

Journal of Physics A: Mathematical and Theoretical



PAPER

OPEN ACCESS

RECEIVED

30 March 2026

REVISED

2 June 2026

ACCEPTED FOR PUBLICATION

8 July 2026

PUBLISHED

30 July 2026

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Nonreciprocal Model B: the role of mobilities and nonreciprocal interfacial forces

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Keywords: phase separation, non-reciprocal interaction, density dependent mobility, travelling states

Supplementary material for this article is available [online](#)

Abstract

We study a non-reciprocal version of Model B, as the continuum theory for non-reciprocal particle mixtures. In contrast to non-reciprocal Cahn–Hilliard models, it is important in this context to consider the dependence of mobility coefficients on the local concentrations. We show that a homogeneous state that is linearly stable for one form of the mobility can be unstable for a different form of mobility, an effect that would be impossible in equilibrium and implies a crucial role for mobilities in non-reciprocal mixtures. For unstable homogeneous states we study the spinodal dynamics governing the onset of phase separation. We find, again in contrast to non-reciprocal Cahn–Hilliard models, that exceptional point transitions between static and oscillatory instabilities are generically avoided by transitions where the spinodal lengthscale changes discontinuously. At these transitions we find intricate spinodal dynamics with two competing lengthscales, one governing a static instability and the other an oscillatory instability, *i.e.* one that generates travelling waves. We demonstrate that, depending on the interaction strengths, more complex transitions can occur in the spinodal dynamics, including coexistence of three lengthscales and lines of discontinuous transitions, terminated by critical points, between distinct static instabilities. Finally, we explore the effects of additional non-reciprocity in the interfacial chemical potentials, which would generically be expected when obtaining Model B by coarse graining from a non-reciprocal particle model. We show that interfacial non-reciprocity can increase the region in the spinodal phase diagram where oscillatory instabilities occur, but only up to a certain boundary that we establish analytically and demonstrate numerically.

1. Introduction

Phase separation is a ubiquitous phenomenon in nature and has been an intense topic of investigation for decades because of its broad relevance. This includes alloys [1] and liquid mixtures [2] in physical systems, but more recently also social phenomena like flocking [3] as well as phase separation in biological systems [4], in the latter context ranging from the formation of biomolecular condensates [5] to genome organization [6], immune response [7, 8], neurodegenerative diseases [9], SARS-Cov2 infections [10] and cancer [11]. When a homogeneous system is brought into an unstable region of its phase diagram, *e.g.* by a quench, *i.e.* a rapid reduction in temperature, fluctuations will start to grow exponentially in a process known as spinodal decomposition, leading eventually to phase separation into two or more phases. At early times in this process, fluctuations grow predominantly at a characteristic length scale. The resulting spinodal patterns initiate the formation of domains of new phases [12] that then grow by Ostwald ripening.

The above processes are well studied for phase separating systems in an equilibrium setting. However, many real-life systems are out of equilibrium, *e.g.* because the interactions are mediated by a non-thermal environment. The resulting effective interactions can be *non-reciprocal* in the sense that they violate the action-reaction symmetry, *i.e.* Newton's third law for interparticle forces [13, 14]. Non-reciprocity is ubiquitous in the natural world in activator-inhibitor reactions, predator-prey dynamics, collective animal behaviour and has been studied in systems such as active colloids [15] and active-passive mixtures [16], as well as by extensive theoretical analysis [16–23].

Recently, the role of non-reciprocal interaction in mixtures of two or more phase separating species has been widely studied in the setting of the Cahn–Hilliard model [24–28]. Non-reciprocity has been shown there to have interesting effects on the dynamics of phase separation, leading *e.g.* to travelling states [29], suppression of coarsening [30], stabilization of mixtures against spinodal decomposition [31] and the emergence of small scale (Turing-like) instabilities [32]. For our purposes it should be noted here that the Cahn–Hilliard model is typically written in a form inspired by magnetic systems. Its non-reciprocal analogue is therefore mostly written with a quartic free energy, while the mobility is taken to be constant [19, 24, 28–33]. Based on experience with reciprocal systems, this choice may seem harmless. One of the key advances of our study will be to show that this is far from the case in non-reciprocal systems: using standard density-dependent mobilities for particle mixtures can alter predictions for spinodal stability and lead to the emergence of qualitatively new features in the spinodal phase diagram, including discontinuous (rather than exceptional point) transitions between static and oscillatory instabilities.

Specifically, we study here the spinodal phase separation dynamics of a non-reciprocal Model B, where the free energy considered has a Flory–Huggins-like form that arises naturally by coarse graining a lattice model for particle mixtures with a solvent. This setting, recently investigated in the context of pattern formation and wetting [34], has no analogue of the Ising-like symmetry under global order parameter sign change that is implicit in the non-reciprocal Cahn–Hilliard model. More importantly, the mobility in Model B naturally acquires a dependence on the local densities of the different particle species, and we will demonstrate that this plays a key role in the phase separation dynamics once non-reciprocity is present.

Studies of non-reciprocal Cahn–Hilliard models [19, 24, 28–33] generally ignore the mobility, taking it as a constant factor. In Model B (and related models, *e.g.* in sociohydrodynamics [35]), however, the mobility generally has to be dependent on local mixture component concentrations [36, 37]. For reciprocal (equilibrium) systems, this mobility only controls the rate at which equilibrium is reached; it does not affect the equilibrium behaviour or the stability of the homogeneous mixture to fluctuations. In the non-equilibrium situation generated by non-reciprocity, we will see that the form of the mobility plays an important role: it can modify not only the nature of the instability of a homogeneous state, but indeed determine whether such a state is unstable at all, thus altering the spinodal phase diagram.

In continuum or field-theoretic descriptions such as non-reciprocal Cahn–Hilliard or non-reciprocal Model B that involve local scalar density variables, but no vectorial order parameters such as local particle orientation, non-reciprocity is encoded in the form of the local chemical potentials. We can then distinguish two kinds of non-reciprocities that can arise. The first of these is bulk non-reciprocity, which captures the asymmetric influence of one species on the chemical potential of another in a bulk phase. This is typically introduced [17–19, 24, 28–33] via a difference between corresponding off-diagonal elements of the interaction matrix, and quantified by a parameter α . With this asymmetry the chemical potentials cannot be derived by taking derivatives of a free energy, thus already implying that the system is inherently out of equilibrium. In addition to bulk non-reciprocity one can then have non-reciprocity in the interfacial contributions to the chemical potentials, which depend on second (or higher) derivatives of local concentrations. This feature is typically (but see [33]) neglected in non-reciprocal continuum models. However, we argue below that interfacial non-reciprocity does arise quite generically when deriving Model B by coarse graining. The resulting contributions to the chemical potentials can, like the mobility, have significant effects on non-reciprocal phase separation dynamics as we will see. This is the second key finding of our study.

We next give an overview of the structure of the paper. In section 2 we define the dynamics of non-reciprocal Model B dynamics and define more explicitly the two kinds of non-reciprocity mentioned above. We also provide details of the linear stability analysis on which most of our results are based. These are discussed in section 3. Focussing on binary mixtures, we demonstrate first that the form of the density-dependent mobility matrix affects the spinodal stability of homogeneous states. When such states are unstable, the mobility also affects the nature of this instability (static vs oscillatory) and hence the qualitative features of the early-time phase separation dynamics (static spinodal patterns vs travelling spinodal waves). We determine the spinodal phase diagrams capturing the dynamics across a range of system compositions and find that in large regions of these, the eigenvalues determining the growth

rate of unstable modes with wavevector q change from real to complex or vice versa as q increases. As a consequence, while a conventional stability analysis focusing on the behaviour for $q \rightarrow 0$ would predict exceptional point transitions between regions of static and travelling spinodal patterns [17], we find that these are generically *avoided* by discontinuous transitions. At these transitions, static and travelling modes with different q compete so that the spinodal patterns consist of a superposition of static and travelling contributions, with distinct associated length scales. For certain interaction parameters we also find more complex scenarios including competition between three modes that are growing equally rapidly. At this dynamical triple-mode point multiple, discontinuous transition lines meet, which we characterise along with the associated critical points. We also demonstrate the emergence of small scale, Turing-like instabilities, which arise for unequal intraspecies interaction strengths and have also been seen in the non-reciprocal Cahn–Hilliard model [30]. Finally, we demonstrate that interfacial non-reciprocity can have similar effects to the mobility by changing the nature of spinodal instabilities, even if the mobility itself is taken as a trivial constant. We explore the associated spinodal phase diagrams and show that the region where travelling spinodal waves occur is in general enlarged by interfacial non-reciprocity, up to a bound that we characterise analytically. We conclude with a discussion and outlook in section 4.

2. Setup of model

2.1. Model definition

The time evolution of the density fields ϕ_i of a phase separating mixture of M components can be mathematically described by Model B [38],

$$\frac{\partial}{\partial t} \phi_i = -\nabla \cdot \mathbf{j}_i \quad (1)$$

$$\mathbf{j}_i = -\sum_{j=1}^M L_{ij} \nabla \mu_j. \quad (2)$$

Here $\mathbf{j}_i$ is the particle current of species i while the L_{ij} are the entries of a symmetric, positive definite mobility matrix that is in general dependent on all densities $\{\phi_i\}$. For equilibrium systems, the chemical potential μ_j of species j is obtained as the functional derivative of the free energy

$$\mu_i = \frac{\delta F[\{\phi_i\}]}{\delta \phi_i}. \quad (3)$$

We will consider specifically a free energy describing the interactions between the particle species $i = 1, \dots, M$, treated at second virial level, while the solvent is assumed to interact only via volume exclusion. This free energy can also be obtained via a Flory–Huggins coarse-graining from a lattice model for particle mixtures [39] and reads

$$F[\{\phi_i\}] = \int d\mathbf{r} \left(T \sum_{i=0}^M \phi_i \log \phi_i + \frac{1}{2} \sum_{i,j=1}^M \epsilon_{ij} \phi_i \phi_j + \frac{1}{2} \sum_{i,j=1}^M K_{ij} \nabla \phi_i \nabla \phi_j \right). \quad (4)$$

Here T is the temperature (we have set $k_B = 1$), while ϵ_{ij} and K_{ij} are coefficients that control the bulk and surface interaction between species i and j . The first term in equation (4) accounts for the entropy of mixing and the excluded volume interaction with the solvent. Here the solvent density is $\phi_0 = 1 - \sum_{i=1}^M \phi_i$ so that the system including the solvent is incompressible. The second term in equation (4) is the bulk free energy of intra- and inter-species interactions, while the last term is the interfacial free energy accounting for the cost of maintaining density gradients as they occur, e.g. at an interface.

From equation (3), the chemical potentials are

$$\mu_i = T \log \phi_i - T \log \phi_0 + \mu_i^{\text{bulk}} + \mu_i^{\text{interf}} \quad (5)$$

$$\mu_i^{\text{bulk}} = \sum_{j=1}^M \epsilon_{ij} \phi_j, \quad \mu_i^{\text{interf}} = -\sum_{j=1}^M K_{ij} \nabla^2 \phi_j \quad (6)$$

where in the second line we have singled out the contributions from the bulk and interfacial interactions. Within the above (equilibrium) formalism, the phenomenon of phase separation is well understood. Starting in an unstable homogeneous state, the system phase separates into a final state consisting

of different phases, thereby minimizing the free energy. Subject to the constraint of particle number conservation this leads to the equality conditions for chemical potentials and pressure and the corresponding common tangent construction [2, 40].

