



Thermodynamically consistent Cahn–Hilliard–Navier–Stokes equations using the metriplectic dynamics formalism

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ARTICLE INFO

Communicated by Y. Zhou

Keywords:

Two phase fluid flow

Cahn–Hilliard

Thermodynamic consistency

Metriplectic dynamics

Hamiltonian structure

ABSTRACT

Cahn–Hilliard–Navier–Stokes (CHNS) systems describe flows with two-phases, e.g., a liquid with bubbles. Obtaining constitutive relations for general dissipative processes for such systems, which are thermodynamically consistent, can be a challenge. We show how the metriplectic 4-bracket formalism (Morrison and Updike, 2024) achieves this in a straightforward, in fact algorithmic, manner. First, from the noncanonical Hamiltonian formulation for the ideal part of a CHNS system we obtain an appropriate Casimir to serve as the entropy in the metriplectic formalism that describes the dissipation (e.g. viscosity, heat conductivity and diffusion effects). General thermodynamics with the concentration variable and its thermodynamics conjugate, the chemical potential, are included. Having expressions for the Hamiltonian (energy), entropy, and Poisson bracket, we describe a procedure for obtaining a metriplectic 4-bracket that describes thermodynamically consistent dissipative effects. The 4-bracket formalism leads naturally to a general CHNS system that allows for anisotropic surface energy effects. This general CHNS system reduces to cases in the literature, to which we can compare.

1. Introduction

The well-known Navier–Stokes equations govern the motion of a single-phase fluid. However, in the case of two-phase fluids, chemical reactions, changes of phase, and migration between substances of phases become significant and cannot be disregarded. J. W. Cahn and J. E. Hilliard were the first to formulate the mathematical equations that describe phase separation in a such a binary fluid [1]. Here we investigate generalizations that combine the Cahn–Hilliard equation with equations that describe the dynamics of fluid flow, referred to as Cahn–Hilliard–Navier–Stokes (CHNS) systems. CHNS systems aim to describe the hydrodynamic properties of a mixture of two phases such as bubbles in a liquid. To narrow down the already broad scope, we assume that the two fluids share the same velocity field, yet we allow for both extended thermodynamics and diffusive interfaces between the two phases.

A substantial hurdle in developing CHNS type systems, systems with a variety of constitutive relations, is to ensure thermodynamic consistency, i.e., adherence to the first law of thermodynamics, which in this context is to produce a set of dynamical equations that conserve energy, and the second law which in this context means the dynamical

production of entropy, ensuring the relaxation (asymptotic stability) to thermodynamic equilibrium. Here we propose an algorithm for constructing such systems, an algorithm that produces a large set of CHNS systems.

The algorithm has four steps: (i) Select a set of dynamical variables. For a CHNS system these will be $\psi := \{\mathbf{m} = \rho\mathbf{v}, \rho, \tilde{c} = \rho c, \sigma = \rho s\}$, which are the momentum density, mass density, volume concentration of one of the constituents, and entropy density, respectively. (ii) The next step is to select energy and entropy functionals, H and S , dependent on the dynamical variables. The choice of these functionals is based on the physics of the phenomena one wishes to describe. (iii) The third step of the algorithm is to obtain the noncanonical Poisson bracket [see 2] of the ideal (nondissipative) part of the theory that has the chosen entropy as a Casimir invariant. Since the work of Morrison and Greene [3], Poisson brackets for a great many systems, including fluid and magnetofluid systems, have been found [e.g. 4–8]. Thus, this step may be immediate. Alternatively, it may be achieved by a coordinate change from a known Hamiltonian theory in order to align with the chosen entropy functional. In either case, we obtain at this stage a noncanonical Hamiltonian system. (iv) The final step is to construct a metriplectic

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<https://doi.org/10.1016/j.physd.2024.134303>

Received 22 February 2024; Received in revised form 17 July 2024; Accepted 19 July 2024

Available online 24 July 2024

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4-bracket as described in [9]. Although there are standard metriplectic 4-bracket constructions, there is freedom at this last step to describe a variety of types of dissipation. However, a natural choice follows upon consideration of the form of an early metriplectic bracket [10]. Given H , S , and the 4-bracket, the dynamical system with thermodynamically consistent dissipation is produced.

We apply the algorithm to two cases. First, in Section 2, we consider a system where the fluid thermodynamics is extended by allowing the internal energy to depend on a concentration variable, with the chemical potential being its thermodynamic dual. Because Gibbs introduced the notion of chemical potential, we refer to the Hamiltonian version of this fluid systems as the Gibbs–Euler (GE) system and the dissipative version as the Gibbs–Navier–Stokes (GNS) system. It is a thermodynamically consistent version of the compressible Navier–Stokes equations with the inclusion of this concentration variable for describing a second phase of the fluid. The GNS system generalizes the early work of Eckart [11,12] and the treatment in [13]; it allows for all possible thermodynamics fluxes. Next, in Section 3, a general form of CHNS system is produced, a form that models surface tension effects and allows for diffuse interfaces. Our work is motivated in large part by the substantial works of Anderson et al. [14] and Guo and Lin [15], which we generalize by obtaining a class of systems that includes theirs as special cases. There is a huge literature on this topic and these papers contain many important references to previous work. [Also, see 16, for a recent review.]

The GNS system of Section 2 serves as a straightforward example of our algorithm. In Section 2.1 we describe the set of dynamical variables, properties of the system, and the energy and entropy functionals H and S . This amounts to the first and second steps of the algorithm. Then in Section 2.2 the Hamiltonian formulation of the dissipation free part of the system is presented. This is the third step of the algorithm where the Poisson bracket is obtained, after a brief review of the noncanonical Hamiltonian formalism. Given the early work of Morrison and Greene [3] and the classification of extensions in [5], this step is immediate. Based on the early and recent works of Morrison [10] and [9] the fourth step of the algorithm is also immediate. In Section 2.3 we first review the metriplectic 4-bracket formalism and present the realization that applies for the GNS system. Thus, the thermodynamically consistent GNS system is determined. In Section 2.4 we obtain the metriplectic 2-bracket equations of motion, and the determined fluxes and affinities, making connection to standard irreversible thermodynamics. Using the results of Section 2, we proceed in Section 3 to obtain the main result of the paper, our general CHNS system that can describe diffuse interface effects. The first and second steps of our algorithm are taken in Section 3.1, while the third step, obtaining the correct Poisson bracket, is undertaken in Section 3.2. In order to complete this step, one must find the Poisson bracket for which the entropy of the second step is a Casimir invariant, which we find can be achieved by a simple coordinate transformation. The fourth step of the algorithm is taken in Section 3.3. Here a choice of metriplectic 4-bracket gives a general class of thermodynamically consistent CHNS systems, a class that contains previous results as special cases. The formalism also shows how one can transform to a simple entropy variable at the expense of a more complicated internal energy. As in Section 2, in Section 3.4 we reduce to the metriplectic 2-bracket. Finally in Section 4 we briefly summarize and make a few comments about ongoing and future work.

2. Metriplectic framework and the Gibbs–Navier–Stokes system

In this section we describe general features of the metriplectic framework in the context of the GNS system, a generalization of the Navier–Stokes equations that includes the dual thermodynamical variables of concentration and chemical potential.

2.1. Description of the Gibbs–Navier–Stokes system

The GNS for 2 phase flow proceeds on familiar ground [11–13]. It amounts to the single phase thermodynamic Navier–Stokes system or as it is sometimes called the Fourier Navier–Stokes system with the dispersed phase described by the addition of a concentration variable, c , giving the set of dynamical variables $\psi = \{\mathbf{v}, \rho, c, s\}$. Here we review global aspects of this known system, before showing how it emerges from the metriplectic formalism.

We suppose the mixture of two phases are contained in a volume Ω , and we consider the following global quantities and their evolution:

$$M = \int_{\Omega} \rho, \quad \dot{M} = 0, \quad (1)$$

$$\mathbf{P} = \int_{\Omega} \rho \mathbf{v}, \quad \dot{\mathbf{P}} = - \int_{\partial\Omega} \bar{\mathbf{J}}_{\mathbf{m}} \cdot \mathbf{n}, \quad (2)$$

$$H = \int_{\Omega} \frac{\rho}{2} |\mathbf{v}|^2 + \rho u(\rho, s, c), \quad \dot{H} = - \int_{\partial\Omega} \mathbf{J}_e \cdot \mathbf{n}, \quad (3)$$

$$C = \int_{\Omega} \rho c, \quad \dot{C} = - \int_{\partial\Omega} \mathbf{J}_c \cdot \mathbf{n}, \quad (4)$$

$$S = \int_{\Omega} \rho s, \quad \dot{S} = - \int_{\partial\Omega} \mathbf{J}_s \cdot \mathbf{n} + \int_{\Omega} \dot{s}^{prod}. \quad (5)$$

Here ρ is the density of the mixture, \mathbf{v} is the mass-averaged velocity of the mixture, s is the specific entropy, and the phase variable c is the specific concentration (dimensionless mass concentration) that determines how much of the dispersed phase of the mixture is present at a point $\mathbf{x} \in \Omega \subset \mathbb{R}^3$. The variable $\bar{c} = \rho c$ is the mass density of the dispersed phase. The functionals M , P , H and S are the total mass, momentum, energy, and entropy of the mixture, respectively, while C is the total mass of one of the constituents. For convenience we omit the incremental volume element for integrations over Ω , i.e., $\int_{\Omega} = \int_{\Omega} d^3x$ and use an over dot to mean the total derivative d/dt . The local thermodynamics of the mixture is described by $u(\rho, s, c)$, the internal energy per unit mass. For convenience the gravitational force is not considered, although its inclusion is straightforward.

Quantities in the time derivatives of the basic functionals are as follows: \mathbf{n} is the unit outward normal vector of the boundary $\partial\Omega$, \mathbf{J}_c is the phase field flux, which depends on the gradient of the chemical potential, $\bar{\mathbf{J}}_{\mathbf{m}}$ is the stress tensor – surface forces – due to pressure and viscosity, \mathbf{J}_e the energy flux that contains the rate of work done by the surface forces (external energy), the rate of heat transfer and the rate of diffusivity in the phase field (internal energy), \mathbf{J}_s is the net entropy flux through the boundary, and \dot{s}^{prod} is the local rate of entropy production. The second law of thermodynamics is expressed by the requirement that \dot{s}^{prod} is non-negative.

For the GNS system the fluxes are given by

$$\mathbf{J}_c = -\bar{D} \cdot \nabla \mu, \quad (6)$$

$$\bar{\mathbf{J}}_{\mathbf{m}} = p \bar{\mathbf{I}} - \bar{\bar{\Lambda}} : \nabla \mathbf{v}, \quad (7)$$

$$\mathbf{J}_e = -\mathbf{v} \cdot \bar{\bar{\Lambda}} : \nabla \mathbf{v} - \bar{\kappa} \cdot \nabla T - \mu \bar{D} \cdot \nabla \mu, \quad (8)$$

$$\mathbf{J}_s = -\frac{\bar{\kappa}}{T} \cdot \nabla T, \quad (9)$$

$$\dot{s}^{prod} = \frac{1}{T} \left[\nabla \mathbf{v} : \bar{\bar{\Lambda}} : \nabla \mathbf{v} + \frac{1}{T} \nabla T \cdot \bar{\kappa} \cdot \nabla T + \nabla \mu \cdot \bar{D} \cdot \nabla \mu \right] \geq 0, \quad (10)$$

where p is the pressure, T is the temperature, $\bar{\mathbf{I}}$ is the unit tensor, $\bar{\kappa}$ is the thermal conductivity tensor, \bar{D} is the diffusion tensor, which along with $\bar{\kappa}$ is assumed to be a symmetric and positive definite 2-tensor, and μ is the chemical potential. We allow the possibility that phenomenological quantities such as $\bar{\kappa}$ and \bar{D} can depend on the dynamical variables. Here, $\bar{\bar{\Lambda}}$ is the viscosity 4-tensor, the usual rank 4 isotropic Cartesian tensor given by

$$A_{ijkl} = \eta \left(\delta_{il} \delta_{jk} + \delta_{jl} \delta_{ik} - \frac{2}{3} \delta_{ij} \delta_{kl} \right) + \zeta \delta_{ij} \delta_{kl}, \quad (11)$$

with viscosity coefficients η and ζ and i, j, k and l taking on values 1, 2, 3. Note, we use boldface as in the fluxes $\mathbf{J}_c, \mathbf{J}_e$, and \mathbf{J}_s to denote

vectors, an over bar as in $\bar{\mathbf{J}}_{\mathbf{m}}$ to denote rank-2 tensors, and a double over bar as in $\bar{\bar{A}}$ to denote rank-4 tensors. A single “.” is used for neighboring contractions, e.g., $(\bar{D} \cdot \nabla \mu)_i = D_{ij} \partial_j \mu$ and we use the double dot convention, e.g., for the stress tensor $(\bar{A} : \nabla \mathbf{v})_{ij} = A_{ijkl} \partial_k v_l$, where repeated indices are summed over.

