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Formulation of strongly non-local, non-isothermal dynamics for heterogeneous solids based on the GENERIC with application to phase-field modeling

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Abstract

The purpose of the current work is the formulation of models for conservative and non-conservative dynamics in solid systems with the help of the General Equation for the Non-Equilibrium Reversible-Irreversible Coupling (GENERIC: e.g., Grmela and Öttinger, Phys. Rev. E 56(6), 6620 (1997); Öttinger and Grmela, Phys. Rev. E 56(6), 6633 (1997)). In this context, the resulting models are inherently spatially strongly non-local (i.e., functional) and non-isothermal in character. They are applicable in particular to the modeling of phase transitions as well as mass and heat transport in multiphase, multicomponent solids. In the last part of the work, the strongly non-local model formulation is reduced to weakly non-local form with the help of generalized gradient approximation of the energy and entropy functionals. On this basis, the current model formulation is shown to be consistent with and reduce to a recent non-isothermal generalization (Gladkov et al., J. Non-Equilib. Thermodyn. 41(2), 131 (2016)) of the well-known phase-field models of Cahn and Hilliard (J. Chem. Phys. 28(2), 258 (1958)) for conservative dynamics and of Allen and Cahn (Acta Metall. 27(6), 1085 (1979)) for non-conservative dynamics. Finally, the current approach is applied to derive a non-isothermal generalization of a phase-field crystal model for binary alloys (see, e.g., Elder et al., Phys. Rev. B 75(6), 064107 (2007)).

Keywords: GENERIC, Non-isothermal, Strongly non-local, Phase field, Multiphase, Multicomponent, Solids

Introduction

Over the years, a number of approaches to the thermodynamic formulation of models for material behavior have been developed. Perhaps the most common of these in the modern era is that of continuum thermodynamics as based on the Clausius-Duhem approach (e.g., Šilhavý 1997). Another is based on the entropy inequality of Müller-Liu (e.g., Liu 1972; Müller 1985) or on extended thermodynamics (e.g., Müller and Ruggeri 1993; Jou et al. 2010). Yet another is based on the so-called General Equation for the Non-Equilibrium Reversible-Irreversible Coupling (GENERIC: e.g., Grmela and Öttinger 1997; Öttinger and Grmela 1997; Öttinger 2005; Grmela 2010). This latter approach is closely related to the bracket formalism (Beris and Edwards 1994). Originally developed for complex fluids, the GENERIC-based approach has more recently been applied to the

formulation of thermodynamic models (Hütter and Tervoort 2008b, c, d; Mielke 2011; Hütter and Svendsen 2011; 2012; Hütter and van Breemen 2012; Hütter and Svendsen 2013; Gladkov et al. 2016; Semkiv and Hütter 2016; Semkiv et al. 2016; Semkiv et al. 2017) for elastic and inelastic solids. Recently, a non-isothermal phase-field model for alloy solidification was formulated based on the bracket formalism (Bollada et al. 2017). Their formulation is closely related to the GENERIC-based treatment of non-isothermal phase-field models and the related temperature equation in Gladkov et al. (2016). In particular, as in the latter case, the formulation of Bollada et al. (2017) is weakly non-local.

The purpose of the current work is to apply the GENERIC-based approach to the formulation of models for the (thermo) dynamics of multicomponent, multiphase solids. In this context, the resulting models for conservative and non-conservative dynamics are inherently spatially strongly non-local (i.e., functional) and non-isothermal in nature. The former is analogous to models formulated in the context of classical density functional theory (CDFT), for example, phase-field crystal modeling, including elastic and plastic deformation and multiple crystal orientations (e.g., Elder and Grant 2004; Elder et al. 2007; Provatas and Elder 2010), as based on the free energy functional. By analogy with CDFT, one can pursue weakly non-local approximations of the energy and entropy functionals of the strongly non-local formulation such as local density or generalized gradient approximations. In particular, in the context of the former, it is shown here that the current formulation simplifies to the corresponding weakly non-local GENERIC-based treatment of Gladkov et al. (2016). In this latter work, it was shown that this is compatible with the non-isothermal generalization of Penrose and Fife (1990, 1993) of the well-known models of Cahn and Hilliard (1958) and Allen and Cahn (1979) for conservative and non-conservative phase field dynamics. Strongly non-local dynamics is also of interest in the context of coarse-grained "slow" dislocation dynamics in terms of ensemble-averaged dislocation densities with long-ranged Peach-Koehler-type interactions (Kooiman et al. 2014, 2015; Zaiser 2015) or more generally for systems with long-range interactions (Giacomin and Lebowitz 1997; Bates 2006; Hütter and Brader 2009).

For completeness, it is worth noting that other thermodynamic frameworks could be, and have been, employed for the formulation of non-local dynamics. For example, the bracket formalism (Beris and Edwards 1994) has been employed to this end in Bollada et al. (2017). A detailed comparison between the single-generator bracket formalism and the double-generator formalism GENERIC has been performed in general terms (Edwards 1998; Edwards et al. 1998). It has been found that the GENERIC is more flexible and has several advantages over the bracket formalism (see p. 19 in Öttinger 2005; Edwards et al. 1998). Therefore, the GENERIC framework is used in this paper.

The paper begins with a brief summary of the GENERIC in the "Brief review of the GENERIC" section. This is followed by the strongly non-local GENERIC-based model formulation in the "Strongly non-local model formulation" section. Connection is made with the existing weakly non-local model formulation of Gladkov et al. (2016) in the "Special case: weakly non-local model formulation" section. Finally, the current approach is applied to derive a non-isothermal generalization of a phase-field crystal model for binary alloys in the "Non-isothermal generalization of a phase-field crystal model for binary alloys" section. The paper ends with a summary and discussion (the "Summary and discussion" section).

In all relations to follow, all operators, such as ∇, $\nabla\cdot \equiv$ div, $\mathcal{L}$, and $\mathcal{M}$, operate on everything to their right (unless otherwise indicated). Position arguments r, r', etc. are given explicitly where required for clarity, but omitted otherwise.