The analysis of phase separation in non-equilibrium mixtures with non-reciprocal interactions between the species has come to the fore relatively recently, but has generated significant research interest [16, 19, 24–33, 41]. Most existing studies of non-reciprocal phase separation consider a Cahn–Hilliard free energy with non-reciprocity only in the bulk interactions terms. The corresponding contributions to the chemical potentials for a binary mixture can then be written as

$$\mu_1^{\text{bulk}} = \epsilon_{11}\phi_1 + (\bar{\epsilon} + \alpha)\phi_2 \quad (7)$$

$$\mu_2^{\text{bulk}} = \epsilon_{22}\phi_2 + (\bar{\epsilon} - \alpha)\phi_1 \quad (8)$$

where α indicates the strength of the bulk non-reciprocity. For $\alpha \neq 0$ the chemical potentials can no longer be derived as derivatives of a free energy, making non-reciprocal systems non-variational in the sense of [30].

As alluded to in the introduction, if one starts from a mixture of particles with non-reciprocal pairwise interaction forces and coarse grains to obtain a continuum theory using Dean’s approach [42], then non-reciprocity appears not only in the bulk free energy but also in the interfacial terms (see appendix E). In a binary mixture the corresponding chemical potential contributions can then be written as

$$\mu_1^{\text{interf}} = -K_{11}\nabla^2\phi_1 - (\bar{K} + \delta)\nabla^2\phi_2 \quad (9)$$

$$\mu_2^{\text{interf}} = -K_{22}\nabla^2\phi_2 - (\bar{K} - \delta)\nabla^2\phi_1 \quad (10)$$

with δ being the strength of interfacial non-reciprocity.

In this paper we will focus on the interplay of the density-dependent mobility in non-reciprocal Model B with bulk non-reciprocity. For this purpose we consider two commonly studied forms of this density dependence [36, 37]

$$L_{ij} = \phi_i\delta_{ij}, \quad L_{ij} = \phi_i\delta_{ij} - \phi_i\phi_j. \quad (11)$$

The diagonal form corresponds to ideal gas limit, whereas the second form with the off diagonal elements is the case for which the system favours interdiffusion between the species. (Interpolating forms between these two extreme forms have also been suggested [36, 37].) We will study specifically the influence of the choice made in equation (11) for the early time phase separation dynamics of non-reciprocal mixtures. After this we will turn to the effects of interfacial non-reciprocity as encoded in equations (9) and (10).

2.2. Linear stability analysis

After a system in a spatially homogeneous state has been brought into an unstable region of its phase diagram, *e.g.* by a quench, phase separation proceeds by spinodal decomposition. In the corresponding time regime, *i.e.* before domains with well-defined interfaces are formed, the dynamics can be described by linearization in the density deviations $\delta\phi_i = \phi_i - \phi_i^0$ from the homogeneous state with ‘parent’ densities ϕ_i^0 . One then finds for the dynamics of the Fourier components $\delta\phi_i(q)$, for a binary system and in vector form,

$$\frac{d}{dt}\delta\phi = -q^2\mathbf{L}\begin{pmatrix} \frac{T}{\phi_1^0} + \frac{T}{\phi_0^0} + \epsilon_{11} + q^2K_{11} & \frac{T}{\phi_0^0} + (\bar{\epsilon} + \alpha) + q^2(\bar{K} + \delta) \\ \frac{T}{\phi_0^0} + (\bar{\epsilon} - \alpha) + q^2(\bar{K} - \delta) & \frac{T}{\phi_2^0} + \frac{T}{\phi_0^0} + \epsilon_{22} + q^2K_{22} \end{pmatrix}\delta\phi \quad (12)$$

$$= -q^2\mathbf{L}(\mathbf{H} + q^2\mathbf{K})\delta\phi = \mathbf{R}\delta\phi \quad (13)$$

where q is the modulus of the Fourier wavevector and $\mathbf{L}$ is the mobility matrix for component densities (ϕ_1^0, ϕ_2^0) . Inside the brackets in equation (13), $\mathbf{H}$ is the matrix containing the bulk interaction coefficients ϵ_{ij} as well as the contribution from the excluded volume interaction, while $\mathbf{K}$ contains the interfacial coefficients. In writing the explicit form (12) we have already inserted the parametrizations from equations (7)–(10).

The nature of the spinodal dynamics is primarily determined by the eigenvalues of the rate matrix $\mathbf{R}$, which are given by

$$\lambda_{1,2} = \frac{\text{tr}\mathbf{R} \pm \sqrt{(\text{tr}\mathbf{R})^2 - 4|\mathbf{R}|}}{2}. \quad (14)$$

in terms of its trace and determinant. All phase diagrams presented below pertain to this spinodal dynamics. Understanding the late-stage dynamics analytically is rather more challenging; we therefore only give an overview of some of the non-trivial phenomenology that is visible in numerical simulations (see section 3.6).

3. Results

3.1. Effect of mobility on global stability

We focus first on the interplay of bulk non-reciprocity (controlled by α) and a density-dependent mobility (11). Beyond overall stability, one question of interest will be the emergence of travelling spinodal waves rather than static spinodal patterns. It has been well documented that such waves can arise from bulk non-reciprocity in Cahn–Hilliard models [24, 28, 29], but as we will see the inclusion of a density-dependent mobility can significantly change the picture.

Considering first the global stability of homogeneous states, which is determined by the sign of the real part of $\lambda_{1/2}$, we show in figure 1 phase diagrams delineating the spinodal phase behaviour for $q \rightarrow 0$, for the two forms of the mobility in equation (11). One clearly observes qualitative difference between the two diagrams. The key observation is that for the mobility $L_{ij} = \phi_i \delta_{ij}$ there is a region of stability, shown in white in figure 1(a), in the regime of dense homogeneous states, *i.e.* large $\phi_1^0 + \phi_2^0$. This region is absent in figure 1(b). The implication is that the kinetics of phase separation, as encoded in the mobility matrix $\mathbf{L}$, can determine whether a non-reciprocal system is unstable or not. This striking effect would be impossible in equilibrium, where stability is determined by properties of the free energy alone, without reference to the kinetics. This can be seen explicitly in our setup by considering the form of the rate matrix,

$$\mathbf{R}(q) = -q^2 \mathbf{L}(\mathbf{H} + q^2 \mathbf{K}). \quad (15)$$

By a similarity transformation, $\mathbf{R}$ has the same eigenvalues as

$$\tilde{\mathbf{R}} = \mathbf{L}^{-1/2} \mathbf{R} \mathbf{L}^{1/2} = -q^2 \mathbf{L}^{1/2} (\mathbf{H} + q^2 \mathbf{K}) \mathbf{L}^{1/2} \quad (16)$$

which for a reciprocal system is a symmetric matrix. The stability criterion that all eigenvalues of $\tilde{\mathbf{R}}$ have to be negative can thus be expressed as $\mathbf{u}^T \tilde{\mathbf{R}} \mathbf{u} < 0$ for any nonzero vector $\mathbf{u}$. Setting $\mathbf{v} = \mathbf{L}^{1/2} \mathbf{u}$ transforms this into $-q^2 \mathbf{v}^T (\mathbf{H} + q^2 \mathbf{K}) \mathbf{v} < 0$ for all nonzero q and $\mathbf{v}$, demonstrating that the stability condition is independent of the mobility $\mathbf{L}$ as claimed.

We now contrast the above general considerations for reciprocal systems to the case of non-reciprocal systems. Taking a specific non-reciprocal example, assume that for some q^2 the bare rate matrix $\mathbf{R}^0 = -q^2 (\mathbf{H} + q^2 \mathbf{K})$ has the form

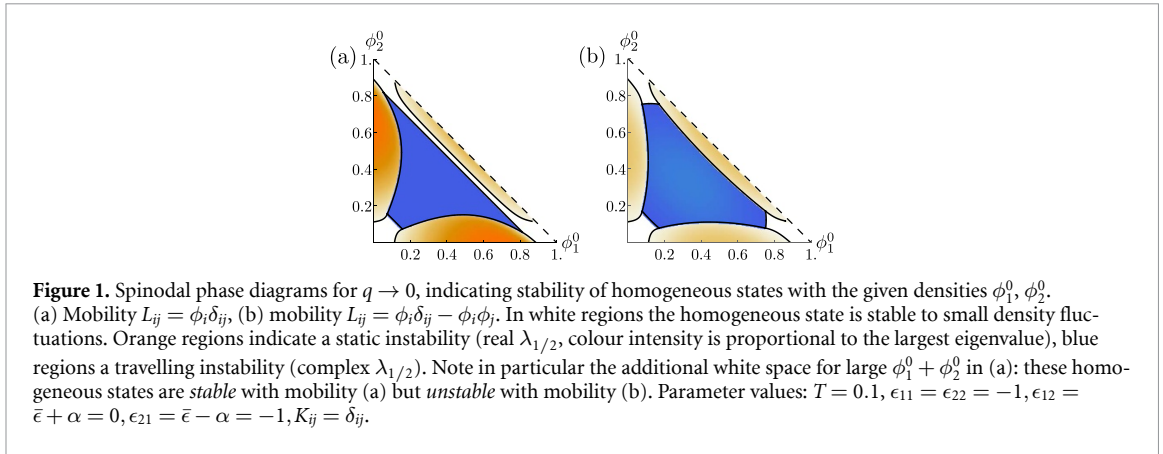
$$\mathbf{R}^0 = \begin{pmatrix} -1 & c \\ 0 & -1 \end{pmatrix}.$$

For the trivial mobility $L_{ij} = \delta_{ij}$ the dynamics is then stable, with both eigenvalues of $\mathbf{R} = \mathbf{R}^0$ being -1 . For a generic mobility, however, $\mathbf{R} = \mathbf{L} \mathbf{R}^0$ acquires eigenvalues with positive real parts whenever

$$\text{tr}(\mathbf{R}) = -L_{11} - L_{22} + cL_{12} > 0.$$

If $c > 2$ this condition can always be met—while maintaining $\mathbf{L}$ positive definite as required—by choosing $L_{12} > (2/c)\sqrt{L_{11}L_{22}}$. The same applies analogously for $c < -2$.

The above analysis can be generalized to arbitrary 2×2 rate matrices: assume that $\text{tr}(\mathbf{R}^0) < 0$ and $|\mathbf{R}^0| > 0$ so that the bare rate matrix $\mathbf{R}^0$ has either two real negative eigenvalues or two complex conjugate eigenvalues with negative real part, making the dynamics for trivial mobility stable. Then one can show that there are positive definite mobility matrices $\mathbf{L}$ so that $\mathbf{R} = \mathbf{L} \mathbf{R}^0$ has eigenvalues with positive real part, thus producing unstable dynamics, if and only if $|\mathbf{R}_s^0| < 0$ where $\mathbf{R}_s^0 = \frac{1}{2}[\mathbf{R}^0 + (\mathbf{R}^0)^T]$ is the symmetric part of $\mathbf{R}^0$. Because $|\mathbf{R}^0| = |\mathbf{R}_s^0| + [(R_{12}^0 - R_{21}^0)/2]^2$ for 2×2 matrices, one again requires a large enough non-reciprocity $|R_{12}^0 - R_{21}^0|$ for the two determinant conditions $|\mathbf{R}^0| > 0$ and $|\mathbf{R}_s^0| < 0$ from above to be met. Summarizing, for strong enough non-reciprocity there are always choices of the mobility matrix that can change the dynamics from stable to unstable.



