Theory of nonequilibrium coexistence with coupled conserved and nonconserved order parameters

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(Received 19 September 2023; accepted 31 October 2025; published 1 December 2025)

Phase separation routinely occurs in both living and synthetic systems. These phases are often complex and distinguished by features including crystallinity, nematic order, and a host of other nonconserved order parameters. For systems at equilibrium, the phase boundaries that characterize these transitions can be straightforwardly determined through the framework of thermodynamics. The prevalence of phase separation in active and driven systems motivates the need for a genuinely nonequilibrium theory for the coexistence of complex phases. Here, we develop a dynamical theory of coexistence when both conserved and nonconserved order parameters are present, casting coexistence criteria into the familiar form of equality of state functions. Our theory generalizes thermodynamic notions such as the chemical potential and Gibbs-Duhem relation to systems out of equilibrium. While these notions may not exist for all nonequilibrium systems, we numerically verify their existence for a variety of systems by introducing phenomenological active field theories. We hope our work aids in the development of a comprehensive theory of high-dimensional nonequilibrium phase diagrams.

DOI: 10.1103/2jgf-yb82

I. INTRODUCTION

Nonequilibrium systems as diverse as reaction-modulated biomolecular condensates [1,2], chiral crystals comprised of motile bacteria [3] or starfish embryos [4], and nematically ordered amoeba cells [5] all can display states of phase coexistence. A hallmark of these nonequilibrium states is the presence of both conserved (e.g., number density) and nonconserved (e.g., crystallinity, fuel concentration, or nematicity as shown in Fig. 1) order parameters, which couple to generate these intriguing states of phase coexistence. In many of these systems, the absence of even a local equilibrium precludes the use of the thermodynamic arguments employed to construct equilibrium phase diagrams. While there is now a growing body of literature on generalizing theories of phase coexistence to nonequilibrium systems with *conserved* order parameters [6–21], there are comparatively fewer studies [22,23] for the construction of nonequilibrium phase diagrams when both conserved and nonconserved order parameters are crucial. A framework for describing these transitions could deepen our understanding of active crystallization [24–31], isotropic-nematic coexistence [32,33], mesa patterns in mass-conserving reaction-diffusion systems [22,34–36], and chemotactic fluid-fluid separation [37,38].

One can always directly determine phase diagrams by directly measuring the order parameters in heterogeneous states of coexistence in simulation or experiment. For systems at equilibrium, thermodynamics allows one to circumvent these measurements and construct *predictive theories* for phase diagrams solely by equating *bulk state* functions. These coexistence criteria are determined variationally from the system's free energy, which is generally ill defined out of equilibrium. What, if any, bulk state functions are equal between coexisting phases is thus an open question for driven systems.

In this article, we explore the questions outlined above by developing an entirely dynamical theory of phase coexistence that is applicable to both equilibrium and nonequilibrium systems with coupled conserved and nonconserved order parameters. In doing so, we extend our mechanical theory of multicomponent coexistence [21] to dynamical contexts, which allows for a natural description of nonconserved order parameters. We find coexistence criteria solely in terms of *bulk equations of state*, just as in equilibrium. While the nonequilibrium coexistence criteria when there is a single conserved order parameter [12,16,39] can always be identified, here we find the criteria to only exist for a subset of nonequilibrium systems. In particular, we find systems in which (1) the criteria are exact and expressed as equality of bulk state functions; (2) the criteria are approximate and expressed as a generalized equal-area Maxwell construction; and (3) no coexistence criteria exist in terms of bulk state functions. To validate our approach, we introduce a generalized version of active model C (AMC) [40–42]. We find excellent agreement between the predictions made by our theory and the phase diagrams obtained through numerical simulation of AMC. We hope our theory provides a practical procedure to construct first-principles phase diagrams of nonequilibrium systems with coupled conserved and nonconserved order parameters.

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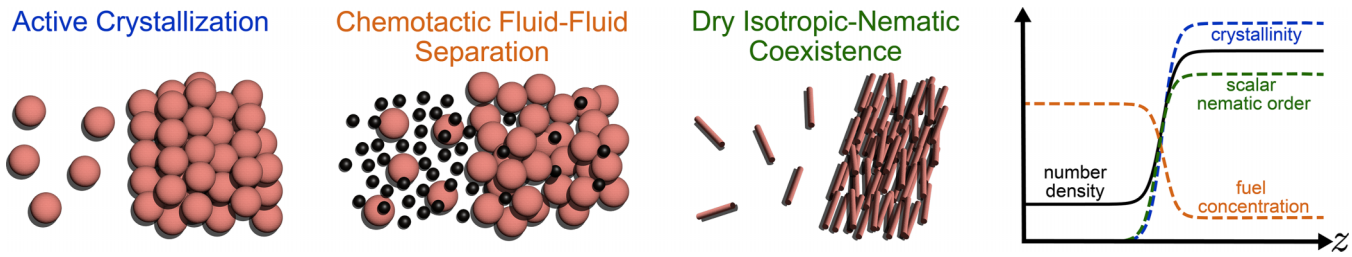


FIG. 1. Three example systems where our theory would apply: active crystallization, chemotactic fluid-fluid separation, and active isotropic-nematic coexistence. A schematic of the order parameter profiles is shown, where the nonconserved order parameter represents the local crystallinity, fuel concentration, and nematic order. While chemoattractant fuel is likely to be subject to a constraint (i.e., mass conservation), common coarse-grained models treat chemotactic fuel as a wholly unconstrained variable [37,38].

II. THEORY OF MULTIPHASE COEXISTENCE

In this section, we describe a dynamical theory of coexistence between n_P phases described by one scalar conserved field, ρ, coupled to one scalar nonconserved field, ψ, that is applicable to both equilibrium and nonequilibrium systems. For a more general treatment, where we extend the coexistence theory to systems with an arbitrary number of conserved and nonconserved order parameters (including order parameters subject to aggregate constraints such as total mass conservation), we refer the reader to Appendix A. We first review the thermodynamically derived equilibrium coexistence criteria in Sec. II A. In Sec. II B, we begin with the evolution equations for each of the order parameters and derive equations of state that are equal in coexisting phases. While not every nonequilibrium system has bulk coexistence criteria, in those that do, we find a pseudopotential and global quantity that are equal in coexisting phases that are analogous to the equilibrium chemical potential and pressure, respectively.

A. Equilibrium criteria from bulk thermodynamics

We consider n_P macroscopic coexisting phases with uniform temperature T. The phases comprise a set $\mathcal{P}$ of length n_P. The overall number density is $\rho^{\text{sys}} = N/V$, where N is the particle number and V is the total system volume. The density field, ρ, can fluctuate in space but is constrained to have a mean of ρ^{sys}. The degree of order in the system is characterized by a nonconserved intensive order parameter, ψ, which, in contrast to ρ, is *unconstrained*.

The bulk (mean-field) free energy density of the system is denoted as $f^{\text{bulk}}(\rho, \psi)$ with the total free energy $F^{\text{bulk}} = V f^{\text{bulk}}(\rho, \psi)$. The absence of a coupling between ψ and ρ in the free energy along with the lack of constraints on ψ will result in each phase having an identical value of ψ: the value that minimizes f^{bulk}. A coupling between ψ and the constrained ρ in the free energy can lead to coexistence between dense and dilute phases with different values of ψ. More specifically, this nontrivial coupling will be reflected in nonadditive contributions of ρ and ψ to the free energy density, i.e., $f^{\text{bulk}}(\rho, \psi) \neq f_\rho^{\text{bulk}}(\rho) + f_\psi^{\text{bulk}}(\psi)$ (and thus the mean-field probability cannot be factorized, i.e., $P(\rho, \psi) \propto \exp[-V f^{\text{bulk}}(\rho, \psi)/k_B T] \neq P_\rho(\rho) P_\psi(\psi)$, where $k_B T$ is the thermal energy). A necessary criterion for equilibrium coexistence with coupled conserved and nonconserved order parameters is thus a nonvanishing mixed derivative, $\partial^2 f^{\text{bulk}}/\partial\rho\partial\psi$.

In the scenario of n_P coexisting phases (e.g., $n_P = 2$ for two-phase coexistence), the total free energy can be expressed as $F^{\text{bulk}} = \sum_{\alpha\in\mathcal{P}} V^\alpha f^{\text{bulk}}(\rho^\alpha, \psi^\alpha)$, where V^α, ρ^α, and ψ^α are the respective volume, number density, and nonconseved order parameter of the α phase and we have neglected the interfacial free energy (the ratio of the interfacial area to the system volume is negligibly small for macroscopic systems). Notably, while the phase volumes and number densities are constrained, there are no constraints on ψ^α (i.e., systems prepared at a given density and total volume can take any value of ψ^α). Minimizing the free energy with respect to each phase's volume, number density, and nonconserved order parameter, subject to the above constraints, results in the equilibrium coexistence criteria:

$$\mu_\rho^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = \mu_\rho^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{1a}$$

$$\mu_\psi^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = 0 \quad \forall \quad \alpha \in \mathcal{P}, \tag{1b}$$

$$p^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = p^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{1c}$$

where $\mu_\rho^{\text{bulk}} \equiv \partial f^{\text{bulk}}/\partial\rho$ is the familiar bulk chemical potential, $\mu_\psi^{\text{bulk}} \equiv \partial f^{\text{bulk}}/\partial\psi$ is the analogous quantity for the nonconserved order parameter, and $p^{\text{bulk}} \equiv \mu_\rho^{\text{bulk}}\rho - f^{\text{bulk}}$ is the bulk pressure. Here, $\mu_\rho^{\text{coexist}}$ and p^{coexist} are constants that must be determined and generally depend on ρ^{sys} and T. Contrasting this, $\mu_\psi^{\text{bulk}} = 0$ during coexistence as ψ is not subject to any constraint by definition. While the criteria following from the constrained minimizations of the free energy with respect to ρ [Eq. (1a)] and V [Eq. (1c)] are familiar for any state of equilibrium n_P-phase coexistence (each providing $n_P - 1$ criteria), the remaining n_P criteria [Eq. (1b)] ensure that within each phase, the optimal value of ψ is selected for the corresponding ρ. Subtracting the number of equations in Eq. (1), $2n_P - 2 + n_P$, from the number of variables describing the coexisting phases, $2n_P$, we find the number of degrees of freedom to be $2 - n_P$, the well-known (constant temperature) Gibbs phase rule [43].

Equation (1c) can be equivalently cast as a Maxwell equal-area construction on the chemical potential vector, resulting in coexistence criteria that do not explicitly contain the thermodynamic pressure:

$$\mu_\rho^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = \mu_\rho^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{2a}$$

$$\mu_\psi^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = 0 \quad \forall \quad \alpha \in \mathcal{P}, \tag{2b}$$

$$\int_{\rho^\alpha}^{\rho^\beta}\left[\mu_\rho^{\text{bulk}}(\rho,\psi)-\mu_\rho^{\text{coexist}}\right]d\rho+\int_{\psi^\alpha}^{\psi^\beta}\mu_\psi^{\text{bulk}}(\rho,\psi)d\psi = 0 \quad \forall \quad \alpha,\beta\in\mathcal{P}, \tag{2c}$$

which is a direct result of the Gibbs-Duhem relation [44,45]:

$$dp^{\text{bulk}} = \rho d\mu_\rho^{\text{bulk}} + \psi d\mu_\psi^{\text{bulk}}. \tag{3}$$

We note that for systems with one order parameter, the coexistence criteria can be expressed entirely with *either* the chemical potential *or* pressure as chemical potential equality can be expressed through the appropriate Maxwell construction on the pressure. For systems with multiple order parameters, this flexibility is lost: There is no way to express the coexistence criteria without the chemical potential vector. However, the path independence of the integrals in Eq. (2c) [its value is $p^{\text{bulk}}(\rho^\alpha,\psi^\alpha)-p^{\text{bulk}}(\rho^\beta,\psi^\beta)$ regardless of the parametrization chosen to evaluate the integrals] allows one to express the criteria in a form that resembles that of one-component systems. We now consider the integration path $\psi^*(\rho)$ such that $\mu_\psi^{\text{bulk}}(\rho,\psi^*(\rho))=0$. While there may be multiple possible solutions ψ^*, one should select the solution that corresponds to the coexistence scenario under consideration [i.e., to describe α-β coexistence, the solution $\psi^*(\rho)$ with end points $\psi^*(\rho^\alpha)=\psi^\alpha$ and $\psi^*(\rho^\beta)=\psi^\beta$ should be chosen]. Along this path, the Maxwell construction resembles that of one-component systems:

$$\mu_\rho^{\text{bulk}}(\rho^\alpha,\psi^\alpha)=\mu_\rho^{\text{coexist}} \quad \forall \quad \alpha\in\mathcal{P}, \tag{4a}$$

$$\int_{\rho^\alpha}^{\rho^\beta}\left[\mu_\rho^{\text{bulk}}(\rho,\psi^*(\rho))-\mu_\rho^{\text{coexist}}\right]d\rho = 0 \quad \forall \quad \alpha,\beta\in\mathcal{P}. \tag{4b}$$

Importantly, $\psi^*(\rho)$ represents the value of ψ that is measurable at a given value of ρ, as $\mu_\rho^{\text{bulk}}(\rho,\psi^*)$ [and equivalently $p^{\text{bulk}}(\rho,\psi^*)$] is what one would measure for a homogeneous system at equilibrium. While the full dependence of μ_ρ^{bulk} on ψ is generally not something one can measure, the path independence of Eq. (2c) renders this inconsequential.

We note that while we have derived these coexistence criteria for systems with coupled conserved and nonconserved fields, the criteria also naturally recover the criteria for coexistence described by a single conserved field (e.g., liquid-gas coexistence) with $\psi^\alpha=\psi^\beta \quad \forall \quad \alpha,\beta\in\mathcal{P}$ and $\rho^\alpha\neq\rho^\beta \quad \forall \quad \alpha\neq\beta\in\mathcal{P}$. Next, we seek to determine the nonequilibrium analogs of the chemical potentials and Gibbs-Duhem relation in order to express the steady-state conditions in Eq. (7) solely in terms of measurable bulk equations of state.

B. Nonequilibrium criteria from dynamics

One may always obtain the phase diagram of a system by determining the full spatial order parameter profiles during coexistence, whether it be through experiment, simulation, or theoretical techniques. This is often difficult, however, and consequently we aim to develop a framework that allows us to circumvent this task, just as in equilibrium, and determine phase diagrams of nonequilibrium systems by equating state functions across phases. The approach proposed below is able to reproduce the criteria derived in the previous section for equilibrium systems while also describing a large class of nonequilibrium systems.

As we look to describe states of macroscopic phase coexistence, where the radius of curvature of the phases far exceeds any other length scale in the system, we consider a quasi-one-dimensional (quasi-1D) geometry where spatial variations only occur in the z dimension [e.g., the direction normal to the interface(s)] with translational invariance in the other dimensions. For macroscopic phase coexistence, the curvature corrections to our coexistence criteria are anticipated to scale inversely with the size of our domains and are thus negligible (e.g., the Laplace pressure jump is negligible in macroscopic equilibrium systems). For this reason, we hereafter neglect the effects of interfacial curvature. Of course, these effects cannot be neglected in the study of *microphase* separation or mesoscale pattern formation [46], which are beyond the scope of the present study. This ensures the z direction will be normal to the interface for phase-separated states.

We begin by considering the spatial and temporal dynamics of each order parameter. The conserved order parameter is subject to an integral constraint, $\int_V d\mathbf{r}\rho(\mathbf{r})=V\rho^{\text{sys}}$, where $\rho(\mathbf{r})$ is its local value at position $\mathbf{r}$ and ρ^{sys} is its space-averaged value. While we explicitly considered a conserved number density in Sec. II A, here we allow ρ to be any conserved order parameter subject to an integral constraint of the above form. Contrasting this, we consider a nonconserved order parameter that is not subject to any constraints and therefore $\int_V d\mathbf{r}\psi(\mathbf{r})$ can take any value. The evolution equations for the order parameters satisfy the balance laws:

$$\partial_t\rho = -\partial_z J_\rho, \tag{5a}$$

$$\partial_t\psi = -\partial_z J_\psi + s_\psi, \tag{5b}$$

where $\partial_t\equiv\partial/\partial t$, $\partial_z\equiv\partial/\partial z$, J_ρ and J_ψ are the respective fluxes of ρ and ψ, and s_ψ is the generation of ψ. The fluxes in the directions orthogonal to z are neglected, consistent with our quasi-1D geometry and translational invariance in these directions. We consider dynamics [Eq. (5)] that are symmetric under the transformation $z\to -z$ (with $\rho\to\rho$ and $\psi\to\psi$). Consequently, $\partial_t\rho$, $\partial_t\psi$, and s_ψ have even spatial parity, while J_ρ and J_ψ have odd spatial parity.

We now require constitutive relations for the fluxes and generation terms appearing in our evolution equations. With precise definitions of the order parameters, we could formulate exact expressions for these quantities (e.g., through an Irving-Kirkwood procedure [47]) and precisely identify *the driving forces* behind them. For example, if the conserved order parameter were the number density, the flux can be directly connected to the linear momentum balance [16]. Here, to maintain generality in the selection of the order parameters, we instead propose Markovian constitutive relations for the dynamics in terms of "flux-driving" and "generation-driving" forces, respectively. Before doing so, to further simplify, we assume that the flux of the nonconserved field is negligible with the generation term controlling the dynamics of ψ at each point in space (i.e., $|\partial_z J_\psi| << |s_\psi|$). Although this is consistent with the commonly invoked "model A" dynamics of a nonconserved order parameter [48], there may be specific

cases in which J_ψ plays a significant role in the dynamics. These cases will be the subject of future work.