Brief review of the GENERIC

As discussed in the introduction, the current framework for non-local thermodynamics is based on the GENERIC (for closed systems: Grmela and Öttinger 1997; Öttinger and Grmela 1997; Öttinger 2005)

$$\dot{x} = \dot{x}|_{\mathrm{rev}} + \dot{x}|_{\mathrm{irr}}, \quad \dot{x}|_{\mathrm{rev}} = \mathcal{L}\,\mathcal{D}_x E, \quad \dot{x}|_{\mathrm{irr}} = \mathcal{M}\,\mathcal{D}_x S, \tag{1}$$

for the reversible (rev) and irreversible (irr) evolution of thermodynamic state variables x. As indicated, this evolution is driven by the gradients $\mathcal{D}_x E$ and $\mathcal{D}_x S$ of total energy $E[x]$ and entropy $S[x]$, respectively, mediated by Poisson $\mathcal{L}$ and "friction" $\mathcal{M}$ operators.

Certain conditions apply to the operators $\mathcal{L}$ and $\mathcal{M}$. With respect to the "scalar" product $\langle \mathcal{D}_x A, \mathcal{D}_x B \rangle$, and $\langle \mathcal{D}_x B, \mathcal{O}^{\mathrm{T}} \mathcal{D}_x A \rangle := \langle \mathcal{D}_x A, \mathcal{O}\, \mathcal{D}_x B \rangle$ defining the operator transpose, one can introduce the brackets

$$\{A, B\} := \langle \mathcal{D}_x A, \mathcal{L}\,\mathcal{D}_x B \rangle, \quad [A, B] := \langle \mathcal{D}_x A, \mathcal{M}\,\mathcal{D}_x B \rangle. \tag{2}$$

In these terms, the conditions on the Poisson operator $\mathcal{L}$ can be written in the form

$$\begin{aligned}
&\{B, A\} = -\{A, B\}, \\
&\{A, S\} = 0, \\
&\{A, \{B, C\}\} + \{B, \{C, A\}\} + \{C, \{A, B\}\} = 0,
\end{aligned} \tag{3}$$

with A, B, and C arbitrary functionals. In particular, the last condition is the Jacobi identity, reflecting the time-structure invariance of the reversible dynamics. The skew-symmetry $(3)_1$ and orthogonality $(3)_2$ conditions can also be written as $\mathcal{L}^{\mathrm{T}} = -\mathcal{L}$ and $\mathcal{L}\,\mathcal{D}_x S = 0$, respectively. Using the bracket $(2)_2$, the conditions for the friction operator $\mathcal{M}$ are

$$\begin{aligned}
&[B, A] = [A, B], \\
&[A, E] = 0, \\
&[A, A] \geq 0,
\end{aligned} \tag{4}$$

with A and B arbitrary functionals. The symmetry $(4)_1$ and orthogonality $(4)_2$ conditions can also be written as $\mathcal{M}^{\mathrm{T}} = \mathcal{M}$ (Onsager-symmetry) and $\mathcal{M}\,\mathcal{D}_x E = 0$, respectively. For the specific case of Casimir, rather than Onsager, symmetry, the reader is referred to Öttinger (2005) for more details.

Based on (1) and (2),

$$\dot{A} = \langle \mathcal{D}_x A, \dot{x} \rangle = \langle \mathcal{D}_x A, \mathcal{L}\,\mathcal{D}_x E \rangle + \langle \mathcal{D}_x A, \mathcal{M}\,\mathcal{D}_x S \rangle = \{A, E\} + [A, S] \tag{5}$$

determines the evolution of an observable $A[x]$. For the specific case of the total energy, $A = E$, one obtains by virtue of $(3)_1$ and $(4)_2$

$$\dot{E} = \{E, E\} + [E, S] = 0, \tag{6}$$

i.e., the total energy is conserved. Conversely, for $A = S$, $(3)_2$ and $(4)_3$ imply

$$\dot{S} = \{S, E\} + [S, S] = [S, S] \geqslant 0, \tag{7}$$

i.e., the entropy is non-decreasing. While conclusions (6) and (7) are definitely meeting the expectations for closed systems, it must be pointed out that the sets of conditions (3) and (4) are much richer than the two scalar conditions (6) and (7). These are employed for model development, as illustrated in the following section.

For more background and details concerning the GENERIC, the interested reader is referred to Grmela and Öttinger (1997); Öttinger and Grmela (1997); Öttinger (2005).

Strongly non-local model formulation

Basic considerations

The GENERIC-based relations summarized in the previous section are abstract and intrinsic in nature. The purpose of this section is to obtain specific forms of these relations relevant to the modeling of the non-local dynamics of multicomponent and multiphase solid systems. In this work, attention is focused on (strong) spatial non-locality (i.e., in the sense of classic density functional theory) alone; temporal non-locality[1] is not considered. Strong spatial non-locality of (1) is modeled in this work via the form[2]

$$\mathcal{G}\,\mathcal{D}_\mathbf{x} A\,(\mathbf{r}) := \int \boldsymbol{G}(\mathbf{r}, \mathbf{r}')\,\mathcal{D}_{\mathbf{x}(\mathbf{r}')}A\,dv(\mathbf{r}') =: \boldsymbol{G} * \mathcal{D}_\mathbf{x} A\,(\mathbf{r}) \tag{8}$$

for the action of $\mathcal{G}$ (i.e., $\mathcal{L}$ or $\mathcal{M}$) on $\mathcal{D}_\mathbf{x} A$ (i.e., $A = E$ or $A = S$) in terms of a corresponding (distribution-function-like) "kernel" operator $\boldsymbol{G}(\mathbf{r}, \mathbf{r}')$. In turn,

$$\langle \mathcal{D}_\mathbf{x} A, \mathcal{G}\,\mathcal{D}_\mathbf{x} B \rangle := \int \mathcal{D}_\mathbf{x} A \cdot \boldsymbol{G} * \mathcal{D}_\mathbf{x} B\,dv = \iint \mathcal{D}_\mathbf{x} A \cdot \boldsymbol{G}\,\mathcal{D}_{\mathbf{x}'} B\,dv\,dv' \tag{9}$$

then holds via (75) for the bilinear operator relations on the right-hand side (r.h.s.) of the Poisson and dissipation brackets (2). In the remainder of this paper, $\mathcal{D}_\mathbf{x} A$ denotes the functional derivative[3] of A.