Returning now to the $q \rightarrow 0$ spinodal phase diagrams of figure 1, we observe static instabilities (real $\lambda_{1/2}$, orange) as well as oscillatory ones (complex $\lambda_{1/2}$, blue) that correspond to travelling spinodal waves. One can check that the transitions between these regions occur, as in the case of non-reciprocal Cahn–Hilliard models [19, 24, 28, 33, 41], via exceptional point transitions. At these exceptional points [17] two distinct real eigenvalues merge and then become complex. The corresponding eigenvectors collapse at the exceptional point, *i.e.* become parallel to each other. We do not explore these exceptional points further because, as we will see next, the presence of a non-trivial mobility means that the nature of instabilities cannot be predicted by considering the limit $q \rightarrow 0$ of small wavevectors.

3.2. Discontinuous transitions between static and oscillatory instabilities

A linear stability analysis of the type above, where only small wavevectors q are considered explicitly, was used also in early studies of non-reciprocal Cahn–Hilliard models [24, 29, 31]. This approach is valid in non-reciprocal Model B when, for example, $L_{ij} \propto K_{ij} \propto \delta_{ij}$. The interfacial $O(q^4)$ contribution to the rate matrix $\mathbf{R}$ is then a multiple of the identity matrix, shifting both eigenvalues $\lambda_{1/2}$ by the corresponding real value. This implies that the nature of the eigenvalues (real or complex) does not change with q and can be deduced from the limit $q \rightarrow 0$. In general, however, finite wavevectors q have to be studied [25, 28, 30, 32, 43].

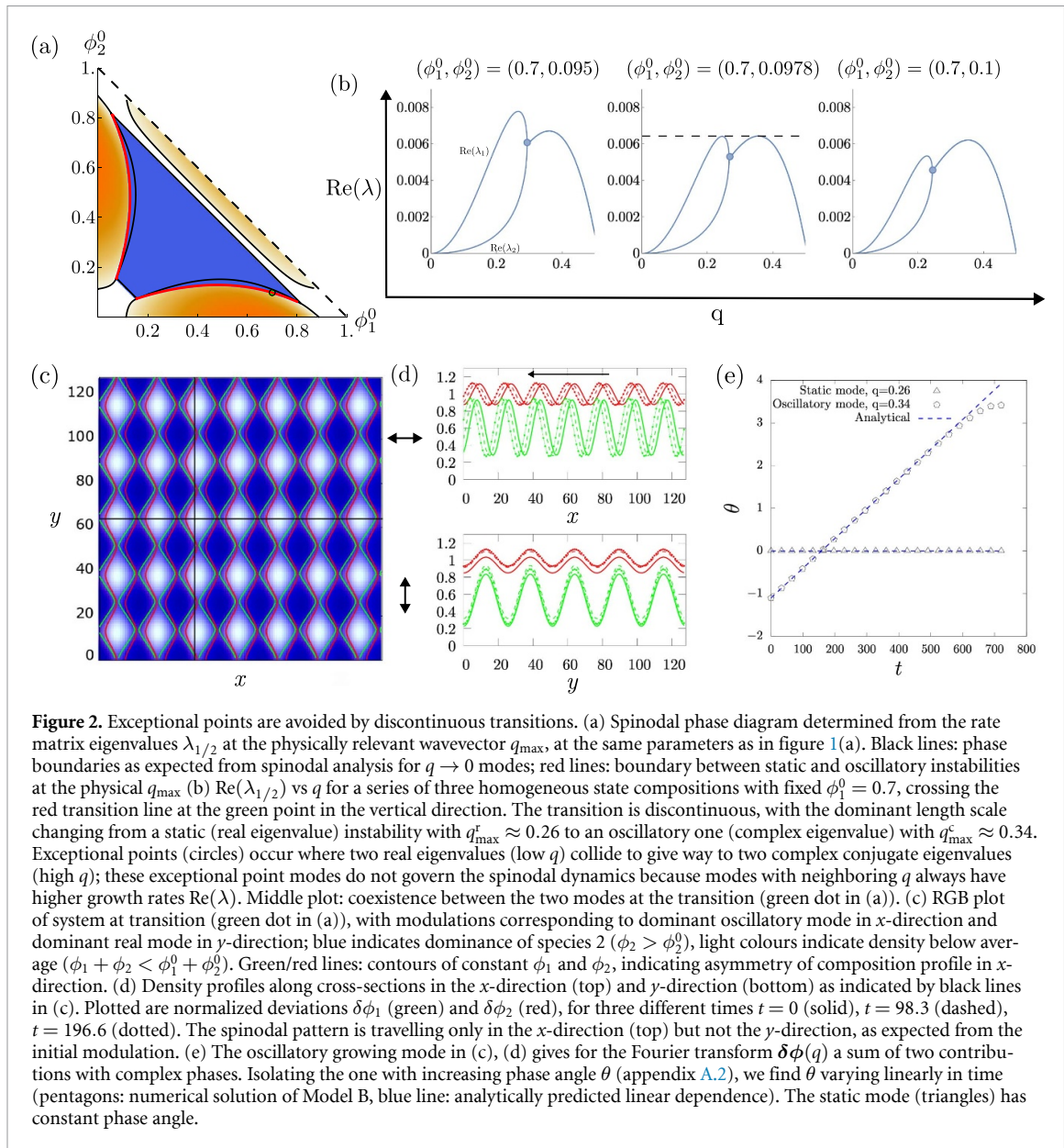
In the case of non-reciprocal Model B considered here, the interfacial contributions to the rate matrix $\mathbf{R}$ will generically have a non-trivial matrix structure arising from the mobility matrix $\mathbf{L}$, even when the interfacial coefficients remain simple ($K_{ij} \propto \delta_{ij}$). For these cases, the eigenvalues $\lambda_{1/2}$ can change from real to complex or vice versa as the wavevector q is varied. The nature of any spinodal instability thus has to be determined explicitly at the wavevector q that is growing most rapidly and hence dominates the spinodal decomposition:

$$q_{\max} = \operatorname{argmax}_q \lambda_r(q) \quad (17)$$

$$\lambda_r(q) = \max\{\operatorname{Re}(\lambda_1(q)), \operatorname{Re}(\lambda_2(q))\}. \quad (18)$$

Figure 2(a) shows the outcome of such an analysis, for the same parameters as in figure 1(a). The overall region of instability in the spinodal phase diagram has not changed, but the region of oscillatory rather than static instabilities has become larger, with the transition between these two types of instability shifted to the red line in figure 2(a). To understand the nature of the transition, we consider three compositions of the homogeneous system lying across the transition line in the vertical direction at the green point in figure 2(a). Figure 2(b) displays the real parts of the eigenvalues $\lambda_{1/2}$ as a function of wavevector modulus q . Where these real parts differ, the eigenvalues themselves are real; when the real parts coincide, the eigenvalues are complex and conjugates of each other, $\lambda_2 = \lambda_1^*$. We observe in figure 2(b) that the transition is *discontinuous*: the fastest growing mode that dominates the spinodal decomposition changes discontinuously, from $q_{\max} \approx 0.26$ and an associated real eigenvalue of the rate matrix, to $q_{\max} \approx 0.34$ with a complex eigenvalue. An exceptional point lying between these two values of q , which in other models is frequently found to govern the transition [17, 19, 24, 28, 29] is thus *avoided*. Instead we have exactly at the transition two competing, equally rapidly growing spinodal modes (figure 2(b) middle), one generating a static pattern and the other an oscillatory one.

To see that the above conclusion is generic within non-reciprocal Model B, *i.e.* that exceptional points are avoided at transitions between instability types in the spinodal phase diagram, we return to the general form of the rate matrix $\mathbf{R}(q)$ from (15). At an exceptional point two real eigenvalues of this



matrix coincide (circles in figure 2(b)) and give way to two complex conjugate ones, which from (14) happens when $(\text{tr}\mathbf{R})^2 = 4|\mathbf{R}|$. When such an exceptional point is crossed into the regime where the eigenvalues are real, the larger one of these eigenvalues grows with a square root dependence from (14) and so is always larger than the one at the exceptional point. This can also be observed in figure 2(b), for values of q just below the exceptional point. Therefore, the exceptional point modes that continuously connect the static (real) and oscillatory (complex) eigenmodes cannot be the ones dominating the spinodal dynamics, and transitions between dominant modes have to be discontinuous instead.

The only scenario where the above reasoning does not apply is the case when an exceptional point cannot be crossed by varying q , i.e. when the eigenvalues of the rate matrix $\mathbf{R}(q)$ cannot change from real to complex by varying q . Referring back to (15), this requires special conditions such as (i) proportionality between $\mathbf{H}$ and $\mathbf{K}$, where q only scales the eigenvalues of $\mathbf{R}(q)$ but does not change the eigenvectors; or (ii) $\mathbf{LK}$ being proportional to the identity, where the interfacial term just shifts the eigenvalues additively. Neither of these conditions applies in non-reciprocal particle mixtures as described by Model B: condition (i) cannot hold generically because the bulk stability matrix $\mathbf{H}$ depends on the density of the homogeneous state while $\mathbf{K}$ does not, and similarly condition (ii) is ruled out by the density-dependence of the mobility matrix $\mathbf{L}$. The typical simplifying assumptions in non-reciprocal Cahn–Hilliard models, on the other hand, are precisely such that one of the non-generic cases (i,ii) from above applies, e.g. by taking $\mathbf{K}$ and $\mathbf{L}$ to be both proportional to the identity matrix (case (ii)). This is why transitions within the spinodal phase diagram are of a qualitatively different nature in non-reciprocal

Model B and in non-reciprocal Cahn–Hilliard models. To what extent this difference is reflected in the long-time dynamics, where nonlinearities are important, is of course an entirely different question.

The above analysis indicates at the discontinuous transitions in the spinodal phase diagram a non-trivial spinodal dynamics with instabilities of different character occurring simultaneously in the system, at two distinct length scales. To demonstrate this, we solve the non-reciprocal Model B equations (1), (5), (7) and (8) numerically in two dimensions for an initial condition corresponding to the green dot in figure 2(a). The initial state is taken as spatially homogeneous but with added small periodic modulations corresponding to the two fastest growing modes (figure 2(b) middle), the complex one in the x -direction and the real one in the y -direction. Figure 2(c) shows a snapshot of the configuration. Following the evolution in time we observe, as predicted, travelling waves in the x -direction while in the y -direction the pattern remains static, see figure 2(d). For ease of visualization we have factored out the exponential growth in time of the density fluctuations, *i.e.* we plot the normalized density fluctuations

$$\delta\tilde{\phi}_i = \delta\phi_i / (3\sigma) \quad (19)$$

where $\sigma^2 = \langle (\delta\phi_1)^2 + (\delta\phi_2)^2 \rangle$ is the time-dependent variance of the two species densities across the system.