The volume density variables are $\psi = (\mathbf{m} := \rho \mathbf{v}, \rho, \bar{c} := \rho c, \sigma := \rho s)$, where \mathbf{m} is the momentum density, σ is entropy per unit volume and \bar{c} is the concentration per unit volume. The local energy per unit volume is given by

$$e = \frac{|\mathbf{m}|^2}{2\rho} + \rho u(\rho, s, c). \quad (12)$$

From the specific internal energy, $u(\rho, s, c)$, we have the thermodynamic relations

$$du = T ds + \frac{p}{\rho^2} d\rho + \mu dc, \quad (13)$$

where

$$T = \frac{\partial u}{\partial s}, \quad p = \rho^2 \frac{\partial u}{\partial \rho}, \quad \mu = \frac{\partial u}{\partial c}. \quad (14)$$

Given the content of this section, we have established the first step of our algorithm for the GNS system, the determination of the dynamical variables $\psi = \{\mathbf{m} = \rho \mathbf{v}, \rho, \bar{c} = \rho c, \sigma = \rho s\}$ or alternatively the set (\mathbf{v}, ρ, c, s) , and the second step of our algorithm by making the choices of Hamiltonian H of (3) and entropy S of (5). In the next section, 2.2, we proceed to the third step of the algorithm by obtaining the Hamiltonian structure for this system. This system without dissipation is the GE system.

2.2. Noncanonical Poisson bracket of the Gibbs-Euler system

Given that the mixture is assumed to be confined in the domain Ω , the Eulerian scalars (volume forms) (ρ, \bar{c}, σ) are functions from space-time $\Omega \mapsto \mathbb{R} \rightarrow \mathbb{R}$, while the vector field \mathbf{m} maps $\Omega \times \mathbb{R} \mapsto T\Omega$, where $T\Omega$ stands for the tangent bundle of the manifold Ω . We will forgo formal geometric considerations and suppose our infinite-dimensional phase space has coordinates $\psi = (\mathbf{m}, \rho, \bar{c}, \sigma)$ and observables are functionals that map $\psi \mapsto \mathbb{R}$ at each fixed time. We will denote the space of such functionals by \mathcal{B} . Then a Poisson bracket is a bilinear operator $\mathcal{B} \times \mathcal{B} \mapsto \mathbb{R}$ that fulfills the Leibniz rule and is a realization of a Lie algebra [see e.g. 17, chap. 14]. The Leibniz rule follows from that for the variational or functional derivative of $F \in \mathcal{B}$, defined by

$$\delta F[\psi; \delta \psi] = \lim_{\epsilon \rightarrow 0} \frac{F(\psi + \epsilon \delta \psi) - F(\psi)}{\epsilon} = \int_{\Omega} \frac{\delta F}{\delta \psi} \delta \psi$$

where $\delta F/\delta \psi$ is the functional derivative. This expression can be viewed as the directional derivative of a functional F at ψ in the direction $\delta \psi$ [see, e.g., 2, for a formal review of these notions].

The appropriate Poisson bracket, defined on two functionals $F, G \in \mathcal{B}$, for the GE system is the following:

$$\begin{aligned} \{F, G\} = & - \int_{\Omega} \mathbf{m} \cdot [F_{\mathbf{m}} \cdot \nabla G_{\mathbf{m}} - G_{\mathbf{m}} \cdot \nabla F_{\mathbf{m}}] + \rho [F_{\rho} \cdot \nabla G_{\rho} - G_{\rho} \cdot \nabla F_{\rho}] \\ & + \sigma [F_{\sigma} \cdot \nabla G_{\sigma} - G_{\sigma} \cdot \nabla F_{\sigma}] + \bar{c} [F_{\bar{c}} \cdot \nabla G_{\bar{c}} - G_{\bar{c}} \cdot \nabla F_{\bar{c}}], \end{aligned} \quad (15)$$

where we compactified our notation by defining $F_{\mathbf{m}} := \delta F/\delta \mathbf{m}$, $F_{\rho} := \delta F/\delta \rho$, etc., the functional derivatives with respect to the various coordinates ψ . That this is the appropriate Poisson bracket is immediate; it is the Lie-Poisson bracket originally given by Morrison and Greene [3] with the addition of the last line of (15) involving the concentration, another volume density variable \bar{c} . Adding such a dynamical variable is common place in the fluid modeling of plasmas over the last decades and fits within the general theory for extension given by Thiffeault and Morrison [5]. By construction we have a Poisson bracket that is a bilinear, antisymmetric, and either by the extension theory or a relatively easy direct calculation using the techniques of [4] it can be shown to satisfy the Jacobi identity, i.e.,

$$\{\{F, G\}, H\} + \{\{H, F\}, G\} + \{\{G, H\}, F\} = 0, \quad (16)$$

for all $F, G, H \in \mathcal{B}$. The Leibniz property, which is required for the Poisson bracket to generate a vector field, is built into the definition of functional derivative.

Upon inserting any functional of ψ , say an observable o , into the Poisson bracket its evolution is determined by

$$\partial_t o = \{o, H\}, \quad (17)$$

where the Hamiltonian functional is the total energy of the system, where we rewrite (3) as follows:

$$H[\rho, \mathbf{m}, \sigma, \bar{c}] = \int_{\Omega} e = \int_{\Omega} \frac{|\mathbf{m}|^2}{2\rho} + \rho u\left(\rho, \frac{\sigma}{\rho}, \frac{\bar{c}}{\rho}\right). \quad (18)$$

In (17) and henceforth we use the shorthand $\partial_t = \partial/\partial t$. Using the following functional derivatives:

$$H_{\rho} = -|\mathbf{v}|^2/2 + u + p/\rho - sT - c\mu, \quad H_{\mathbf{m}} = \mathbf{v}, \quad H_{\sigma} = T, \quad H_{\bar{c}} = \mu, \quad (19)$$

the bracket form of (17) gives the ideal two-phase flow system

$$\partial_t \mathbf{v} = \{\mathbf{v}, H\} = -\mathbf{v} \cdot \nabla \mathbf{v} - \nabla p/\rho, \quad (20)$$

$$\partial_t \rho = \{\rho, H\} = -\mathbf{v} \cdot \nabla \rho - \rho \nabla \cdot \mathbf{v}, \quad (21)$$

$$\partial_t \bar{c} = \{\bar{c}, H\} = -\mathbf{v} \cdot \nabla \bar{c} - \bar{c} \nabla \cdot \mathbf{v}, \quad (22)$$

$$\partial_t \sigma = \{\sigma, H\} = -\mathbf{v} \cdot \nabla \sigma - \sigma \nabla \cdot \mathbf{v}. \quad (23)$$

Here we have dropped surface terms arising from integration by parts and have used $\delta \rho(\mathbf{x})/\delta \rho(\mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}')$. Eqs. (20)–(22) can also be written easily using, e.g., $D\rho/Dt := \partial\rho/\partial t + \mathbf{v} \cdot \nabla \rho$. These equations comprise the GE system.

Casimir invariants are special functionals \mathcal{C} that satisfy

$$\{F, \mathcal{C}\} = 0 \quad \forall F \in \mathcal{B}, \quad (24)$$

and thus are constants of motion for any Hamiltonian. From (15) we obtain the following equations that a Casimir functional \mathcal{C} must satisfy:

$$\nabla \cdot (\rho \mathcal{C}_{\mathbf{m}}) = \nabla \cdot (\sigma \mathcal{C}_{\sigma}) = \nabla \cdot (\bar{c} \mathcal{C}_{\bar{c}}) = 0, \quad (25)$$

and

$$m_j \nabla \mathcal{C}_{m_j} + \partial_j (\mathbf{m} \mathcal{C}_{m_j}) + \rho \nabla \mathcal{C}_{\rho} + \sigma \nabla \mathcal{C}_{\sigma} + \bar{c} \nabla \mathcal{C}_{\bar{c}} = 0, \quad (26)$$

where we use the shorthand $\delta \mathcal{C}/\delta \mathbf{m} := \mathcal{C}_{\mathbf{m}}$, $\delta \mathcal{C}/\delta \rho := \mathcal{C}_{\rho}$, etc. and summation of repeated indices is assumed. For the purpose at hand we assume \mathcal{C} is independent of \mathbf{m} , yielding the single condition

$$\rho \nabla \mathcal{C}_{\rho} + \sigma \nabla \mathcal{C}_{\sigma} + \bar{c} \nabla \mathcal{C}_{\bar{c}} = 0. \quad (27)$$

Eq. (27) is satisfied by

$$\mathcal{C} = \int_{\Omega} C(\rho, \sigma, \bar{c}) \quad (28)$$

for any C that is Euler homogeneous of degree one, i.e., satisfies

$$C(\lambda \rho, \lambda \sigma, \lambda \bar{c}) = \lambda C(\rho, \sigma, \bar{c}). \quad (29)$$

The proof of this is straightforward.

To complete the third step of our algorithm, the entropy functional must be chosen from the set of Casimir invariants. Writing the Euler homogeneous integrand as

$$C(\rho, \sigma, \bar{c}) = \rho f(\sigma/\rho, \bar{c}/\rho)$$

it is clear that

$$S = \int_{\Omega} \rho s = \int_{\Omega} \sigma \quad (30)$$

lies in our set of Casimirs. This quantity was first shown to be a Casimir for the ideal fluid in [4] and used for the thermodynamically consistent Navier–Stokes metriplectic system in [10]. We note in passing, for other theories that might have a nontraditional dynamical equilibrium playing the role of thermodynamic equilibrium, one may wish to choose another Casimir.

2.3. Metriplectic 4-bracket for the Gibbs-Navier–Stokes system

Now let us turn to our fourth and final step of the algorithm, construction of the metriplectic 4-bracket. To this end we review the formalism of [9] in general terms and then apply it to the GNS system, which is a generalization of an example given in that work.

2.3.1. General metriplectic 4-bracket dynamics

The metriplectic 4-bracket theory was introduced by Morrison and Updike [9] to describe dissipative dynamics. Let us briefly recall the metriplectic 4-bracket description in infinite dimensions. In this description, we consider the dynamics of classical field theories with multi-component fields

$$\chi(z, t) = (\chi^1(z, t), \chi^2(z, t), \dots, \chi^M(z, t)) \quad (31)$$

defined on $z = (z^1, z^2, \dots, z^N)$ for times $t \in \mathbb{R}$. Here we use z to be a label space coordinate with the volume element $d^N z$, but with the domain unspecified. In fluid mechanics this domain would be Ω , the 3-dimensional domain occupied by the fluid and recall we used x for the coordinate of this point. In general we suppose that χ^1, \dots, χ^M are real-valued functions of z and t . Given the space of functionals of χ , B , we define 4-bracket as an operator

$$(\cdot, \cdot; \cdot, \cdot) : B \times B \times B \times B \rightarrow B \quad (32)$$

such that for any four functionals $F, K, G, N \in B$ we have

$$(F, G; K, N) = \int d^N z \int d^N z' \int d^N z'' \int d^N z''' \hat{R}^{\alpha\beta\gamma\delta} \times \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z')} \frac{\delta K}{\delta \chi^\gamma(z'')} \frac{\delta N}{\delta \chi^\delta(z''')}, \quad (33)$$

where $\hat{R}^{\alpha\beta\gamma\delta}(z, z', z'', z''')$ is a 4-tensor functional operator with coordinate form given by the following integral kernel:

$$\hat{R}^{\alpha\beta\gamma\delta}(z, z', z'', z''')[\chi] = \hat{R}(\mathbf{d}\chi^\alpha(z), \mathbf{d}\chi^\beta(z'), \mathbf{d}\chi^\gamma(z''), \mathbf{d}\chi^\delta(z'''))[\chi(z)], \quad (34)$$

where $\alpha, \beta, \gamma, \delta$ range over $1, 2, \dots, M$. The 4-bracket is assumed to satisfy the following proprieties:

(i) Linearity in all arguments, e.g., for all $\lambda \in \mathbb{R}$

$$(F + \lambda H, K; G, N) = (F, K; G, N) + \lambda(H, K; G, N) \quad (35)$$

(ii) The algebraic symmetries

$$(F, K; G, N) = -(K, F; G, N) \quad (36)$$

$$(F, K; G, N) = -(F, K; N, G) \quad (37)$$

$$(F, K; G, N) = (G, N; F, K) \quad (38)$$

(iii) Derivation in all arguments, e.g.,

$$(FH, K; G, N) = F(H, K; G, N) + (F, K; G, N)H. \quad (39)$$

Here, as usual, FH denotes point-wise multiplication. In addition, to ensure entropy production we require

$$\dot{S} = (S, H; S, H) \geq 0. \quad (40)$$

Metriplectic 4-brackets that satisfy (35)–(40) are called *minimal metriplectic*. In Section 2.3.2 we will give a construction that ensures such appropriate positive semidefiniteness.