The strongly non-local formulation studied in this paper, (8) and (9), is a realization of the GENERIC (Grmela and Öttinger 1997; Öttinger and Grmela 1997), involving generalized contractions, i.e., summations over both discrete and continuous indices, the latter leading to integrations (see Section 2.2.2 in Öttinger 2005). It has been shown that systematic coarse-graining using non-equilibrium statistical mechanics, which has been employed to derive the GENERIC in general (see Chapter 6 in Öttinger 2005), results in non-local expressions for the Poisson and friction operators (see Section 5 in Hütter and Tervoort 2008a). For the special case that the entries in the Poisson and friction operators can be expressed in terms of the Dirac delta function, the strongly non-local formulation reduces to its local counterpart (see Section 2.2.2 in Öttinger 2005; Section 5 in Hütter and Tervoort 2008a).

Given these basic relations for the functional-based formulation of the GENERIC, (8) and (9), we are now in a position to apply these to the case of multicomponent, multiphase solid systems.

GENERIC variables and basic functionals

The strongly non-local form of the GENERIC formulated in the last section is now applied to the formulation of a model for a non-isothermal, heat-conducting mixture of displacively transforming thermoelastic solid phases and diffusing chemical constituents. Basic mixture fields include the mass density ρ, the deformation $\boldsymbol{\chi} \equiv (\chi_1, \chi_2, \chi_3)$, the velocity $\boldsymbol{v} \equiv (v_1, v_2, v_3)$, the momentum density $\boldsymbol{m} = \rho\,\boldsymbol{v} \equiv (m_1, m_2, m_3)$, and a "thermal"

variable ς chosen in what follows in order to obtain the simplest possible formulation. Lastly, $\boldsymbol{\varphi} = (\varphi_1, \ldots, \varphi_p)$ represents an array of p scalar phase fields of both conservative and non-conservative type. These will be specified in more detail later. On this basis,

$$\boldsymbol{x}^\varsigma := (\boldsymbol{\chi}, \boldsymbol{m}, \varsigma, \boldsymbol{\varphi}) \equiv (\chi_1, \chi_2, \chi_3, m_1, m_2, m_3, \varsigma, \varphi_1, \ldots, \varphi_p) \tag{10}$$

represents the complete set of GENERIC-based variables for the current constitutive class. All these fields are referential or Lagrangian with respect to the mixture. In this case, mixture mass conservation implies $\dot{\rho} = 0$ and $\boldsymbol{v} = \dot{\boldsymbol{\chi}}$ holds. In the context of (10), then, $E[\boldsymbol{x}^\varsigma]$ and $S[\boldsymbol{x}^\varsigma]$ take the forms

$$E[\boldsymbol{x}^\varsigma] = \int e[\boldsymbol{x}^\varsigma]\ dv, \quad e[\boldsymbol{x}^\varsigma] = \frac{|\boldsymbol{m}|^2}{2\rho} + \varepsilon[\boldsymbol{x}^\varsigma], \quad S[\boldsymbol{x}^\varsigma] = \int \eta[\boldsymbol{x}^\varsigma]\ dv, \tag{11}$$

respectively, in terms of the corresponding densities of internal energy ε and of entropy η. Like E and S, ε and η are in general functionals of the "components" (10) of $\boldsymbol{x}^\varsigma$, the only exception to this being that both ε and η do not depend of the momentum density $\boldsymbol{m}$.

Poisson operator and reversible dynamics

As shown elsewhere (e.g., Öttinger 2005), the choice

$$\varsigma \equiv \eta \tag{12}$$

results in the simplest possible formulation of reversible dynamics $(1)_2$. Then[4]

$$\begin{aligned}
\mathcal{D}_{\boldsymbol{x}^\eta} E &= (\mathcal{D}_{\boldsymbol{\chi}} E, \mathcal{D}_{\boldsymbol{m}} E, \mathcal{D}_\eta E, \mathcal{D}_{\boldsymbol{\varphi}} E) = (\mathcal{D}_{\boldsymbol{\chi}} E, \boldsymbol{m}/\rho, \theta, \mathcal{D}_{\boldsymbol{\varphi}} E), \\
\mathcal{D}_{\boldsymbol{x}^\eta} S &= (\mathcal{D}_{\boldsymbol{\chi}} S, \mathcal{D}_{\boldsymbol{m}} S, \mathcal{D}_\eta S, \mathcal{D}_{\boldsymbol{\varphi}} S) = (\boldsymbol{0}, \boldsymbol{0}, 1, \boldsymbol{0}),
\end{aligned} \tag{13}$$

follow from (11) and the fact that η is in the set of independent variables. In particular, note that

$$\mathcal{D}_{\eta(\boldsymbol{r})} S = \int \mathcal{D}_{\eta(\boldsymbol{r})} \eta(\boldsymbol{r}')\ dv(\boldsymbol{r}') = \int \delta(\boldsymbol{r} - \boldsymbol{r}')\ dv(\boldsymbol{r}') = 1 \tag{14}$$

follows from (11), and

$$\theta(\boldsymbol{r}) := \mathcal{D}_{\eta(\boldsymbol{r})} E = \int \mathcal{D}_{\eta(\boldsymbol{r})} \varepsilon(\boldsymbol{r}')\ dv(\boldsymbol{r}') \tag{15}$$

has units[5] of temperature.

In the context of (8), the current model for strongly non-local reversible dynamics $(1)_2$ takes the form

$$\dot{\boldsymbol{x}}^\eta|_{\text{rev}}(\boldsymbol{r}) = \mathcal{L}\mathcal{D}_{\boldsymbol{x}^\eta} E(\boldsymbol{r}) = \int \boldsymbol{L}(\boldsymbol{r}, \boldsymbol{r}')\ \mathcal{D}_{\boldsymbol{x}^\eta(\boldsymbol{r}')} E\ dv(\boldsymbol{r}') = \boldsymbol{L} * \mathcal{D}_{\boldsymbol{x}^\eta} E(\boldsymbol{r}), \tag{16}$$

with $\mathcal{L}$, and so $\boldsymbol{L}$, subject to the conditions (3). Since the evolution of the phase field $\boldsymbol{\varphi}$ is due only to irreversible processes like transport and relaxation, the dynamics of these fields is purely irreversible (i.e., in the current Lagrangian or referential setting). In this case, the only variables with non-vanishing reversible dynamics are $\boldsymbol{\chi}$, $\boldsymbol{m}$, and $\varsigma = \eta$. The first two have been treated in (Hütter and Svendsen 2011; 2012) for the purely local and weakly non-local cases. Generalizing their approach to the current context, $\boldsymbol{L}$ in (16) takes the form

$$\boldsymbol{L} = \begin{bmatrix} L_{\chi\chi} & L_{\chi m} & L_{\chi\eta} & L_{\chi\varphi} \\ L_{m\chi} & L_{mm} & L_{m\eta} & L_{m\varphi} \\ L_{\eta\chi} & L_{\eta m} & L_{\eta\eta} & L_{\eta\varphi} \\ L_{\varphi\chi} & L_{\varphi m} & L_{\varphi\eta} & L_{\varphi\varphi} \end{bmatrix} = \begin{bmatrix} 0 & \delta\boldsymbol{I} & 0 & 0 \\ -\delta\boldsymbol{I} & 0 & L_{m\eta} & 0 \\ 0 & L_{\eta m} & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}. \tag{17}$$