The oscillatory nature of the dominant complex mode can also be confirmed from the time evolution of the Fourier transform $\delta\phi(q)$ of the two species densities. This is a sum of two terms proportional to the two right eigenvectors of $\mathbf{R}(q)$, each containing a phase factor $e^{\pm i\omega t}$. If we project $\delta\phi(q)$ onto the first contribution, its phase angle θ therefore has to vary linearly in time. This is confirmed within the spinodal time scale (until $t \approx 500$) in figure 2(e).

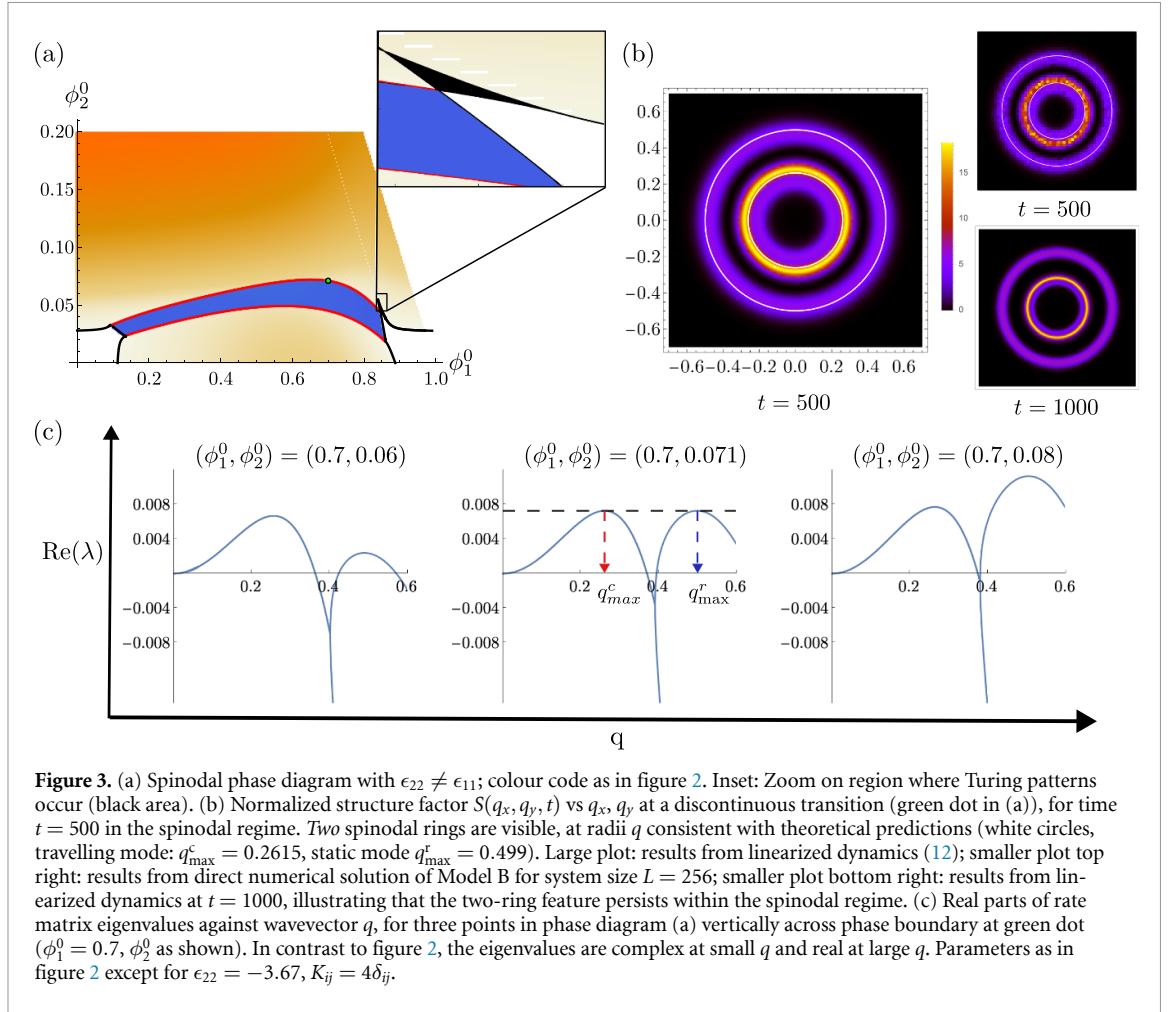
3.3. Discontinuous transition in the nature of instability with asymmetric ϵ

We next show that rather more complex spinodal phase diagram topologies are possible for interaction matrices with unequal intraspecies interactions, $\epsilon_{11} \neq \epsilon_{22}$. In figure 3(a) we show the spinodal phase diagram for the case $\epsilon_{22} = -3.67$. All other parameters are as in figures 1(a) and 2, with the exception of the interfacial coefficients, which we increase to $K_{ij} = 4\delta_{ij}$ to keep the characteristic wavevectors of the same order as before. As in figure 2 we observe discontinuous transitions between static and travelling instabilities (red lines in figure 3). Plots of the real parts of the rate matrix eigenvalues against q are shown in figure 3(c) for three points across one of the transition lines. In contrast to figure 2(b), here the eigenvalues change from complex to real with growing q , rather than the other way around. Physically, this means that in the present case, instabilities are oscillatory for small q (larger length scales) and static for larger q . This is the opposite of what we observed in figure 2, thus illustrating that the relative ordering of the lengthscales of dominant static and travelling spinodal modes depends in a subtle manner on the interplay of non-reciprocity and mobility as well as on the location within the spinodal phase diagram. Additionally, the black region in the inset of figure 3(a) indicates where the linear stability analysis predicts a Turing instability. The real parts of the rate matrix eigenvalues for this region are non-negative only above some non-zero threshold for the wavevector q . Hence, fluctuations corresponding (only) to small length scales are excited, suggesting the existence of Turing patterns. Turing patterns have also been observed in non-reciprocal Cahn–Hilliard models [32].

We finally illustrate for the current set of parameters an experimentally observable consequence of the discontinuous nature transition in the spinodal dynamics: at the transition, *two* distinct spinodal rings can be seen in the structure factor, one each corresponding to the wavevectors of the modes dominating the static and travelling spinodal patterns, respectively. We demonstrate this effect in figure 3(b), both by evaluating the linearized theory (equation (12)) and by a numerical solution of the non-reciprocal Model B equations. In both cases we start from an initial condition with small, spatially uncorrelated fluctuations in both densities ϕ_i around the homogeneous state. Specifically we plot in figure 3(b) the structure factor (with $q = (q_x^2 + q_y^2)^{1/2}$)

$$S(q_x, q_y, t) \equiv S(q, t) \propto \langle |\delta\phi(q, t)|^2 \rangle \quad (20)$$

against (q_x, q_y) . More precisely, $S(q, t)$ here denotes the trace of the matrix of partial equal-time structure factors. The proportionality factor in (20) is chosen so that the average over all (q_x, q_y) of the structure factor is unity. Note that the two-ring pattern in figure 3 is generic in the sense that it will occur at all discontinuous transitions in the spinodal phase diagram, including *e.g.* the ones in figure 2. The location of the transitions can also be detected by changes in $S(q, t)$, as illustrated in appendix B.



3.4. Competition of three spinodal modes

We next demonstrate that yet further topologies of the spinodal phase diagram are possible in non-reciprocal Model B depending on the interaction parameters, revealing in particular homogeneous states for which the initial phase separation dynamics exhibits three competing spinodal lengthscales.

Consider *e.g.* a system with two-species interaction matrix

$$\begin{pmatrix} \epsilon_{11} & \epsilon_{12} \\ \epsilon_{21} & \epsilon_{22} \end{pmatrix} = - \begin{pmatrix} 1 & 1/6 \\ 5/6 & 3.37 \end{pmatrix} \quad (21)$$

and all other parameters as in the previous subsection. The resulting spinodal phase diagram, displayed in figures 4(a) and (b), features several branches of discontinuous transitions between instabilities with different lengthscales. There are in addition two triple-mode points, where three of these lines meet and consequently three spinodal modes compete, one travelling and two static, all growing exponentially with the same λ_r but with different associated wavevectors q . For the triple-mode point at $\phi_1^0 \approx 0.53$ (left green dot) in figure 4(a), the corresponding rate matrix spectrum is shown in figure 4(c) and demonstrates the coexistence of three distinct dominant spinodal modes, two of them static and one oscillatory.

A further new feature apparent from figures 4(a) and (b) is that, from each triple-mode point, we have one transition line emanating that indicates a discontinuous transition between two distinct *static* spinodal modes. Each such static-static discontinuous transition line ends in a critical point, indicated by a purple circle in figures 4(a) and (b). The corresponding wavevector dependence of the rate matrix eigenvalues is displayed in figure 4(c) for three points along one of the static-static transition lines. Note finally that, related to the existence of the two triple-mode points, the region in the spinodal phase diagram where travelling patterns appear (marked blue in figures 4(a) and (b)) lies entirely within the instability area, *i.e.* does not border the region where homogeneous states are stable.