The remaining quantities governing the dynamics of the order parameter fields are now just J_ρ and s_ψ. Stationarity of the conserved field ($\partial_t \rho = 0$) combined with flux-free boundary conditions results in $J_\rho = 0$. Additionally, stationarity of the nonconserved field ($\partial_t \psi = 0$) directly implies $s_\psi = 0$. We now propose that these steady-state conditions correspond to a "force" balance with respect to "flux-driving" and "generation-driving" forces. The most general linear model of this form is

$$J_\rho = L f_\rho, \tag{6a}$$

$$s_\psi = M f_\psi, \tag{6b}$$

where f_ρ is the force driving the flux of the conserved field and f_ψ is the force driving the generation of the nonconserved field. Here, f_ρ and f_ψ are generally functionals of the complete spatial profile of the order parameters, $\rho(z)$ and $\psi(z)$. Both L and M are transport coefficients that must be positive to, respectively, ensure the flux acts in the same direction as its driving force and that ψ is generated with positive forces and destroyed with negative forces. Notably, L and M both depend on the local value of ρ and ψ but have no explicit spatial dependence, and thus J_ρ and s_ψ, respectively, inherit the spatial parity of f_ρ (odd) and f_ψ (even). As f_ρ is spatially odd, it cannot enter s_ψ, which must be spatially even; however, in principle $\partial_z f_\psi$ could enter J_ρ. It will be clear that including this coupling will be inconsequential in determining the ultimate coexistence criteria. Moreover, there is a freedom in defining f_ρ and f_ψ, as one can always scale them by a constant and appropriately rescale L and M. However, this freedom does not affect the coexistence criteria.

For passive systems, the driving forces can be identified through the framework of linear irreversible thermodynamics [49]. In the absence of nonconserved fields, the driving force for the flux can be equivalently expressed using either the chemical potential or pressure, $f_\rho = -\partial_z p = -\rho \partial_z \mu_\rho$, which are related through the Gibbs-Duhem relation [Eq. (3)]. Note that this "pressure" is the mechanical pressure when the conserved field is the number density. When a nonconserved field is present, the flux of the conserved field is still driven by pressure gradients; however, the Gibbs-Duhem relation now contains additional terms related to μ_ψ. Relaxational dynamics with $f_\psi = -\mu_\psi$ ensures that the equilibrium condition of $\mu_\psi = 0$ is reached.

We note that the linearity of the constitutive relations proposed above limits their application to small forces. For the context of coexistence, the crucial aspect of these relations is that they identify the correct driving forces so that we may describe the stationary state. If these forces are identified, we can use the steady-state conditions found above to immediately identify that both forces must vanish:

$$f_\rho = 0, \tag{7a}$$

$$f_\psi = 0, \tag{7b}$$

which are exact conditions for any system following the constitutive relations in Eq. (6).

One may microscopically derive Eq. (6) for a given system following an Irving-Kirkwood procedure [47] and identify exact microscopic forms for f_ρ and f_ψ. Alternatively, these expressions can be derived variationally for equilibrium systems using classical density functional theory [50]. The resulting forms for f_ρ and f_ψ are often integro-differential equations with nonlocal contributions from long-ranged interparticle interactions and correlations. We aim to circumvent the often difficult task of solving these equations for the complete spatial steady-state order parameter profiles.

To proceed, we perform a general expansion of f_ρ and f_ψ with respect to spatial gradients of the order parameters. We retain spatial gradients up to third order in our expansion of f_ρ (which has odd parity) and up to second order in our expansion of f_ψ (which has even parity). It will become apparent that the parity of these expansions plays a prominent role in the development of coexistence criteria. The accuracy of these expansions will depend on the degree to which any nonlocal terms in these forces can be approximated with local terms.

We first expand the generation-driving force:

$$f_\psi \approx f_\psi^{\text{bulk}} + f_\psi^{\text{int}}, \tag{8a}$$

where

$$\begin{aligned} f_{\psi_i}^{\text{int}} = &- f_{\psi\rho\rho}^{(2,1)} (\partial_z \rho)^2 - 2 f_{\psi\rho\psi}^{(2,1)} \partial_z \rho \partial_z \psi - f_{\psi\psi\psi}^{(2,1)} (\partial_z \psi)^2 \\ &- f_{\psi\rho}^{(2,2)} \partial_{zz}^2 \rho - f_{\psi\psi}^{(2,2)} \partial_{zz}^2 \psi. \end{aligned} \tag{8b}$$

Here, f_ψ^{bulk} is an equation of state that we assume has at least one zero such that a homogeneous steady state is possible. The coefficients $f_{\psi ij}^{(2,1)}$ and $f_{\psi i}^{(2,2)}$ for $i, j \in \{\rho, \psi\}$ are each equations of state that generally depend on the local value of ρ and ψ but not their spatial gradients. We have defined $f_{\psi\rho\psi}^{(2,1)} = f_{\psi\psi\rho}^{(2,1)}$ as antisymmetric contributions do not enter f_ψ^{int}.

We now expand the flux-driving force:

$$f_\rho \approx f_\rho^{(1)} + f_\rho^{(3)}, \tag{9a}$$

where

$$f_\rho^{(1)} = f_{\rho\rho}^{(1,1)} \partial_z \rho + f_{\rho\psi}^{(1,1)} \partial_z \psi, \tag{9b}$$

$$\begin{aligned} f_\rho^{(3)} = &- f_{\rho\rho\rho\rho}^{(3,1)} (\partial_z \rho)^3 - 3 f_{\rho\rho\rho\psi}^{(3,1)} (\partial_z \rho)^2 \partial_z \psi \\ &- 3 f_{\rho\rho\psi\psi}^{(3,1)} (\partial_z \psi)^2 \partial_z \rho - f_{\rho\psi\psi\psi}^{(3,1)} (\partial_z \psi)^3 \\ &- f_{\rho\rho\rho}^{(3,2)} \partial_{zz}^2 \rho \partial_z \rho - f_{\rho\rho\psi}^{(3,2)} \partial_{zz}^2 \rho \partial_z \psi - f_{\rho\psi\rho}^{(3,2)} \partial_{zz}^2 \psi \partial_z \rho \\ &- f_{\rho\psi\psi}^{(3,2)} \partial_{zz}^2 \psi \partial_z \psi - f_{\rho\rho}^{(3,3)} \partial_{zzz}^3 \rho - f_{\rho\psi}^{(3,3)} \partial_{zzz}^3 \psi. \end{aligned} \tag{9c}$$

Each $f_{\rho i}^{(1,1)}$, $f_{\rho i}^{(3,3)}$, $f_{\rho ij}^{(3,2)}$, and $f_{\rho ijk}^{(3,1)}$ for $i, j, k \in \{\rho, \psi\}$ is generally an equation of state that depends on ρ and ψ but not their spatial gradients. Equation (9c) is unaffected by antisymmetric contributions to $f_{\rho ijk}^{(3,1)}$ (with respect to permuting i, j, and k), and thus we define $f_{\rho ijk}^{(3,1)}$ to be symmetric under permuting i, j, and k.

Our goal is to extract bulk coexistence criteria from the steady-state conditions that all forces must vanish. Broadly, forces with even spatial parity, such as f_ψ, contain bulk contributions that do not necessarily vanish in the absence of spatial gradients. It is precisely these bulk contributions that will appear in our coexistence criteria. Conversely, forces with odd spatial parity do not contain a bulk contribution and hence one cannot straightforwardly extract coexistence criteria from

a spatially uniform odd quantity. It is for this reason that coexistence criteria for the $f_\psi = 0$ condition can be immediately obtained, as will be shown, while the coexistence criteria that result from $f_\rho = 0$ require the conversion of this odd force to a quantity with even spatial parity.

We immediately identify n_P coexistence criteria from the force balance on the nonconserved field, $f_\psi = 0$. Substituting the absence of spatial variations of the order parameters in the bulk phases into Eq. (8), we have

$$f_\psi^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = 0 \quad \forall \quad \alpha \in \mathcal{P}. \tag{10}$$

This set of criteria applies to any coexistence scenario with a nonconserved order parameter and, analogously to that in equilibrium [Eq. (1)], corresponds to a generation-free state.

It is now apparent that the conserved field, and consequently f_ρ, is what makes determining the phase diagrams of these systems a nontrivial task. f_ρ is odd and must be transformed to an even quantity in order to extract bulk coexistence criteria. We therefore seek a transformation that allows us to equivalently express zero flux-driving force as spatial uniformity of an even potential-like quantity:

$$f_\rho = 0 \rightarrow \partial_z u_\rho = 0, \tag{11}$$

where we have defined u_ρ to be a "pseudopotential." If such a transformation can be made, we will find that u_ρ is spatially uniform with even spatial parity and therefore coexistence criteria can be extracted from this condition. Before detailing the transformation between the flux-driving force and u_ρ, we expand u_ρ to second order in spatial gradients of the order parameters, consistent with our expansions of f_ρ and f_ψ:

$$u_\rho \approx u_\rho^{\text{bulk}} + u_\rho^{\text{int}}, \tag{12a}$$

where

$$\begin{aligned} u_\rho^{\text{int}} = &- u_{\rho\rho\rho}^{(2,1)}(\partial_z\rho)^2 - 2u_{\rho\rho\psi}^{(2,1)}\partial_z\rho\partial_z\psi - u_{\rho\psi\psi}^{(2,1)}(\partial_z\psi)^2 \\ &- u_{\rho\rho}^{(2,2)}\partial_{zz}^2\rho - u_{\rho\psi}^{(2,2)}\partial_{zz}^2\psi. \end{aligned} \tag{12b}$$

This expansion has the same form as that of f_ψ [Eq. (8)], containing a total of six equations of state in the scalar u_ρ^{bulk}, each $u_{\rho ij}^{(2,1)}$, and each $u_{\rho i}^{(2,2)}$ for $i, j \in \{\rho, \psi\}$. As was the case for $f_{\psi\rho\psi}^{(2,1)}$, we define $u_{\rho\rho\psi}^{(2,1)} = u_{\rho\psi\rho}^{(2,1)}$ as any antisymmetric contributions do not impact u_ρ^{int}.

If we can find a transformation of the form of Eq. (11), we can simply integrate the steady-state condition $\partial_z u_\rho = 0$ to determine coexistence criteria. By again noting that the order parameters are spatially uniform in the bulk phases, we find the following $n_P - 1$ criteria from $\partial_z u_\rho = 0$:

$$u_\rho^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = u_\rho^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{13}$$

where u_ρ^{coexist} is the coexistence value of u_ρ, which is a constant that must be determined. Equation (13) is the nonequilibrium analog of equality of chemical potentials in coexisting phases [Eq. (1)].

We now look to explicitly determine the form of the transformation in Eq. (11) to extract the coexistence criteria in Eq. (13). This transformation must be bijective such that there is a one-to-one mapping between zero flux-driving force and constant u_ρ. To maintain generality, we allow u_ρ to contain contributions from the generation-driving force. Allowing these contributions will prove necessary to ensure one can identify $u_\rho = \mu_\rho$ in equilibrium when the conserved field is the number density. The simplest bijective transformation that contains contributions from the generation-driving force is [21]

$$\mathcal{T}_{\rho\rho}\overline{f}_\rho + \mathcal{T}_{\rho\psi}\psi\partial_z f_\psi = \partial_z u_\rho, \tag{14a}$$

$$\mathcal{T}_{\psi\rho}\overline{f}_\rho + \mathcal{T}_{\psi\psi}\psi\partial_z f_\psi = \partial_z u_\psi, \tag{14b}$$

where $\mathcal{T}_{\rho\rho}$, $\mathcal{T}_{\rho\psi}$, $\mathcal{T}_{\psi\rho}$, and $\mathcal{T}_{\psi\psi}$ are each an equation of state that depends on ρ and ψ but not their spatial gradients and composes the generally asymmetric transformation tensor, $\boldsymbol{\mathcal{T}}$. Importantly, the factor of ψ in front of $\partial_z f_\psi$ can be absorbed into $\mathcal{T}_{\psi\psi}$ and $\mathcal{T}_{\rho\psi}$; however, here we include it to ensure the elements of $\boldsymbol{\mathcal{T}}$ all have the same units. We have introduced a pseudopotential of the nonconserved field, u_ψ, which is simply the negative of the generation-driving force, $u_\psi = -f_\psi$. Consequently, $\mathcal{T}_{\psi\rho} = 0$ and $\mathcal{T}_{\psi\psi} = -\psi^{-1}$. We identify each of the terms in u_ψ as

$$u_\psi \approx u_\psi^{\text{bulk}} + u_\psi^{\text{int}}, \tag{15a}$$

where

$$\begin{aligned} u_\psi^{\text{int}} = &- u_{\psi\rho\rho}^{(2,1)}(\partial_z\rho)^2 - 2u_{\psi\rho\psi}^{(2,1)}\partial_z\rho\partial_z\psi - u_{\psi\psi\psi}^{(2,1)}(\partial_z\psi)^2 \\ &- u_{\psi\rho}^{(2,2)}\partial_{zz}^2\rho - u_{\psi\psi}^{(2,2)}\partial_{zz}^2\psi, \end{aligned} \tag{15b}$$

with

$$u_\psi^{\text{bulk}} \equiv -f_\psi^{\text{bulk}}, \tag{15c}$$

$$u_{\psi ij}^{(2,1)} \equiv -f_{\psi ij}^{(2,1)} \quad \forall \quad i, j \in \{\rho, \psi\}, \tag{15d}$$

$$u_{\psi i}^{(2,2)} \equiv -f_{\psi i}^{(2,2)} \quad \forall \quad i \in \{\rho, \psi\}. \tag{15e}$$

From our definition of u_ψ, we only need to determine u_ρ, $\mathcal{T}_{\rho\rho}$, and $\mathcal{T}_{\rho\psi}$. Therefore, we can focus solely on Eq. (14a), noting that $\mathcal{T}_{\rho\rho}$ must be nonzero for the transformation in Eq. (14a) to be bijective.

In equilibrium and when the conserved field is the number density, u_ρ can be identified as either the chemical potential or pressure. If one identifies u_ρ as the pressure, which is straightforward beginning from our force balance, then $\mathcal{T}_{\rho\rho} = -1$ and $\mathcal{T}_{\rho\psi} = 0$. Alternatively, if one identifies u_ρ as the chemical potential, Eq. (14a) encodes the Gibbs-Duhem relation [Eq. (3)] such that $\mathcal{T}_{\rho\rho} = -1/\rho$ and $\mathcal{T}_{\rho\psi} = 1/\rho$. Hereafter, we use $u_\rho = \mu_\rho$ in equilibrium to simplify the following analysis.

Substituting the expansions for f_ρ, f_ψ, and u_ρ into Eq. (14a), we match terms and find a number of relations that must hold (as detailed in Appendix A 2):

$$\mathcal{T}_{\rho\rho}f_{\rho i}^{(1,1)} + \mathcal{T}_{\rho\psi}\overline{f}_{\psi i}^{(1,1)} = \partial_i u_\rho^{\text{bulk}}, \tag{16a}$$

$$\mathcal{T}_{\rho\rho}f_{\rho ijk}^{(3,1)} + \mathcal{T}_{\rho\psi}\overline{f}_{\psi ijk}^{(3,1)} = \frac{1}{3}\left(\partial_k u_{\rho ij}^{(2,1)} + \partial_j u_{\rho ki}^{(2,1)} + \partial_i u_{\rho jk}^{(2,1)}\right), \tag{16b}$$

$$\mathcal{T}_{\rho\rho}f_{\rho ij}^{(3,2)} + \mathcal{T}_{\rho\psi}\overline{f}_{\psi ij}^{(3,2)} = 2u_{\rho ij}^{(2,1)} + \partial_j u_{\rho i}^{(2,2)}, \tag{16c}$$

$$\partial_j\left(\mathcal{T}_{\rho\rho}f_{\rho i}^{(3,3)} + \mathcal{T}_{\rho\psi}\overline{f}_{\psi i}^{(3,3)}\right) = \partial_j u_{\rho i}^{(2,2)}, \tag{16d}$$

where $\partial_i \equiv \partial/\partial i$ and we have defined

$$\overline{f}_{\psi i}^{(1,1)} = \psi\partial_i f_\psi^{\text{bulk}}, \tag{17a}$$

$$\overline{f}^{(3,1)}_{\psi ijk} = \frac{\psi}{3}\left(\partial_k f^{(2,1)}_{\psi ij} + \partial_j f^{(2,1)}_{\psi ki} + \partial_i f^{(2,1)}_{\psi jk}\right), \tag{17b}$$

$$\overline{f}^{(3,2)}_{\psi ij} = 2\psi f^{(2,1)}_{\psi ij} + \psi \partial_j f^{(2,2)}_{\psi i}, \tag{17c}$$

$$\overline{f}^{(3,3)}_{\psi i} = \psi f^{(2,2)}_{\psi i}, \tag{17d}$$

for $i, j, k \in \{\rho, \psi\}$. We again note that Eq. (16) can be expressed more concisely in vector notation, which is applicable for any n_C and n_N [see Eq. (A12)]. Equation (16) is an overdetermined system of 14 linear, first-order, homogeneous partial differential equations (PDEs) for eight unknown functions ($\mathcal{T}_{\rho\rho}$, $\mathcal{T}_{\rho\psi}$, and each coefficient in u_ρ). We see that the number of equations exceeds the number of unknown functions and hence a solution is *not guaranteed* to exist. This makes clear that the coexistence theories developed for a single conserved field [12,16,39] represent a special case where the system of PDEs in Eq. (16) becomes an exactly determined (and consequently solveable) system of ordinary differential equations. For this reason, the single-field coexistence theories in Refs. [12,16] represent an exceptional case where it is guaranteed that u_ρ exists.