In this case, the upper-left block of $L(r, r')$ is determined in particular by the three-dimensional delta function $\delta(r - r')$ and I the second-order identity tensor. Given (17), (16) reduces to

$$
\begin{aligned}
\dot{\chi}|_{\text{rev}} &= L_{\chi m} * \mathcal{D}_m E & &= m/\rho \,, \\
\dot{m}|_{\text{rev}} &= L_{m\chi} * \mathcal{D}_\chi E + L_{m\eta} * \mathcal{D}_\eta E &&= -\mathcal{D}_\chi E + L_{m\eta} * \theta \,, \\
\dot{\eta}|_{\text{rev}} &= L_{\eta m} * \mathcal{D}_m E & &= L_{\eta m} * \dot{\chi} \,, \\
\dot{\varphi}|_{\text{rev}} &= \mathbf{0} \,,
\end{aligned}
\tag{18}
$$

via $(13)_1$. Furthermore, using $(13)_2$, the only non-trivial contribution to the orthogonality condition $(3)_2$, in the form $0 = \mathcal{L} \mathcal{D}_{\chi\eta} S(r) = L * \mathcal{D}_{\chi\eta} S(r)$, is given by

$$
\mathbf{0} = L_{m\eta} * \mathcal{D}_\eta S(r) = \int L_{m\eta}(r, r') \, dv(r')
\tag{19}
$$

via (14). This condition is solved by

$$
L_{m\eta} = \mathbf{0} \,, \quad L_{\eta m} = -L_{m\eta}^{\mathrm{T}} = \mathbf{0} \,,
\tag{20}
$$

where in the second equality the skew-symmetry condition $(3)_1$ has been employed. Given these results, (18) reduce to

$$
\dot{\chi}|_{\text{rev}} = m/\rho \,, \quad \dot{m}|_{\text{rev}} = -\mathcal{D}_\chi E \,, \quad \dot{\eta}|_{\text{rev}} = 0 \,, \quad \dot{\varphi}|_{\text{rev}} = \mathbf{0} \,,
\tag{21}
$$

for reversible dynamics. The expected form $(21)_3$ for the reversible dynamics of the entropy density η serves in hindsight as a justification of the choice (20). In order to complete the formulation of the reversible dynamics, it can be shown by a straightforward calculation that the Poisson operator (17) with (20) satisfies the Jacobi identity $(3)_3$.

Friction operator and irreversible dynamics

For irreversible dynamics $(1)_3$, the choice

$$
\varsigma \equiv \varepsilon
\tag{22}
$$

results in the simplest formulation, and so the complete set of variables is denoted by x^ε. In this case,

$$
\begin{aligned}
\mathcal{D}_{x^\varepsilon} E &= (\mathcal{D}_\chi E, \mathcal{D}_m E, \mathcal{D}_\varepsilon E, \mathcal{D}_\varphi E) = (\mathbf{0}, m/\rho, 1, \mathbf{0}) \,, \\
\mathcal{D}_{x^\varepsilon} S &= (\mathcal{D}_\chi S, \mathcal{D}_m S, \mathcal{D}_\varepsilon S, \mathcal{D}_\varphi S) = (\mathcal{D}_\chi S, \mathbf{0}, \vartheta, \mathcal{D}_\varphi S) \,,
\end{aligned}
\tag{23}
$$

is obtained from (11), since the internal energy density ε is in the set of variables. Here,

$$
\mathcal{D}_{\varepsilon(r)} E = \int \mathcal{D}_{\varepsilon(r)} \varepsilon(r') \, dv(r') = \int \delta(r - r') \, dv(r') = 1 \,,
\tag{24}
$$

analogous to (14), and

$$
\vartheta(r) := \mathcal{D}_{\varepsilon(r)} S = \int \mathcal{D}_{\varepsilon(r)} \eta(r') \, dv(r')
\tag{25}
$$

has units of inverse temperature or "coldness."

Analogous to (16) in the reversible case, the model relation

$$
\dot{x}^\varepsilon|_{\text{irr}}(r) = \mathcal{M} \mathcal{D}_{x^\varepsilon} S(r) = \int M(r, r') \mathcal{D}_{x^\varepsilon(r')} S \, dv(r') = M * \mathcal{D}_{x^\varepsilon} S(r)
\tag{26}
$$

is assumed for $(1)_3$ in the context of (8) together with the choice (22) for ς. For the current class of materials, $\dot{x}|_{\text{irr}}$ from $(1)_3$ is assumed to be influenced in general by (irreversible, diffusive) mass, momentum and energy (heat) transport, chemical reactions, and

(micro)structural rearrangement (e.g., phase transformation). For simplicity, the following treatment is restricted to thermoelastic phases, and irreversible momentum transport is neglected. In this case, there is no irreversible contribution to momentum evolution. Since the kinematic relation $(21)_1$ is not affected by irreversible processes, this together with (1) implies that the χ and m rows of M vanish identically. On this basis, the reduced form

$$
M = \begin{bmatrix} M_{\chi\chi} & M_{\chi m} & M_{\chi\varepsilon} & M_{\chi\varphi} \\ M_{m\chi} & M_{mm} & M_{m\varepsilon} & M_{m\varphi} \\ M_{\varepsilon\chi} & M_{\varepsilon m} & M_{\varepsilon\varepsilon} & M_{\varepsilon\varphi} \\ M_{\varphi\chi} & M_{\varphi m} & M_{\varphi\varepsilon} & M_{\varphi\varphi} \end{bmatrix} = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & M_{\varepsilon\varepsilon} & M_{\varepsilon\varphi} \\ 0 & 0 & M_{\varphi\varepsilon} & M_{\varphi\varphi} \end{bmatrix}
\tag{27}
$$