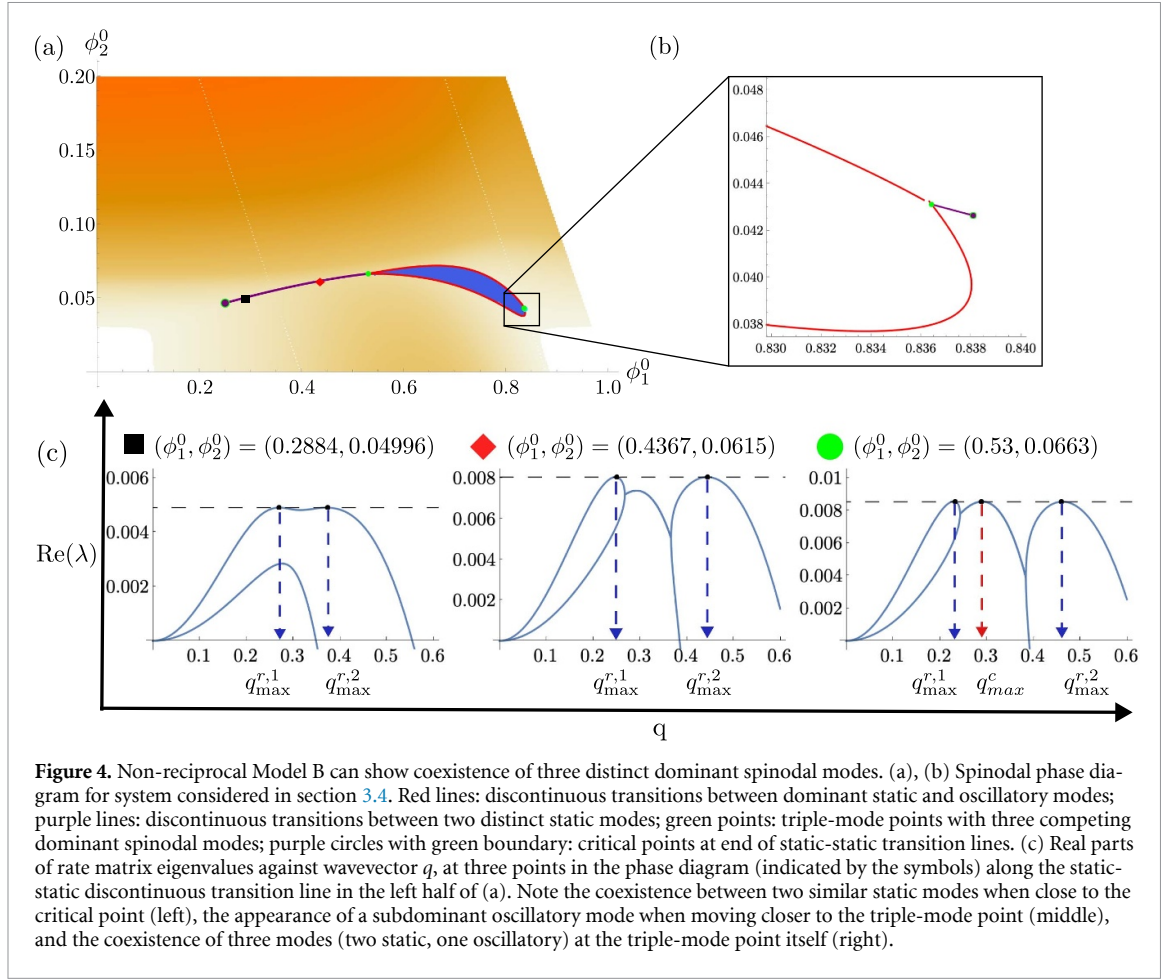


Figure 4. Non-reciprocal Model B can show coexistence of three distinct dominant spinodal modes. (a), (b) Spinodal phase diagram for system considered in section 3.4. Red lines: discontinuous transitions between dominant static and oscillatory modes; purple lines: discontinuous transitions between two distinct static modes; green points: triple-mode points with three competing dominant spinodal modes; purple circles with green boundary: critical points at end of static-static transition lines. (c) Real parts of rate matrix eigenvalues against wavevector q , at three points in the phase diagram (indicated by the symbols) along the static-static discontinuous transition line in the left half of (a). Note the coexistence between two similar static modes when close to the critical point (left), the appearance of a subdominant oscillatory mode when moving closer to the triple-mode point (middle), and the coexistence of three modes (two static, one oscillatory) at the triple-mode point itself (right).

3.5. Effect of interfacial non-reciprocity on spinodal behaviour

So far we have investigated parameter settings in non-reciprocal Model B where the interfacial contributions to the chemical potentials in equations (9) and (10) remain reciprocal ($\bar{K} = \delta = 0$). However, as argued in appendix E, interfacial non-reciprocity is generically expected for continuum models of particle systems with non-reciprocal pairwise forces; explicit expressions for the coefficients K_{ij} as found by coarse-graining can be found in equations (E8). We therefore next consider this case.

To understand the effects of interfacial non-reciprocity we return to the rate matrix (12) for the linearised dynamics,

$$R(q) = -q^2 L(H + q^2 K) = -q^2 \tilde{H} - q^4 \tilde{K}. \quad (22)$$

Here we have abbreviated by $\tilde{H} = LH$ and $\tilde{K} = LK$ the overall contribution coming from bulk and interfacial terms, respectively. Asking now specifically when travelling spinodal patterns arise, we require the rate matrix to have complex eigenvalues with positive real part. From (14) one sees that this requirement translates to

$$(\text{tr} R)^2 - 4|R| < 0 \quad \text{and} \quad \text{tr} R > 0. \quad (23)$$

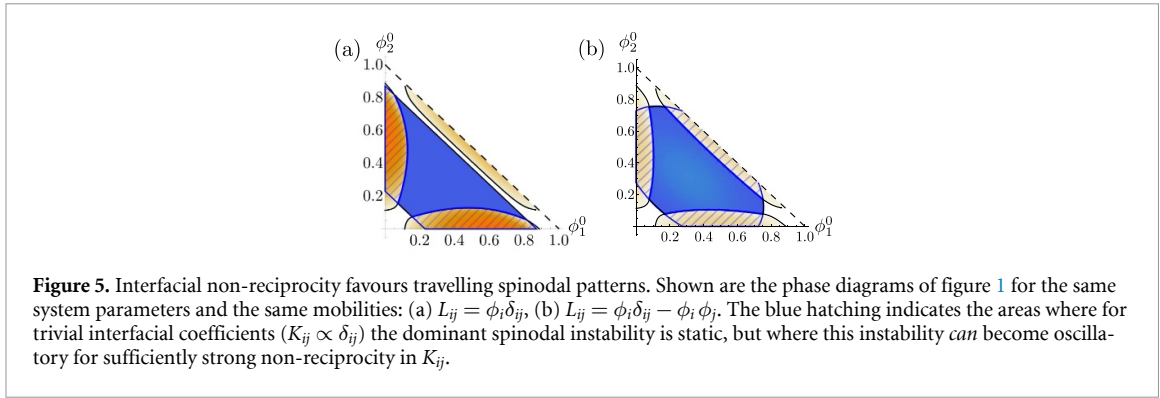
These conditions can be written explicitly as

$$[(\tilde{H}_{11} - \tilde{H}_{22}) + q^2(\tilde{K}_{11} - \tilde{K}_{22})]^2 + 4(\tilde{H}_{12} + \tilde{K}_{12}q^2)(\tilde{H}_{21} + \tilde{K}_{21}q^2) < 0, \quad (24)$$

$$\tilde{H}_{11} + \tilde{H}_{22} < -q^2(\tilde{K}_{11} + \tilde{K}_{22}). \quad (25)$$

The question is now when these conditions can be satisfied by tuning $\tilde{K}$. Here we have to bear in mind that for the continuum model to make sense, there must be no instabilities for $q \rightarrow \infty$, which from (22) means that all eigenvalues of $\tilde{K}$ need to have positive real parts. This constraint is equivalent to

$$\text{tr} \tilde{K} > 0 \quad \text{and} \quad |\tilde{K}| > 0 \quad (26)$$



and the equivalence applies whether or not the eigenvalues of $\tilde{\mathbf{K}}$ are real or complex, as one can confirm from the generic expression (14) for the eigenvalues of 2×2 matrices.

The determinant constraint in (26) can always be satisfied by making $\tilde{K}_{12}$ and $\tilde{K}_{21}$ large and of opposite sign. The first condition (24) for the occurrence of travelling spinodal patterns is then automatically satisfied because the second term becomes negative and large, dominating over the first term. But given the trace constraint on $\tilde{\mathbf{K}}$ in (26), the second condition (25) always has a negative right hand side. Thus, a *necessary* condition for travelling spinodal patterns is that

$$\text{tr} \tilde{\mathbf{H}} = \tilde{H}_{11} + \tilde{H}_{22} < 0. \quad (27)$$

When this condition is satisfied, travelling spinodal modes always exist for some choice of $\tilde{\mathbf{K}}$ respecting (26), for wavevectors $q < (-\text{tr} \tilde{\mathbf{H}} / \text{tr} \tilde{\mathbf{K}})^{1/2}$ as can be seen from (25).

We show the areas in the spinodal phase diagram where (27) holds by blue hatching in figure 5, for the same system parameters as in figure 1. One observes that, like the overall spinodal phase diagram, also the areas where travelling spinodal waves can occur in the presence of interfacial non-reciprocity depend crucially on the form of the mobility. Of course, as the condition (27) is only a necessary one, then for a given choice of $\mathbf{K}$ and the resulting $\tilde{\mathbf{K}}$, the region in the phase diagram where the dominant spinodal modes are oscillatory, *i.e.* produce travelling patterns, will generically be smaller than the hatched areas. We illustrate in appendix D.1, however, that for suitable choices of $\mathbf{K}$ the difference can be made rather small.

3.6. Long time behaviour with bulk NR

So far we have focussed on the phase separation dynamics in non-reciprocal Model B in the spinodal time regime, where the dynamics can be described by linearization around the initial homogeneous state. Making analytical predictions about the long-time fate of the system is a more serious challenge [26, 43, 44]. We therefore use numerical simulations of the non-reciprocal Model B equations to illustrate some of the fascinating long-time behaviours that can emerge, leaving a comprehensive study for future work.

3.6.1. Coexistence of stable and transient domains

We simulate the system starting from a homogeneous state with parent density $(\phi_1^0, \phi_2^0) = (0.15, 0.15)$, with added small fluctuations (see appendix A.3). The system parameters are the same as in figure 1(a). At long times, well beyond the spinodal regime, we observe the appearance of patterns where one phase forms one or several stable domains; in figure 6 this phase contains mainly particles of species 1 so appears red. A second phase also occurs, containing predominantly particles of species 2 (blue/purple in figure 6), but the domains where this is found keep breaking apart as is visible in the series of snapshots in figure 6. Similar patterns have recently been observed in non-reciprocal Cahn–Hilliard models [43]. Intuitively, species 1 (red) ‘chases’ species 2, causing it to invade domains rich in species 2 (blue), while species 2 ‘runs away’ from this invasion leading to the break-up of the blue domains. Species 1 then moves back to its original (red) domain to aggregate there, allowing domains enriched in species 2 to form again so that the process can repeat.

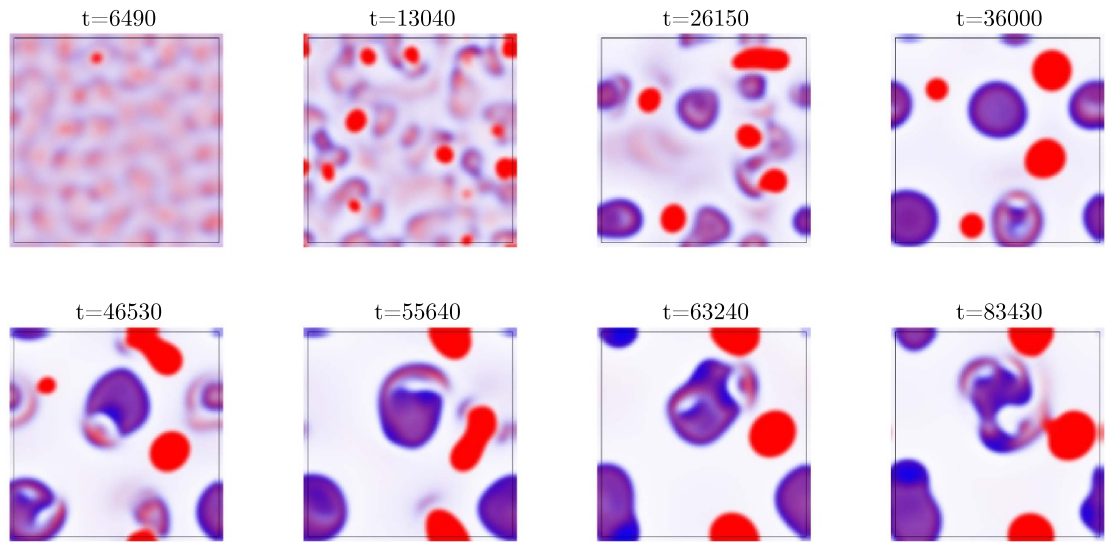


Figure 6. Long-time behaviour for system parameters as in figure 1 and initial homogeneous composition $(\phi_1^0, \phi_2^0) = (0.15, 0.15)$. The later snapshots show formation of stable domains of one phase (red, consisting mainly of particles of species 1) and transient domains of a second phase (blue/purple, mainly species 2). The latter keep breaking up after being ‘invaded’ by the first phase.