The minimal metriplectic properties of metriplectic 4-brackets are reminiscent of the algebraic properties possessed by a curvature tensor. In fact, every Riemannian manifold naturally has a metriplectic 4-bracket, and $(S, H; S, H)$ provides a notion of sectional curvature [see 9].

From the metriplectic 4-bracket (33), the dissipative dynamics of an observable o is generated as follows:

$$\partial_t o = (o, H; S, H) = \int d^N z \int d^N z' \int d^N z'' \int d^N z''' \hat{R}^{\alpha\beta\gamma\delta}$$

$$\times \frac{\delta o}{\delta \chi^\alpha(z)} \frac{\delta H}{\delta \chi^\beta(z')} \frac{\delta S}{\delta \chi^\gamma(z'')} \frac{\delta H}{\delta \chi^\delta(z''')}. \quad (41)$$

If we choose o to be the Hamiltonian H , then $\dot{H} = (H, H; S, H) \equiv 0$ by the antisymmetry condition of (36). If we choose o to be the entropy S , then $\dot{S} = (S, H; S, H) \geq 0$ by (40).

The dissipative dynamics generated by the 4-bracket on our set of field variables χ is given by

$$\partial_t \chi^\alpha(z) = (\chi^\alpha, H; S, H) = \int d^N z'' G^{\alpha\beta}(z, z'') \frac{\delta S}{\delta \chi^\beta(z'')}, \quad (42)$$

where the G -metric is given as follows:

$$G^{\alpha\gamma}(z, z'') := \int d^N z' \int d^N z''' R^{\alpha\beta\gamma\delta}(z, z', z'', z''') \frac{\delta H}{\delta \chi^\beta(z')} \frac{\delta H}{\delta \chi^\delta(z''')}. \quad (43)$$

For the full metriplectic dynamics we would add the Poisson bracket contribution to the above. Eq. (42) is written so as to show that it amounts to a gradient system with the entropy S as generator.

2.3.2. General Kulkarni–Nomizu construction

We can easily create specific metriplectic 4-brackets that have the minimal metriplectic properties: the requisite symmetries and the positive semidefiniteness $(S, H; S, H)$. We do this by using the Kulkarni–Nomizu (K-N) product [18,19]. See also [20] for relevant theorems. Consistent with the bracket formulation of (33), we deviate from the conventional K-N product by working on the dual. Given two symmetric operator fields, say Σ and M , operating on the variational derivatives; we again use the subscript notation when convenient,

$$F_\chi := \frac{\delta F}{\delta \chi} = \left(\frac{\delta F}{\delta \chi^1}, \frac{\delta F}{\delta \chi^2}, \dots, \frac{\delta F}{\delta \chi^M} \right),$$

the K-N product is defined as follows:

$$\begin{aligned} (\Sigma \wedge M)(dF, dK, dG, dN) &= \Sigma(dF, dG) M(dK, dN) \\ &\quad - \Sigma(dF, dN) M(dK, dG) \\ &\quad + M(dF, dG) \Sigma(dK, dN) \\ &\quad - M(dF, dN) \Sigma(dK, dG). \end{aligned} \quad (44)$$

A finite-dimensional form of Σ would be a symmetric contravariant 2-tensor, say γ , and this would give the term

$$\gamma(df, dg) = \gamma^{ij} \frac{\partial f}{\partial z^i} \frac{\partial g}{\partial z^j}. \quad (45)$$

A conventional form of K-N product would involve rank 2 covariant tensors. The form of (45) suggests a general form in infinite dimensions would be

$$\Sigma(dF, dG) = \int d^N z \int d^N z' \Sigma^{\alpha\beta}(z, z') \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z')}, \quad (46)$$

where $\Sigma^{\alpha\beta}(z, z')$ is symmetric in both α, β and z, z' and operates to the right on both functional derivatives. For example,

$$\Sigma^{\alpha\beta}(z, z') = L_{ab}^{\alpha\beta}(z, z') \mathcal{L}^a \mathcal{L}^b, \quad (47)$$

where $L_{ab}^{\alpha\beta}$ is symmetric and \mathcal{L}^a is a differential operator. This implies, e.g.,

$$\Sigma(dF, dG) = \int d^N z \int d^N z' L_{ab}^{\alpha\beta}(z, z') \mathcal{L}^a \frac{\delta F}{\delta \chi^\alpha(z)} \mathcal{L}^b \frac{\delta G}{\delta \chi^\beta(z')}. \quad (48)$$

With an expression for M similar to (46), a term in the K-N decomposition would have the following form:

$$\begin{aligned} &\int d^N z \int d^N z' \int d^N z'' \int d^N z''' \Sigma^{\alpha\beta}(z, z') M^{\gamma\delta}(z'', z''') \\ &\quad \times \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z')} \frac{\delta K}{\delta \chi^\gamma(z'')} \frac{\delta N}{\delta \chi^\delta(z''')}, \end{aligned} \quad (49)$$

which could be generalized further by adding filtering kernels.

It is easy to see that brackets constructed with this K-N product will have all of the algebraic symmetries described in Section 2.3.1. In addition, it is shown in the Appendix using the Cauchy–Schwarz

inequality that positivity of $(S, H; S, H)$ is satisfied, if both Σ and M are positive semidefinite. Moreover, if one of Σ or M is positive definite, defining an inner product, then the sectional curvature of (37) satisfies $(S, H; S, H) \geq 0$ with equality if and only if $\delta S / \delta \chi \propto \delta H / \delta \chi$. Thus, it is not difficult to build minimal metriplectic 4-brackets.

Alternative to (46) we can define $\Sigma(dF, dG)$ pointwise as

$$\begin{aligned} \Sigma(dF, dG)(z) &:= \int d^N z' \Sigma^{\alpha\beta}(z, z') \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z')} \\ &= A^{\alpha\beta}(z) \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z)}, \end{aligned} \quad (50)$$

which could follow from (46) if we added an additional argument to Σ . Then, with a corresponding form for M the algebraic curvature symmetries would be induced in the integrand. This is the case for our present purposes, where we assume the specific K-N form given in [9], viz. where the 4-bracket is given by

$$(F, K; G, N) = \int d^N z W(\Sigma \wedge M)(dF, dK, dG, dN),$$

where W is an arbitrary weight, possibly depending on χ and z , that multiplies $(\Sigma \wedge M)$ where all of the functional derivatives are evaluated at the same point, z . (See (52) and (53) below.) In Section 2.3.3 we will see that this form of 4-bracket is sufficient for the CHNS systems of interest.

2.3.3. Metriplectic 4-bracket for the GNS system

Now suppose our multi-component field variable χ is that for the multiphase fluid, i.e.,

$$\psi(\mathbf{x}, t) = (\mathbf{m}(\mathbf{x}, t), \rho(\mathbf{x}, t), \tilde{c}(\mathbf{x}, t), \sigma(\mathbf{x}, t)) \quad (51)$$

and consider a specific, but still quite general, form of the K-N construction, one adaptable to the GNS type of system. For multi-component fields ψ of our fluid we could choose

$$M(dF, dG) = F_{\psi^\gamma} A^{\gamma\delta} G_{\psi^\delta}, \quad (52)$$

$$\Sigma(dF, dG) = \nabla F_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla G_{\psi^\beta}, \quad (53)$$

where repeated indices are to be summed, $A^{\gamma\delta} = A^{\delta\gamma}$, and in coordinates $B_{ij}^{\alpha\beta}$ is symmetric in both $i, j = 1, 2, 3$ and $\alpha, \beta = 1, \dots, 6$, which are indices that range over the six fields of ψ . Here, the nablas are contracted on i and j . With the choices of (52) and (53), the metriplectic 4-bracket is

$$\begin{aligned} (F, K; G, N) &= \int_{\Omega} \nabla F_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla G_{\psi^\beta} K_{\psi^\gamma} A^{\gamma\delta} N_{\psi^\delta} \\ &\quad - \nabla F_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla N_{\psi^\beta} K_{\psi^\gamma} A^{\gamma\delta} G_{\psi^\delta} \\ &\quad + \nabla K_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla N_{\psi^\beta} F_{\psi^\gamma} A^{\gamma\delta} G_{\psi^\delta} \\ &\quad - \nabla K_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla G_{\psi^\beta} F_{\psi^\gamma} A^{\gamma\delta} N_{\psi^\delta}. \end{aligned} \quad (54)$$

Entropy production is governed by

$$\begin{aligned} (S, H; S, H) &= \int_{\Omega} \nabla S_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla S_{\psi^\beta} H_{\psi^\gamma} A^{\gamma\delta} H_{\psi^\delta} \\ &\quad + \nabla H_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla H_{\psi^\beta} S_{\psi^\gamma} A^{\gamma\delta} S_{\psi^\delta} \\ &\quad - 2 \nabla H_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla S_{\psi^\beta} S_{\psi^\gamma} A^{\gamma\delta} H_{\psi^\delta}. \end{aligned} \quad (55)$$

From the general results of the Appendix it follows that $(S, H; S, H) \geq 0$ for this special case if $A^{\alpha\beta}$ and $B^{\alpha\beta}$ are positive semidefinite.

Observe that (53) could be replaced by the more general expression

$$\Sigma(dF, dG) = \mathcal{L} F_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \mathcal{L} G_{\psi^\beta}, \quad (56)$$

where \mathcal{L} is contained within a general class of pseudodifferential operators. Later we will see an example of this.