for M follows, where the symmetry $(4)_1$ of the friction operator has been employed. We then have the following non-trivial contributions

$$
\begin{aligned}
\dot{\varepsilon}|_{\mathrm{irr}} &= M_{\varepsilon\varepsilon} * \mathcal{D}_\varepsilon S + M_{\varepsilon\varphi} * \mathcal{D}_\varphi S, \\
\dot{\varphi}|_{\mathrm{irr}} &= M_{\varphi\varepsilon} * \mathcal{D}_\varepsilon S + M_{\varphi\varphi} * \mathcal{D}_\varphi S,
\end{aligned}
\tag{28}
$$

to the evolution equations from (26). Furthermore, using $(23)_1$, the only non-trivial contributions to the orthogonality condition $(4)_2$, in the form $0 = \mathcal{M} \mathcal{D}_{\chi^\varepsilon} E(r) = M * \mathcal{D}_{\chi^\varepsilon} E(r)$, are given by

$$
\begin{aligned}
0 = M_{\varepsilon\varepsilon} * \mathcal{D}_\varepsilon E(r) &= \int M_{\varepsilon\varepsilon}(r, r')\, dv(r'), \\
\mathbf{0} = M_{\varphi\varepsilon} * \mathcal{D}_\varepsilon E(r) &= \int M_{\varphi\varepsilon}(r, r')\, dv(r'),
\end{aligned}
\tag{29}
$$

via (24). It is clear that the choices $M_{\varepsilon\varepsilon} = 0$ and $M_{\varphi\varepsilon} = \mathbf{0}$ are physically not meaningful, since the symmetry condition $(4)_1$ would imply $M_{\varepsilon\varphi} = \mathbf{0}$, and therefore, $\dot{\varepsilon}|_{\mathrm{irr}} = 0$ according to $(28)_1$, which is incompatible with heat conduction. This is discussed in more detail in the sequel.

Decomposition of the friction operator

A refined model formulation is obtained with the help of the multiplicative decomposition

$$
\begin{aligned}
M(r, r') &= \iint C(r, r'') D(r'', r''') C^{\mathrm{T}}(r''', r')\, dv(r'')\, dv(r'''), \\
&= C * D * C^{\mathrm{T}}(r, r'),
\end{aligned}
\tag{30}
$$

(e.g., Edwards 1998; Öttinger 2005) relating the flux-force-based kinetic coefficients D (material properties) to the friction operator M in terms of the operator C and its adjoint C^{T}. Given (30), (26) takes the split form

$$
\dot{x}^\varepsilon|_{\mathrm{irr}} = C * j, \quad j := D * f, \quad f := C^{\mathrm{T}} * \mathcal{D}_{\chi^\varepsilon} S,
\tag{31}
$$

in terms of thermodynamic forces f and fluxes j. As well, (30) induces the form

$$\dot{S} = \int \mathcal{D}_{\chi^\varepsilon} S \cdot \dot{\boldsymbol{x}}^\varepsilon \, dv = \int \mathcal{D}_{\chi^\varepsilon} S \cdot \boldsymbol{C} * \boldsymbol{D} * \boldsymbol{C}^\mathrm{T} * \mathcal{D}_{\chi^\varepsilon} S \, dv$$

$$= \int (\boldsymbol{C}^\mathrm{T} * \mathcal{D}_{\chi^\varepsilon} S) \cdot \boldsymbol{D} * (\boldsymbol{C}^\mathrm{T} * \mathcal{D}_{\chi^\varepsilon} S) \, dv \qquad (32)$$

$$= \int \boldsymbol{f} \cdot \boldsymbol{D} * \boldsymbol{f} \, dv$$

of the entropy production-rate via (7), which is in general quasi-quadratic in f (i.e., $\boldsymbol{D}$ may in general depend on f). The symmetry and non-negative definiteness of $\boldsymbol{D}$, i.e.,

$$\boldsymbol{D}^\mathrm{T} = \boldsymbol{D}, \quad \int \boldsymbol{f} \cdot \boldsymbol{D} * \boldsymbol{f} \, dv \geqslant 0, \qquad (33)$$

respectively, induce the analogous restrictions

$$\boldsymbol{M}^\mathrm{T} = \boldsymbol{M}, \quad \int \mathcal{D}_{\chi^\varepsilon} A \cdot \boldsymbol{M} * \mathcal{D}_{\chi^\varepsilon} A \, dv \geqslant 0, \qquad (34)$$

on $\boldsymbol{M}$ via (30), and so those (4)$_{1,3}$ on $\mathcal{M}$. As well, the orthogonality condition (4)$_2$ becomes

$$\mathcal{M} \mathcal{D}_{\chi^\varepsilon} E = \boldsymbol{C} * \boldsymbol{D} * \boldsymbol{C}^\mathrm{T} * \mathcal{D}_{\chi^\varepsilon} E = 0, \qquad (35)$$

which is satisfied identically if

$$\boldsymbol{C}^\mathrm{T} * \mathcal{D}_{\chi^\varepsilon} E = 0. \qquad (36)$$

Experience with the structure of numerous non-equilibrium thermodynamics models has shown that, in addition to the roles of $\boldsymbol{C}$ in (31)$_3$ and (31)$_1$, the condition (36) is useful for determination of the operator $\boldsymbol{C}$ (e.g., Edwards 1998). In the context of (8) and (30), the conditions implied by (4) on the friction operator $\mathcal{M}$ are satisfied mathematically if (i) $\boldsymbol{D}$ is symmetric and non-negative-definite and (ii) $\boldsymbol{C}^\mathrm{T}$ is orthogonal to $\mathcal{D}_{\chi^\varepsilon} E$, i.e., (36). Any physical choice for the "components" of $\boldsymbol{M}$ must satisfy these.