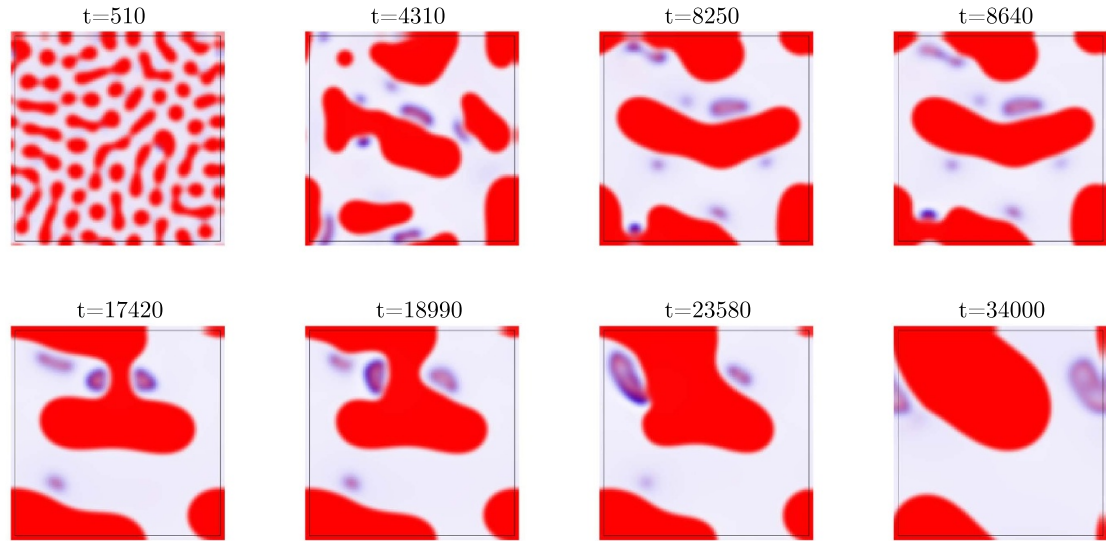


Figure 7. Long-time behaviour for system parameters as in figure 1 and homogeneous parent composition $(\phi_1^0, \phi_2^0) = (0.05, 0.4)$. A single domain of mainly species 1 forms (red); species 2 particles are concentrated in domains moving along the interface of the species 1 domain and locally deforming its interface.

3.6.2. Chase-and-run behaviour along interfaces

With the same parameters as above but a different initial composition of $(\phi_1^0, \phi_2^0) \equiv (0.05, 0.4)$, our analysis correctly predicts that, initially, static spinodal patterns appear. These look essentially indistinguishable from those in standard reciprocal Model B (figure 7). However, at late times, the (bulk) non-reciprocity becomes manifest as can be seen from figure 7. We observe again a single domain containing predominantly species 1 particles (red). Particles of species 2 are now predominantly found in small domains that exhibit chase-and-run dynamics along the *interface* of the species 1 domain, reminiscent of behaviour analysed in [45]. The effect of species 1 wanting to ‘chase’ species 2 here leads to the interface of species 1 domains (red) being locally deformed when a domain rich in species 2 (blue) is close, which in turn causes the species 2 domain to ‘run’ away along the interface.

4. Discussion

We have explored non-reciprocal Model B, the appropriate continuum theory for non-reciprocal particle mixtures. We have demonstrated in particular that the non-trivial density dependence of the mobility in Model B, along with interfacial non-reciprocity that we argue has to be present quite generically, can cause significant differences in the phase separation dynamics compared to *e.g.* non-reciprocal Cahn–Hilliard models.

We have focused mostly on the spinodal dynamics after a quench into a region of the phase diagram where a spatially homogeneous system is unstable, and have analysed the corresponding spinodal phase diagrams. Our key conclusions are as follows. Firstly, the form of the density-dependent mobility is crucial in non-reciprocal Model B. In contrast to equilibrium systems, the mobility can determine whether or not a homogeneous state is stable to small density fluctuations; when it is unstable, the mobility can also determine whether this instability produces static spinodal patterns or travelling ones. The latter arise from dominant spinodal modes that have associated complex eigenvalues and therefore an oscillatory time dependence.

While in the spinodal phase diagrams of non-reciprocal models [17, 46, 47] including non-reciprocal Cahn–Hilliard models [19, 24, 28, 33, 41], the transitions between static and travelling instabilities generically occur at exceptional points, we showed that in non-reciprocal Model B these exceptional points are generally avoided by discontinuous transitions. Crossing these transitions, the length scale of the spinodal patterns changes discontinuously. Directly at the discontinuous transitions point, two dominant spinodal modes with distinct lengthscales compete, one static and one oscillatory, resulting in intricate spinodal dynamics where travelling patterns are superimposed onto static ones. We demonstrated these effects in a system initialized with the two relevant fluctuation lengthscales in orthogonal spatial directions. For random initial density fluctuations, we showed that the same physics shows up in the structure factor, accessible *e.g.* via scattering experiments, as a double spinodal ring.

We demonstrated further that more complex spinodal phase diagram topologies can result depending on the specific form of the non-reciprocal bulk interactions. These include discontinuous transition lines between two static spinodal patterns of different lengthscales, which are terminated by critical points. Three discontinuous transition lines can also meet, resulting in a triple-mode point where three lengthscales compete in the spinodal dynamics.

We argued based on a simple coarse-graining of the non-reciprocal Dean equation that *interfacial* non-reciprocity should be generic when coarse-graining non-reciprocal particle mixtures. We showed that such interfacial non-reciprocity can increase the region in the spinodal phase diagram where travelling spinodal patterns occur, up to a simple bound that we derived analytically and confirmed numerically.

Finally we provided numerical evidence for non-trivial phenomena that arise in the long-time behaviour of non-reciprocal Model B, in particular coexistence of stable and transient domains as well as chase-and-run dynamics along domain interfaces. We emphasize here that such late stage nonlinear dynamics, especially for the *chase and run* case, cannot be predicted robustly from the linear stability analysis we focused on. This is illustrated by the fact that interfacial chase-and-run case appears at a location in the phase diagram where the spinodal instability only produces static patterns.

In future work on the spinodal dynamics of non-reciprocal Model B, it will be interesting to explore comprehensively whether yet more complex phase diagrams can occur, with possibly even more than three competing spinodal lengthscales. Also the effects of more general forms of the Model B mobility [36, 37] will be interesting to study. Beyond this, investigating the long-time behaviour of non-reciprocal Model B is of obvious interest, *e.g.* by quantitatively characterizing the domain patterns that arise, studying their coarsening dynamics, and analysing the role of finite size effects on the long time dynamics. One would in particular like to understand for which parameter settings the steady states can be mapped to those of appropriate non-reciprocal Cahn–Hilliard models, and conversely where novel classes of steady state behaviour appear.

Acknowledgements

This work was supported by the German Research Foundation (DFG) under Grant Number SO 1790/1-1. BS also thanks IMPRS-PBCS Excellence fellowship for financial support. RM acknowledges support from the ANRF, India through PMECRG (Project ANRF/ECRG/2024/002036/PMS). We thank Prof. Matthias Krüger, Prof. Uwe Thiele and Prof. Erwin Frey for helpful discussions.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: <https://doi.org/10.25625/QGUIT9> [52].

2 dominant length scales available at: <https://doi.org/10.1088/1751-8121/ae8809/data1>.

Stable+transient domains available at: <https://doi.org/10.1088/1751-8121/ae8809/data2>.

Interface chase and run available at: <https://doi.org/10.1088/1751-8121/ae8809/data3>.

Dynamics with interfacial NR available at: <https://doi.org/10.1088/1751-8121/ae8809/data4>.

Appendix A. Numerical methods

A.1. Discontinuous transitions in spinodal phase diagrams

Figures 2(a), 3(a) and 4(a) show discontinuous transitions within the spinodal diagrams (red lines), where two spinodal modes with different q compete because their growth is governed by rate matrix eigenvalues with identical real part $\lambda_r(q)$. To locate these transitions precisely, we impose the required condition that λ_r has two distinct maxima at wavevectors q_1 and q_2 at equal height, *i.e.*

$$\frac{d\lambda_r(q_1)}{dq} = \frac{d\lambda_r(q_2)}{dq} = 0 \quad \text{and} \quad \lambda_r(q_1) = \lambda_r(q_2). \quad (\text{A1})$$

We fix ϕ_1^0 and then solve these three conditions numerically for q_1 , q_2 and ϕ_2^0 to locate the discontinuous transition, and then vary ϕ_1^0 . One can of course conversely fix ϕ_2^0 and solve for q_1 , q_2 , ϕ_1^0 ; we use whichever is more convenient numerically.

A.2. Analysis of oscillatory spinodal modes

To obtain the phase angle, θ , of an oscillatory density Fourier mode, we proceed as follows. For fixed wavevector q , the solution of the linearized dynamics (13) for density Fourier mode $\delta\phi$ is

$$\delta\phi(t) = e^{Rt} \delta\phi(0) = \sum_{a=1}^M e^{\lambda_a t} \mathbf{r}_a \mathbf{l}_a^T \delta\phi(0) \quad (\text{A2})$$

where a labels the eigenvalues λ_a and right and left eigenvectors $\mathbf{r}_a$, $\mathbf{l}_a$ of the rate matrix $\mathbf{R}$. For $M=2$ species and complex conjugate eigenvalues $\lambda_{1,2} = \lambda_r \pm i\omega$ one obtains two contributions with increasing and decreasing complex phases. Using the bi-orthogonality $\mathbf{l}_a^T \mathbf{r}_b = \delta_{ab}$ of the eigenvectors, we can isolate each component by taking the inner product with the corresponding left eigenvector, *e.g.*

$$\mathbf{l}_1^T \phi(t) = A e^{\lambda_1 t} = |A| e^{\lambda_r t} e^{i(\varphi + \omega t)} \quad (\text{A3})$$

where $A = \mathbf{l}_1^T \delta\phi(0) = |A| e^{i\varphi}$. Defining $\hat{\phi}_r$ and $\hat{\phi}_i$ as the real and imaginary parts of $\mathbf{l}_1^T \phi(t)$, respectively, the phase angle $\theta = \varphi + \omega t$ can then be extracted as

$$\theta = \arctan(\hat{\phi}_i / \hat{\phi}_r) \quad (\text{A4})$$

The slope of θ versus t then gives the imaginary part ω of the eigenvalue λ_1 , *i.e.* the oscillation frequency of the mode. The corresponding (phase) velocity of the travelling spinodal wave is ω/q . For static spinodal modes, the same procedure gives a constant phase angle $\theta = \varphi$, corresponding to $\omega = 0$.

A.3. Numerical solution of non-reciprocal Model B

For the numerical simulations, we consider a two-dimensional square domain with periodic boundary conditions, discretized into 128×128 grid points, except where specified otherwise. To evaluate the spatial derivatives appearing in the Model B evolution equation, we use a Fourier pseudo-spectral scheme. For time stepping we use a semi-implicit Euler step, which contains a correction term of $\mathcal{O}(q^4)$ in the denominator in the update step [48, 49]. The system is initialized with a homogeneous parent density (ϕ_1^0, ϕ_2^0) with small random (independent Gaussian) fluctuations added at every grid point.