One approach to solving Eq. (16) may be to eliminate the coefficients of u_ρ from each equation through a series of linear operations, obtaining a set of equations that solely contain $\mathcal{T}_{\rho\rho}$, $\mathcal{T}_{\rho\psi}$, and the known force coefficients. If one does this, for $\mathcal{T}_{\rho\rho}$, $\mathcal{T}_{\rho\psi}$, and the coefficients in u_ρ to be continuous and differentiable functions of ρ and ψ, it must be ensured that partial differentiation of these unknown functions with respect to the order parameters is commutative (this is formally known as involutivity [51]). A solution that satisfies Eq. (16) may not have this property. We therefore must determine additional conditions that the solution must satisfy to ensure the commutativity of partial differentiation (these additional equations are formally referred to as compatibility conditions [51]). Determining these conditions is a difficult task but, nevertheless, Eq. (16) serves as the starting point in determining solutions for $\mathcal{T}_{\rho\rho}$, $\mathcal{T}_{\rho\psi}$, and u_ρ. Importantly, a solution to Eq. (16) can be subsequently checked to ensure partial differentiation is commutative. This commutativity is satisfied in passive systems when the pseudopotential of each order parameter is its associated chemical potential, $u_\rho = \mu_\rho$ and $u_\psi = \mu_\psi$, and the transformation tensor has elements $\mathcal{T}_{\psi\psi} = -1/\psi$, $\mathcal{T}_{\psi\rho} = 0$, $\mathcal{T}_{\rho\rho} = -1/\rho$, and $\mathcal{T}_{\rho\psi} = 1/\rho$.

When a full solution to Eq. (16) (one with a full-rank transformation tensor that respects the commutativity of partial differentiation) exists, we can identify u_ρ as

$$u^{\text{bulk}}_\rho = \int \left(\mathcal{T}_{\rho\rho} f^{(1,1)}_{\rho\rho} + \mathcal{T}_{\rho\psi}\overline{f}^{(1,1)}_{\psi\rho}\right) d\rho + \int \left(\mathcal{T}_{\rho\rho} f^{(1,1)}_{\rho\psi} + \mathcal{T}_{\rho\psi}\overline{f}^{(1,1)}_{\psi\psi}\right) d\psi, \tag{18a}$$

$$u^{(2,1)}_{\rho ij} = \frac{1}{2}\left(\mathcal{T}_{\rho\rho} f^{(3,2)}_{\rho ij} + \mathcal{T}_{\rho\psi}\overline{f}^{(3,2)}_{\psi ij} - \partial_j\left[\mathcal{T}_{\rho\rho} f^{(3,3)}_{\rho i} + \mathcal{T}_{\rho\psi}\overline{f}^{(3,3)}_{\psi i}\right]\right), \tag{18b}$$

$$u^{(2,2)}_{\rho i} = \mathcal{T}_{\rho\rho} f^{(3,3)}_{\rho i} + \mathcal{T}_{\rho\psi}\overline{f}^{(3,3)}_{\psi i}, \tag{18c}$$

where $i, j \in \{\rho, \psi\}$. We note that Eq. (18) can again be expressed more compactly in vector notation [see Eq. (A17)]. Combining Eq. (18) with Eq. (15), we now have an expression for both pseudopotentials.

In systems where a full solution to Eq. (16) can be found, we still require an additional set of coexistence criteria, analogous to the equality of pressures in coexisting phases in equilibrium. To identify the remaining criteria, we introduce a generalized Gibbs-Duhem relation [21] that maps differentials in the pseudopotentials to differentials in a global quantity with even spatial parity, $\mathcal{G}$:

$$\mathcal{E}_\rho du_\rho + \mathcal{E}_\psi du_\psi = d\mathcal{G}, \tag{19}$$

where we have introduced the Maxwell construction variables, $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$, which are each an equation of state that depends on ρ and ψ but not their spatial gradients. The connection between these variables and a Maxwell construction will later become apparent; however, we can appreciate that the equilibrium Gibbs-Duhem relation [Eq. (3)] allows us to express equality of pressures across phases [Eq. (1c)] as the equilibrium Maxwell construction [Eq. (2c) or equivalently (4b)].

We now expand $\mathcal{G}$ to second order:

$$\mathcal{G} \approx \mathcal{G}^{\text{bulk}} + \mathcal{G}^{\text{int}}, \tag{20a}$$

where

$$\mathcal{G}^{\text{int}} = -\mathcal{G}^{(2,1)}_{\rho\rho}(\partial_z\rho)^2 - 2\mathcal{G}^{(2,1)}_{\rho\psi}\partial_z\rho\partial_z\psi - \mathcal{G}^{(2,1)}_{\psi\psi}(\partial_z\psi)^2 - \mathcal{G}^{(2,2)}_{\rho}\partial^2_{zz}\rho - \mathcal{G}^{(2,2)}_{\psi}\partial^2_{zz}\psi, \tag{20b}$$

which can again be expressed more concisely in vector notation [see Eq. (A18)]. Each of $\mathcal{G}^{\text{bulk}}$, $\mathcal{G}^{(2,2)}_i$, and $\mathcal{G}^{(2,1)}_{ij}$ for $i, j \in \{\rho, \psi\}$ are equations of state that depend on ρ and ψ but not their spatial gradients. As was the case for $f^{(2,1)}_{\psi ij}$ and $u^{(2,1)}_{\rho ij}$, we define $\mathcal{G}^{(2,1)}_{ij}$ to be symmetric as only the sum $\mathcal{G}^{(2,1)}_{\psi\rho} + \mathcal{G}^{(2,1)}_{\rho\psi}$ impacts $\mathcal{G}^{\text{int}}$.

If a global quantity can be found, our remaining coexistence criteria follow from substituting the spatial uniformity of the pseudopotentials ($\partial_z u_\rho = \partial_z u_\psi = 0$) into the generalized Gibbs-Duhem relation [Eq. (19)], which informs us that $\mathcal{G}$ is spatially constant. As the order parameters are homogeneous within the bulk phases, from Eq. (20) we identify that $\mathcal{G}^{\text{bulk}}$ is equal in coexisting phases:

$$\mathcal{G}^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = \mathcal{G}^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{21}$$

providing our final $n_P - 1$ coexistence criteria, which, in equilibrium, correspond to equality of pressures [Eq. (1c)] between phases.

We now look to determine the components of the Maxwell construction vector. Substituting the expressions for u_ρ, u_ψ, and $\mathcal{G}$ into the generalized Gibbs-Duhem relation [Eq. (19)], we identify a number of relations that must hold (as detailed in Appendix A 3):

$$\mathcal{E}_\rho\partial_i u^{\text{bulk}}_\rho + \mathcal{E}_\psi\partial_i u^{\text{bulk}}_\psi = \partial_i\mathcal{G}^{\text{bulk}}, \tag{22a}$$

$$\partial_j\left(\mathcal{E}_\rho u^{(2,2)}_{\rho i} + \mathcal{E}_\psi u^{(2,2)}_{\psi i}\right) = \partial_j\mathcal{G}^{(2,2)}_i, \tag{22b}$$

$$\mathcal{E}_\rho\left(2u^{(2,1)}_{\rho ij} + \partial_j u^{(2,2)}_{\rho i}\right) + \mathcal{E}_\psi\left(2u^{(2,1)}_{\psi ij} + \partial_j u^{(2,2)}_{\psi i}\right) = 2\mathcal{G}^{(2,1)}_{ij} + \partial_j\mathcal{G}^{(2,2)}_i, \tag{22c}$$

$$\mathcal{E}_\rho\left(\partial_k u^{(2,1)}_{\rho ij} + \partial_j u^{(2,1)}_{\rho ki} + \partial_i u^{(2,1)}_{\rho jk}\right) + \mathcal{E}_\psi\left(\partial_k u^{(2,1)}_{\psi ij} + \partial_j u^{(2,1)}_{\psi ki} + \partial_i u^{(2,1)}_{\psi jk}\right) = \partial_k \mathcal{G}^{(2,1)}_{ij} + \partial_j \mathcal{G}^{(2,1)}_{ki} + \partial_i \mathcal{G}^{(2,1)}_{jk}, \tag{22d}$$

where $i, j, k \in \{\rho, \psi\}$. Just as was the case with Eq. (16), Eq. (22) can be expressed in vector notation [see Eq. (A23)] and is overdetermined with 14 linear, first-order, homogeneous PDEs for eight unknown functions ($\mathcal{E}_\rho$, $\mathcal{E}_\psi$, and each coefficient in $\mathcal{G}$). We can again solve Eq. (22) for $\mathcal{E}_\rho$, $\mathcal{E}_\psi$, and $\mathcal{G}$ and subsequently check if partial differentiation with respect to the order parameters is commutative. In the Supplemental Material [52], we differentiate Eq. (22) to find the conditions that ensure partial differentiation is commutative (this is formally known as prolongation [51]), gaining additional PDEs (compatibility conditions) that must be satisfied. We emphasize that even though we may find the additional equations that ensure partial differentiation is commutative, the system of PDEs is overdetermined and hence one may not be able to find a consistent solution to every equation: A solution is *not guaranteed*. When a full solution to Eq. (22) exists, we can identify the form of $\mathcal{G}$:

$$\mathcal{G}^{\text{bulk}} = \mathcal{E}_\rho u^{\text{bulk}}_\rho + \mathcal{E}_\psi u^{\text{bulk}}_\psi - \int u^{\text{bulk}}_\rho d\mathcal{E}_\rho - \int u^{\text{bulk}}_\psi d\mathcal{E}_\psi, \tag{23a}$$

$$\mathcal{G}^{(2,1)}_{ij} = \mathcal{E}_\rho u^{(2,1)}_{\rho ij} + \mathcal{E}_\psi u^{(2,1)}_{\psi ij} - \frac{1}{2}\left(\partial_i \mathcal{E}_\rho u^{(2,2)}_{\rho j} + \partial_i \mathcal{E}_\psi u^{(2,2)}_{\psi j}\right), \tag{23b}$$

$$\mathcal{G}^{(2,2)}_{i} = \mathcal{E}_\rho u^{(2,2)}_{\rho i} + \mathcal{E}_\psi u^{(2,2)}_{\psi i}, \tag{23c}$$

where $i, j \in \{\rho, \psi\}$. Expressions for these coefficients in vector notation can be found in Appendix A 3 in Eq. (A27). For Eq. (21) to provide a distinct criterion, G^{bulk} must not be expressible solely in terms of u^{bulk}_ρ and u^{bulk}_ψ (formally, G^{bulk} cannot factor through u^{bulk}_ρ and u^{bulk}_ψ). Two examples where equality of $\mathcal{G}^{\text{bulk}}$ is not distinct from equality of u^{bulk}_ρ and $u^{\text{bulk}}_\psi = 0$ are (1) when $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are constants, which leads to $\mathcal{G}^{\text{bulk}}$ being a linear combination of the bulk pseudopotentials, and (2) when $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are linear functions of u^{bulk}_ρ and u^{bulk}_ψ, which causes G^{bulk} to be a quadratic function of the bulk pseudopotentials.

In equilibrium, the generalized Gibbs-Duhem relation [Eq. (19)] reduces to its equilibrium form [Eq. (3)], where the pseudopotentials are the chemical potentials, $u_\rho = \mu_\rho$ and $u_\psi = \mu_\psi$, the global quantity is the pressure, $\mathcal{G} = p$, and the Maxwell construction variables are the order parameters, $\mathcal{E}_\rho = \rho$ and $\mathcal{E}_\psi = \psi$. Notably, the global quantity appears to only have a clear mechanical interpretation in equilibrium.

Our final nonequilibrium coexistence criteria are

$$u^{\text{bulk}}_\rho(\rho^\alpha, \psi^\alpha) = u^{\text{coexist}}_\rho \quad \forall \quad \alpha \in \mathcal{P}, \tag{24a}$$

$$u^{\text{bulk}}_\psi(\rho^\alpha, \psi^\alpha) = 0 \quad \forall \quad \alpha \in \mathcal{P}, \tag{24b}$$

$$\mathcal{G}^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = \mathcal{G}^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{24c}$$

where u^{coexist}_ρ and $\mathcal{G}^{\text{coexist}}$ are generally nonvanishing and must be determined by equating u^{bulk}_ρ and $\mathcal{G}^{\text{bulk}}$ across phases, respectively. Subtracting the number of equations in Eq. (24), $2n_P - 2 + n_P$, from the number of variables describing the coexisting phases, $2n_P$, we find the number of degrees of freedom to be $2 - n_P$, recovering the equilibrium Gibbs phase rule as expected [21].

Importantly, the coexistence criteria in Eq. (24) are a form of the steady-state conditions [Eq. (7)] and thus may describe even an unstable steady state. Once the values of ρ and ψ in the bulk phases are determined, the stability of these phases can be determined through linear stability analysis [53–55]. When one or more phases are unstable, this can lead to the coexistence of stationary, spatially uniform phases with lamellar and temporally oscillating phases [18]. Furthermore, the interface between coexisting phases can be unstable toward, for example, drifting states [17,56] or states with small-scale patterning along the interface [53]. We emphasize that our theory, and the criteria in Eq. (24), can serve as the starting point for describing these kinds of systems in addition to enabling the prediction of the binodals that characterize stable coexisting stationary homogeneous phases.

C. Alternative forms of nonequilibrium criteria

In an analogous manner to the equilibrium Maxwell construction [Eq. (2c) or equivalently Eq. (4b)], we can use the generalized Gibbs-Duhem relation [Eq. (19)] to re-express equality of $\mathcal{G}^{\text{bulk}}$ across phases as a generalized equal-area Maxwell construction:

$$\int_{\mathcal{E}^\alpha_\rho}^{\mathcal{E}^\beta_\rho} \left[u^{\text{bulk}}_\rho(\rho, \psi) - u^{\text{coexist}}_\rho\right] d\mathcal{E}_\rho + \int_{\mathcal{E}^\alpha_\psi}^{\mathcal{E}^\beta_\psi} u^{\text{bulk}}_\psi(\rho, \psi) d\mathcal{E}_\psi = 0 \quad \forall \quad \alpha, \beta \in \mathcal{P}, \tag{25a}$$

where the integration is path-independent as it is equivalent to an integral over $-d\mathcal{G}^{\text{bulk}}$ between the α and β phases. This path independence allows us to select the most convenient path to integrate, which is often $\psi^*(\rho)$ satisfying $u_\psi(\rho, \psi^*) = 0$ for all ρ considered. This path selection reduces Eq. (25a) to a one-dimensional integral:

$$\int_{\mathcal{E}^\alpha_\rho}^{\mathcal{E}^\beta_\rho} \left[u^{\text{bulk}}_\rho(\rho, \psi^*(\rho)) - u^{\text{coexist}}_\rho\right] d\mathcal{E}_\rho = 0 \quad \forall \quad \alpha, \beta \in \mathcal{P}. \tag{25b}$$

Equation (25) is the nonequilibrium generalization of Eqs. (2c) and (4b).

If $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are not strictly monotonic functions of ρ and ψ, there is ambiguity in evaluating the generalized Maxwell constructions in Eq. (25). This ambiguity is eliminated by expressing these criteria as "weighted-area" constructions, substituting $d\mathcal{E}_\rho = \partial_\rho \mathcal{E}_\rho d\rho + \partial_\psi \mathcal{E}_\rho d\psi$ and $d\mathcal{E}_\psi = \partial_\rho \mathcal{E}_\psi d\rho + \partial_\psi \mathcal{E}_\psi d\psi$ into Eq. (25a):

$$\int_{\rho^\alpha}^{\rho^\beta} \left[u^{\text{bulk}}_\rho(\rho, \psi) - u^{\text{coexist}}_\rho\right] \partial_\rho \mathcal{E}_\rho d\rho + \int_{\psi^\alpha}^{\psi^\beta} \left[u^{\text{bulk}}_\rho(\rho, \psi) - u^{\text{coexist}}_\rho\right] \partial_\psi \mathcal{E}_\rho d\psi + \int_{\rho^\alpha}^{\rho^\beta} u^{\text{bulk}}_\psi(\rho, \psi) \partial_\rho \mathcal{E}_\psi d\rho + \int_{\psi^\alpha}^{\psi^\beta} u^{\text{bulk}}_\psi(\rho, \psi) \partial_\psi \mathcal{E}_\psi d\psi = 0, \tag{26a}$$

for $\alpha, \beta \in \mathcal{P}$, which [see Eq. (A21b) for a more compact form of this expression], along the path $\psi^*(\rho)$, reduces to

$$\int_{\rho^\alpha}^{\rho^\beta} \left[u_\rho^{\text{bulk}}(\rho, \psi(\rho)^*) - u_\rho^{\text{coexist}}\right]\left(\partial_\rho \mathcal{E}_\rho + \partial_\psi \mathcal{E}_\rho \frac{d\psi^*}{d\rho}\right) d\rho = 0. \tag{26b}$$

When both $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are nonconstant, Eq. (24) can be straightforwardly recast as a generalized common tangent construction on a suitably defined effective bulk free energy, $w^{\text{bulk}} \equiv \int u_\rho^{\text{bulk}} d\mathcal{E}_\rho + \int u_\psi^{\text{bulk}} d\mathcal{E}_\psi$, with respect to $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$:

$$\left.\frac{\partial w^{\text{bulk}}}{\partial \mathcal{E}_\rho}\right|_{\rho=\rho^\alpha, \psi=\psi^\alpha} = u_\rho^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}, \tag{27a}$$

$$\left.\frac{\partial w^{\text{bulk}}}{\partial \mathcal{E}_\psi}\right|_{\rho=\rho^\alpha, \psi=\psi^\alpha} = 0 \quad \forall \quad \alpha \in \mathcal{P}, \tag{27b}$$

$$u_\rho^{\text{coexist}} \mathcal{E}_\rho(\rho^\alpha, \psi^\alpha) - w^{\text{bulk}}(\rho^\alpha, \psi^\alpha) = \mathcal{G}^{\text{coexist}} \quad \forall \quad \alpha \in \mathcal{P}. \tag{27c}$$

The definition of w^{bulk} requires its Hessian to be symmetric, which is guaranteed if the Hessian of $\mathcal{G}^{\text{bulk}}$ is symmetric. It is then clear that when the generalized Gibbs-Duhem relation is satisfied and both $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are nonconstant, the coexistence criteria can be expressed as a common tangent construction on w^{bulk} with respect to $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$. While Eq. (27) is an appealing form of the criteria, only in equilibrium can it be derived from an extremization of w^{bulk} subject to the appropriate physical constraints. It is clear from Ref. [21] (see Appendix C in that work) that the conditions for defining an effective free energy functional, $\mathcal{W}[\rho(z), \psi(z)]$ such that $u_i = \delta\mathcal{W}[\rho(z), \psi(z)]/\delta\mathcal{E}_i$ for $i \in \{\rho, \psi\}$, are more restrictive than the conditions necessary for $\mathcal{G}$ to exist [Eq. (22)]. This contrasts the case in which there is a single (conserved) order parameter, where Solon *et al.* [12] demonstrated that a free energy functional can be readily defined.