Now consider an even more restrictive K-N product, a special case of (54) with what appears to be the simplest K-N options. As discussed

earlier, we do not expect our 4-bracket to depend on functional derivatives with respect to ρ , which could produce density diffusion. Thus, for M we take

$$M(dF, dG) = F_\sigma G_\sigma. \quad (57)$$

The placement of the ∇ in (54) leads to a diffusive type of relaxation, so this is natural, and the simplest case would be to select Σ with no cross terms, i.e.,

$$\Sigma(dF, dG) = \nabla F_{\mathbf{m}} : \bar{\Lambda}_1 : \nabla G_{\mathbf{m}} + \nabla F_\sigma \cdot \bar{\Lambda}_2 \cdot \nabla G_\sigma + \nabla F_{\tilde{c}} \cdot \bar{\Lambda}_3 \cdot \nabla G_{\tilde{c}}, \quad (58)$$

where the 4-tensor $\bar{\Lambda}_1$ and the symmetric 2-tensors $\bar{\Lambda}_2$ and $\bar{\Lambda}_3$ are to be determined. We make the following choices

$$\bar{\Lambda}_1 = \frac{\bar{\Lambda}}{T}, \quad \bar{\Lambda}_2 = \frac{\bar{\kappa}}{T^2}, \quad \bar{\Lambda}_3 = \frac{\bar{D}}{T}, \quad (59)$$

where $\bar{\Lambda}$ is the isotropic Cartesian 4-tensor given by (11), $\bar{\Lambda}_{2,3}$ are symmetric positive definite 2-tensors defined by the previously introduced $\bar{\kappa}$ and \bar{D} . We take the weight W to be the Lagrange multiplier defined in Section 2.4.1 i.e. $W = 1$. Then, the 4-bracket reads

$$\begin{aligned} (F, K; G, N) &= \int_{\Omega} \frac{1}{T} \left[[K_\sigma \nabla F_{\mathbf{m}} - F_\sigma \nabla K_{\mathbf{m}}] : \bar{\Lambda} : [N_\sigma \nabla G_{\mathbf{m}} - G_\sigma \nabla N_{\mathbf{m}}] \right. \\ &\quad + \frac{1}{T} [K_\sigma \nabla F_\sigma - F_\sigma \nabla K_\sigma] \cdot \bar{\kappa} \cdot [N_\sigma \nabla G_\sigma - G_\sigma \nabla N_\sigma] \\ &\quad \left. + [K_\sigma \nabla F_{\tilde{c}} - F_\sigma \nabla K_{\tilde{c}}] \cdot \bar{D} \cdot [N_\sigma \nabla G_{\tilde{c}} - G_\sigma \nabla N_{\tilde{c}}] \right]. \end{aligned} \quad (60)$$

Upon insertion of H as given by (18) and S from the set of Casimirs of Section 2.2 to be as in (30), the dynamics is given by

$$\partial_t \psi^\alpha = \{\psi^\alpha, H\} + (\psi^\alpha, H; S, H). \quad (61)$$

Using $H_{\mathbf{m}} = \mathbf{v}$, $H_\sigma = T$, and $S_\sigma = 1$, the following GNS system is produced:

$$\partial_t \mathbf{v} = \{\mathbf{v}, H\} + (\mathbf{v}, H; S, H) = -\mathbf{v} \cdot \nabla \mathbf{v} - \nabla p / \rho + \frac{1}{\rho} \nabla \cdot (\bar{\Lambda} : \nabla \mathbf{v}), \quad (62)$$

$$\partial_t \rho = \{\rho, H\} + (\rho, H; S, H) = -\mathbf{v} \cdot \nabla \rho - \rho \nabla \cdot \mathbf{v}, \quad (63)$$

$$\partial_t \tilde{c} = \{\tilde{c}, H\} + (\tilde{c}, H; S, H) = -\mathbf{v} \cdot \nabla \tilde{c} - \tilde{c} \nabla \cdot \mathbf{v} + \nabla \cdot (\bar{D} \cdot \nabla \mu), \quad (64)$$

$$\begin{aligned} \partial_t \sigma &= \{\sigma, H\} + (\sigma, H; S, H) \\ &= -\mathbf{v} \cdot \nabla \sigma - \sigma \nabla \cdot \mathbf{v} + \nabla \cdot \left(\frac{\bar{\kappa}}{T} \cdot \nabla T \right) + \frac{1}{T^2} \nabla T \cdot \bar{\kappa} \cdot \nabla T \\ &\quad + \frac{1}{T} \nabla \mathbf{v} : \bar{\Lambda} : \nabla \mathbf{v} + \frac{1}{T} \nabla \mu \cdot \bar{D} \cdot \nabla \mu. \end{aligned} \quad (65)$$

By construction we automatically have energy conservation, i.e., for (18) $\dot{H} = 0$, and entropy production

$$\begin{aligned} \dot{S} &= (S, H; S, H) \\ &= \int_{\Omega} \frac{1}{T} \left[\nabla \mathbf{v} : \bar{\Lambda} : \nabla \mathbf{v} + \frac{1}{T} \nabla T \cdot \bar{\kappa} \cdot \nabla T + \nabla \mu \cdot \bar{D} \cdot \nabla \mu \right] \geq 0. \end{aligned} \quad (66)$$

2.4. GNS metriplectic 2-bracket and conventional fluxes and affinities

For completeness we demonstrate two things in this subsection: how the metriplectic 4-bracket formalism relates to the original binary metriplectic formalism given in [10,21,22] and how it relates to conventional nonequilibrium thermodynamics, making the connection between the 4-bracket K-N construction and the phenomenology of thermodynamics fluxes and affinities (sometimes called thermodynamic forces).

2.4.1. GNS metriplectic 2-bracket

As noted above we are concerned with the metriplectic dynamics introduced in [10,21,22] [see also 23,24], but we mention that other binary brackets for describing dissipation were presented over the years [e.g. 25–31]. In addition we mention a recent alternative approach to multiphase fluids, one based on constrained variational principles, that is given in [32]. We refer the reader to [9] for comparisons

with other formulations and how they emerge from the metriplectic 4-bracket.

Metriplectic dynamics was introduced as a means of building thermodynamically consistent theories in terms of a binary bracket, which we now call the metriplectic 2-bracket. The theory applies to a wide class of dynamical systems, including both ordinary and partial differential equations. Evolution of an observable o using the metriplectic 2-bracket has the following form:

$$\partial_t o = \{o, F\} - (o, F)_H, \quad (67)$$

where as before $\{\cdot, \cdot\}$ is the noncanonical Poisson bracket that generates the ideal part of the dynamics, while now $(F, G)_H$, the metriplectic 2-bracket, generates the dissipative part. The functional F represents the global Helmholtz free energy of the system, and is given by:

$$F = H - TS, \quad (68)$$

where again H is the Hamiltonian and S the entropy selected from the set of Casimirs of the noncanonical Poisson bracket (ensuring $\{F, S\} = 0$ for any functional F), and T is a uniform nonnegative constant (a global temperature). The metriplectic 2-bracket (\cdot, \cdot) is assumed to be bilinear, symmetric, and satisfies

$$(F, H)_H \equiv 0 \quad \text{for any functional } F. \quad (69)$$

Thus, metriplectic systems are thermodynamically consistent:

First law (energy conservation):

$$\begin{aligned} \dot{H} &= \{H, F\} - (H, F)_H \\ &= \{H, H\} + \mathcal{T}(H, S)_H = 0; \end{aligned} \quad (70)$$

Second law (entropy production):

$$\begin{aligned} \dot{S} &= \{S, F\} - (S, F)_H \\ &= -(S, H)_H + \mathcal{T}(S, S)_H = \mathcal{T}(S, S)_H \geq 0, \end{aligned} \quad (71)$$

which follows because $\{S, F\} \equiv 0$ and $(S, H)_H \equiv 0$. As shown in [9] the metriplectic 2-bracket emerges from the 4-bracket as follows:

$$(F, G)_H = (F, H; G, H), \quad (72)$$

where for convenience here and henceforth we set $\mathcal{T} = 1$. Because of the minimal metriplectic properties of the 4-bracket, we are assured to have the thermodynamic consistency of (70) and (71).

The 2-bracket that emerges from the general 4-bracket of (54) is the following:

$$\begin{aligned} (F, G)_H &= (F, H; G, H) \\ &= \int_{\Omega} \nabla F_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla G_{\psi^\beta} H_{\psi^\gamma} A^{\gamma\delta} H_{\psi^\delta} \\ &\quad - \nabla F_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla H_{\psi^\beta} H_{\psi^\gamma} A^{\gamma\delta} G_{\psi^\delta} \\ &\quad + \nabla H_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla H_{\psi^\beta} F_{\psi^\gamma} A^{\gamma\delta} G_{\psi^\delta} \\ &\quad - \nabla H_{\psi^\alpha} \cdot B^{\alpha\beta} \cdot \nabla G_{\psi^\beta} F_{\psi^\gamma} A^{\gamma\delta} H_{\psi^\delta}, \end{aligned} \quad (73)$$

which in light of the K-N product satisfies $(F, H)_H = 0$ for all F . This will be true for any choice of the Hamiltonian H . Indeed, a special case of this was used in (60) to obtain the thermodynamically consistent set of Eqs. (62)–(64). Recall, for this case M and Σ were chosen as in (57) and (58), special cases of (52) and (53).

Another 2-bracket can be obtained from (54) by making a convenient choice of variables; viz., instead of the variables ψ of (134) in (73) we choose the following:

$$\xi(\mathbf{x}, t) := (\mathbf{m}(\mathbf{x}, t), \rho(\mathbf{x}, t), \tilde{c}(\mathbf{x}, t), e(\mathbf{x}, t)), \quad (74)$$

where the total energy density is used instead of the entropy density as one of our dynamical variables. That this is possible is well known in thermodynamics because the entropy must be a monotonic increasing

function of the internal energy, which allows via the inverse function theorem transformation between the extensive energy or extensive entropy representations [33]. We will denote these density variables in order by ξ_α , $\alpha = 1, \dots, 6$. With this choice the Hamiltonian (18) is given by

$$H = \int_{\Omega} e = \int_{\Omega} \xi_6, \quad (75)$$

$\delta H / \delta \xi_\alpha = \delta_{\alpha 6}$, the Kronecker delta, and $\nabla \delta H / \delta \xi_\alpha \equiv 0$. Thus, (73) reduces to

$$(F, G)_H = (F, H; G, H) = \int_{\Omega} \nabla F_{\xi_\alpha} \cdot L_{\alpha\beta} \cdot \nabla G_{\xi_\beta}, \quad (76)$$

where without loss of generality we set $A^{66} = 1$ and $B^{\alpha\beta} = L_{\alpha\beta}$ for this case. In Section 2.4.2 we will follow [24] and show how the bracket of (76) fits into the framework of conventional nonequilibrium thermodynamics as, e.g., described in [13]. In this way we will physically identify the meaning of $L_{\alpha\beta}$.

2.4.2. Fluxes and affinities for the GNS system

A fundamental equations of nonequilibrium thermodynamics is the general thermodynamic identity

$$d\sigma = X^\alpha d\xi_\alpha, \quad (77)$$

which relates σ , the entropy density, to the ξ_α densities associated with conserved extensive properties and to $X^\alpha := \partial\sigma / \partial\xi_\alpha$, quantities called affinities (or thermodynamic forces). All the densities are characterized by the following conservation equations:

$$\partial_t \xi_\alpha + \nabla \cdot \mathcal{J}_\alpha = 0, \quad (78)$$

where \mathcal{J}_α is at present an unknown flux associated with the density ξ_α . Then, the evolution of the entropy is given by

$$\partial_t \sigma + \nabla \cdot (X^\alpha \mathcal{J}_\alpha) = \mathcal{J}_\alpha \cdot \nabla X^\alpha. \quad (79)$$

The righthand side of (79) is the dissipative term, which is the sum of the fluxes \mathcal{J}_α contracted with ∇X^α . The linear assumption of nonequilibrium processes amounts to relating fluxes and affinities according to

$$\mathcal{J}_\alpha = L_{\alpha\beta} X^\beta. \quad (80)$$

If we identify the $L_{\alpha\beta}$ of (80) with that of (76), we see how metriplectic brackets are related to the flux-affinity relations. Onsager symmetry, assumed to arise from microscopic reversibility, amounts to the symmetry $L_{\alpha\beta} = L_{\beta\alpha}$ and the semi-definiteness property assures the second law, i.e., entropy growth. It remains to identify how the fluxes of Section 2.1 enter the picture.

To further identify the meaning of $L_{\alpha\beta}$ we revisit the thermodynamics of (13), in light of our choice of the variables ξ of (134). Thus, we rewrite the thermodynamic relation (13) upon changing variables,

$$T d\sigma = de - \mathbf{v} \cdot d\mathbf{m} - \mu d\tilde{c} - g d\rho, \quad (81)$$

where e is the energy density of (12) and g is a modified specific Gibbs free energy, viz.

$$g := u - Ts + p/\rho - \mu c - |\mathbf{v}|^2/2. \quad (82)$$

We have assumed in (1) that there is no flux associated with ρ , i.e., in CHNS chemical reactions and/or particle creation and annihilation are ignored. Thus, the phase space for the thermodynamics is smaller than that for the Hamiltonian dynamics, because the variable ρ as seen e.g. in (63) has no dissipative terms. This leads us to focus on the thermodynamic variables $(e, \mathbf{m}, \tilde{c})$ and (81) reduces to

$$T d\sigma = de - \mathbf{v} \cdot d\mathbf{m} - \mu d\tilde{c}. \quad (83)$$

Comparison of (77) and (83) suggests we require the affinities associated with \mathbf{m} , e , and \tilde{c} . The conventional choices for these affinities are $\nabla(1/T)$, $\nabla(-\mathbf{v}/T)$, and $\nabla(-\mu/T)$, respectively [13]. However,

examination of (10) or (66) suggests using instead ∇T , $\nabla \mathbf{v}$, and $\nabla \mu$, as was done in [24].

