we have derived an amplitude equation valid in the vicinity of a conserved-Hopf bifurcation and as well at related bifurcations of higher codimension. It qualitatively captures transitions generically occurring in the wide variety of out-of-equilibrium systems that feature two conservation laws. Note that close to the conserved-Hopf instability it also provides a rather good quantitative description of the bifurcation structure. This is exemplified in Sec. 4 of the Supplemental Material [65] where the amplitude equation is derived and analyzed in comparison with the full system for the relatively simple case of a three-component reaction-diffusion system with two conservation laws. As the latter reduce the local phase space (defined as in Ref. [42]) to one dimension, the emerging behavior will be much less complex than seen in the Min system [41] and other high dimensional cases [49].

The derived equation forms part of the hierarchy of universal amplitude equations for the above discussed eight basic instabilities. Thus, its relevance for the classification of pattern forming behavior close to the onset of instabilities resembles that of the complex Ginzburg-Landau equation that describes the universal bifurcation behavior in the vicinity of a standard Hopf instability in systems without conservation laws [24–26,72]. However, one has to add restrictively that the large number of parameters of the derived generic model might limit its practical use as a complete parametric study of all generic behaviors is prohibitively costly. Still its study has already started to form a valuable bridge between the analysis of the many specific models and the set of amplitude equations on a lower hierarchy level (that still needs completion). In cases where the primary bifurcation is subcritical (e.g., for the Min oscillations [41]), even higher order amplitude equations might be insufficient to faithfully predict the

spatiotemporal behavior. Then weakly and fully nonlinear approaches should be employed in a complementary manner.

Although it is known that the conserved-Hopf instability is related to phenomena that are not covered by the complex Ginzburg-Landau equation [56] only very few studies have considered its (weakly) nonlinear behavior by corresponding amplitude equations, normally, in special cases [57,73]. On the one hand, Ref. [57] restricts its focus to amplitude equations for spatially periodic traveling and standing waves, and on the other hand, Ref. [73] deals with a particular case without reflection symmetry where one of the two conservation laws is weakly broken. The universal character of the model derived here, implies that literature results on the onset of motion and oscillations [16–18] and as well potentially on the suppression of coarsening and the existence of localized states [18,23] may be applied to the class of out-of-equilibrium systems that undergo a conserved-Hopf instability. In consequence, spatiotemporal patterns occurring in a wide spectrum of systems from protein dynamics within cells and on membranes [41,50], chemotactic systems of organisms [47], coupled cytoskeleton and cytosol dynamics [48], multicomponent phase-separating reactive, surface-active or active systems [49,51,52], to two-layer liquid films with heating or mass transfer [53–55] should be further studied to identify their common universal features as out-of-equilibrium systems with conservation laws as well as characterizing differences that may prompt a further development of the hierarchy of amplitude equations.

Note that the present work has entirely focused on isotropic homogeneous systems described by scalar fields, implying that systems like the active Ising model in [74,75] are not covered as they involve a pseudoscalar. The dispersion relations of such systems with conservation laws have properties different from the ones considered here. It would be highly interesting to produce a systematics similar to the one proposed here for systems involving pseudoscalars. To our knowledge, so far only a few cases have been treated by weakly nonlinear theory.

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