The remainder of the formulation is based on the split $\boldsymbol{\varphi} = (\boldsymbol{\varrho}, \boldsymbol{\phi})$ of $\boldsymbol{\varphi}$ into m conservative $\boldsymbol{\varrho} = (\varrho_1, \ldots, \varrho_m)$ and s non-conservative $\boldsymbol{\phi} = (\phi_1, \ldots, \phi_s)$ parts, with $p = m + s$. To determine the elements of $\boldsymbol{C}$, we aim in particular at $\dot{\boldsymbol{\chi}}|_{\mathrm{irr}} = \boldsymbol{0}$ and $\dot{\boldsymbol{m}}|_{\mathrm{irr}} = \boldsymbol{0}$ (see argument above). In the context of (31)$_1$, then, $\boldsymbol{C}$ reduces to

$$\boldsymbol{C} = \begin{bmatrix} \boldsymbol{0} & \boldsymbol{0} & \boldsymbol{0} \\ \boldsymbol{0} & \boldsymbol{0} & \boldsymbol{0} \\ C_{\varepsilon j_\varepsilon} & C_{\varepsilon j_\varrho} & C_{\varepsilon j_\phi} \\ C_{\varrho j_\varepsilon} & C_{\varrho j_\varrho} & C_{\varrho j_\phi} \\ C_{\phi j_\varepsilon} & C_{\phi j_\varrho} & C_{\phi j_\phi} \end{bmatrix}. \qquad (37)$$

For the further specification of the elements of $\boldsymbol{C}$, it is noted that the split (30) of $\boldsymbol{M}$ is not unique, even when using the conditions on $\boldsymbol{C}$ and $\boldsymbol{D}$ mentioned above. Nevertheless, the split (30) can be used if the operator $\boldsymbol{C}$ is determined by a more specific interpretation of the fluxes j, as explained in the following. With the interpretation that j_ε and j_ϱ are diffusive fluxes (i.e., heat and mass, respectively), whereas j_ϕ is a (generally non-conservative) "displacive" flux related to structural rearrangement, $-\nabla \cdot j_\varepsilon$, $-\nabla \cdot j_\varrho$, and j_ϕ,

respectively, are the corresponding contributions to irreversible[6] dynamics. From these, follow

$$C_{\varepsilon j_\varepsilon}(r, r'') = -\nabla \delta(r - r''),$$
$$C_{\varrho j_\varrho}(r, r'') = -\nabla \delta(r - r'') \, I, \tag{38}$$
$$C_{\phi j_\phi}(r, r'') = \delta(r - r'') \, I,$$

for the "diagonal" components of C. As before, δ is the three-dimensional Dirac delta function. Here and in what follows, ∇ is the gradient operator with respect to r, ∇' that with respect to r', and so on. The non-square structure of C follows from the fact that $(31)_1$ transforms *three* thermodynamic fluxes j_ε, j_ϱ, and j_ϕ into *five* relations for the irreversible dynamics of χ, m, ε, ϱ, and ϕ.

While the components (38) of C contain negative derivatives of the Dirac delta function with respect to the first position argument of C, in turn, the components of C^T must contain negative derivatives with respect to the second position argument of C^T. This leads to

$$C_{\varepsilon j_\varepsilon}^T(r''', r') = -\nabla' \delta(r''' - r'),$$
$$C_{\varrho j_\varrho}^T(r''', r') = -\nabla' \delta(r''' - r') \, I, \tag{39}$$
$$C_{\phi j_\phi}^T(r''', r') = \delta(r''' - r') \, I,$$

for the components of C^T related to (38). Further elements in (37) are constrained by the orthogonality condition (36). In particular,

$$0 = C_{\varepsilon j_\varepsilon}^T * D_\varepsilon E(r''') = -\int \nabla' \delta(r''' - r') \, dv(r'),$$
$$0 = C_{\varepsilon j_\varrho}^T * D_\varepsilon E(r''') = \int C_{\varepsilon j_\varrho}^T(r''', r') \, dv(r'), \tag{40}$$
$$0 = C_{\varepsilon j_\phi}^T * D_\varepsilon E(r''') = \int C_{\varepsilon j_\phi}^T(r''', r') \, dv(r'),$$

follow from $(23)_1$ and $(39)_1$. Clearly, $(40)_1$ is identically fulfilled. Sufficient for satisfaction of the other two are

$$C_{\varepsilon j_\varrho} = 0, \quad C_{\varepsilon j_\phi} = 0, \tag{41}$$

which we work with in what follows. The remaining elements in (37) can be determined by the physical assumptions that there are no other contributions to the irreversible dynamics of ϱ and ϕ than $-\nabla \cdot j_\varrho$ and j_ϕ, respectively, i.e.,

$$C_{\varrho j_\varepsilon} = 0, \quad C_{\varrho j_\phi} = 0, \quad C_{\phi j_\varepsilon} = 0, \quad C_{\phi j_\varrho} = 0. \tag{42}$$

These physical assumptions, together with (38), imply

$$C(r, r'') = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ -\nabla \delta(r - r'') & 0 & 0 \\ 0 & -\nabla \delta(r - r'') \, I & 0 \\ 0 & 0 & \delta(r - r'') \, I \end{bmatrix}, \tag{43}$$

and the irreversible contributions $(31)_1$ to the evolution equations become

$$\dot{\chi}|_{\mathrm{irr}} = 0, \quad \dot{m}|_{\mathrm{irr}} = 0, \quad \dot{\varepsilon}|_{\mathrm{irr}} = -\nabla \cdot j_\varepsilon, \quad \dot{\varrho}|_{\mathrm{irr}} = -\nabla \cdot j_\varrho, \quad \dot{\phi}|_{\mathrm{irr}} = j_\phi. \tag{44}$$

Calculating the operator adjoint to C given by (43), using (39),

$$
C^{\mathrm{T}}(r''', r') = \begin{bmatrix} \mathbf{0} & \mathbf{0} & -\nabla'\delta(r''' - r') & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & -\nabla'\delta(r''' - r')\,\mathbf{I} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} & \delta(r''' - r')\,\mathbf{I} \end{bmatrix}, \tag{45}
$$

the thermodynamic forces $f = (f_\varepsilon, f_\varrho, f_\phi)$ given by $(31)_3$ for irreversible dynamics take the form

$$
f_\varepsilon = \nabla \mathcal{D}_\varepsilon S = \nabla \vartheta, \quad f_\varrho = \nabla \mathcal{D}_\varrho S, \quad f_\phi = \mathcal{D}_\phi S. \tag{46}
$$

To complete the thermodynamic modeling of the irreversible dynamics (44) with thermodynamic forces (46), one requires a constitutive relation between the thermodynamic forces f and fluxes j, i.e., a realization of $(31)_2$. If the corresponding (in general non-diagonal and non-local) operator D is symmetric and non-negative definite according to (33), then all GENERIC conditions (4) are respected, since the orthogonality condition $(4)_2$ has been taken care of via (41).