The relationship between the flux-affinity relations in terms of these two choices of bases are given by the following:

$$\begin{aligned} \bar{\mathbf{J}}_{\mathbf{m}} &= L_{\mathbf{m}e} \cdot \nabla \left(\frac{1}{T} \right) + L_{\mathbf{m}\mathbf{m}} : \nabla \left(\frac{-\mathbf{v}}{T} \right) + L_{\mathbf{m}\bar{c}} \cdot \nabla \left(\frac{-\mu}{T} \right) \\ &= -\bar{\bar{\Lambda}} : \nabla \mathbf{v}, \end{aligned} \quad (84)$$

$$\begin{aligned} \mathbf{J}_e &= L_{ee} \cdot \nabla \left(\frac{1}{T} \right) + L_{e\mathbf{m}} : \nabla \left(\frac{-\mathbf{v}}{T} \right) + L_{e\bar{c}} \cdot \nabla \left(\frac{-\mu}{T} \right) \\ &= -\mathbf{v} \cdot (\bar{\bar{\Lambda}} : \nabla \mathbf{v}) - \bar{\kappa} \cdot \nabla T - \mu \bar{\mathbf{D}} \cdot \nabla \mu, \end{aligned} \quad (85)$$

$$\begin{aligned} \mathbf{J}_{\bar{c}} &= L_{\bar{c}e} \cdot \nabla \left(\frac{1}{T} \right) + L_{\bar{c}\mathbf{m}} : \nabla \left(\frac{-\mathbf{v}}{T} \right) + L_{\bar{c}\bar{c}} \cdot \nabla \left(\frac{-\mu}{T} \right) \\ &= -\bar{\mathbf{D}} \cdot \nabla \mu. \end{aligned} \quad (86)$$

Recall $\bar{\mathbf{J}}_{\mathbf{m}}$ is a 2-tensor, thus $L_{\mathbf{m}e} = L_{e\mathbf{m}}$ is a 3-tensor, $L_{\mathbf{m}\mathbf{m}}$ is a 4-tensor, and $L_{\mathbf{m}\bar{c}} = L_{\bar{c}\mathbf{m}}$ is a 3-tensor. Since \mathbf{J}_e and $\mathbf{J}_{\bar{c}}$ are vectors, $L_{ee}, L_{e\bar{c}} = L_{\bar{c}e}$, and $L_{\bar{c}\bar{c}}$ are 2-tensors. From (84), (85), and (86), we identify the components of $L_{\alpha\beta}$ as follows:

$$\begin{aligned} L_{\mathbf{m}e} &= T \bar{\bar{\Lambda}} \cdot \mathbf{v}, \quad L_{\mathbf{m}\mathbf{m}} = T \bar{\bar{\Lambda}}, \quad L_{\mathbf{m}\bar{c}} = 0, \quad L_{e\bar{c}} = T \mu \bar{\mathbf{D}}', \\ L_{ee} &= T^2 \bar{\kappa} + T \mathbf{v} \cdot \bar{\bar{\Lambda}} \cdot \mathbf{v} + T \mu^2 \bar{\mathbf{D}}, \quad L_{\bar{c}\bar{c}} = T \bar{\mathbf{D}}. \end{aligned} \quad (87)$$

The metricplectic 2-bracket in terms of the ξ -variables is given by

$$\begin{aligned} (F, G)_H &= \int_{\Omega} T \left[\nabla F_e \cdot (T \bar{\kappa} + \mathbf{v} \cdot \bar{\bar{\Lambda}} \cdot \mathbf{v} + \mu^2 \bar{\mathbf{D}}) \cdot \nabla G_e \right. \\ &\quad + \nabla F_{\mathbf{m}} : \bar{\bar{\Lambda}} : \nabla G_{\mathbf{m}} + \nabla F_{\bar{c}} \cdot \bar{\mathbf{D}} \cdot \nabla G_{\bar{c}} \\ &\quad + \nabla F_e \cdot (\bar{\bar{\Lambda}} \cdot \mathbf{v}) : \nabla G_{\mathbf{m}} + \nabla G_e \cdot (\bar{\bar{\Lambda}} \cdot \mathbf{v}) : \nabla F_{\mathbf{m}} \\ &\quad \left. + \mu \nabla F_e \cdot \bar{\mathbf{D}} \cdot \nabla G_{\bar{c}} + \mu \nabla G_e \cdot \bar{\mathbf{D}} \cdot \nabla F_{\bar{c}} \right]. \end{aligned} \quad (88)$$

Upon writing

$$S = \int_{\Omega} \sigma(\rho, e, \bar{c}, \mathbf{m}) \quad (89)$$

and using standard thermodynamic manipulations we obtain

$$S_e = 1/T, \quad S_{\mathbf{m}} = -\mathbf{v}/T, \quad \text{and} \quad S_{\bar{c}} = -\mu/T. \quad (90)$$

Inserting these into $(\sigma, S)_H$ using the 2-bracket of (88) yields the dissipative terms of (62), (64), and with the manipulations of transforming from e to σ , those of the entropy Eq. (65). By direct calculation, as well as by construction, we obtain $(H, S)_H = 0$ and $\dot{S} = (S, S)_H = (S, H; S, H) \geq 0$ which reproduces (66).

To close the circle we transform the bracket of (88) in terms of the variables (e, \mathbf{m}, \bar{c}) to one in terms of $(\sigma, \mathbf{m}, \bar{c})$ via the following chain rule formulas:

$$G_e \rightarrow G_{\sigma}/T, \quad G_{\mathbf{m}} \rightarrow G_{\mathbf{m}} - \mathbf{v} \cdot G_{\sigma}/T, \quad G_{\bar{c}} \rightarrow G_{\bar{c}} - \mu G_{\sigma}/T. \quad (91)$$

This calculation gives precisely the bracket of (60).

3. The Cahn-Hilliard-Navier-Stokes system

Now we apply our algorithm to obtain Cahn-Hilliard-Navier-Stokes (CHNS) systems allows for diffuse-interfaces. We follow the steps in order, just as in Section 2. However, here we have the additional step of aligning the desired entropy functional with the Poisson bracket, so that it is indeed a Casimir invariant.

3.1. Hamiltonian and entropy functional forms

The phenomenon of material transport along an interface is known as the Marangoni effect. The presence of a surface tension gradient naturally induces the migration of particles, moving from regions of low tension to those of high tension. This gradient can be triggered by a concentration gradient (or also a temperature gradient). In two-phase theory the interface between phases is regarded as being diffuse. According to the work of Taylor and Cahn [34], one can model the

diffuse interface by a single order parameter, say ϕ , and with a free energy functional,

$$\mathfrak{F} = \int_{\Omega} \frac{\epsilon}{2} \Gamma^2 (\nabla \phi) + \frac{1}{\epsilon} V(\phi), \quad (92)$$

with Γ being a homogeneous function of degree one; further details on this will be provided later. Here V can be any non-negative function that equals zero at $\phi = \pm 1$ and ϵ is a small parameter that goes to zero in the sharp-interface limit. We choose the order parameter ϕ to be the concentration.

In the isotropic surface energy case [15] develop a phase-field model for two-phase flow, which is thermodynamically consistent. The modeling is based on a non-classical choice of energy and entropy, given respectively by

$$H_{GL} = \int_{\Omega} \frac{\rho}{2} |\mathbf{v}|^2 + \rho u(\rho, s, c) + \frac{\rho}{2} \lambda_u |\nabla c|^2, \quad (93)$$

$$S_{GL} = \int_{\Omega} \rho s + \frac{\rho}{2} \lambda_s |\nabla c|^2, \quad (94)$$

where u and s stand for the classical specific internal energy and entropy, respectively, while the coefficients λ_s and λ_u are constant parameters.

Alternatively, [14] propose a model of phase-field of solidification with convection, the model permits the interface to have an anisotropic surface energy. The choice of energy and entropy are given by

$$H_{AMW} = \int_{\Omega} \frac{\rho}{2} |\mathbf{v}|^2 + \rho u(\rho, s, c) + \frac{\epsilon^2 E}{2} \Gamma^2 (\nabla c), \quad (95)$$

$$S_{AMW} = \int_{\Omega} \rho s - \frac{\epsilon^2 S}{2} \Gamma^2 (\nabla c), \quad (96)$$

where the coefficients ϵ_S and ϵ_E are assumed to be constant and Γ is a homogeneous function of degree one that takes a vector to a real number.

In this section, we explore a choice of energy and entropy functionals, from which the previously mentioned choices are special cases, and we consider the associated functionals, viz.

$$H^a = \int_{\Omega} \frac{\rho}{2} |\mathbf{v}|^2 + \rho u + \frac{\rho^a}{2} \lambda_u \Gamma^2 (\nabla c) =: \int_{\Omega} e_{\text{Total}}^a, \quad (97)$$

$$S^a = \int_{\Omega} \rho s + \frac{\rho^a}{2} \lambda_s \Gamma^2 (\nabla c) =: \int_{\Omega} \sigma_{\text{Total}}^a, \quad (98)$$

$$\mathfrak{F}^a = \int_{\Omega} \rho f + \frac{\rho^a}{2} \lambda_f(T) \Gamma^2 (\nabla c), \quad (99)$$

where u , s and f stand for the classical specific internal energy, entropy, and free energy, respectively, the coefficients λ_s and λ_u are constant parameters, and $\lambda_f(T)$ is a parameter depending on the temperature that will lead to anisotropic surface energy effects. We have defined the total densities e_{Total}^a and σ_{Total}^a for later use. The parameter a takes on two values: $a = 0$ reduces (97) and (98) to the expressions of Anderson et al. [14], where we set $\epsilon_E^2 = \lambda_u$ and $\epsilon_S^2 = -\lambda_s$, while for $a = 1$ they reduce to those used by Guo and Lin [15] provided the choice of an isotropic surface energy is assumed, viz., $\Gamma(\nabla c) = |\nabla c|$. Thus, as is clear from (97), (98), and (99), the dimensions of λ_f , λ_s , and λ_u are either specific or volumetric depending on the case. As usual, we have the thermodynamic relation

$$f = u - T s, \quad (100)$$

which allows us to assume the relationship between the coefficients

$$\lambda_f(T) = \lambda_u - T \lambda_s \quad \text{and} \quad \frac{d\lambda_f(T)}{dT} = -\lambda_s, \quad (101)$$

To summarize, our expressions (97) and (98) generalize the model studied by Guo and Lin [15] by including Γ , which accounts for anisotropic surface energy effects, while our expressions generalize the model of Anderson et al. [14] by including the factors of ρ in the integrands making all quantities in the integrands specific quantities multiplied by the density, giving rise to more general sources of energy.