D. Approximations to nonequilibrium criteria

While the criteria described in Secs. II B and II C constitute, to the best of our knowledge, the first set of genuinely nonequilibrium coexistence criteria with coupled conserved and nonconserved order parameters, exact solutions for u_ρ and $\mathcal{G}$ may not always be possible. For systems in which formally exact solutions cannot be obtained but macroscopic phase separation remains a steady state, we look to develop *approximate* coexistence criteria that one can use to estimate the phase boundaries. To do so, we consider systems where u_ρ, $\mathcal{T}_{\rho\rho}$, and $\mathcal{T}_{\rho\psi}$ can be found [and thus a solution to Eq. (16) exists]. In these systems, the final set of coexistence criteria corresponds to equality of $\mathcal{G}^{\text{bulk}}$ across phases and relies on a solution to Eq. (22), which does not necessarily exist. In systems where we cannot find a full solution to Eq. (22), we propose partitioning the generalized Gibbs-Duhem relation into bulk and interfacial contributions (as detailed in Appendix A 4):

$$\mathcal{E}_\rho^{\text{bulk}} du_\rho^{\text{bulk}} + \mathcal{E}_\psi^{\text{bulk}} du_\psi^{\text{bulk}} = d\mathcal{G}^{\text{bulk}}, \tag{28a}$$

$$\mathcal{E}_\rho^{\text{int}} du_\rho^{\text{bulk}} + \mathcal{E}_\psi^{\text{int}} du_\psi^{\text{bulk}} = d\mathcal{G}^{\text{int}}, \tag{28b}$$

such that $\mathcal{E}_\rho^{\text{bulk}}$ and $\mathcal{E}_\psi^{\text{bulk}}$ satisfy Eq. (22a) and $\mathcal{E}_\rho^{\text{int}}$ and $\mathcal{E}_\psi^{\text{int}}$ satisfy Eqs. (22b)–(22d). Only when $\mathcal{E}_i = \mathcal{E}_i^{\text{bulk}} = \mathcal{E}_i^{\text{int}}$ for $i \in \{\rho, \psi\}$ is the global quantity rigorously defined with Eq. (22). When this is the case, the generalized Gibbs-Duhem relation [Eq. (19)] holds and $\mathcal{G}^{\text{bulk}}$ [Eq. (23a) with $\mathcal{E}_\rho \to \mathcal{E}_\rho^{\text{bulk}}$ and $\mathcal{E}_\psi \to \mathcal{E}_\psi^{\text{bulk}}$] is equal in coexisting phases. Otherwise, when either $\mathcal{E}_\rho^{\text{bulk}} \neq \mathcal{E}_\rho^{\text{int}}$ or $\mathcal{E}_\psi^{\text{bulk}} \neq \mathcal{E}_\psi^{\text{int}}$, $\mathcal{G}^{\text{bulk}}$ generally takes a different value in each of the coexisting phases.

As shown in Appendix A 5, the weighted-area construction [Eq. (26a)] using $\mathcal{E}_\rho^{\text{int}}$ and $\mathcal{E}_\psi^{\text{int}}$ is guaranteed to vanish when evaluated along the spatial coexistence profiles that connect coexisting α and β phases, $\rho^{\text{c}}(z)$ and $\psi^{\text{c}}(z)$, if a solution for $\mathcal{E}_\rho^{\text{int}}$ and $\mathcal{E}_\psi^{\text{int}}$ [Eqs. (22b)–(22d)] can be found:

$$\int_{z^\alpha}^{z^\beta} \left[\left(u_\rho^{\text{bulk}}(\rho^{\text{c}}, \psi^{\text{c}}) - u_\rho^{\text{coexist}}\right)\left(\partial_\rho \mathcal{E}_\rho^{\text{int}} \partial_z \rho^{\text{c}} + \partial_\psi \mathcal{E}_\rho^{\text{int}} \partial_z \psi^{\text{c}}\right) + u_\psi^{\text{bulk}}(\rho^{\text{c}}, \psi^{\text{c}})\left(\partial_\rho \mathcal{E}_\psi^{\text{int}} \partial_z \rho^{\text{c}} + \partial_\psi \mathcal{E}_\psi^{\text{int}} \partial_z \psi^{\text{c}}\right)\right] dz = 0, \tag{29}$$

where z^α and z^β are positions deep in the homogeneous α and β phases, respectively, and $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are each evaluated along $\rho^{\text{c}}(z)$ and $\psi^{\text{c}}(z)$. A more concise form of this expression can again be found in Eq. (A31), where vector notation is used. The value of this integral is now path-dependent, as it is no longer the difference in $\mathcal{G}^{\text{bulk}}$ between phases. Importantly, $\rho^{\text{c}}(z)$ and $\psi^{\text{c}}(z)$ are the full spatial coexistence profiles that we set out to avoid determining. We can then gain approximate coexistence criteria by performing the weighted-area construction [Eq. (26b)] with $\mathcal{E}_\rho \to \mathcal{E}_\rho^{\text{int}}$, evaluating the integral along a path other than the generally unknown $(\rho^{\text{c}}, \psi^{\text{c}})$ such as $(\rho, \psi^*(\rho))$ for any ρ connecting ρ^α and ρ^β. However, the weighted-area construction is not guaranteed to vanish along these other paths, where its value quantifies the departure of the criteria from exactness. Importantly, the approximation gains accuracy as the selected integration path approaches the actual coexistence profiles.

The approximate criteria in Eq. (29) makes use of $\mathcal{E}_\rho^{\text{int}}$ and $\mathcal{E}_\psi^{\text{int}}$; however, we can alternatively approximate the final set of criteria by setting $\mathcal{G}^{\text{bulk}}$ equal across phases, i.e., using the weighted-area construction [Eq. (26a)] with $\mathcal{E}_\rho \to \mathcal{E}_\rho^{\text{bulk}}$ and $\mathcal{E}_\psi \to \mathcal{E}_\psi^{\text{bulk}}$. When using $\mathcal{E}_\rho \to \mathcal{E}_\rho^{\text{bulk}}$ and $\mathcal{E}_\psi \to \mathcal{E}_\psi^{\text{bulk}}$, the weighted-area construction is path-independent as it is equal to the difference in $\mathcal{G}^{\text{bulk}}$ between phases; however, this difference is generally finite when $\mathcal{E}_i^{\text{bulk}} \neq \mathcal{E}_i^{\text{int}}$ for any $i \in \{\rho, \psi\}$. This approach, in contrast to Eq. (29), does not inherently contain a degree of freedom (such as the integration path) to change the quality of the approximation.

In the event that one can access the coexistence profile to check the approximation, the difference in $\mathcal{G}^{\text{bulk}}$ between α and β phases can be determined (as shown in Appendix A 6) with

$$\begin{aligned}\Delta^{\alpha\beta}\mathcal{G}^{\text{bulk}} = \int_{z^\alpha}^{z^\beta} \Big[&\left(u_\rho^{\text{bulk}}(\rho^{\text{c}}, \psi^{\text{c}}) - u_\rho^{\text{coexist}}\right) \\ &\times \left(\partial_\rho\left(\mathcal{E}_\rho^{\text{bulk}} - \mathcal{E}_\rho^{\text{int}}\right)\partial_z \rho^{\text{c}} + \partial_\psi\left(\mathcal{E}_\rho^{\text{bulk}} - \mathcal{E}_\rho^{\text{int}}\right)\partial_z \psi^{\text{c}}\right) \\ &+ u_\psi^{\text{bulk}}(\rho^{\text{c}}, \psi^{\text{c}})\left(\partial_\rho\left(\mathcal{E}_\psi^{\text{bulk}} - \mathcal{E}_\psi^{\text{int}}\right)\partial_z \rho^{\text{c}}\right. \\ &\left. + \partial_\psi\left(\mathcal{E}_\psi^{\text{bulk}} - \mathcal{E}_\psi^{\text{int}}\right)\partial_z \psi^{\text{c}}\right)\Big] dz,\end{aligned} \tag{30}$$

where we again refer the reader to Eq. (A32) for a more compact form of this expression. When $\Delta^{\alpha\beta}\mathcal{G}^{\text{bulk}} \approx 0$ for every pair of coexisting phases, equating $\mathcal{G}^{\text{bulk}}$ across phases is a good approximation for the final set of coexistence criteria.

III. APPLICATION TO NONEQUILIBRIUM MODELS

In this section, we introduce phenomenological models of nonequilibrium field dynamics and numerically obtain the phase diagrams of these systems by determining the complete spatial order parameter profiles. This allows us to directly evaluate our theory of nonequilibrium coexistence, which enables the construction of phase diagrams solely from bulk equations of state.

A number of phenomenological nonequilibrium field theories have been proposed to describe systems with a single nonconserved field, such as active model A [57,58], a single conserved field, such as active model B+[9,46,57], and coupled conserved fields, such as the nonreciprocal Cahn-Hilliard (NRCH) model [56,59]. Additionally, active model C [40–42] has been used to study a conserved field coupled to a nonconserved field in active nematics.

Here, we consider a more general form of AMC, where the deterministic (noise will not affect the coexistence criteria) dynamics of ρ and ψ are

$$\partial_t \rho = \partial_z (L \partial_z u_\rho), \tag{31a}$$

$$\partial_t \psi = -M u_\psi. \tag{31b}$$

It is evident that we have chosen $\mathcal{T}_{\rho\rho} = 1$ and $\mathcal{T}_{\rho\psi} = 0$, however, as L can be any positive equation of state, this model can describe any system where $\mathcal{T}_{\rho\rho}(\rho, \psi) > 0$ and $\mathcal{T}_{\rho\psi} = 0$ by redefining $L \to L/\mathcal{T}_{\rho\rho}$. The pseudopotentials take the form

$$u_\rho = \frac{\delta \mathcal{F}}{\delta \rho} + \tau_\rho^N - \lambda_{\rho\rho\rho}(\partial_z\rho)^2 - 2\lambda_{\rho\rho\psi}\partial_z\rho\partial_z\psi - \lambda_{\rho\psi\psi}(\partial_z\psi)^2 - \pi^{\text{A}}\partial_{zz}^2\psi, \tag{31c}$$

$$u_\psi = \frac{\delta \mathcal{F}}{\delta \psi} + \tau_\psi^N - \lambda_{\psi\rho\rho}(\partial_z\rho)^2 - 2\lambda_{\psi\rho\psi}\partial_z\rho\partial_z\psi - \lambda_{\psi\psi\psi}(\partial_z\psi)^2 + \pi^{\text{A}}\partial_{zz}^2\rho, \tag{31d}$$

$$\mathcal{F}[\rho(z), \psi(z)] = \int_V d\mathbf{r}\Big[f^{\text{bulk}} + \frac{\kappa_{\rho\rho}}{2}(\partial_z\rho)^2 + \kappa_{\rho\psi}\partial_z\rho\partial_z\psi + \frac{\kappa_{\psi\psi}}{2}(\partial_z\psi)^2\Big], \tag{31e}$$

where f^{bulk}, π^{A}, and each κ_{ij}, τ_i^N, and λ_{ijk} (for $i, j, k \in \{\rho, \psi\}$) are generally equations of state that depend on ρ and ψ but not their spatial gradients.

Each pseudopotential, for both the conserved and nonconserved field, contains four contributions. The first is variational in that it is derived from a free energy functional, $\mathcal{F}$, which would be the system free energy in the absence of the other terms. The next three contributions are nonvariational, the first of which is each τ_i^N that captures contributions to u_i^{bulk} that cannot be integrated into the bulk portion of the would-be free energy, f^{bulk}. Each nonzero λ_{ijk} breaks the relationship between interfacial terms in the associated u_i^{int} that result from the functional derivative. Finally, π^{A} captures nonreciprocity in the interfacial coefficients of the pseudopotentials that cannot result from a functional derivative. Reference [21] considers species pseduopotentials in multicomponent active model B that are analogous to those in Eq. (31). In the Supplemental Material of that work, it is shown that this form of the pseudopotentials preserves clear limits of gradient and passive dynamics while capturing all possible bulk and second-order contributions to the *immediately identifiable* pseudopotentials (i.e., beginning with $\mathcal{T}_{\rho\rho} = 1$ and $\mathcal{T}_{\rho\psi} = 0$).

Passive model C dynamics [48] are achieved by setting $\pi^{\text{A}} = 0$, $\tau_i^N = 0$, and $\lambda_{ijk} = 0$ for every $i, j, k \in \{\rho, \psi\}$. If any of these terms are nonzero, the passive structure of the dynamics is broken.

We consider the bulk contributions to the pseudopotentials to result from

$$f^{\text{bulk}} = -0.6\rho^2 + \frac{\psi^2}{2} + \rho^4 + \chi\rho\psi, \tag{32a}$$

$$\tau_\rho^N = \alpha\psi, \tag{32b}$$

$$\tau_\psi^N = -\alpha\rho, \tag{32c}$$

where f^{bulk} appears in the free energy functional [see Eq. (31e)] and χ and α are constants. We assume that π^{A} and every κ_{ij} and λ_{ijk} (where $i, j, k \in \{\rho, \psi\}$) are constant and, moreover, that every $\lambda_{\psi jk} = 0$. For convenience, we define $\overline{\kappa}_{\rho\psi} = \kappa_{\rho\psi} + \pi^{\text{A}}$, $\overline{\kappa}_{\rho\psi} = \kappa_{\rho\psi} - \pi^{\text{A}}$, $\overline{\kappa}_{\rho\rho} = \kappa_{\rho\rho}$, and $\overline{\kappa}_{\psi\psi} = \kappa_{\psi\psi}$.

We first consider systems where every $\lambda_{\rho jk} = 0$ and therefore nonequilibrium effects are solely due to $\alpha \neq 0$ and $\overline{\kappa}_{\rho\psi} \neq \overline{\kappa}_{\psi\rho}$. These dynamics resemble the two-component NRCH model [56,59] except one of the components is nonconserved. When the parameters of this model are constrained such that $\overline{\kappa}_{\rho\psi}(\chi - \alpha) = \overline{\kappa}_{\psi\rho}(\chi + \alpha)$, it has been shown that the pseudopotentials can be redefined such that the equilibrium coexistence criteria [Eq. (1)] are exact [17–19] despite the nonequilibrium nature of the system dynamics. This is due to the fact that these systems have "spurious" gradient dynamics [23]. We aim to recover this result using our theory while extending the coexistence criteria to systems where $\overline{\kappa}_{\rho\psi}(\chi - \alpha) \neq \overline{\kappa}_{\psi\rho}(\chi + \alpha)$.

We now look to solve Eq. (22) for $\boldsymbol{\mathcal{E}} \equiv [\mathcal{E}_\rho \quad \mathcal{E}_\psi]^T$ and $\mathcal{G}$, augmented with the compatibility conditions provided in the Supplemental Material [52]. For arbitrary values of the parameters, these equations do not have a general solution. However, solving for $\boldsymbol{\mathcal{E}}^{\text{bulk}}$ and $\boldsymbol{\mathcal{E}}^{\text{int}}$ independently (as described in Appendix A 4), where $\boldsymbol{\mathcal{E}}^\gamma \equiv [\mathcal{E}_\rho^\gamma \quad \mathcal{E}_\psi^\gamma]^T$ for $\gamma \in \{\text{bulk}, \text{int}\}$, we find

$$\boldsymbol{\mathcal{E}}^{\text{bulk}} = \Big[\rho \quad \tfrac{\chi+\alpha}{\chi-\alpha}\psi\Big]^T, \tag{33a}$$

$$\boldsymbol{\mathcal{E}}^{\text{int}} = \Big[\rho \quad \tfrac{\overline{\kappa}_{\rho\psi}}{\overline{\kappa}_{\psi\rho}}\psi\Big]^T. \tag{33b}$$

If $\overline{\kappa}_{\rho\psi}(\chi - \alpha) = \overline{\kappa}_{\psi\rho}(\chi + \alpha)$, it is clear that $\boldsymbol{\mathcal{E}}^{\text{bulk}} = \boldsymbol{\mathcal{E}}^{\text{int}}$ and the coexistence criteria in Eq. (24) are exact. This is the limit where the system experiences spurious gradient dynamics [18,23] and where our coexistence criteria are equivalent to those in Refs. [17–19]. If $\overline{\kappa}_{\rho\psi}(\chi - \alpha) \neq \overline{\kappa}_{\psi\rho}(\chi + \alpha)$, we approximate the criterion corresponding to equality of $\mathcal{G}^{\text{bulk}}$ ($n_P = 2$ and hence this is a single criterion) [Eq. (24c)] by defining $\mathcal{G}^{\text{bulk}}$ through Eq. (23a) and using $\boldsymbol{\mathcal{E}} \to \boldsymbol{\mathcal{E}}^{\text{bulk}}$.