Complete model: combination of reversible and irreversible dynamics

The formulations of the reversible (the "Poisson operator and reversible dynamics" section) and irreversible (the "Friction operator and irreversible dynamics" and "Decomposition of the friction operator" sections) dynamics did not make use of the same set of variables. This has been done for reasons of keeping the application of the GENERIC framework transparent. However, this discrepancy in the sets of variables, x^η and x^ε, comes at the cost of additional work required to formulate the composed model, combining the reversible and irreversible dynamics. This is the topic of this section.

Combining (21) and (44), one obtains for the dynamics

$$
\dot{\chi} = m/\rho, \quad \dot{m} = -\mathcal{D}_\chi E[x^\eta], \quad \dot{\varrho} = -\nabla \cdot j_\varrho, \quad \dot{\phi} = j_\phi, \tag{47}
$$

where the thermodynamic fluxes j are related to the thermodynamic forces

$$
f_\varepsilon = \nabla \vartheta, \qquad f_\varrho = \nabla \mathcal{D}_\varrho S[x^\varepsilon], \qquad f_\phi = \mathcal{D}_\phi S[x^\varepsilon], \tag{48}
$$

from (46) via the non-local flux-force relation $(31)_2$. The notation (see Footnote 4 above) $\mathcal{D}_{x_i^\eta} E[x^\eta]$ and $\mathcal{D}_{x_i^\varepsilon} S[x^\varepsilon]$ for partial functional derivatives emphasizes the fact that different sets of arguments (i.e., x^η and x^ε) are involved and being held fixed.

To make this model usable for an arbitrary choice of the thermal variable ς, two steps need to be taken. To simplify notation in relation to the transformation of variables, the following abbreviations are introduced, as an amendment to the operator-related notation (8),

$$
a * \left(\mathcal{D}_{x_i} b\right)^\alpha (r) := \int a(r') \left(\mathcal{D}_{x_i(r')} b(r)\right)^\alpha dv(r'),
$$

$$
\left(\mathcal{D}_{x_i} b\right)^\alpha * a\,(r) := \int \left(\mathcal{D}_{x_i(r)} b(r')\right)^\alpha a(r')\, dv(r'), \tag{49}
$$

for arbitrary function(al)s a and b with $\alpha = \pm 1$. For the first step in transforming the dynamic model to the set of variables x^ς, it is useful to employ the following identities,

via the chain rule for functional differentiation (e.g., Engel and Dreizler 2011, (A.38)),

$$
\begin{aligned}
\mathcal{D}_{\chi} E[\, x^{\eta}\,] &= \mathcal{D}_{\chi} E[\, x^{\varsigma}\,] - \left(\mathcal{D}_{\chi} \eta[\, x^{\varsigma}\,] \right) * \theta\,, \\
\mathcal{D}_{\varrho} S[\, x^{\varepsilon}\,] &= \mathcal{D}_{\varrho} S[\, x^{\varsigma}\,] - \left(\mathcal{D}_{\varrho} \varepsilon[\, x^{\varsigma}\,] \right) * \vartheta\,, \\
\mathcal{D}_{\phi} S[\, x^{\varepsilon}\,] &= \mathcal{D}_{\phi} S[\, x^{\varsigma}\,] - \left(\mathcal{D}_{\phi} \varepsilon[\, x^{\varsigma}\,] \right) * \vartheta\,,
\end{aligned}
\tag{50}
$$

with θ from (15), ϑ from (25), and now

$$
\begin{aligned}
\theta &= \mathcal{D}_{\eta} E[\, x^{\eta}\,] = \left(\mathcal{D}_{\varsigma} E[\, x^{\varsigma}\,] \right) * \left(\mathcal{D}_{\varsigma} \eta[\, x^{\varsigma}\,] \right)^{-1}\,, \\
\vartheta &= \mathcal{D}_{\varepsilon} S[\, x^{\varepsilon}\,] = \left(\mathcal{D}_{\varsigma} S[\, x^{\varsigma}\,] \right) * \left(\mathcal{D}_{\varsigma} \varepsilon[\, x^{\varsigma}\,] \right)^{-1}\,.
\end{aligned}
\tag{51}
$$

Here, the notation $(\mathcal{D}_{\varsigma(r')} a[\, x^{\varsigma}\,](r))^{-1} := \mathcal{D}_{a(r)} \varsigma[\, x^{a}\,](r')$ has been used for $a = \eta$ and $a = \varepsilon$. For the second step in transforming the dynamic model to the set of variables x^{ς}, the dynamics $(21)_3$ and $(44)_3$ are combined to obtain an evolution equation for the thermal variable ς. In particular, one can show that

$$
\begin{aligned}
\dot{\varsigma}|_{\mathrm{rev}} * \mathcal{D}_{\varsigma} \eta[\, x^{\varsigma}\,] &= -\dot{\chi} * \mathcal{D}_{\chi} \eta[\, x^{\varsigma}\,]\,, \\
\dot{\varsigma}|_{\mathrm{irr}} * \mathcal{D}_{\varsigma} \varepsilon[\, x^{\varsigma}\,] &= -\nabla \cdot \boldsymbol{j}_{\varepsilon} + \sum_{k=1}^{m} \left(\nabla \cdot \boldsymbol{j}_{\varrho_k} \right) * \mathcal{D}_{\varrho_k} \varepsilon[\, x^{\varsigma}\,] - \sum_{l=1}^{s} \boldsymbol{j}_{\phi_l} * \mathcal{D}_{\phi_l} \varepsilon[\, x^{\varsigma}\,]\,.
\end{aligned}
\tag{52}
$$

Using the chain rule $\mathcal{D}_{\varsigma} \varepsilon[\, x^{\varsigma}\,] = \mathcal{D}_{\varsigma} \eta[\, x^{\varsigma}\,] * \mathcal{D}_{\eta} \varepsilon[\, x^{\eta}\,]$, the two equations in (52) combine to yield

$$
\begin{aligned}
\dot{\varsigma} * \mathcal{D}_{\varsigma} \varepsilon[\, x^{\varsigma}\,] &= \left(-\dot{\chi} * \mathcal{D}_{\chi} \eta[\, x^{\varsigma}\,] \right) * \mathcal{D}_{\eta} \varepsilon[\, x^{\eta}\,] \\
&\quad - \nabla \cdot \boldsymbol{j}_{\varepsilon} + \sum_{k=1}^{m} \left(\nabla \cdot \boldsymbol{j}_{\varrho_k} \right) * \mathcal{D}_{\varrho_k} \varepsilon[\, x^{\varsigma}\,] - \sum_{l=1}^{s} \boldsymbol{j}_{\phi_l} * \mathcal{D}_{\phi_l} \varepsilon[\, x^{\varsigma}\,]\,,
\end{aligned}
\tag{53}
$$

with the reversible and irreversible contributions on the first and second lines on the r.h.s., respectively. For the particular choice $\varsigma = \eta$, the reversible contributions vanish, in agreement with $(21)_3$. Alternatively, for the special choice $\varsigma = \varepsilon$, the irreversible contribution reduces to $(44)_3$. Yet another choice for ς will be discussed in the following section, when reducing the above model to the the weakly non-local case.