Because Γ is a homogeneous function of degree unity,

$$\Gamma(\lambda \mathbf{p}) = \lambda \Gamma(\mathbf{p}) \text{ for all } \lambda > 0. \quad (102)$$

Differentiating (102) with respect to λ and then setting $\lambda = 1$ yields the fundamental relation

$$\Gamma(\mathbf{p}) = \mathbf{p} \cdot \boldsymbol{\xi} := p_j \frac{\partial \Gamma(\mathbf{p})}{\partial p_j}. \quad (103)$$

Then, differentiating (103) gives a second well-known relation,

$$\frac{\partial \Gamma}{\partial p_i} = \frac{\partial}{\partial p_i} (\boldsymbol{\xi} \cdot \mathbf{p}) = \xi_i + \frac{\partial^2 \Gamma}{\partial p_i \partial p_j} p_j = \xi_i, \quad (104)$$

where evidently p_j must be a null eigenvector of the matrix $\partial^2 \Gamma / \partial p_i \partial p_j$. Henceforth we will assume the argument of Γ to be ∇c . For the case of isotropic surface energy, where $\Gamma(\nabla c) = |\nabla c|$, the associated homogeneous function of degree zero is given by

$$\boldsymbol{\xi} = \nabla c / |\nabla c|. \quad (105)$$

From (99) we can obtain a generalized chemical potential

$$\mu_\Gamma^a := \frac{\delta \tilde{\sigma}^a}{\delta \tilde{c}} = \rho \frac{\partial \mu}{\partial \tilde{c}} - \nabla \cdot (\lambda_f \rho^a \Gamma \nabla c) = \mu - \frac{1}{\rho} \nabla \cdot (\lambda_f \rho^a \Gamma \boldsymbol{\xi}), \quad (106)$$

where recall $\tilde{c} = \rho c$. For the case of isotropic surface energy, this becomes

$$\mu_{|\nabla c|}^a = \mu - \frac{1}{\rho} \nabla \cdot (\lambda_f \rho^a \nabla c). \quad (107)$$

Upon setting $a = 1$ [the case of 15], this reduces to

$$\mu_{|\nabla c|}^1 = \mu - \frac{1}{\rho} \nabla \cdot (\lambda_f \rho \nabla c), \quad (108)$$

an expression that differs from that in [15] unless $\lambda_{f\rho}$ is constant. If this is the case and we choose a classical $\mu = c^3 - c$, corresponding to the quartic Landau potential, we obtain

$$\mu_{CH} = c^3 - c - \lambda_f \nabla^2 c, \quad (109)$$

the chemical potential of Cahn and Hilliard who indeed make these assumptions [cf. page 267 of 1]. For $\alpha = 0$ [the case of 14], we have

$$\mu_\Gamma^0 = \mu - \frac{1}{\rho} \nabla \cdot (\lambda_f \Gamma \boldsymbol{\xi}), \quad (110)$$

which allows for the weighted mean curvature effects of anisotropy.

Maintaining the same set of dynamical variables as in (51) (or an equivalent set) and making the choices of energy and entropy functionals of (97) and (98), we have completed the first two steps of the algorithm.

3.2. Noncanonical Poisson bracket of the Cahn-Hilliard-Euler system

To complete the next step of the algorithm, the third step, we need to manufacture a bracket that has (98) as a Casimir invariant. We do this by starting from the bracket of (15) in terms of the original variables $\psi = \{\mathbf{m}, \rho, \tilde{c}, \sigma\}$ and then transforming it to a new set of dynamical variables $\hat{\psi}^a := \{\hat{\mathbf{m}}, \hat{\rho}, \hat{\tilde{c}}, \hat{\sigma}^a\}$, giving the same bracket in terms of new coordinates. We have included the superscript a because in effect we have two sets of coordinates, corresponding to the desired entropies of (98) for $a = 0$ and $a = 1$. To distinguish the old from the new, we write the bracket in the transformed variables as $\{\hat{F}, \hat{G}\}^a$. Because of coordinate invariance, $\{C, F\} = \{C^a, \hat{F}\}^a = 0$, where $F[\psi] = \hat{F}[\hat{\psi}^a]$ is any functional written in one or the other coordinates. The Casimir $S = \int_\Omega \sigma$ in our original coordinates is transformed into a different form in the new coordinates. Specifically, we change the variables as follows:

$$\mathbf{m} = \hat{\mathbf{m}}, \quad \rho = \hat{\rho}, \quad \tilde{c} = \hat{\tilde{c}}, \quad \sigma = \hat{\sigma}^a + \frac{\hat{\rho}^a}{2} \lambda_s \Gamma^2(\nabla \hat{c}), \quad (111)$$

where $\hat{c} = \hat{\rho} \hat{\tilde{c}}$. Consequently, the entropy S in the old coordinates written in terms of the new coordinates will, by design, become the following Casimir for the Poisson bracket in the new coordinates:

$$\hat{S}^a = \int_\Omega \hat{\sigma}^a + \frac{\hat{\rho}^a}{2} \lambda_s \Gamma^2(\nabla \hat{c}). \quad (112)$$

Thus we have manufactured a bracket with the entropy expression of (98) as a Casimir.

Transformation of the Poisson bracket (15) requires use of the functional chain rule. For convenience we use

$$\hat{\sigma}^a = \sigma - \frac{\rho^a}{2} \lambda_s \Gamma^2(\nabla c) \quad (113)$$

and consider the variation of any functional of the new variables. Thus we use $\delta \rho = \delta \hat{\rho}$, $\delta \mathbf{m} = \delta \hat{\mathbf{m}}$, $\delta \tilde{c} = \delta \hat{\tilde{c}}$, and for the entropy variable

$$\begin{aligned} \delta \hat{\sigma}^a &= \delta \sigma - \frac{1}{2} a \rho^{a-1} \lambda_s \Gamma^2(\nabla c) \delta \rho - \rho^a \lambda_s \Gamma \boldsymbol{\xi} \cdot \nabla \delta \left(\frac{\tilde{c}}{\rho} \right) \\ &= \delta \sigma - \frac{1}{2} a \rho^{a-1} \lambda_s \Gamma^2(\nabla c) \delta \rho - \rho^a \lambda_s \Gamma \boldsymbol{\xi} \cdot \nabla \left(\frac{\delta \tilde{c}}{\rho} \right) \\ &\quad + \rho^a \lambda_s \Gamma \boldsymbol{\xi} \cdot \nabla \left(\frac{\tilde{c}}{\rho^2} \delta \rho \right), \end{aligned} \quad (114)$$

where use has been made of (104). Now let F be an arbitrary functional of the original variables and \hat{F} the same functional in terms of the new variables. Thus,

$$\begin{aligned} \int_\Omega \hat{F}_{\hat{\mathbf{m}}} \cdot \delta \hat{\mathbf{m}} + \hat{F}_{\hat{\rho}} \delta \hat{\rho} + \hat{F}_{\hat{\tilde{c}}} \delta \hat{\tilde{c}} + \hat{F}_{\hat{\sigma}^a} \delta \hat{\sigma}^a + \hat{F}_{\tilde{c}} \delta \tilde{c} \\ = \int_\Omega F_{\mathbf{m}} \cdot \delta \mathbf{m} + F_{\rho} \delta \rho + F_{\sigma} \delta \sigma + F_{\tilde{c}} \delta \tilde{c}. \end{aligned} \quad (115)$$

Note, no sum over a is to be done. By identification of terms we obtain

$$\begin{aligned} F_{\mathbf{m}} &= \hat{F}_{\hat{\mathbf{m}}}, \quad F_{\sigma} = \hat{F}_{\hat{\sigma}^a}, \\ F_{\rho} &= \hat{F}_{\hat{\rho}} - \frac{a}{2} \hat{\rho}^{a-1} \lambda_s \Gamma^2 \hat{F}_{\hat{\sigma}^a} - \frac{\hat{\tilde{c}}}{\hat{\rho}^2} \nabla \cdot (\hat{\rho}^a \lambda_s \Gamma \boldsymbol{\xi} \hat{F}_{\hat{\sigma}^a}), \\ F_{\tilde{c}} &= \hat{F}_{\hat{\tilde{c}}} + \frac{1}{\hat{\rho}} \nabla \cdot (\hat{\rho}^a \lambda_s \Gamma \boldsymbol{\xi} \hat{F}_{\hat{\sigma}^a}). \end{aligned} \quad (116)$$

The transformed Poisson bracket is obtained by inserting the expressions of (116) into (15), writing it entirely in terms of the hat variables. Upon doing this and then dropping the hats, we get for any functionals F and G the following Poisson bracket:

$$\begin{aligned} \{F, G\}^a &= - \int_\Omega \mathbf{m} \cdot \left[F_{\mathbf{m}} \cdot \nabla G_{\mathbf{m}} - G_{\mathbf{m}} \cdot \nabla F_{\mathbf{m}} \right] \\ &\quad + \rho \left[F_{\rho} \cdot \nabla \left(G_{\rho} - a \rho^{a-1} \lambda_s \Gamma^2 G_{\sigma^a} / 2 - \tilde{c} \nabla \cdot (\rho^a \lambda_s \Gamma \boldsymbol{\xi} G_{\sigma^a}) / \rho^2 \right) \right. \\ &\quad \left. - G_{\rho} \cdot \nabla \left(F_{\rho} - a \rho^{a-1} \lambda_s \Gamma^2 F_{\sigma^a} / 2 - \tilde{c} \nabla \cdot (\rho^a \lambda_s \Gamma \boldsymbol{\xi} F_{\sigma^a}) / \rho^2 \right) \right] \\ &\quad + (\sigma^a + \rho^a \lambda_s \Gamma^2 / 2) \left[F_{\mathbf{m}} \cdot \nabla G_{\sigma^a} - G_{\mathbf{m}} \cdot \nabla F_{\sigma^a} \right] \\ &\quad + \tilde{c} \left[F_{\mathbf{m}} \cdot \nabla (G_{\tilde{c}} + \nabla \cdot (\rho^a \lambda_s \Gamma \boldsymbol{\xi} G_{\sigma^a}) / \rho) \right. \\ &\quad \left. - G_{\mathbf{m}} \cdot \nabla (F_{\tilde{c}} + \nabla \cdot (\rho^a \lambda_s \Gamma \boldsymbol{\xi} F_{\sigma^a}) / \rho) \right]. \end{aligned} \quad (117)$$

This bracket is clearly bilinear and skew-symmetric. Because it was derived from the bracket (15) by a change of variables, satisfaction of the Jacobi identity is assured. We note, as before, strong boundary conditions are assumed such that all integrations by parts produce vanishing boundary terms.

Thus we have completed the third part of our algorithm, the construction of a Poisson bracket that has the entropy functional of (98) in the set of its Casimir invariants. Recall the integrand of the entropy is given by

$$\sigma_{\text{Total}}^a := \sigma^a + \frac{\rho^a}{2} \lambda_s \Gamma^2(\nabla c); \quad (118)$$

so we find

$$\begin{aligned} \frac{\delta S^a}{\delta \sigma^a} &= 1, \quad \frac{\delta S^a}{\delta \tilde{c}} = -\frac{1}{\rho} \nabla \cdot (\rho^a \lambda_s \Gamma \boldsymbol{\xi}) \\ \frac{\delta S^a}{\delta \rho} &= \frac{a}{2} \rho^{a-1} \lambda_s \Gamma^2 + \frac{\tilde{c}}{\rho^2} \nabla \cdot (\rho^a \lambda_s \Gamma \boldsymbol{\xi}). \end{aligned} \quad (119)$$

Using (119) one can easily check that $\{F, S^a\} = 0$ for all F , which by construction had to be the case. Now we are free to choose any Hamiltonian we desire in (117) to obtain the evolution of any observable o as follows:

$$\partial_t o = \{o, H^a\}^a, \quad (120)$$

In Section 3.1 we proposed the Hamiltonian functional of (97), which we rewrite as follows in order to make all arguments clear:

$$H^a[\rho, \mathbf{m}, \sigma^a, \tilde{c}] = \int_{\Omega} \frac{|\mathbf{m}|^2}{2\rho} + \rho u \left(\rho, \frac{\sigma^a}{\rho}, \frac{\tilde{c}}{\rho} \right) + \frac{\rho^a}{2} \lambda_u \Gamma^2 \left(\nabla \left(\frac{\tilde{c}}{\rho} \right) \right). \quad (121)$$

Using the functional derivatives of this Hamiltonian,

$$\begin{aligned} H_{\mathbf{m}}^a &= \mathbf{v}, & H_{\sigma^a} &= T, \\ H_{\rho}^a &= -|\mathbf{v}|^2/2 + u + p/\rho - sT - c\mu + a\rho^{a-1}\lambda_u\Gamma^2/2 + c\nabla \cdot (\rho^a\lambda_u\Gamma\xi)/\rho, \\ H_{\tilde{c}}^a &= \mu - \nabla \cdot (\rho^a\lambda_u\Gamma\xi)/\rho, \end{aligned} \quad (122)$$

in the bracket (117) gives the equations of motion in the form of (120). At this point we could write this out and display a general system of equations that includes both cases, but we choose to consider them separately because the general system is unwieldy and not particularly perspicuous.