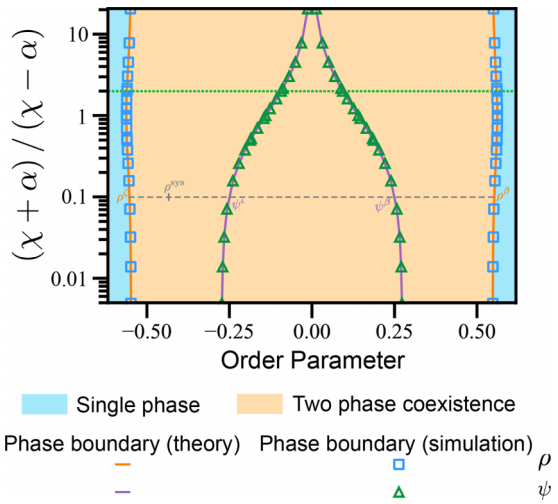


FIG. 2. Phase diagram of AMC, displaying the coexistence values of each order parameter (ρ and ψ) as a function of the ratio $(\chi+\alpha)/(\chi-\alpha)$. We set $\overline{\kappa}_{\rho\rho}=\overline{\kappa}_{\psi\psi}=\overline{\kappa}_{\rho\psi}=0.01$, $\overline{\kappa}_{\psi\rho}=0.005$, and $\chi=1/2$. The dotted green line indicates the limit where $\overline{\kappa}_{\rho\psi}(\chi-\alpha)=\overline{\kappa}_{\psi\rho}(\chi+\alpha)=2$ and the coexistence criteria are exact. A tieline at $\alpha=-0.45/1.1$ is shown in a dashed gray line, where the predicted values of ρ and ψ in the ζ and β phases (defined by $\rho^\beta>\rho^\zeta$) are, respectively, shown in orange and purple. An example global system density, $\rho^{\rm sys}$, where phase separation would occur is shown in gray. The volume fraction of the ζ phase, ϕ^ζ (by definition $\phi^\beta=1-\phi^\zeta$), is set through the level rule $\phi^\zeta=(\rho^\beta-\rho^{\rm sys})/(\rho^\beta-\rho^\zeta)$.

Using our exact and approximate coexistence criterion, we predict the phase diagram of this system and compare it to that obtained by numerically determining the full coexistence profiles, $\rho^c(z)$ and $\psi^c(z)$, following Refs. [52,60–62]. Figure 2 shows the coexistence values of the order parameters as a function of the ratio $(\chi+\alpha)/(\chi-\alpha)$, demonstrating that our exact and approximate criterion both provide highly accurate predictions. To understand the accuracy of the approximate criterion, in Appendix B 1 we compute $\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk}$ [Eq. (30)]:

$$\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk}=\left(\frac{\chi+\alpha}{\chi-\alpha}-\frac{\overline{\kappa}_{\rho\psi}}{\overline{\kappa}_{\psi\rho}}\right)\int_{z^\alpha}^{z^\beta}u_\psi^{\rm bulk}(\rho^{\rm c},\psi^{\rm c})\partial_z\psi^{\rm c}dz. \tag{34}$$

We find $\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk}$ to be approximately zero, justifying equating $\mathcal{G}^{\rm bulk}$ across phases as an approximate criterion when $\overline{\kappa}_{\rho\psi}(\chi-\alpha)\neq\overline{\kappa}_{\psi\rho}(\chi+\alpha)$.

While the system in Fig. 2 provided a minimal platform to explore nonequilibrium coexistence criteria, both exact and approximate, each element of the Maxwell construction vector(s) is linear in the order parameters, just as with passive systems. We seek a system where the Maxwell construction vector is a nonlinear function of the order parameters to further validate our theory. This is the case when $\overline{\kappa}_{\rho\psi}=\overline{\kappa}_{\psi\rho}=\lambda_{\rho\psi\psi}=\lambda_{\rho\rho\psi}=0$:

$$\boldsymbol{\mathcal{E}}^{\rm bulk}=\left[\exp\left(\tfrac{2\lambda_{\rho\rho\rho}}{\overline{\kappa}_{\rho\rho}}\rho\right)\quad\tfrac{\chi+\alpha}{\chi-\alpha}\exp\left(\tfrac{2\lambda_{\rho\rho\rho}}{\overline{\kappa}_{\rho\rho}}\rho\right)\right]^T, \tag{35a}$$

$$\boldsymbol{\mathcal{E}}^{\rm int}=\left[\exp\left(\tfrac{2\lambda_{\rho\rho\rho}}{\overline{\kappa}_{\rho\rho}}\rho\right)\quad 0\right]^T. \tag{35b}$$

We see that $\boldsymbol{\mathcal{E}}^{\rm bulk}=\boldsymbol{\mathcal{E}}^{\rm int}$ when $\alpha=-\chi$ and hence the coexistence criteria are exact. We predict the phase diagram using our criteria when $\boldsymbol{\mathcal{E}}^{\rm bulk}=\boldsymbol{\mathcal{E}}^{\rm int}$ in Fig. 3(a), finding excellent agreement compared to numerical results. In Appendix B 2, we offer further support that our coexistence criteria are exact in this system by keeping the ratio $\lambda_{\rho\rho\rho}/\overline{\kappa}_{\rho\rho}$ fixed while tuning the raw value of these interfacial parameters, finding that the shape of the interface changes but the binodals do not.

We require an approximate criterion when $\alpha\neq-\chi$ (correspondingly $\boldsymbol{\mathcal{E}}^{\rm bulk}\neq\boldsymbol{\mathcal{E}}^{\rm int}$), for which we may use $\boldsymbol{\mathcal{E}}^{\rm int}$ in a weighted-area construction [Eq. (29)] along the path $\psi^*(\rho)$ [63]. We use this as the approximate coexistence criterion, again finding excellent agreement with the numerically determined phase diagram as shown in Fig. 3(b). In Appendix B 3, we provide numerical support for using this approximate criterion by demonstrating that the weighted-area construction in Eq. (29) is similar in value when evaluated along $\psi^*(\rho)$ and the full spatial coexistence profile, $\rho^{\rm c}(z)$ and $\psi^{\rm c}(z)$.

When $\overline{\kappa}_{\rho\psi}\neq0$ or $\lambda_{\rho jk}\neq$ for more elements than just $j=k=\rho$, $\boldsymbol{\mathcal{E}}^{\rm int}$ cannot be determined. There is no well-defined approximate final criterion in this case; however, the system retains coexistence as a steady state. Strikingly, using a form of $\boldsymbol{\mathcal{E}}^{\rm int}$ that only solves a subset of the required equations [i.e., not every equation in Eqs. (22b)–(22d) is satisfied] in the weighted-area construction still provides quantitatively accurate predictions as shown in Fig. 3(c). It is clear from Fig. 4 that these predictions are accurate within 2% of the binodals and are approximately 20 times more accurate than the predictions made using the equilibrium Maxwell construction. Moreover, Fig. 4 shows that the exact and (well-defined) approximate predictions in Figs. 3(a) and 3(b) are an order of magnitude more accurate than the predictions in Fig. 3(c), respectively, having percent errors within <0.2% and <0.8%, while the predictions made using the equilibrium Maxwell construction can reach errors close to 40%. This demonstrates that, as anticipated, the predictions made using the exact final coexistence criterion [Fig. 3(a)] are more accurate than those made using the well-defined approximate criterion [Fig. 3(b)], which are more accurate than those made using the poorly defined approximate criterion [Fig. 3(c)]. We emphasize that the predictions made using our dynamical theory, even when approximate, are significantly more accurate than those made using the equilibrium Maxwell construction.

The phase diagram in Fig. 2 is symmetric about the vertical line at $\rho=\psi=0$. This is due to the symmetry of the model when every $\lambda_{ijk}=0$, where inverting the order parameter values, $\rho\to-\rho$ and $\psi\to-\psi$, always results in inverting the pseudopotentials, $u_\rho\to-u_\rho$ and $u_\psi\to-u_\psi$. When $\lambda_{ijk}\neq0$ for any $i,j,k\in\{\rho,\psi\}$, the inversion symmetry of the pseudopotentials is broken and inverting the order parameters no longer implies $u_\rho\to-u_\rho$ and $u_\psi\to-u_\psi$. Consequently, the phase diagrams in Fig. 3 are not symmetric about $\rho=\psi=0$.

IV. DISCUSSION AND CONCLUSIONS

We have outlined a procedure to determine the phase diagram of systems described by coupled conserved and nonconserved fields, both in and out of equilibrium. Beginning from a force-balance representation of steady-state conditions, our theory allows one to derive a pseudopotential for the

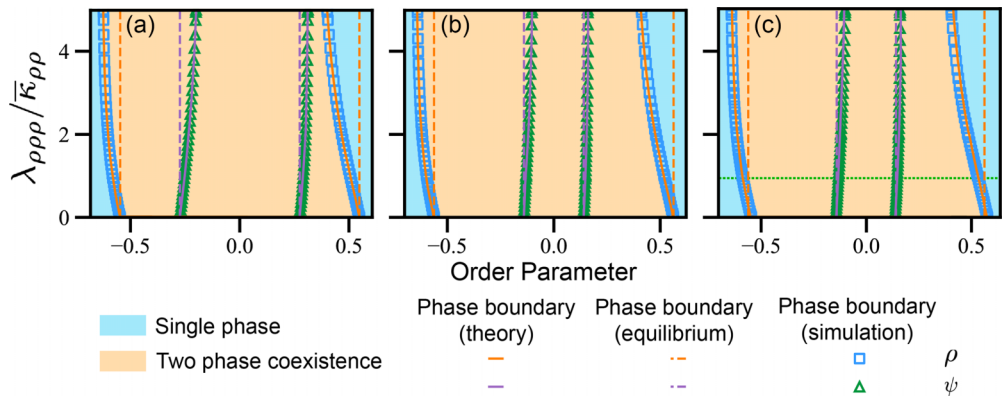


FIG. 3. Phase diagrams of AMC, displaying the coexistence values of each order parameter (ρ and ψ) as a function of $\lambda_{\rho\rho\rho}/\overline{\kappa}_{\rho\rho}$. We set $\overline{\kappa}_{\psi\rho} = 0$ and $\overline{\kappa}_{\rho\rho} = \overline{\kappa}_{\psi\psi} = 0.01$ in every case. We consider systems with (a) exact coexistence criteria by setting $\chi = -\alpha = 1/4$ and $\overline{\kappa}_{\rho\psi} = \lambda_{\rho\psi\rho} = \lambda_{\rho\rho\psi} = \lambda_{\rho\psi\psi} = 0$, (b) approximate coexistence criteria by setting $\chi = 1/2$, $\alpha = 0$, and $\overline{\kappa}_{\rho\psi} = \lambda_{\rho\psi\rho} = \lambda_{\rho\rho\psi} = \lambda_{\rho\psi\psi} = 0$, and (c) no coexistence criteria within our theory by setting $\chi = 1/2$, $\alpha = 0$, and $\overline{\kappa}_{\rho\psi} = \lambda_{\rho\psi\rho} = \lambda_{\rho\rho\psi} = \lambda_{\rho\psi\psi} = 0.01$. The dotted green line in panel (c) indicates where $\lambda_{\rho\rho\rho} = \lambda_{\rho\psi\psi}$ and well-defined approximate coexistence criteria can be formulated. When this is the case, the interfacial Maxwell construction vector takes the form $\boldsymbol{\mathcal{E}}^{\text{int}} = [\exp(2\lambda_{\rho\rho\rho}\rho/\overline{\kappa}_{\rho\rho} + 2\lambda_{\rho\rho\psi}\psi/\overline{\kappa}_{\rho\psi}) \quad 0]^T$. When $\lambda_{\rho\rho\rho} \neq \lambda_{\rho\psi\psi}$, a solution to Eqs. (22b)–(22d) does not exist. In this case, we continue to use the weighted-area construction in Eq. (29) to predict the phase diagram in panel (c) even though it is not a well-defined approximation here. In all phase diagrams, the volume fraction of each phase can be determined using the lever rule illustrated in Fig. 2. Just as was the case in Fig. 2, defining ζ and β phases such that $\rho^\zeta < \rho^\beta$, we find $\psi^\zeta > \psi^\beta$ in every system considered.

conserved field that must be equal in coexisting phases, which, in equilibrium, is the chemical potential. With this pseudopotential, one can introduce a generalized Gibbs-Duhem relation to derive the final set of coexistence criteria, the equality of a global quantity across phases. This global quantity becomes the pressure in equilibrium. These coexistence criteria can sometimes be expressed as a generalized common tangent construction on an effective free energy. When a solution to the generalized Gibbs-Duhem relation does not exist, we introduce and justify two approximate forms of the criteria that one may use to estimate the phase diagram. In equilibrium systems, the coexistence criteria are always equality of the chemical potential of the conserved field across phases, zero chemical potential of the nonconserved field in each phase, and equality of pressures across phases. Out of equilibrium, however, the functional form of the analogous state functions follow from system-specific steady-state force balances. We validate our theory by introducing a generalized form of active model C and predicting the phase diagrams of various systems, which we find to be in excellent agreement with the phase diagrams obtained by numerically determining the full spatial order parameter profiles during coexistence.

To use our theory, a number of equations of state in the flux-driving and generation-driving forces must be determined. In equilibrium, there are a number of techniques, both experimental and computational, one can use to determine these equations of state. For example, there are structural expressions for both the bulk and interfacial contributions to the free energy [50,64], allowing one to determine the passive forces given the microscopic structure of a system. While determining the functional dependence of the generation-driving forces of some nonconserved fields may prove challenging (e.g., a crystallinity field), $\mu_\rho(\rho, \psi^*)$ and $p(\rho, \psi^*)$ are more easily measured. Due to the path independence of the equilibrium Maxwell construction, only one of either $\mu_\rho(\rho, \psi^*)$ or $p(\rho, \psi^*)$, along with the parametrization $\psi^*(\rho)$, is required to determine the phase diagram of a system, and thus the difficulty in determining generation-driving forces is not an obstacle in equilibrium. It is clear in a number of contexts that theoretical, computational, and experimental approaches are consistent in equilibrium. Out of equilibrium, a similar understanding is still absent.

If the order parameters are known, there are theoretical techniques (such as an Irving-Kirkwood procedure [47]) to formally determine the coefficients in our forces. The coefficients resulting from these techniques generally depend on the microscopic degrees of freedom in the system and therefore can be determined from particle-based simulations. Experimentally, microscopic information is not always accessible and thus we desire a continuum-level procedure to determine the coefficients in our forces. One potential route toward this is to make dynamical measurements that can be compared to our constitutive relations in Eq. (6). As was suggested in Ref. [21], measuring the flux of the conserved quantity may allow the form of the flux-driving force to be determined. The dynamics of the nonconserved field is generally less straightforward to measure than that of the conserved field; however, there is growing work to determine the coefficients in our forces using machine learning approaches [65–67].

There are macroscopically phase-separating systems that cannot be described by our theory, such as the system in Fig. 3(c). It remains an open question as to whether there is a coexistence theory that can describe these systems. Moreover, elucidating the structure of AMC and how the coexistence criteria change by introducing nonzero elements of the various nonequilibrium terms would facilitate a continuum-level understanding of how to tune a given nonequilibrium system such that a desired phase diagram is achieved.

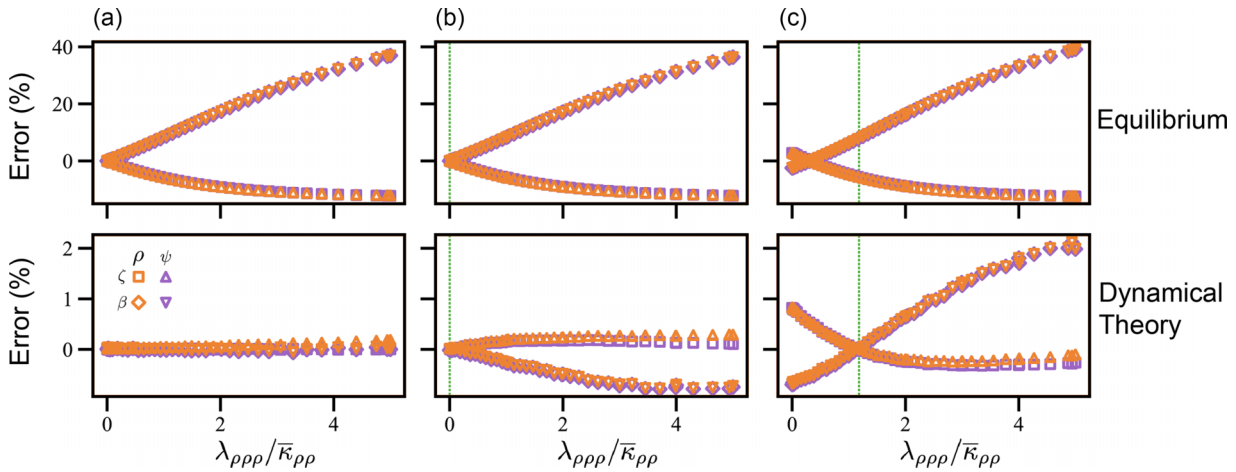


FIG. 4. Percent error of binodals in the ζ and β phases (defined by $\rho^{\beta} > \rho^{\zeta}$) predicted using the equilibrium Maxwell construction (top) and the dynamical theory (bottom) in Fig. 3 compared to the steady-state numerical solutions of AMC. All parameters match those in Figs. 3(a)–3(c), covering systems where the final coexistence criterion is (a) exact, (b) well defined but approximate, and (c) poorly defined and approximate. The dotted green lines in panels (b) and (c) indicate the passive limit and the limit where there is a solution for $\boldsymbol{\mathcal{E}}^{\text{int}}$ (and hence where the criteria are well defined but approximate), respectively.