For the case with interfacial non-reciprocity, we needed to use adaptive time stepping. To do this, we perform an update with time step Δt and compare with the result of two successive updates with time step $\Delta t/2$. The deviation between the two results is measured as the mean-squared deviation across the grid. If the deviation is above a set tolerance, we reduce Δt for the next time step. This optimizes computational effort while maintaining a desired level of accuracy via the chosen error tolerance [50].

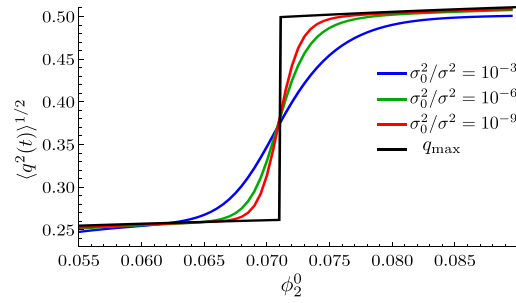


Figure A1. Root-mean-square wavevector $\langle q^2(t) \rangle^{1/2}$ plotted against ϕ_2^0 across the transition point $\phi_2^0 = 0.071$ for different values of σ_0^2/σ^2 (blue, green and red lines); black line shows theoretical predictions for $q_{\max}$. Parameters are as in figure 3, with additionally ϕ_1^0 fixed to 0.7.

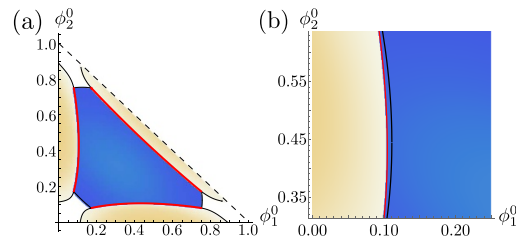


Figure A2. (a) Full (finite q) spinodal phase diagram for system parameters as in figure 1(b), i.e. analog of figure 2(a) but with mobility $L_{ij} = \phi_i \delta_{ij} - \phi_i \phi_j$. As there, we find that exceptional point transitions (black lines between blue and yellow regions) are avoided by discontinuous transitions (red lines). (b) Zoom on the transition line close to the ϕ_2^0 -axis, to show the (small) separation between the two types of transition line.

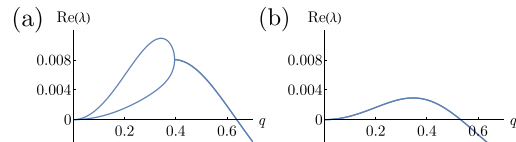


Figure A3. Rate matrix spectra $\text{Re}(\lambda)$ vs q : (a) for mobility $L_{ij} = \phi_i \delta_{ij}$, corresponding to figure 2(a); (b) for mobility $L_{ij} = \phi_i \delta_{ij} - \phi_i \phi_j$, corresponding to figure A2. Homogeneous state composition $(\phi_1^0, \phi_2^0) = (0.12, 0.45)$ in both cases. The different mobility is seen to lead to a dominant static spinodal mode in (a) but an oscillatory one in (b).

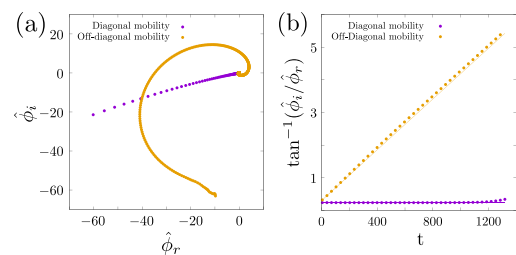


Figure A4. (a) Real and imaginary part of (projected) density Fourier mode at wavevector $q_{\max}$ eigenvalue from simulations, for system parameters corresponding to figure A3(a) (diagonal mobility, $L_{ij} = \phi_i \delta_{ij}$) and figure A3(b) (off-diagonal mobility, $L_{ij} = \phi_i \delta_{ij} - \phi_i \phi_j$).

A.4. Colour code for numerical simulation results

In snapshots of the system state resulting from the numerical simulations described above we show the local composition by the RGB colour code $(\text{RGB}) = 255(\phi_1, \phi_0, \phi_2)$ where $\phi_0 = 1 - \phi_1 - \phi_2$ is the vacancy density. Areas with high density of species 1 or 2 thus appear red or blue, respectively.

To make the spinodal patterns visible in figure 2(c), we rescale the deviations from the homogeneous state as defined in (19) and show the corresponding densities $\tilde{\phi}_i = \text{squash}(\phi_i^0 + \delta\tilde{\phi}_i)$. Here the squashing function $\text{squash}(x) = \min(\max(x, 0), 1)$ restricts the values to the physical range $(0, 1)$.

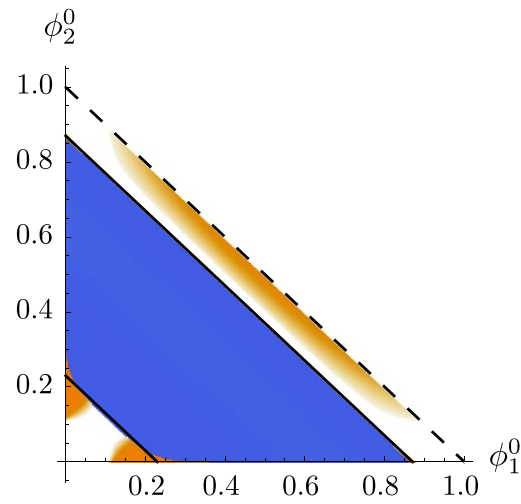


Figure A5. Phase diagram with highly non-reciprocal interface matrix $\mathbf{K}$ given by equation (D1). Blue region: travelling instabilities, caused by dominant spinodal modes with complex eigenvalues. Black lines: boundary of region where such instabilities can be generated by surface nonreciprocity.

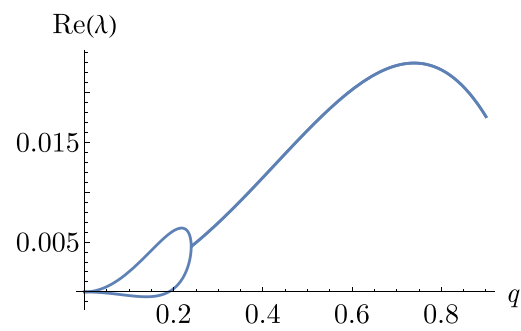


Figure A6. Rate matrix spectrum $\text{Re}(\lambda)$ vs. q for the system from figure A5 at composition $(\phi_1^0, \phi_2^0) = (0.05, 0.4)$.

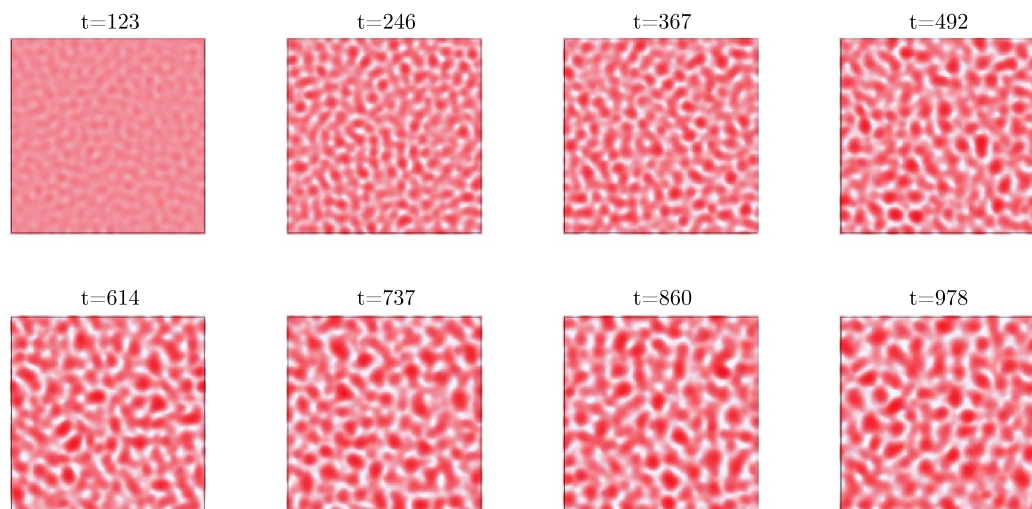


Figure A7. Snapshots of numerical simulations with interfacial non-reciprocity, for the system from figure A5 with $(\phi_1^0, \phi_2^0) = (0.05, 0.4)$. Travelling spinodal patterns are observed, as predicted.

Appendix B. Detecting discontinuous transitions in spinodal regime from scattering data

In section 3.2 we focused our analysis on the spinodal modes with the fastest growth rates, i.e. those that dominate the dynamics. This leads to discontinuous transitions in the spinodal phase diagram as we showed, where the dominant spinodal wave number $q_{\max}$ has a jump. Here, we discuss how these transitions would manifest themselves in average structure factor data that could be measured in scattering experiments.

Consider for example the following observable:

$$\langle q^2(t) \rangle = \frac{\int d\mathbf{q} q^2 S(\mathbf{q}, t)}{\int d\mathbf{q} S(\mathbf{q}, t)} = \frac{\int d\mathbf{r} \sum_i \langle (\nabla \delta \phi_i(\mathbf{r}, t))^2 \rangle}{\int d\mathbf{r} \sum_i \langle (\delta \phi_i(\mathbf{r}, t))^2 \rangle} \quad (\text{B1})$$

where as in the main text $S(\mathbf{q}, t)$ is the trace of the equal-time partial structure factor matrix. Within the spinodal regime, as t grows, the above average will be increasingly dominated by values of q around $q_{\max}$ such that $\langle q^2(t) \rangle^{1/2} \rightarrow q_{\max}$. The spinodal regime ends when the density fluctuations become comparable to the densities (ϕ_1^0, ϕ_2^0) of the homogeneous state, at which point the nonlinearities becomes significant. This can be quantified by fixing $\sigma^2 = \sum_i \langle (\delta \phi_i(\mathbf{r}, t))^2 \rangle$ to be some small constant of order unity. Then the time t it takes to reach fluctuations of this size grows logarithmically with the inverse of the initial fluctuations, $\sigma_0^2 = \sum_i \langle (\delta \phi_i(\mathbf{r}, 0))^2 \rangle$. As $\sigma_0^2/\sigma^2 \rightarrow 0$ this time diverges and hence $\langle q^2(t) \rangle^{1/2} \rightarrow q_{\max}$. A plot of $\langle q^2(t) \rangle^{1/2}$ against one of the parameters of the spinodal phase diagram, such as the density of one of the components in the homogeneous states, thus approaches the discontinuous dependence predicted for $q_{\max}$. At any nonzero value of σ_0^2/σ^2 , the plot remains smooth but its slope at the transition will increase linearly with t , i.e. logarithmically with $(\sigma_0^2/\sigma^2)^{-1}$.

The plot below exemplifies the above discussion. We use the parameters from in figure 3, fix $\phi_1^0 = 0.7$ and vary ϕ_2^0 across the transition point in the spinodal phase diagram, for various values of σ_0^2/σ^2 . As expected, $\langle q^2(t) \rangle^{1/2}$ vs ϕ_2^0 shows an increasingly steep variation as σ_0^2/σ^2 decreases, approaching the theoretically predicted curve $q_{\max}(\phi_1^0, \phi_2^0)$.