Let us first consider the simplified version of our derived Poisson bracket for the case $a = 1$, which is as follows:

$$\begin{aligned} \{F, G\}^1 &= - \int_{\Omega} \mathbf{m} \cdot [F_{\mathbf{m}} \cdot \nabla G_{\mathbf{m}} - G_{\mathbf{m}} \cdot \nabla F_{\mathbf{m}}] + \rho [F_{\mathbf{m}} \cdot \nabla G_{\rho} - G_{\mathbf{m}} \cdot \nabla F_{\rho}] \\ &\quad - \lambda_s [F_{\mathbf{m}} \cdot \nabla \cdot (\rho G_{\sigma^1} \Gamma \xi \otimes \nabla c) - G_{\mathbf{m}} \cdot \nabla \cdot (\rho F_{\sigma^1} \Gamma \xi \otimes \nabla c)] \\ &\quad + \sigma^1 [F_{\mathbf{m}} \cdot \nabla G_{\sigma^1} - G_{\mathbf{m}} \cdot \nabla F_{\sigma^1}] + \tilde{c} [F_{\mathbf{m}} \cdot \nabla G_{\tilde{c}} - G_{\mathbf{m}} \cdot \nabla F_{\tilde{c}}], \end{aligned} \quad (123)$$

where \otimes denotes tensor product of two vectors and consistent with our convention we have

$$\nabla \cdot (\mathbf{u} \otimes \mathbf{v}) = (\nabla \cdot \mathbf{u})\mathbf{v} + \mathbf{u} \cdot \nabla \mathbf{v}.$$

Using (122), the bracket form of (120) gives the ideal diffuse two-phase flow system

$$\partial_t \mathbf{v} = \{\mathbf{v}, H^1\}^1 = -\mathbf{v} \cdot \nabla \mathbf{v} - \frac{1}{\rho} \nabla \cdot (p\bar{\mathbf{I}} + \rho\lambda_f\Gamma\xi \otimes \nabla c), \quad (124)$$

$$\partial_t \rho = \{\rho, H^1\}^1 = -\mathbf{v} \cdot \nabla \rho - \rho \nabla \cdot \mathbf{v}, \quad (125)$$

$$\partial_t \tilde{c} = \{\tilde{c}, H^1\}^1 = -\mathbf{v} \cdot \nabla \tilde{c} - \tilde{c} \nabla \cdot \mathbf{v}, \quad (126)$$

$$\partial_t \sigma_{\text{Total}}^1 = \{\sigma_{\text{Total}}^1, H^1\}^1 = -\mathbf{v} \cdot \nabla \sigma_{\text{Total}}^1 - \sigma_{\text{Total}}^1 \nabla \cdot \mathbf{v}, \quad (127)$$

where $\bar{\mathbf{I}}$ is the unit tensor. Observe in (127) we have chosen the observable σ_{Total}^1 instead of σ^1 , in order to demonstrate its conservation.

Similarly, for the case where $a = 0$, the Poisson bracket has the following form:

$$\begin{aligned} \{F, G\}^0 &= - \int_{\Omega} \mathbf{m} \cdot [F_{\mathbf{m}} \cdot \nabla G_{\mathbf{m}} - G_{\mathbf{m}} \cdot \nabla F_{\mathbf{m}}] + \rho [F_{\mathbf{m}} \cdot \nabla G_{\rho} - G_{\mathbf{m}} \cdot \nabla F_{\rho}] \\ &\quad - \lambda_s [F_{\mathbf{m}} \cdot \nabla \cdot (G_{\sigma^0} \Gamma \xi \otimes \nabla c) - G_{\mathbf{m}} \cdot \nabla \cdot (F_{\sigma^0} \Gamma \xi \otimes \nabla c)] \\ &\quad + \lambda_s [F_{\mathbf{m}} \cdot \nabla (\Gamma^2 G_{\sigma^0}) - G_{\mathbf{m}} \cdot \nabla (\Gamma^2 F_{\sigma^0})] / 2 \\ &\quad + \sigma^0 [F_{\mathbf{m}} \cdot \nabla G_{\sigma^0} - G_{\mathbf{m}} \cdot \nabla F_{\sigma^0}] + \tilde{c} [F_{\mathbf{m}} \cdot \nabla G_{\tilde{c}} - G_{\mathbf{m}} \cdot \nabla F_{\tilde{c}}]. \end{aligned} \quad (128)$$

Same as above, using (122), the ideal diffuse two-phase flow system is produced

$$\begin{aligned} \partial_t \mathbf{v} &= \{\mathbf{v}, H^0\}^0 \\ &= -\mathbf{v} \cdot \nabla \mathbf{v} - \frac{1}{\rho} \nabla \cdot \left[(p - \lambda_f \Gamma^2 / 2) \bar{\mathbf{I}} + \lambda_f \Gamma \xi \otimes \nabla c \right], \end{aligned} \quad (129)$$

$$\partial_t \rho = \{\rho, H^0\}^0 = -\mathbf{v} \cdot \nabla \rho - \rho \nabla \cdot \mathbf{v}, \quad (130)$$

$$\partial_t \tilde{c} = \{\tilde{c}, H^0\}^0 = -\mathbf{v} \cdot \nabla \tilde{c} - \tilde{c} \nabla \cdot \mathbf{v}, \quad (131)$$

$$\partial_t \sigma_{\text{Total}}^0 = \{\sigma_{\text{Total}}^0, H^0\}^0 = -\mathbf{v} \cdot \nabla \sigma_{\text{Total}}^0 - \sigma_{\text{Total}}^0 \nabla \cdot \mathbf{v}, \quad (132)$$

where recall from (103), $\xi = \partial\Gamma(\mathbf{p})/\partial\mathbf{p}$.

Let us now comment on these two Hamiltonian systems. By construction both the $a = 1$ and $a = 0$ systems conserve their Hamiltonians and entropies, as given by (97) and (98) with $a = 1$ and $a = 0$, respectively. Both systems have momentum equations containing a term describing anisotropic surface energy (capillary) effects. The $a = 0$ system of (129)–(132) is identical to the ideal limit of that given in the work of Anderson et al. [14]. Upon choosing $\Gamma(\nabla c) = |\nabla c|$, the

$a = 1$ system of (124)–(127) should correspond to the ideal limit of that of Guo and Lin [15], but it does not. In fact the system of Guo and Lin [15] in this limit does not conserve energy. Moreover, the capillary effect in their momentum equation (equation (3.40)), which should be replaced by (124) with $\Gamma(\nabla c) = |\nabla c|$, vanishes in the one-dimensional limit. Since such surface effects are determined by mean or weighted mean curvature [35], it is clear that this is physically untenable. Fortunately, our method provides a simple fix to their equations, while showing how to generalize them to include anisotropic surface effects.

An alternative but equivalent Hamiltonian formulation of the above systems exists, in fact, one that has a standard entropy functional of the form of (30). Given that the bracket of (117) was obtained via a transformation of the bracket of (15), we can transform it back from one that has (98) as a Casimir to the original that has (5) as a Casimir. However, to generate equivalent equations of motion, we would have to transform the Hamiltonian of (97) into a more complicated form. Tracing back through our transformations, we would replace the coordinate σ^a in the Hamiltonian by $\sigma - \rho^a \lambda_s \Gamma^2 / 2$, which means the internal energy becomes

$$u \mapsto u(\rho, (\sigma - \rho^a \lambda_s \Gamma^2 / 2) / \rho, \tilde{c} / \rho), \quad (133)$$

while otherwise the Hamiltonian remains the same. Just as with finite-dimensional Hamiltonian systems, one can change coordinates and arrive at equivalent systems with different Poisson brackets and Hamiltonians, and in the noncanonical case different expressions for the Casimir invariants. Often one has the options of a simple bracket and complicated Hamiltonian or vice versa.

3.3. Metriplectic 4-bracket for the Cahn-Hilliard-Navier-Stokes system

Now we turn to the 4th and final step of our algorithm. Just as in Section 2.3.3 we build a metriplectic 4-bracket using the K-N construction. We suppose our multi-component field variable is

$$\psi(\mathbf{x}, t) = (\mathbf{m}(\mathbf{x}, t), \rho(\mathbf{x}, t), \tilde{c}(\mathbf{x}, t), \sigma^a(\mathbf{x}, t)) \quad (134)$$

and in the K-N construction we use a more general expression for Σ akin to that mentioned in (56), viz.

$$M(dF, dG) = F_{\sigma^a} G_{\sigma^a}, \quad (135)$$

$$\begin{aligned} \Sigma(dF, dG) &= \nabla F_{\mathbf{m}} : \bar{\bar{\Lambda}}_1 : \nabla G_{\mathbf{m}} + \nabla F_{\sigma^a} \cdot \bar{\bar{\Lambda}}_2 \cdot \nabla G_{\sigma^a} \\ &\quad + \nabla \mathcal{L}_{\tilde{c}}^a(F) \cdot \bar{\bar{\Lambda}}_3 \cdot \mathcal{L}_{\tilde{c}}^a(G), \end{aligned} \quad (136)$$

where a , of course, is not to be summed over and the pseudodifferential operator $\mathcal{L}_{\tilde{c}}^a$ has the following form:

$$\mathcal{L}_{\tilde{c}}^a(F) := \nabla (F_{\tilde{c}} + \nabla \cdot (\rho^a \lambda_s \Gamma \xi F_{\sigma^a}) / \rho). \quad (137)$$

Here the tensors $\bar{\bar{\Lambda}}_1$, $\bar{\bar{\Lambda}}_2$ and $\bar{\bar{\Lambda}}_3$ are defined by (59). Then, the 4-bracket reads

$$\begin{aligned} (F, K; G, N)^a &= \int_{\Omega} \frac{1}{T} \left[[K_{\sigma^a} \nabla F_{\mathbf{m}} - F_{\sigma^a} \nabla K_{\mathbf{m}}] : \bar{\bar{\Lambda}} : [N_{\sigma^a} \nabla G_{\mathbf{m}} - G_{\sigma^a} \nabla N_{\mathbf{m}}] \right. \\ &\quad + \frac{1}{T} [K_{\sigma^a} \nabla F_{\sigma^a} - F_{\sigma^a} \nabla K_{\sigma^a}] \cdot \bar{\bar{\kappa}} \cdot [N_{\sigma^a} \nabla G_{\sigma^a} - G_{\sigma^a} \nabla N_{\sigma^a}] \\ &\quad \left. + [K_{\sigma^a} \mathcal{L}_{\tilde{c}}^a(F) - F_{\sigma^a} \mathcal{L}_{\tilde{c}}^a(K)] \cdot \bar{\bar{D}} \cdot [N_{\sigma^a} \mathcal{L}_{\tilde{c}}^a(G) - G_{\sigma^a} \mathcal{L}_{\tilde{c}}^a(N)] \right]. \end{aligned} \quad (138)$$

Observe, with the exception of the last line this bracket is identical to that of (60).

Upon insertion of H^a as given by (97) and S from the set of Casimirs to be as in (98), the dynamics is given by

$$\partial_t \psi^a = \{\psi^a, H^a\}^a + (\psi^a, H^a; S^a, H^a)^a. \quad (139)$$

Using $\mathcal{L}_{\tilde{c}}^a(H^a) = \nabla \mu_{\tilde{c}}^a$, $H_{\mathbf{m}}^a = \mathbf{v}$, $H_{\sigma^a}^a = T$, $S_{\sigma^a}^a = 1$ and $\mathcal{L}_{\tilde{c}}^a(S^a) = 0$, the following diffuse-interface CHNS system for $a = 1$ is produced:

$$\begin{aligned} \partial_t \mathbf{v} &= \{\mathbf{v}, H^1\}^1 + (\mathbf{v}, H^1; S^1, H^1)^1 \\ &= -\mathbf{v} \cdot \nabla \mathbf{v} - \frac{1}{\rho} \nabla \cdot [p\bar{\mathbf{I}} + \lambda_f \rho \Gamma \xi \otimes \nabla c] \end{aligned}$$

$$+ \frac{1}{\rho} \nabla \cdot (\bar{\Lambda} : \nabla \mathbf{v}), \quad (140)$$

$$\begin{aligned} \partial_t \rho &= \{\rho, H^1\}^1 + (\rho, H^1; S^1, H^1)^1 \\ &= -\mathbf{v} \cdot \nabla \rho - \rho \nabla \cdot \mathbf{v}, \end{aligned} \quad (141)$$

$$\begin{aligned} \partial_t \bar{c} &= \{\bar{c}, H^1\}^1 + (\bar{c}, H^1; S^1, H^1)^1 \\ &= -\mathbf{v} \cdot \nabla \bar{c} - \bar{c} \nabla \cdot \mathbf{v} + \nabla \cdot (\bar{D} \cdot \nabla \mu_F^1), \end{aligned} \quad (142)$$

$$\begin{aligned} \partial_t \sigma_{\text{Total}}^1 &= \{\sigma_{\text{Total}}^1, H^1\}^1 + (\sigma_{\text{Total}}^1, H^1; S^1, H^1)^1 \\ &= -\mathbf{v} \cdot \nabla \sigma_{\text{Total}}^1 - \sigma_{\text{Total}}^1 \nabla \cdot \mathbf{v} \\ &\quad + \nabla \cdot \left(\frac{\bar{\kappa}}{T} \cdot \nabla T \right) + \frac{1}{T^2} \nabla T \cdot \bar{\kappa} \cdot \nabla T \\ &\quad + \frac{1}{T} \nabla \mathbf{v} : \bar{\Lambda} : \nabla \mathbf{v} + \frac{1}{T} \nabla \mu_F^1 \cdot \bar{D} \cdot \nabla \mu_F^1. \end{aligned} \quad (143)$$