This work focused on systems described by coupled conserved and nonconserved order parameters where the dynamics of the nonconserved fields can solely be described by a generation term. We aim to extend this work to systems where fluxes of nonconserved order parameters (not considered here) play an important role in the dynamics. It is our hope that this theory lays the groundwork to determine the phase diagram of complex nonequilibrium systems, including multicomponent crystallization [68], nematic mixtures [69,70], and reaction-diffusion systems with multiple conservation laws [71].

ACKNOWLEDGMENTS

We thank Yizhi Shen, Dimitrios Fraggedakis, Yu-Jen Chiu, Luke Langford, Davide Toffenetti, and Beatrice Nettuno for helpful discussions and feedback on this manuscript. We acknowledge support from the Laboratory Directed Research and Development Program of Lawrence Berkeley National Laboratory under U.S. Department of Energy Contract No. DE-AC02-05CH11231, the UC Berkeley College of Engineering, and the U.S. Department of Defense through the National Defense Science and Engineering Graduate Fellowship Program. Publication made possible in part by support from the Berkeley Research Impact Initiative (BRII) sponsored by the UC Berkeley Library.

DATA AVAILABILITY

The data that support the findings of this study are openly available [72].

APPENDIX A: DERIVATION OF CRITERIA WITH ANY NUMBER OF ORDER PARAMETERS

In this Appendix, we extend the coexistence theory presented in Secs. II B–II D to systems described by n_C conserved order parameters coupled to n_N nonconserved order parameters. First, in Appendix A 1, we discuss the constitutive dynamics we consider and demonstrate how reaction-diffusion systems can be described by our theory. We then extend the transformations in Eqs. (14) and (19) to systems with any number of order parameters in Appendixes A 2 and A 3, respectively. As there are many systems that display coexistence yet these transformations cannot be performed, we extend the partial transformations in Eq. (28) in Appendix A 4 and then describe the resulting approximations in Appendixes A 5 and A 6.

1. Constitutive dynamics

We consider systems with n_C scalar conserved fields coupled to n_N scalar nonconserved fields in a quasi-1D geometry where spatial variations only occur in the z dimension. The ith conserved and nonconserved fields are, respectively, denoted ρ_i (for $i \in \mathcal{C}$) and ψ_i (for $i \in \mathcal{N}$) where $\mathcal{C}$ and $\mathcal{N}$ are their respective sets of indices. We define a vector of order parameters, $\mathbf{X} \equiv [\rho_1 \quad \rho_2 \quad \cdots \quad \psi_{n_C} \quad \psi_1 \quad \psi_2 \quad \cdots \quad \psi_{n_N}]^T$, whose indices comprise a set $\mathcal{X}$ of length $n_C + n_N$ and are the symbols of the order parameters themselves (e.g., $X_{\rho_i} = \rho_i \quad \forall \quad i \in \mathcal{C}$ and $X_{\psi_i} = \psi_i \quad \forall \quad i \in \mathcal{N}$).

The evolution equations for the order parameters satisfy the balance laws:

$$\partial_t \mathbf{X} = -\partial_z \mathbf{J} + \mathbf{s}, \tag{A1}$$

where $\mathbf{J} \equiv [J_{\rho_1} \quad \cdots \quad J_{\rho_{n_C}} \quad J_{\psi_1} \quad \cdots \quad J_{\psi_{n_N}}]^T$ is the flux of each order parameter in the z direction, $\mathbf{s} \equiv [s_{\rho_1} \quad \cdots \quad s_{\rho_{n_C}} \quad s_{\psi_1} \quad \cdots \quad s_{\psi_{n_N}}]^T$ is the generation term associated with each order parameter, $\partial_t \equiv \partial/\partial t$, and $\partial_z \equiv \partial/\partial z$. As each ρ_i is conserved, $s_{\rho_i} = 0 \ \forall \, i \in \mathcal{C}$ by definition. We consider dynamics [Eq. (A1)] that are symmetric under the transformation $z \to -z$ (with $\mathbf{X} \to \mathbf{X}$). Consequently, $\partial_t \mathbf{X}$ and $\mathbf{s}$ have even spatial parity while $\mathbf{J}$ has odd spatial parity.

The conserved order parameters are each subject to an integral constraint, $\int_V d\mathbf{r}\rho_i(\mathbf{r}) = V\rho_i^{\text{sys}} \ \forall\, i \in \mathcal{C}$, where $\rho_i(\mathbf{r})$ is its local value at position $\mathbf{r}$, ρ_i^{sys} is its space-averaged value, and V is the system volume that we take to be fixed. We require that the nonconserved order parameters are wholly unconstrained, i.e., each $\int_V d\mathbf{r}\psi_i(\mathbf{r}) \quad \forall \quad i \in \mathcal{N}$ is not fixed to a specific value. There may be order parameters that are not directly subject to a conservation law but are instead subject to *aggregate constraints*, such as species concentrations in mass-conserving reaction-diffusion systems. We will now demonstrate how to apply our theory to these systems.

Consider a reaction-diffusion systems with $n_C^{\text{non-rxn}}$ nonreacting species and n_N^{rxn} reacting species. The generation term of the ith reacting species can be expressed as $\sum_r^{n_r} \nu_{ir} j_r$, where j_r is the rate of the rth reaction (of n_r) and ν_{ir} is the stoichiometry of the ith reacting species in reaction r. These reactions can, for example, correspond to birth and death or involve interconversion between species. The matrix $\mathbf{C}$ whose rows are the n_C^{rxn} left null vectors of $\boldsymbol{\nu}$ (whose ir component is ν_{ir}) can be used to identify n_C^{rxn} quantities that are conserved by the reactions as $\rho_i^{\text{rxn}} = \sum_j^{n_N^{\text{rxn}}} C_{ij}\psi_j$ where ψ_j is the concentration of the jth reacting species. We then identify that the conserved order parameters are the concentrations of the $n_C^{\text{non-rxn}}$ nonreacting species in addition to these n_C^{rxn} combinations of reacting species concentrations. One can find an orthogonal complement to these n_C^{rxn} conserved combinations of species, consisting of $n_N = n_N^{\text{rxn}} - n_C^{\text{rxn}}$ truly nonconserved order parameters that are not subject to any conservation law. By changing the basis of the reacting species concentrations to one where conservation laws (or their absence) are transparent, reaction-diffusion systems can be described by our theory.

We now require constitutive relations for the fluxes and generation terms appearing in our evolution equations. We assume the dynamics of the nonconserved order parameters are dominated by their generation term, $|\partial_z J_{\psi_i}| << |s_{\psi_i}| \quad \forall \quad i \in \mathcal{N}$. Furthermore, we assume fluxes and generation terms are driven by their respective driving forces, extending the dynamics in Eq. (6) to find

$$J_{\rho_i} = \sum_{j\in\mathcal{C}} L_{ij} f_{\rho_j} \quad \forall \quad i \in \mathcal{C}, \tag{A2a}$$

$$s_{\psi_i} = \sum_{j\in\mathcal{N}} M_{ij} f_{\psi_j} \quad \forall \quad i \in \mathcal{N}, \tag{A2b}$$

where f_{ρ_j} is the direct force driving the flux of the jth conserved field and f_{ψ_j} is the direct force driving the generation of the jth nonconserved field. Here, every f_{ρ_i} and f_{ψ_i}, respectively, have odd and even spatial parity, and each is generally a functional of the complete spatial profile of the order parameters, $\mathbf{X}(z)$. L_{ij} and M_{ij} are, respectively, the ij components of generally asymmetric $n_C \times n_C$ and $n_N \times n_N$ full-rank matrices of transport coefficients whose symmetric parts are positive definite, ensuring fluxes act in the same direction as their driving forces and that nonconserved fields are generated with positive forces and destroyed with negative forces. Each L_{ij} and M_{ij} depend on the local value of $\mathbf{X}$ but not its spatial gradients. Notably, the spatial parities of f_{ρ_j} and f_{ψ_i} prevent any f_{ρ_j} from entering s_{ψ_i}; however, in principle, the spatial gradient of each f_{ψ_j} could enter J_{ρ_i}. As will be clear in Appendix A 2, including couplings to each f_{ψ_i} in J_{ρ_i} will not affect the coexistence criteria. Moreover, there is ambiguity in the definition of each L_{ij} and f_{ρ_j}, as well as each M_{ij} and f_{ψ_i}, that will also be inconsequential in the development of coexistence criteria as we will ultimately take the stationary limit.

In flux-free boundary conditions, the steady-state conditions $J_i = 0 \quad \forall \quad i \in \mathcal{C}$ and $s_i = 0 \quad \forall \quad i \in \mathcal{N}$ imply that all forces must vanish:

$$f_{\rho_i} = 0 \quad \forall \quad i \in \mathcal{C}, \tag{A3a}$$

$$f_{\psi_i} = 0 \quad \forall \quad i \in \mathcal{N}, \tag{A3b}$$

which are exact conditions for any system following the constitutive relations in Eq. (A2).

We perform a general gradient expansion of each f_{ρ_i} and f_{ψ_i} with respect to spatial gradients of the order parameters, first expanding the generation-driving forces:

$$f_{\psi_i} \approx f_{\psi_i}^{\text{bulk}} + f_{\psi_i}^{\text{int}} \quad \forall \quad i \in \mathcal{N}, \tag{A4a}$$

where

$$f_{\psi_i}^{\text{int}} = -\mathbf{f}_{\psi_i}^{(2,1)} : \partial_z\mathbf{X}\partial_z\mathbf{X} - \mathbf{f}_{\psi_i}^{(2,2)} \cdot \partial_{zz}^2\mathbf{X} \quad \forall \quad i \in \mathcal{N}. \tag{A4b}$$

Here, $f_{\psi_i}^{\text{bulk}} \quad \forall \quad i \in \mathcal{N}$ are each equations of state that we assume have at least one zero and each $\mathbf{f}_{\psi_i}^{(2,1)}$ and $\mathbf{f}_{\psi_i}^{(2,2)}$ are, respectively, rank 2 and 1 tensors of equations of state. Without loss of generality, we define each $\mathbf{f}_{\psi_i}^{(2,1)}$ to be symmetric as antisymmetric contributions do not impact $f_{\psi_i}^{\text{int}}$. In total, the expanded generation-driving forces contain $n_N + n_N(n_N + n_C) + ([n_N + n_C]^2 + n_N + n_C)/2$ equations of state. Noting that the order parameters are spatially uniform in the coexisting phases, the first $n_N n_P$ coexistence criteria immediately follow from substituting Eq. (A4) into Eq. (A3b):

$$f_{\psi_i}^{\text{bulk}}(\mathbf{X}^\alpha) = 0 \quad \forall \quad i \in \mathcal{N},\ \alpha \in \mathcal{P}. \tag{A5}$$

Appendixes A 2 and A 3 demonstrate how to attain the remaining coexistence criteria, when possible.

We now expand the flux-driving forces:

$$f_{\rho_i} \approx f_{\rho_i}^{(1)} + f_{\rho_i}^{(3)} \ \forall\, i \in \mathcal{C}, \tag{A6a}$$

where

$$f_{\rho_i}^{(1)} = \mathbf{f}_{\rho}^{(1,1)} \cdot \partial_z\mathbf{X}, \tag{A6b}$$

$$f_{\rho_i}^{(3)} = -\mathbf{f}_{\rho}^{(3,1)} \vdots \partial_z\mathbf{X}\partial_z\mathbf{X}\partial_z\mathbf{X} - \mathbf{f}_{\rho}^{(3,2)} : \partial_{zz}^2\mathbf{X}\partial_z\mathbf{X} - \mathbf{f}_{\rho}^{(3,3)} \cdot \partial_{zzz}^3\mathbf{X}. \tag{A6c}$$

Every element of each $\mathbf{f}_{\rho_i}^{(1,1)}$, $\mathbf{f}_{\rho_i}^{(3,1)}$, $\mathbf{f}_{\rho_i}^{(3,2)}$, and $\mathbf{f}_{\rho_i}^{(3,3)}$ (rank 1, 3, 2, and 1 tensors, respectively) are generally equations of state that depend on $\mathbf{X}$ but not its spatial gradients. Equation (A6c) is unaffected by antisymmetric contributions to $\mathbf{f}_{\rho_i}^{(3,1)}$ (with respect to permuting a triplet of indices) and thus we define each $\mathbf{f}_{\rho_i}^{(3,1)}$ to be symmetric under permuting each of its three indices. In total, the flux-driving forces contain $2n_C(n_N + n_C) + (n_N + n_C)^2 + \frac{1}{6}(n_N + n_C)(n_N + n_C + 1)(n_N + n_C + 2)$ equations of state.

As will be clear in Appendix A 2, it is worthwhile to define the force vector $\bar{\mathbf{f}} \equiv [f_{\rho_1} \ \cdots \ f_{\rho_{n_C}} \ \psi_1\partial_z f_{\psi_1} \ \cdots \ \psi_{n_N}\partial_z f_{\psi_{n_N}}]^T$, which is expanded similarly to each $\mathbf{f}_{\rho_i}$:

$$\bar{\mathbf{f}} \approx \bar{\mathbf{f}}^{(1)} + \bar{\mathbf{f}}^{(3)}, \tag{A7a}$$

where

$$\bar{\mathbf{f}}^{(1)} = \bar{\mathbf{f}}^{(1,1)} \cdot \partial_z \mathbf{X}, \tag{A7b}$$

$$\bar{\mathbf{f}}^{(3)} = -\bar{\mathbf{f}}_\rho^{(3,1)} \vdots \partial_z \mathbf{X} \partial_z \mathbf{X} \partial_z \mathbf{X} - \bar{\mathbf{f}}_\rho^{(3,2)} : \partial_{zz}^2 \mathbf{X} \partial_z \mathbf{X} - \bar{\mathbf{f}}_\rho^{(3,3)} \cdot \partial_{zzz}^3 \mathbf{X}. \tag{A7c}$$

Every element of the rank 2, 4, 3, and 2 tensors, i.e., $\bar{\mathbf{f}}^{(1,1)}$, $\bar{\mathbf{f}}^{(3,1)}$, $\bar{\mathbf{f}}^{(3,2)}$, and $\bar{\mathbf{f}}^{(3,3)}$ are generally equations of state that depend on $\mathbf{X}$ but not its spatial gradients, where we again define $\bar{\mathbf{f}}^{(3,1)}$ to be symmetric upon permuting its rightmost three indices. We directly identify $\bar{f}_i = f_i \quad \forall \quad i \in \mathcal{C}$ along with

$$\bar{f}_{ij}^{(1,1)} = \psi_i \partial_j f_i^{\text{bulk}} \quad \forall \quad i \in \mathcal{N},\ j \in \mathcal{X}, \tag{A8a}$$

$$\bar{f}_{ij}^{(3,3)} = \psi_i f_{ij}^{(2,2)} \quad \forall \quad i \in \mathcal{N},\ j \in \mathcal{X}, \tag{A8b}$$

$$\bar{f}_{ijk}^{(3,2)} = \psi_i \big(\partial_k f_{ij}^{(2,2)} + 2 f_{ijk}^{2,1}\big) \quad \forall \quad i \in \mathcal{N},\ j,k \in \mathcal{X}, \tag{A8c}$$

$$\bar{f}_{ijkl}^{(3,1)} = \psi_i \tfrac{1}{3} \big(\partial_l f_{ijk}^{(2,1)} + \partial_k f_{ijl}^{(2,1)} + \partial_j f_{ilk}^{(2,1)}\big) \quad \forall \quad i \in \mathcal{N},\ j,k,l \in \mathcal{X}, \tag{A8d}$$

where $\partial_j \equiv \partial/\partial j$ for $j \in \mathcal{X}$.

2. Transformation tensor

The next $n_C(n_P - 1)$ criteria [in addition to the $n_N n_P$ in Eq. (A5)] follow from the transformation $\bar{\mathbf{f}} = \mathbf{0} \to \partial_z \mathbf{u} = \mathbf{0}$ that generalizes Eq. (11), where the vector of pseudopotentials, $\mathbf{u}$, is spatially even and expanded as

$$\mathbf{u} \approx \mathbf{u}^{\text{bulk}} + \mathbf{u}^{\text{int}}, \tag{A9a}$$

with

$$\mathbf{u}^{\text{int}} = -\mathbf{u}^{(2,1)} : \partial_z \mathbf{X} \partial_z \mathbf{X} - \mathbf{u}^{(2,2)} \cdot \partial_{zz}^2 \mathbf{X}. \tag{A9b}$$

Antisymmetric (with respect to exchanging j and k) contributions to $u_{ijk}^{(2,1)}$ do not impact the value of u_i^{int} and hence we define each $u_{ijk}^{(2,1)}$ to be symmetric.

The transformation tensor, $\mathcal{T}$, is defined to convert $\bar{\mathbf{f}}$ into $\partial_z \mathbf{u}$ through a linear, bijective transformation:

$$\mathcal{T} \cdot \bar{\mathbf{f}} = \partial_z \mathbf{u}. \tag{A10a}$$

We define the pseudopotentials of the nonconserved fields to be the negative of their generation-driving forces, $u_i = -f_i \quad \forall \quad i \in \mathcal{N}$, and thus $\mathcal{T}_{ij} = -\delta_{ij}\psi_i^{-1} \quad \forall \quad i \in \mathcal{N},\ j \in \mathcal{X}$. We then only need to determine the rows of the transformation tensor that correspond to conserved order parameters, $\mathcal{T}_{ij} \quad \forall \quad i \in \mathcal{C},\ j \in \mathcal{X}$, and their associated pseudopotentials, $u_i \quad \forall \quad i \in \mathcal{C}$:

$$\sum_{j \in \mathcal{X}} \mathcal{T}_{ij} \bar{f}_j = \partial_z u_i \quad \forall \quad i \in \mathcal{C}. \tag{A10b}$$

It is clear that introducing a global multiplicative constant into each $\mathcal{T}_{ij}$ simply rescales u_i, i.e., $\mathcal{T}_{ij} \to c\mathcal{T}_{ij} \Rightarrow u_i \to c u_i$, and thus the condition of uniformity of u_i is unaffected. Introducing an additive constant, $\mathcal{T}_{ij} \to \mathcal{T}_{ij} + c$, *does* nontrivially affect u_i, however.