Appendix C. Spinodal phase diagram for mobility $L_{ij} = \phi_i \delta_{ij} - \phi_i \phi_j$

C.1. Discontinuous transitions

In section 3.2 we demonstrated that, for non-reciprocal Model B with mobility $L_{ij} = \phi_i \delta_{ij}$, the exceptional point transitions predicted by an analysis for $q \rightarrow 0$ are avoided by discontinuous transitions when the full q -dependence of the rate matrix eigenvalues is taken into account. Figure A2 demonstrates that the same behaviour is observed for the mobility $L_{ij} = \phi_i \delta_{ij} - \phi_i \phi_j$, with the only modification that the quantitative differences between the two types of transition line are now smaller.

C.2. Effect of mobility on nature of dominant spinodal modes

A comparison between figures 2(a) and A2 shows that, as expected from the $q \rightarrow 0$ analysis in figure 1, the form of the mobility in non-reciprocal Model B can affect whether the spinodal dynamics is dominated by static or oscillatory modes. As an example we show in figure A3, for the location $(\phi_1^0, \phi_2^0) = (0.12, 0.45)$ in the spinodal phase diagrams of figures 2(a) and A2, respectively, the corresponding rate matrix spectra. These plots demonstrate that the dominant spinodal mode is static in figure A3(a), but oscillatory in figure A3(b), with the only difference between the two situations being the mobility.

C.3. Numerical simulations with the two mobilities

We confirm the predictions from the linearized dynamics for the two systems in figure A3 by direct numerical simulation of the full nonlinear non-reciprocal Model B equations for the parent composition $(\phi_1^0, \phi_2^0) = (0.12, 0.45)$. We project the Fourier transform of the density with wavevector $q_{\max}$ onto one of the two eigenvectors determined from the linearized dynamics as explained in appendix A.2, and then obtain the real and imaginary parts $\hat{\phi}_r$ and $\hat{\phi}_i$. The results are shown in figure A4.

One observes that for the diagonal mobility $L_{ij} = \phi_i \delta_{ij}$, where from figure A3(a) we expect a static dominant spinodal mode, the trajectory of $(\hat{\phi}_r, \hat{\phi}_i)$, corresponding to the complex plane of the (projected) density Fourier mode, follows a straight line in the spinodal regime. The corresponding phase angle (figure A4(b)) is therefore constant until nonlinear effects set in.

For the case of figure A3(b), on the other hand, the dominant spinodal mode is predicted to be oscillatory. Figure A4(a) confirms this, with $(\hat{\phi}_r, \hat{\phi}_i)$ tracing out an exponential spiral. The corresponding phase angle (figure A4(b)) grows linearly in time, with a prefactor that agrees with the imaginary part of the rate matrix eigenvalue as it should (thin line).

Appendix D. Interfacial non-reciprocity

D.1. Spinodal phase diagram

In section 3.5, we predict the maximal region in the spinodal phase diagram where we can get travelling spinodal waves in the presence of interfacial non-reciprocity. As a concrete example, we consider the set of highly nonreciprocal interfacial coefficients

$$\mathbf{K} = \begin{pmatrix} 0.34 & 0 \\ -80 & 0.34 \end{pmatrix} \quad (\text{D1})$$

and otherwise choose the same system parameters as in figure 1(a). The resulting spinodal phase diagram is shown in figure A5. In accordance with our theoretical expectation, the region where spinodal travelling waves occur (blue) is significantly enlarged compared to figure 1(a). In fact it almost fills the maximal area (hatched in figure 5(a), solid black lines in figure A5) where spinodal travelling waves can occur at all.

D.2. Numerical simulations

To illustrate the occurrence of travelling spinodal waves due to interfacial non-reciprocity, we perform numerical simulations at composition $(\phi_1^0, \phi_2^0) = (0.05, 0.4)$. With interfacial coefficients $K_{ij} = \delta_{ij}$ as in figure 2, *i.e.* without interfacial non-reciprocity, the spinodal dynamics then produces static patterns (cf figure 2(a)).

With interfacial non-reciprocity defined by (D1), however, we predict a travelling instability for the above composition, as can be seen from figure A5 and the corresponding rate matrix spectrum in figure A6. Careful inspection of the simulation snapshots in figure A7 confirms that the spinodal patterns do indeed travel, as expected.

Appendix E. Non-reciprocal Dean equation and coarse-graining

An exact equation for dynamics of the density fields describing a mixture with non-reciprocal two-body interactions can be derived following Dean's original approach [42]. We use in this appendix Greek letters α, β for the particle species, to have the Roman indices i, j available for the particles.

Assuming overdamped dynamics, the Langevin equation for a particle i of species α is

$$\gamma \dot{\mathbf{r}}_i = \sum_{\beta} \sum_{j \in \beta} \mathbf{F}_{\alpha\beta}(\mathbf{r}_i - \mathbf{r}_j) + \boldsymbol{\xi}_i \quad (\text{E1})$$

where $\mathbf{F}_{\alpha\beta}$ is the force exerted by a particle of species β on a particle of species α , γ is the friction coefficient and $\boldsymbol{\xi}_i$ is thermal (Gaussian) white noise, with variance $2T\gamma$. With non-reciprocal interactions one has $\mathbf{F}_{\alpha\beta} \neq \mathbf{F}_{\beta\alpha}$. The density fields are defined as

$$\phi_{\alpha}(\mathbf{r}, t) = \sum_{i \in \alpha} w(\mathbf{r} - \mathbf{r}_i(t)) \quad (\text{E2})$$

where $w(\mathbf{r})$ is a smearing function obeying $\int d\mathbf{r} w(\mathbf{r}) = 1$. Taking specifically the limit of no smearing, $w(\mathbf{r}) = \delta(\mathbf{r})$, one then finds by following the steps in [42] the equations of motion

$$\frac{\partial \phi_{\alpha}(\mathbf{r}, t)}{\partial t} = -\nabla \cdot \mathbf{j}_{\alpha}(\mathbf{r}, t) \quad (\text{E3})$$

with current

$$\begin{aligned} \mathbf{j}_{\alpha}(\mathbf{r}, t) = & -D \nabla \phi_{\alpha}(\mathbf{r}, t) + \gamma^{-1} \phi_{\alpha}(\mathbf{r}, t) \sum_{\beta} \int d\mathbf{r}' \mathbf{F}_{\alpha\beta}(\mathbf{r} - \mathbf{r}') \phi_{\beta}(\mathbf{r}', t) \\ & + (2D \phi_{\alpha}(\mathbf{r}, t))^{1/2} \boldsymbol{\eta}_{\alpha}(\mathbf{r}, t) \end{aligned} \quad (\text{E4})$$

where $D = T/\gamma$ and $\boldsymbol{\eta}_\alpha$ is unit variance Gaussian white noise. The second line contains the effect of the non-reciprocal interactions, via the density of ϕ_α of species α times the velocity $\gamma^{-1} \int d\mathbf{r}' \mathbf{F}_{\alpha\beta}(\mathbf{r} - \mathbf{r}') \phi_\beta(\mathbf{r}', t)$ arising from the interactions.

While equation (E3) is formally exact, for further analysis the $\phi_\alpha(\mathbf{r}, t)$ are often treated as smooth density fields. Adopting this approximation and assuming that the interaction forces $\mathbf{F}_{\alpha\beta}(\mathbf{r}) = -\nabla V_{\alpha\beta}(\mathbf{r})$ are derived from (non-reciprocal) potentials $V_{\alpha\beta}(\mathbf{r})$ that are central ($V_{\alpha\beta}(\mathbf{r}) = V_{\alpha\beta}(|\mathbf{r}|)$), one finds within a gradient expansion to second order, *i.e.*

$$\phi_\beta(\mathbf{r}') = \phi_\beta(\mathbf{r}) + (\mathbf{r}' - \mathbf{r}) \cdot \nabla \phi_\beta(\mathbf{r}) + \frac{1}{2} [(\mathbf{r}' - \mathbf{r}) \cdot \nabla]^2 \phi_\beta(\mathbf{r}) \quad (\text{E5})$$

for the second line of (E4)

$$-\gamma^{-1} \phi_\alpha \sum_\beta \nabla (\epsilon_{\alpha\beta} \phi_\beta - K_{\alpha\beta} \nabla^2 \phi_\beta) \quad (\text{E6})$$

with

$$\epsilon_{\alpha\beta} = \int d\mathbf{r} V_{\alpha\beta}(\mathbf{r}) \quad (\text{E7})$$

$$K_{\alpha\beta} = -\frac{1}{2d} \int d\mathbf{r} V_{\alpha\beta}(\mathbf{r}) \mathbf{r}^2 \quad (\text{E8})$$

where d is the spatial dimension. Inserting into (E3) gives contributions to the equations of motion for the densities identical to those (up to the species index relabelling $\alpha, \beta \rightarrow i, j$) from the interaction chemical potentials (6), with mobility $L_{\alpha\beta} = \gamma^{-1} \phi_i \delta_{ij}$ in (2). While this coarse-graining approach is clearly naive, given that *e.g.* it ignores the effects of short-range repulsion between particles and consequently becomes undefined for interaction potentials $V_{\alpha\beta}(\mathbf{r})$ that diverge sufficiently strongly for $\mathbf{r} \rightarrow 0$, it does show that non-reciprocal interactions $V_{\alpha\beta}(\mathbf{r})$ will generically produce not only non-reciprocal bulk interaction coefficients $\epsilon_{\alpha\beta}$, but also non-reciprocal interfacial terms $K_{\alpha\beta}$. Indeed, avoiding non-reciprocity in the interfacial coefficients $K_{\alpha\beta}$ from (E8) while maintaining it in the bulk terms $\epsilon_{\alpha\beta}$ from (E7) would require exquisite fine tuning of the non-reciprocal pair potentials $V_{\alpha\beta}(\mathbf{r})$. The same argument will apply in more sophisticated coarse-graining schemes because bulk and interfacial coefficients are, by construction, sensitive to different aspects of the distance dependence of the $V_{\alpha\beta}(\mathbf{r})$. Our conclusion is therefore that interfacial non-reciprocity is generic in particle mixtures interacting via non-reciprocal pair potentials.

An interesting open question is how the above reasoning extends to non-reciprocal many-body interactions. One example in this class is quorum sensing, where particles respond to local densities and where microscopically non-reciprocal interactions can, under certain conditions, lead to a reciprocal macroscopic description [51].

Appendix F. Movies from simulations

The movies from our numerical simulations can be found on the supplementary data files. The movies show dynamics of the phases (following the RGB scheme mentioned in the main text) for different values of the parameters.

- MovieS1.mp4: two competing length scales for parameters as mentioned in figure 2(c) for parent composition a
- MovieS2.mp4: long time behaviour (coexistence of stable and transient domains) for the parameters as in figure 1 for homogeneous parent composition b
- MovieS3.mp4: long time behaviour (chase and run behaviour along interfaces) for the parameters as in figure 1 for homogeneous parent composition c
- MovieS4.mp4: spinodal dynamics with interfacial nonreciprocity for the parameters as mentioned in figure A7.

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