Similarly, for $a = 0$ we obtain

$$\begin{aligned} \partial_t \mathbf{v} &= \{\mathbf{v}, H^0\}^0 + (\mathbf{v}, H^0; S^0, H^0)^0 \\ &= -\mathbf{v} \cdot \nabla \mathbf{v} - \frac{1}{\rho} \nabla \cdot \left[(p - \lambda_f \Gamma^2 / 2) \mathbf{I} + \lambda_f \Gamma \xi \otimes \nabla c \right] \\ &\quad + \frac{1}{\rho} \nabla \cdot (\bar{\Lambda} : \nabla \mathbf{v}), \end{aligned} \quad (144)$$

$$\begin{aligned} \partial_t \rho &= \{\rho, H^0\}^0 + (\rho, H^0; S^0, H^0)^0 \\ &= -\mathbf{v} \cdot \nabla \rho - \rho \nabla \cdot \mathbf{v}, \end{aligned} \quad (145)$$

$$\begin{aligned} \partial_t \bar{c} &= \{\bar{c}, H^0\}^0 + (\bar{c}, H^0; S^0, H^0)^0 \\ &= -\mathbf{v} \cdot \nabla \bar{c} - \bar{c} \nabla \cdot \mathbf{v} + \nabla \cdot (\bar{D} \cdot \nabla \mu_F^0), \end{aligned} \quad (146)$$

$$\begin{aligned} \partial_t \sigma_{\text{Total}}^0 &= \{\sigma_{\text{Total}}^0, H^0\}^0 + (\sigma_{\text{Total}}^0, H^0; S^0, H^0)^0 \\ &= -\mathbf{v} \cdot \nabla \sigma_{\text{Total}}^0 - \sigma_{\text{Total}}^0 \nabla \cdot \mathbf{v} \\ &\quad + \nabla \cdot \left(\frac{\bar{\kappa}}{T} \cdot \nabla T \right) + \frac{1}{T^2} \nabla T \cdot \bar{\kappa} \cdot \nabla T \\ &\quad + \frac{1}{T} \nabla \mathbf{v} : \bar{\Lambda} : \nabla \mathbf{v} + \frac{1}{T} \nabla \mu_F^0 \cdot \bar{D} \cdot \nabla \mu_F^0. \end{aligned} \quad (147)$$

Thus we have extracted from our general system with arbitrary a , two thermodynamically consistent CHNS systems. By construction both the $a = 1$ and $a = 0$ systems must conserve energy and both must produce entropy, which we find is governed by the following:

$$\begin{aligned} S^a &= (S^a, H^a; S^a, H^a)^a \\ &= \int_{\Omega} \frac{1}{T} \left[\nabla \mathbf{v} : \bar{\Lambda} : \nabla \mathbf{v} + \frac{1}{T} \nabla T \cdot \bar{\kappa} \cdot \nabla T + \nabla \mu_F^a \cdot \bar{D} \cdot \nabla \mu_F^a \right] \geq 0. \end{aligned} \quad (148)$$

3.4. Metriplectic 2-bracket for the Cahn-Hilliard-Navier-Stokes system

Proceeding as in the previous section, using $\mathcal{L}_c^a(H^a) = \nabla(\mu - \frac{1}{\rho} \nabla \cdot (\lambda_f \rho^a \Gamma \xi)) = \nabla \mu_F^a$, $H_{\mathbf{m}}^a = \mathbf{v}$, $H_{\sigma^a}^a = T$, the metriplectic 2-bracket emerges directly from the 4-bracket as follows:

$$\begin{aligned} (F, G)_{H^a}^a &= (F, H^a; G, H^a)^a \\ &= \int_{\Omega} \frac{1}{T} \left[[T \nabla F_{\mathbf{m}} - F_{\sigma^a} \nabla \mathbf{v}] : \bar{\Lambda} : [T \nabla G_{\mathbf{m}} - G_{\sigma^a} \nabla \mathbf{v}] \right. \\ &\quad + \frac{1}{T} [T \nabla F_{\sigma^a} - F_{\sigma^a} \nabla T] \cdot \bar{\kappa} \cdot [T \nabla G_{\sigma^a} - G_{\sigma^a} \nabla T] \\ &\quad \left. + [T \mathcal{L}_c^a(F) - F_{\sigma^a} \nabla \mu_F^a] \cdot \bar{D} \cdot [T \mathcal{L}_c^a(G) - G_{\sigma^a} \nabla \mu_F^a] \right], \end{aligned} \quad (149)$$

which is the analog of (60) for the GNS system in Section 2.3.3.

Now we could proceed as in Section 2.4.1 and obtain the analog of $L_{\alpha\beta}$ by transforming to the coordinates $\xi^a := (\mathbf{m}, \rho, \bar{c}, e_{\text{Total}}^a)$, where e_{Total}^a is the total energy density defined in (97). This would lead to a metriplectic 2-bracket analogous to (76) and the concomitant flux and affinity relations analogous to (87) would emerge. Because this is hardly more enlightening than (149), we do not record this result here.

4. Summary and conclusions

In this paper we have described the metriplectic 4-bracket formalism and how it can algorithmically be used in the context of multiphase fluids to construct thermodynamically consistent models, ones that

conserve energy and produce entropy. In particular, we have used it in Section 2 to obtain the GE Hamiltonian system, which adds a concentration variable to conventional Eulerian fluid mechanics, and to the GNS system, a generalization of the Navier-Stokes system that is thermodynamically consistent with the collection of thermodynamic fluxes. Then, in Section 3 we used the algorithm to obtain a class of Hamiltonian fluid systems that allow for anisotropic surface effects, followed by the construction of a general class of CHNS systems that couple Cahn-Hilliard physics with that of Navier-Stokes dynamics, in a thermodynamically consistent way. The systems we obtain generalizes previous work by including anisotropic effects in the surface tension and all phenomenological parameters.

A cornerstone of Hamiltonian dynamics is its geometric invariance under coordinate changes. Because the minimal metriplectic properties are algebraic and geometric, they too are invariant under coordinate changes. Thus, we can write our CHNS class of dissipative systems with a standard entropy functional of the form of (30), but with a more complicated Hamiltonian using (133).

From the examples presented, it is clear that the 4-bracket formalism can be applied to obtain a wide variety of dynamical systems in various fields. In fact it was recently applied to obtain generalized collision operators in kinetic theory [36] and a thermodynamically consistent model for radiation hydrodynamics [37]. Although incompressible flows do not have the usual thermodynamics associated with compression and pressure, they can be included in the metriplectic formalism by using the techniques of Chandre et al. [38].

The metriplectic 4-bracket formalism also provides an avenue for designing structure preserving numerical algorithms [see e.g. 39]. Any discretization that preserves the symmetries of the 4-bracket, which is not a difficult task, will be thermodynamically consistent on the semi-discrete level, i.e. produce a set of ordinary differential equations that conserve energy and produce entropy.

CRedit authorship contribution statement

Azeddine Zaidni: Writing – review & editing, Writing – original draft, Formal analysis, Conceptualization. **Philip J. Morrison:** Writing – review & editing, Writing – original draft, Supervision, Investigation, Formal analysis, Conceptualization. **Saad Benjelloun:** Writing – review & editing, Validation, Supervision.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Philip J. Morrison reports financial support was provided by US Department of Energy. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

A.Z. acknowledges support from the Mohammed VI Polytechnic University for supporting an internship at the University of Texas at Austin. P.J.M. acknowledges support from the DOE Office of Fusion Energy Sciences under DE-FG02-04ER-54742, and from a Forschungspreis from the Alexander von Humboldt Foundation.

Appendix. Semidefinite curvature

We define the binary operations $\langle \cdot, \cdot \rangle_\Sigma$ and $\langle \cdot, \cdot \rangle_M$ that satisfy all of the axioms of an inner product space, except the non-degeneracy condition

$$\langle F, G \rangle_\Sigma := \int d^N z \int d^N z' \Sigma^{\alpha\beta}(z, z') \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z')},$$

$$\langle F, G \rangle_M := \int d^N z'' \int d^N z''' M^{\gamma\delta}(z'', z''') \frac{\delta F}{\delta \chi^\gamma(z'')} \frac{\delta G}{\delta \chi^\delta(z''')},$$

where Σ and M are positive semi-definites. We have the Cauchy-Schwarz inequality

$$|\langle F, G \rangle_\Sigma| \leq \sqrt{\langle F, F \rangle_\Sigma} \sqrt{\langle G, G \rangle_\Sigma} = \|F\|_\Sigma \|G\|_\Sigma.$$

Lemma 1. *A metriplectic quadravector constructed using the K - N product, has non-negative sectional curvature,*

$$K(F, G) = \int d^N z \int d^N z' \int d^N z'' \int d^N z''' \Sigma^{ij}(z, z') M^{kl}(z'', z''') \times \frac{\delta F}{\delta \chi^\alpha(z)} \frac{\delta G}{\delta \chi^\beta(z')} \frac{\delta F}{\delta \chi^\gamma(z'')} \frac{\delta G}{\delta \chi^\delta(z''')} + \text{other terms.}$$

Proof. Direct calculation gives

$$K(F, G) = \|F\|_\Sigma^2 \|G\|_M^2 - 2\langle F, G \rangle_\Sigma \langle F, G \rangle_M + \|G\|_\Sigma^2 \|F\|_M^2.$$

The following inequality

$$(\|F\|_\Sigma \|G\|_M - \|G\|_\Sigma \|F\|_M)^2 \geq 0$$

implies

$$\begin{aligned} \|F\|_\Sigma^2 \|G\|_M^2 + \|G\|_\Sigma^2 \|F\|_M^2 &\geq 2\|F\|_M \|F\|_\Sigma \|G\|_M \|G\|_\Sigma \\ &\geq 2|\langle F, G \rangle_\Sigma| |\langle F, G \rangle_M| \\ &\geq 2\langle F, G \rangle_\Sigma \langle F, G \rangle_M, \end{aligned}$$

where the second inequality follows from the Cauchy-Schwarz inequality. Evidently, the last inequality implies $K(F, G) \geq 0$ for all F and G .

Lemma 2. *We suppose that Σ is positive definite, defining an inner product. Given any two Σ -arbitrary linearly independent $\delta F/\delta \chi$ and $\delta G/\delta \chi$, then the sectional curvature is strictly positive ($K(F, G) > 0$).*

Proof. Since $\delta F/\delta \chi$ and $\delta G/\delta \chi$ are Σ -Linearly independent, the Cauchy-Schwarz inequality given by

$$|\langle F, G \rangle_\Sigma| < \|F\|_\Sigma \|G\|_\Sigma.$$

In the same way we have

$$(\|F\|_\Sigma \|G\|_M - \|G\|_\Sigma \|F\|_M)^2 \geq 0$$

implies

$$\begin{aligned} \|F\|_\Sigma^2 \|G\|_M^2 + \|G\|_\Sigma^2 \|F\|_M^2 &\geq 2\|F\|_M \|F\|_\Sigma \|G\|_M \|G\|_\Sigma \\ &> 2|\langle F, G \rangle_\Sigma| |\langle F, G \rangle_M| \\ &> 2\langle F, G \rangle_\Sigma \langle F, G \rangle_M. \end{aligned}$$

Hence, we deduce that $K(F, G) > 0$.

Finite-dimensional versions of these two lemmas were first reported in [9].

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