Differentiating u_i in the z direction, we have

$$\begin{aligned}\partial_z u_i = {} & \sum_{j \in \mathcal{X}} \partial_j u_i^{\text{bulk}} \partial_z X_j - \sum_{j \in \mathcal{X}} u_{ij}^{(2,2)} \partial_{zzz}^3 X_j \\ & - \sum_{j \in \mathcal{X}} \sum_{k \in \mathcal{X}} \big(\partial_k u_{\rho j}^{(2,2)} + 2 u_{\rho jk}^{(2,1)}\big) \partial_{zz}^2 X_j \partial_z X_k \\ & - \frac{1}{3} \sum_{j \in \mathcal{X}} \sum_{k \in \mathcal{X}} \sum_{l \in \mathcal{X}} \big(\partial_l u_{\rho jk}^{(2,1)} + \partial_k u_{\rho jl}^{(2,1)} \\ & + \partial_j u_{\rho lk}^{(2,1)}\big) \partial_z X_j \partial_z X_k \partial_z X_l. \end{aligned} \tag{A11}$$

Substituting this into Eq. (A10a) along with Eq. (A7), matching terms, and differentiating until every algebraic equation is first order, we find an overdetermined system of $n_C(n_N + n_C) + 2n_C(n_N + n_C)^2 + n_C(n_N + n_C)(n_N + n_C + 1)(n_N + n_C + 2)/6$ linear, first-order, homogeneous PDEs for $n_C + 2n_C(n_N + n_C) + n_C(n_N + n_C)(n_N + n_C + 1)/2$ unknown functions (each $\mathcal{T}_{ij}$ and every coefficient in each u_i):

$$\sum_{j \in \mathcal{X}} \mathcal{T}_{ij} \bar{f}_{jk}^{(1,1)} = \partial_k u_i^{\text{bulk}} \quad \forall \quad i \in \mathcal{C},\ k \in \mathcal{X}, \tag{A12a}$$

$$\sum_{j \in \mathcal{X}} \mathcal{T}_{ij} \bar{f}_{jklm}^{(3,1)} = \frac{1}{3} \big(\partial_m u_{ikl}^{(2,1)} + \partial_l u_{ikm}^{(2,1)} + \partial_k u_{iml}^{(2,1)}\big) \quad \forall \quad i \in \mathcal{C},\ k,l,m \in \mathcal{X}, \tag{A12b}$$

$$\sum_{j \in \mathcal{X}} \mathcal{T}_{ij} \bar{f}_{jkl}^{(3,2)} = \partial_l u_{ik}^{(2,2)} + 2 u_{ikl}^{(2,1)} \quad \forall \quad i \in \mathcal{C},\ k,l \in \mathcal{X}, \tag{A12c}$$

$$\sum_{j \in \mathcal{X}} \partial_l \big(\mathcal{T}_{ij} \bar{f}_{jk}^{(3,3)}\big) = \partial_l u_{ik}^{(2,2)} \quad \forall \quad i \in \mathcal{C},\ k,l \in \mathcal{X}. \tag{A12d}$$

To make further progress, we look to eliminate the coefficients of u_i from Eq. (A12) such that we obtain a system of equations for each $\mathcal{T}_{ij}$ solely in terms of the known force coefficients. If one does this, care must be taken to ensure the commutativity of partial differentiation of each of the unknown functions with respect to the order parameters. This procedure to obtain a system of PDEs where commutativity is ensured is known as "completion to involution" [51]. Here, we demonstrate how to eliminate the coefficients of u_i from Eq. (A12), noting that a solution to the resulting equations can be checked subsequently to ensure the commutativity of partial differentiation is indeed respected.

The first relation we find follows from Eq. (A12a) and identifying that each $\partial_{jk}^2 u_i^{\text{bulk}}$ must be symmetric with respect to exchanging j and k:

$$\sum_{j \in \mathcal{X}} \big[\partial_l \mathcal{T}_{ij} \bar{f}_{jk}^{(1,1)} - \partial_k \mathcal{T}_{ij} \bar{f}_{jl}^{(1,1)} + \mathcal{T}_{ij} \partial_l \bar{f}_{jk}^{(1,1)} - \mathcal{T}_{ij} \partial_k \bar{f}_{jl}^{(1,1)}\big] = 0 \quad \forall \quad i \in \mathcal{C},\ k,l \in \mathcal{X}. \tag{A13}$$

Next, we note each $u^{(2,1)}_{ijk}$ must also be symmetric with respect to exchanging j and k and therefore

$$\sum_{j\in\mathcal{X}}\big[\mathcal{T}_{ij}\overline{f}^{(3,2)}_{jkl}-\mathcal{T}_{ij}\overline{f}^{(3,2)}_{jlk}-\partial_l\mathcal{T}_{ij}\overline{f}^{(3,3)}_{jk}+\partial_k\mathcal{T}_{ij}\overline{f}^{(3,3)}_{jl}-\mathcal{T}_{ij}\partial_l\overline{f}^{(3,3)}_{jk}+\mathcal{T}_{ij}\partial_k\overline{f}^{(3,3)}_{jl}\big]=0\quad\forall\quad i\in\mathcal{C},\ k,l\in\mathcal{X}. \tag{A14}$$

Our last relation ensures $u^{(2,1)}_{ijk}$ and $u^{(2,2)}_{ij}$ appear consistently in Eqs. (A12b)–(A12d). Differentiating Eqs. (A12c) and (A12d) and substituting the result into Eq. (A12b), we find

$$\sum_{j\in\mathcal{X}}\big[6\mathcal{T}_{ij}\overline{f}^{(3,1)}_{jklm}-\partial_m\big(\mathcal{T}_{ij}\overline{f}^{(3,2)}_{jkl}\big)-\partial_l\big(\mathcal{T}_{ij}\overline{f}^{(3,2)}_{jkm}\big)-\partial_k\big(\mathcal{T}_{ij}\overline{f}^{(3,2)}_{jlm}\big)+2\partial^2_{lm}\big(\mathcal{T}_{ij}\overline{f}^{(3,3)}_{jk}\big)+\partial^2_{kl}\big(\mathcal{T}_{ij}\overline{f}^{(3,3)}_{jm}\big)\big]=0\quad\forall\quad i\in\mathcal{C},\ k,l,m\in\mathcal{X}. \tag{A15}$$

Importantly, the system of PDEs in Eqs. (A13), (A14), and (A15) does not contain the compatibility conditions that ensure partial differentiation is commutative. To demonstrate how to ensure this commutativity, we perform the procedure for the system of PDEs that result from the generalized Gibbs-Duhem relation (derived in Appendix A 3) when $n_C = n_N = 1$ in the Supplemental Material [52].

When the transformation from $\overline{\mathbf{f}}$ to $\mathbf{u}$ can be made, we identify $n_N n_P + n_C(n_P-1)$ coexistence criteria from the steady-state conditions in Eq. (A3) as

$$\mathbf{u}^{\text{bulk}}(\mathbf{X}^\alpha)=\mathbf{u}^{\text{coexist}}\quad\forall\quad\alpha\in\mathcal{P}, \tag{A16}$$

where the coexistence pseudopotential of the nonconserved fields is zero, $u_i^{\text{coexist}}=0\quad\forall\quad i\in\mathcal{N}$ [these correspond to the criteria in Eq. (A5)]. The coefficients of $\mathbf{u}$ can be identified as

$$\mathbf{u}^{\text{bulk}}=\int\boldsymbol{\mathcal{T}}\cdot\overline{\mathbf{f}}^{(1,1)}\cdot d\mathbf{X}, \tag{A17a}$$

$$\mathbf{u}^{(2,2)}=\boldsymbol{\mathcal{T}}\cdot\overline{\mathbf{f}}^{(3,3)}, \tag{A17b}$$

$$\mathbf{u}^{(2,1)}=\frac{1}{2}(\boldsymbol{\mathcal{T}}\cdot\overline{\mathbf{f}}^{(3,2)}-\partial_{\mathbf{X}}[\boldsymbol{\mathcal{T}}\cdot\overline{\mathbf{f}}^{(3,3)}]), \tag{A17c}$$

where $\partial_{\mathbf{X}}\equiv\partial/\partial\mathbf{X}$.

3. Maxwell Construction Vector

The final n_P-1 criteria, if they exist, follow from equality of a global quantity, $\mathcal{G}$, that is expanded as

$$\mathcal{G}\approx\mathcal{G}^{\text{bulk}}+\mathcal{G}^{\text{int}}, \tag{A18a}$$

where

$$\mathcal{G}^{\text{int}}=-\boldsymbol{\mathcal{G}}^{(2,1)}:\partial_z\mathbf{X}\partial_z\mathbf{X}-\boldsymbol{\mathcal{G}}^{(2,2)}\cdot\partial^2_{zz}\mathbf{X}. \tag{A18b}$$

Again, antisymmetric contributions to $\boldsymbol{\mathcal{G}}^{(2,2)}$ do not impact the value of $\mathcal{G}^{\text{int}}$.

The generalized Gibbs-Duhem relation is

$$\boldsymbol{\mathcal{E}}\cdot d\mathbf{u}=d\mathcal{G}, \tag{A19}$$

where we have introduced the Maxwell construction vector $\boldsymbol{\mathcal{E}}$, which depends on $\mathbf{X}$ but not its spatial gradients. Here, the expansion of $\mathbf{u}$ in Eq. (A9) is used. We can immediately appreciate that introducing a global multiplicative constant into the Maxwell construction vector simply rescales $\mathcal{G}$, $\boldsymbol{\mathcal{E}}\to c\boldsymbol{\mathcal{E}}\Rightarrow\mathcal{G}\to c\mathcal{G}$, and hence does not alter the spatial uniformity of $\mathcal{G}$. Furthermore, this uniformity is also unaffected by introducing a vector of additive constants into the Maxwell construction vector, $\boldsymbol{\mathcal{E}}\to\boldsymbol{\mathcal{E}}+\mathbf{c}$, as this simply adds a linear combination of the pseudopotentials to the global quantity, $\mathcal{G}\to\mathcal{G}+\mathbf{c}\cdot\mathbf{u}$. As $\mathbf{u}$ is spatially uniform, $\mathbf{c}\cdot\mathbf{u}$ trivially is as well. When $\boldsymbol{\mathcal{E}}$ and $\mathcal{G}$ can be found, the final n_P-1 coexistence criteria follow as

$$\mathcal{G}^{\text{bulk}}(\mathbf{X}^\alpha)=\mathcal{G}^{\text{coexist}}\quad\forall\quad\alpha\in\mathcal{P}, \tag{A20}$$

which, when combined with Eq. (13), recovers the Gibbs phase rule where there are n_C-n_P+1 degrees of freedom. Using the generalized Gibbs-Duhem relation [Eq. (A19)], we can express the criteria in Eq. (A20) as a generalized Maxwell construction:

$$\int_{\boldsymbol{\mathcal{E}}^\alpha}^{\boldsymbol{\mathcal{E}}^\beta}(\mathbf{u}^{\text{bulk}}(\mathbf{X})-\mathbf{u}^{\text{coexist}})\cdot d\boldsymbol{\mathcal{E}}(\mathbf{X})=0\quad\forall\quad\alpha,\beta\in\mathcal{P}. \tag{A21a}$$

When $\boldsymbol{\mathcal{E}}$ is not a monotonic function of $\mathbf{X}$, there is ambiguity in evaluating this generalized Maxwell construction. This can be resolved by expressing Eq. (A21a) as a weighted-area construction:

$$\int_{\mathbf{X}^\alpha}^{\mathbf{X}^\beta}(\mathbf{u}^{\text{bulk}}(\mathbf{X})-\mathbf{u}^{\text{coexist}})\cdot\mathbf{E}(\mathbf{X})\cdot d\mathbf{X}=0\quad\forall\quad\alpha,\beta\in\mathcal{P}, \tag{A21b}$$

where $\mathbf{E}\equiv\partial_{\mathbf{X}}\boldsymbol{\mathcal{E}}$. The integrals in Eqs. (A21a) and (A21b) are path-independent and thus one may evaluate them along the path that is most convenient. For example, one convenient choice is the path $\mathbf{X}^*$ that causes the bulk part of the pseudopotentials of the nonconserved fields to be zero at all points during the integration, i.e., $\mathbf{X}^*$ satisfies $u_i^{\text{bulk}}(\mathbf{X}^*)=0\quad\forall\quad i\in\mathcal{N}$ where the nonconserved fields are functions of the conserved fields, $\psi_i^*=\psi_i^*(\{\rho_j^*\})\quad\forall\quad i\in\mathcal{N}$.

Differentiating $\mathcal{G}$ in the z direction, we have

$$\partial_z\mathcal{G}=\sum_{j\in\mathcal{X}}\partial_j\mathcal{G}^{\text{bulk}}\partial_z X_j-\sum_{j\in\mathcal{X}}\mathcal{G}^{(2,2)}_j\partial^3_{zzz}X_j-\sum_{j\in\mathcal{X}}\sum_{k\in\mathcal{X}}\big(\partial_k\mathcal{G}^{(2,2)}_j+2\mathcal{G}^{(2,1)}_{jk}\big)\partial^2_{zz}X_j\partial_z X_k-\frac{1}{3}\sum_{j\in\mathcal{X}}\sum_{k\in\mathcal{X}}\sum_{l\in\mathcal{X}}\big(\partial_l\mathcal{G}^{(2,1)}_{jk}+\partial_k\mathcal{G}^{(2,1)}_{jl}+\partial_j\mathcal{G}^{(2,1)}_{lk}\big)\partial_z X_j\partial_z X_k\partial_z X_l. \tag{A22}$$

Substituting $\partial_z u_i$ [Eq. (A11)] and $\partial_z\mathcal{G}$ [Eq. (A22)] into Eq. (A19) and matching terms, we find an overdetermined system of $n_N+n_C+2(n_N+n_C)^2+\frac{1}{6}(n_N+n_C)(n_N+n_C+1)(n_N+n_C+2)$ linear, first-order, homogeneous PDEs for $1+2(n_N+n_C)+(n_N+n_C)(n_N+n_C+1)/2$ unknown functions (each component of $\boldsymbol{\mathcal{E}}$ and each coefficient in $\mathcal{G}$):

$$\sum_{i\in\mathcal{X}}\mathcal{E}_i\partial_j u_i^{\text{bulk}}=\partial_j\mathcal{G}^{\text{bulk}}\quad\forall\quad j\in\mathcal{X}, \tag{A23a}$$

$$\sum_{i\in\mathcal{X}} \partial_k\left(\mathcal{E}_i u_{ij}^{(2,2)}\right) = \partial_k \mathcal{G}_j^{(2,2)} \quad \forall \quad j,k \in \mathcal{X}, \tag{A23b}$$

$$\sum_{i\in\mathcal{X}} \mathcal{E}_i\left(2u_{ijk}^{(2,1)} + \partial_k u_{ij}^{(2,2)}\right) = 2\mathcal{G}_{jk}^{(2,1)} + \partial_k \mathcal{G}_j^{(2,2)} \quad \forall \quad j,k \in \mathcal{X}, \tag{A23c}$$

$$\sum_{i\in\mathcal{X}} \mathcal{E}_i\left(\partial_l u_{ijk}^{(2,1)} + \partial_k u_{ijl}^{(2,1)} + \partial_j u_{ilk}^{(2,1)}\right) = \partial_l \mathcal{G}_{jk}^{(2,1)} + \partial_k \mathcal{G}_{jl}^{(2,1)} + \partial_j \mathcal{G}_{lk}^{(2,1)} \quad \forall \quad j,k,l \in \mathcal{X}. \tag{A23d}$$

To proceed, we seek equations for the components of $\boldsymbol{\mathcal{E}}$ solely in terms of coefficients in $\mathbf{u}$, i.e., we look to eliminate the coefficients of $\mathcal{G}$ from Eq. (A23). Just as was the case for the transformation tensor, one generally must find compatibility conditions that ensure partial differentiation is commutative. Here, we eliminate the coefficients of $\mathcal{G}$ from Eq. (A23), finding equations one can solve and subsequently check if partial differentiation is commutative. In the Supplemental Material [52], we follow a formal procedure (completion to involution [51]) when $n_C = n_N = 1$ to find the compatibility conditions one needs to augment Eq. (A23) with to ensure partial differentiation is commutative.

The first set of relationships we find follows from Eq. (A23a), noting $\partial_{jk}^2 \mathcal{G}^{\text{bulk}}$ must be symmetric upon exchanging j and k:

$$\sum_{i\in\mathcal{X}} \left[E_{ij}\partial_k u_i^{\text{bulk}} - E_{ik}\partial_j u_i^{\text{bulk}}\right] = 0 \quad \forall \quad j,k \in \mathcal{X}, \tag{A24}$$

where $E_{ij} \equiv \partial_j \mathcal{E}_i$. Additionally, $\mathcal{G}_{jk}^{(2,1)}$ must be symmetric with respect to exchanging j and k:

$$\sum_{i\in\mathcal{X}} \left[u_{ij}^{(2,2)} E_{ik} - u_{ik}^{(2,2)} E_{ij}\right] = 0 \quad \forall \quad j,k \in \mathcal{X}. \tag{A25}$$

Our last set of equations ensures $\mathcal{G}_{jk}^{(2,1)}$ and $\mathcal{G}_j^{(2,2)}$ appear consistently in Eqs. (A23b)–(A23d). Differentiating Eqs. (A23b) and (A23c) and substituting the result into Eq. (A23d), we find

$$\sum_{i\in\mathcal{X}} \left[2E_{il}u_{ijk}^{(2,1)} + 2E_{ik}u_{ijl}^{(2,1)} + 2E_{ij}u_{ilk}^{(2,1)} - \partial_l\left(E_{ik}u_{ij}^{(2,2)}\right) - \partial_k\left(E_{il}u_{ij}^{(2,2)}\right) - \partial_j\left(E_{ik}u_{il}^{(2,2)}\right)\right] = 0 \quad \forall \quad j,k,l \in \mathcal{X}. \tag{A26}$$

When these equations can be solved, the coefficients in $\mathcal{G}$ can be identified as

$$\mathcal{G}^{\text{bulk}} = \mathbf{u}^{\text{bulk}} \cdot \boldsymbol{\mathcal{E}} - \int \mathbf{u}^{\text{bulk}} \cdot d\boldsymbol{\mathcal{E}}, \tag{A27a}$$

$$\boldsymbol{\mathcal{G}}^{(2,2)} = \boldsymbol{\mathcal{E}} \cdot \mathbf{u}^{(2,2)}, \tag{A27b}$$

$$\boldsymbol{\mathcal{G}}^{(2,1)} = \boldsymbol{\mathcal{E}} \cdot \mathbf{u}^{(2,1)} - \frac{1}{2}\mathbf{E}^T \cdot \mathbf{u}^{(2,2)}. \tag{A27c}$$

4. Partial solutions to the generalized Gibbs-Duhem relation

We now consider systems where $\mathcal{T}$ and $\mathbf{u}$ can be determined, yet a full solution to the generalized Gibbs-Duhem relation [Eq. (A19)] does not exist. We propose splitting the generalized Gibbs-Duhem relation into bulk and interfacial portions:

$$\boldsymbol{\mathcal{E}}^{\text{bulk}} \cdot \mathbf{u}^{\text{bulk}} = d\mathcal{G}^{\text{bulk}}, \tag{A28a}$$

$$\boldsymbol{\mathcal{E}}^{\text{int}} \cdot \mathbf{u}^{\text{int}} = d\mathcal{G}^{\text{int}}. \tag{A28b}$$

Here, $\boldsymbol{\mathcal{E}}^{\text{bulk}}$ is defined to satisfy Eq. (A23a):

$$\sum_{i\in\mathcal{X}} \mathcal{E}_i^{\text{bulk}} \partial_j u_i^{\text{bulk}} = \partial_j \mathcal{G}^{\text{bulk}} \quad \forall \quad j \in \mathcal{X}, \tag{A29}$$

while $\boldsymbol{\mathcal{E}}^{\text{int}}$ is defined to satisfy Eqs. (A23b)–(A23d):

$$\sum_{i\in\mathcal{X}} \partial_k\left(\mathcal{E}_i u_{ij}^{(2,2)}\right) = \partial_k \mathcal{G}_j^{(2,2)} \quad \forall \quad j,k \in \mathcal{X}, \tag{A30a}$$

$$\sum_{i\in\mathcal{X}} \mathcal{E}_i\left(2u_{ijk}^{(2,1)} + \partial_k u_{ij}^{(2,2)}\right) = 2\mathcal{G}_{jk}^{(2,1)} + \partial_j \mathcal{G}_j^{(2,2)} \quad \forall \quad j,k \in \mathcal{X}, \tag{A30b}$$

$$\sum_{i\in\mathcal{X}} \mathcal{E}_i\left(\partial_l u_{ijk}^{(2,1)} + \partial_k u_{ijl}^{(2,1)} + \partial_j u_{ilk}^{(2,1)}\right) = \partial_l \mathcal{G}_{jk}^{(2,1)} + \partial_k \mathcal{G}_{jl}^{(2,1)} + \partial_j \mathcal{G}_{lk}^{(2,1)} \quad \forall \quad j,k,l \in \mathcal{X}. \tag{A30c}$$

When these equations can be solved, $\mathcal{G}$ can be defined; however, it is not necessarily spatially uniform in a steady state.

5. Approximate weighted-area construction

We now derive an approximate set of coexistence criteria when the interfacial generalized Gibbs-Duhem relation [Eq. (A28b) and correspondingly Eq. (A30)] can be solved for $\boldsymbol{\mathcal{E}}^{\text{int}}$, replacing Eq. (A20). The following result is independent of a solution to the bulk generalized Gibbs-Duhem relation [Eq. (A28a) and correspondingly Eq. (A29)].

The interfacial generalized Gibbs-Duhem relation [Eq. (A28b)] can be expressed as $\partial_z \mathcal{G}^{\text{int}} = \partial_z(\boldsymbol{\mathcal{E}}^{\text{int}} \cdot \mathbf{u}^{\text{int}}) - \mathbf{u}^{\text{int}} \cdot \mathbf{E}^{\text{int}} \cdot \partial_z \mathbf{X}$ (where $\mathbf{E}^{\text{int}} \equiv \partial_{\mathbf{X}} \boldsymbol{\mathcal{E}}^{\text{int}}$) and therefore $\mathbf{u}^{\text{int}} \cdot \mathbf{E}^{\text{int}} \cdot \partial_z \mathbf{X}$ must be the z derivative of a contribution to $\mathcal{G}^{\text{int}}$. Importantly, every contribution to $\mathcal{G}^{\text{int}}$ vanishes in the homogeneous phases by definition and thus the spatial integral of $\mathbf{u}^{\text{int}} \cdot \mathbf{E}^{\text{int}} \cdot \partial_z \mathbf{X}$ over the interfacial profile is guaranteed to vanish. In other words, the weighted-area construction [Eq. (A21b)] with $\mathbf{E} \to \mathbf{E}^{\text{int}}$ vanishes when evaluated along $\mathbf{X}^{\text{c}}(z)$:

$$\int_{z^\alpha}^{z^\beta} (\mathbf{u}^{\text{bulk}}(\mathbf{X}^{\text{c}}) - \mathbf{u}^{\text{coexist}}) \cdot \mathbf{E}^{\text{int}}(\mathbf{X}^{\text{c}}) \cdot \partial_z \mathbf{X}^{\text{c}} dz = 0 \quad \forall \quad \alpha,\beta \in \mathcal{P}, \tag{A31}$$

where the value of this integral is *path-dependent* as it is not equal to the difference in $\mathcal{G}^{\text{bulk}}$ between phases if $\boldsymbol{\mathcal{E}}^{\text{bulk}} \neq \boldsymbol{\mathcal{E}}^{\text{int}}$. The full spatial coexistence profile, $\mathbf{X}^{\text{c}}(z)$, is what we aim to avoid determining. We then see that as an approximate set of coexistence criteria, we may evaluate the integral in Eq. (A31) along a different path, such as $\mathbf{X}^*(\rho)$. This approximation gains accuracy as the selected integration path approaches the coexistence profile.

6. Approximate equality of the bulk global quantity

We now derive a second approximate form of the final set of coexistence criteria [replacing Eq. (A20)], applicable to systems where the bulk generalized Gibbs-Duhem relation [Eq. (A28a) and correspondingly Eq. (A23a)] have solutions.

Rearranging Eq. (A31) and identifying $\mathcal{G}^{\rm bulk} = \boldsymbol{\mathcal{E}}^{\rm bulk} \cdot \mathbf{u}^{\rm bulk} - \int \mathbf{u}^{\rm bulk} \cdot d\boldsymbol{\mathcal{E}}^{\rm bulk}$ [from Eq. (A29)], we find

$$\begin{aligned}\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk} &\equiv \mathcal{G}^{\rm bulk}(\mathbf{X}^\alpha) - \mathcal{G}^{\rm bulk}(\mathbf{X}^\beta) \\ &= \int_{\mathbf{X}^\alpha}^{\mathbf{X}^\beta} (\mathbf{u}^{\rm bulk}(\mathbf{X}) - \mathbf{u}^{\rm coexist}) \cdot (\mathbf{E}^{\rm bulk}(\mathbf{X}) \\ &\quad - \mathbf{E}^{\rm int}(\mathbf{X})) \cdot d\mathbf{X} \quad \forall \quad \alpha, \beta \in \mathcal{P}. \end{aligned} \tag{A32}$$

We then see that one may equate $\mathcal{G}^{\rm bulk}$ across coexisting phases as an approximate set of coexistence criteria, with the approximation gaining accuracy as the weighted-area construction with $\mathbf{E} = \mathbf{E}^{\rm bulk} - \mathbf{E}^{\rm int}$ [evaluated along $\mathbf{X}^{\rm c}(z)$] gets closer to zero.

APPENDIX B: NUMERICAL SUPPORT FOR COEXISTENCE CRITERIA

1. Accuracy of equating $\mathcal{G}^{\rm bulk}$ as an approximate criterion

We now confirm that the approximate criterion used to construct Fig. 2 (equating $\mathcal{G}^{\rm bulk} = \mathbf{u}^{\rm bulk} \cdot \boldsymbol{\mathcal{E}}^{\rm bulk} - \int \mathbf{u}^{\rm bulk} \cdot d\boldsymbol{\mathcal{E}}^{\rm int}$ across phases) is indeed a good approximation. To do so, we compute the difference in $\mathcal{G}^{\rm bulk}$ between the two phases [Eq. (30)]:

$$\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk} = \left(\frac{\chi+\alpha}{\chi-\alpha} - \frac{\overline{\kappa}_{\rho\psi}}{\overline{\kappa}_{\psi\rho}}\right) \int_{z^\alpha}^{z^\beta} dz u_\psi^{\rm bulk}(\mathbf{X}^{\rm c}) \partial_z \psi^{\rm c}. \tag{B1}$$

It is not clear from Eq. (B1) what a "small" or "large" value of $\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk}$ is and thus when equating it across phases is a good approximation or not, respectively. We therefore desire to provide a scale to Eq. (B1). One way of doing so is to treat Eq. (B1) as a mean of sorts by dividing by the system length in the z direction, L_z:

$$\begin{aligned}\left\langle \Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk} \right\rangle &\equiv \frac{\Delta^{\alpha\beta}\mathcal{G}^{\rm bulk}}{L_z} \\ &= \left(\frac{\chi+\alpha}{\chi-\alpha} - \frac{\overline{\kappa}_{\rho\psi}}{\overline{\kappa}_{\psi\rho}}\right) \int_{z^\alpha}^{z^\beta} \frac{u_\psi^{\rm bulk}(\mathbf{X}^{\rm c}) \partial_z \psi^{\rm c}}{L_z} dz, \end{aligned} \tag{B2a}$$

with the analogous second moment

$$\begin{aligned}&\left\langle \left(\Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk}\right)^2 \right\rangle \\ &= \left(\frac{\chi+\alpha}{\chi-\alpha} - \frac{\overline{\kappa}_{\rho\psi}}{\overline{\kappa}_{\psi\rho}}\right)^2 \int_{z^\alpha}^{z^\beta} \frac{\left(u_\psi^{\rm bulk}(\mathbf{X}^{\rm c}) \partial_z \psi^{\rm c}\right)^2}{L_z} dz. \end{aligned} \tag{B2b}$$

We can now define a variance as

$$\mathrm{Var}\left(\Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk}\right) \equiv \left\langle \left(\Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk}\right)^2 \right\rangle - \left\langle \Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk} \right\rangle^2, \tag{B3a}$$

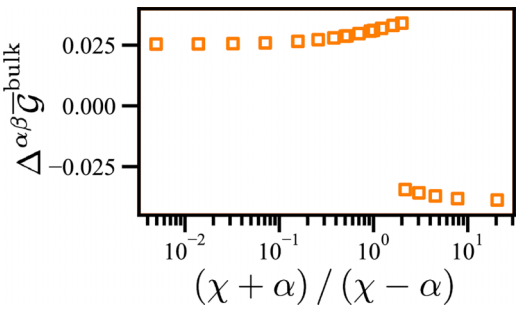


FIG. 5. $\Delta^{\alpha\beta}\overline{\mathcal{G}}^{\rm bulk}$ for the system in Fig. 2.

to define the dimensionless

$$\Delta^{\alpha\beta}\overline{\mathcal{G}}^{\rm bulk} \equiv \left\langle \Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk} \right\rangle \Big/ \sqrt{\mathrm{Var}\left(\Delta^{\alpha\beta}\mathcal{G}_{L_z}^{\rm bulk}\right)}, \tag{B3b}$$

which we plot in Fig. 5.

The consistently low $\Delta^{\alpha\beta}\overline{\mathcal{G}}^{\rm bulk}$ across parameter values suggests equating $\mathcal{G}^{\rm bulk}$ across phases is a serviceable approximate coexistence criterion, as expected from the accuracy of the predictions in Fig. 2.

2. Further demonstration of exact criteria

We now offer further support for the coexistence criteria presented in Secs. II B and II C. Our theory predicts that the binodals in Fig. 3(a) are a function of the ratio $\lambda_{\rho\rho\rho}/\overline{\kappa}_{\rho\rho}$. In Fig. 6, we numerically determine the coexistence profiles for this system, tuning $\lambda_{\rho\rho\rho}$ and $\overline{\kappa}_{\rho\rho}$ while keeping the ratio between the two fixed. As expected, we find the shape of the interface changes with the raw values of $\lambda_{\rho\rho\rho}$ and $\overline{\kappa}_{\rho\rho}$; however, the values of the order parameters in the bulk phases remain unchanged for fixed $\lambda_{\rho\rho\rho}/\overline{\kappa}_{\rho\rho}$. This demonstrates that our theory correctly identifies the relevant parameters that control the phase diagram of a system.

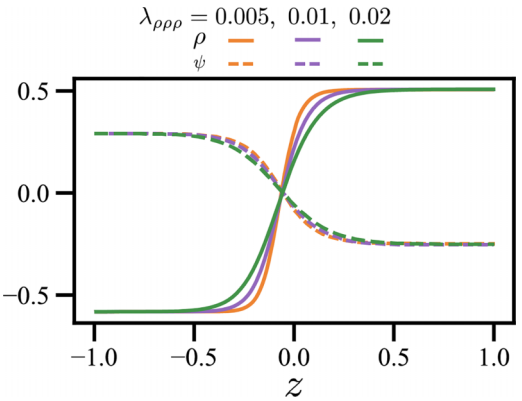


FIG. 6. Coexistence profiles of systems with exact coexistence criteria, the phase diagram of which are shown in Fig. 3(a). We set $\chi = -\alpha = 1/4$, $\overline{\kappa}_{\psi\rho} = \overline{\kappa}_{\rho\psi} = \lambda_{\rho\psi\rho} = \lambda_{\rho\rho\psi} = \lambda_{\rho\psi\psi} = 0$, and $\overline{\kappa}_{\psi\psi} = 0.01$ in every case. We determine three profiles with $\overline{\kappa}_{\rho\rho} = \lambda_{\rho\rho\rho}$ while varying $\lambda_{\rho\rho\rho}$.

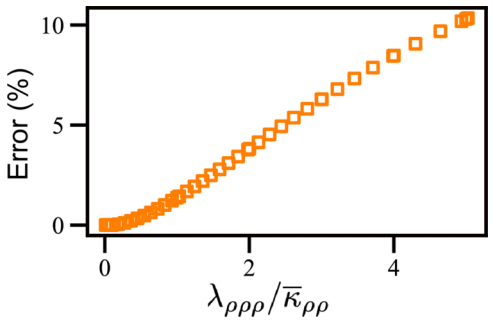


FIG. 7. Percent error of $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^*]$ compared to $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^c]$ for the system in Fig. 3(b).

3. Influence of integration path in approximate weighted-area construction

We now examine how the selected integration path influences the weighted-area construction:

$$\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}] \equiv \int_{\mathbf{X}^\alpha}^{\mathbf{X}^\beta} (\mathbf{u}^{\text{bulk}}(\mathbf{X}) - \mathbf{u}^{\text{coexist}}) \cdot \mathbf{E}^{\text{int}}(\mathbf{X}) \cdot d\mathbf{X}, \tag{B4a}$$

where $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^c] = 0$, i.e., the weighted-area construction vanishes when evaluated along the the spatial coexistence profile, $\mathbf{X}^c$. We aim to avoid determining this profile and hence we can evaluate $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}]$ along another integration path and equate the result to zero as an approximate coexistence criterion. Here, we consider the system in Fig. 3(b) where $\boldsymbol{\mathcal{E}}^{\text{int}} = [\exp(2\lambda_{\rho\rho\rho}\rho/\overline{\kappa}_{\rho\rho}) \quad 0]^T$ and therefore the approximate criterion becomes

$$\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}] = \int_{\rho^\alpha}^{\rho^\beta} \left(u_\rho^{\text{bulk}}(\mathbf{X}) - u_\rho^{\text{coexist}}\right) e^{2\frac{\lambda_{\rho\rho\rho}}{\overline{\kappa}_{\rho\rho}}\rho} d\rho = 0. \tag{B4b}$$

We construct Fig. 3(b) using $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^*] = 0$. To understand the accuracy of this approximation, we compute the percent error of $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}$ evaluated along $\mathbf{X}^*$ instead of $\mathbf{X}^c$:

$$\text{error } (\%) \equiv \frac{\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^*] - \Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^c]}{\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^c]}, \tag{B5}$$

as shown in Fig. 7. While $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^c]$ is theoretically zero, in practice it takes a very small numerical value that we use here to construct the percent error. We see that the error goes to zero in the passive limit, $\lambda_{\rho\rho\rho}/\overline{\kappa}_{\rho\rho} \to 0$, and monotonically increases with $\lambda_{\rho\rho\rho}/\overline{\kappa}_{\rho\rho}$. Upon comparison to Fig. 4(b), it is clear that the error in the predicted binodals increases with the error in $\Delta^{\alpha\beta}\mathcal{G}^{\text{int}}[\mathbf{X}^*]$.

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