Note that the left-hand side (l.h.s.) of the evolution equation (53) is not a simple time derivative but rather involves in general a spatial integration (convolution). In order to solve this evolution equation, one of the following two alternatives can be used. On the one hand, one can see the functional derivative $\mathcal{D}_{\varsigma} \varepsilon[\, x^{\varsigma}\,]$ on the l.h.s. of (53) as an operator. If its inverse exists, the operator inverse can be applied to both sides of (53), which will lead to an explicit equation for the change in the thermal variable, $\dot{\varsigma}$. More generally, forward-Euler numerical time integration of both sides of (53) yields a coupled functional relation between the current temperature and deformation fields. Given the latter from forward-Euler-based solution of $(47)_{1,2}$, (53) can be solved in fixed-point fashion for the current temperature.

Note that the driving forces (50) for the reversible and irreversible dynamics can be simplified, if the quantities θ and ϑ in (51), respectively, satisfy certain conditions. Specifically,

$$
\begin{aligned}
\mathcal{D}_{\chi} \theta[\, x^{\varsigma}\,] &= 0\,, &\Rightarrow& &\mathcal{D}_{\chi} E[\, x^{\eta}\,] &= \mathcal{D}_{\chi} \Psi[\, x^{\varsigma}\,]\,, \\
\mathcal{D}_{\varrho} \vartheta[\, x^{\varsigma}\,] &= 0\,, &\Rightarrow& &\mathcal{D}_{\varrho} S[\, x^{\varepsilon}\,] &= \mathcal{D}_{\varrho} \hat{\Psi}[\, x^{\varsigma}\,]\,, \\
\mathcal{D}_{\phi} \vartheta[\, x^{\varsigma}\,] &= 0\,, &\Rightarrow& &\mathcal{D}_{\phi} S[\, x^{\varepsilon}\,] &= \mathcal{D}_{\phi} \hat{\Psi}[\, x^{\varsigma}\,]\,,
\end{aligned}
\tag{54}
$$

in terms of the Helmholtz free energy Ψ with density ψ, and the free entropy $\hat{\Psi}$, also known as the Massieu function, with density $\hat{\psi}$, defined as

$$
\begin{aligned}
\Psi &= \int \psi \, dv, \text{ with } \psi := \varepsilon - \theta \, \eta, \\
\hat{\Psi} &= \int \hat{\psi} \, dv, \text{ with } \hat{\psi} := \eta - \vartheta \, \varepsilon.
\end{aligned}
\tag{55}
$$

The conclusion that the reversible and irreversible dynamics are driven by the derivatives of the Helmholtz free energy and free entropy, respectively, is in agreement with earlier observations (Mielke 2011). As special cases of (54), one observes that the condition in $(54)_1$ is trivially satisfied if $\theta[x^\varsigma]$ depends only on ς, while the conditions in $(54)_{2,3}$ are trivially satisfied if $\vartheta[x^\varsigma]$ depends only on ς.

In summary, the main result of this paper is a non-isothermal strongly non-local model formulation for the dynamics of the GENERIC variables (10), including a split of the phase fields into conservative and non-conservative parts, as discussed in the "Decomposition of the friction operator" section. The complete model is given by the evolution Eqs. (47) and (53) together with the forces (48) and force-flux relations $(31)_2$ for an arbitrary thermal variable ς, where the relevant functional derivatives are to be calculated with the help of (50) and (51).

Special case: weakly non-local model formulation

Analogous to the case of classical density functional theory (CDFT), consider now the approximation of the above strongly non-local formulation by a weakly non-local one. As in the case of CDFT, this is based in particular on the approximation

$$
\varepsilon[x^\varsigma] \approx \varepsilon(\tilde{x}^\varsigma), \quad \eta[x^\varsigma] \approx \eta(\tilde{x}^\varsigma),
\tag{56}
$$

of the internal energy and entropy density *functionals* in (11) by *functions* of

$$
\tilde{x}^\varsigma = (\nabla \chi, \varsigma, \varrho, \nabla \varrho, \phi, \nabla \phi) \, .
\tag{57}
$$

More specifically, in the language of CDFT, this is analogous to the generalized gradient approximation (GGA). Further neglect of the dependence on $\nabla \varrho$ and $\nabla \phi$ would result in the so-called local density approximation (LDA).

For any functional A (e.g., total energy E or entropy S) of the form

$$
A[x] = \int a(x, \nabla x) \, dv,
\tag{58}
$$

with the density a being a (local) function of the variables x and their spatial gradients ∇x, the partial functional derivative of A assumes the form

$$
\mathcal{D}_{x_i} A = \partial_{x_i} a - \nabla \cdot \partial_{\nabla x_i} a =: \delta_{x_i} a,
\tag{59}
$$

in terms of the (first-order) partial variational derivative $\delta_{x_i} a$ of its density. The first equality in (59) can be used to derive

$$
\mathcal{D}_{x_i(r)} a(r') = \left(\partial_{x_i(r)} a(r) \right) \delta(r - r') - \nabla \cdot \left[\left(\partial_{\nabla x_i(r)} a(r) \right) \delta(r - r') \right],
\tag{60}
$$

by applying (59) to the functional $a(r') = \int a(r'') \delta(r'' - r') \, dv(r'')$. Both (59) and (60) will be used repeatedly in this section when reducing the model of the "Complete model: combination of reversible and irreversible dynamics" section to the weakly non-local case.

Choosing now $\varsigma = \theta$ as the thermal variable in (56), one obtains

$$
\left(\mathcal{D}_\varsigma E[\, \boldsymbol{x}^\varsigma \,] \right) * \left(\mathcal{D}_\varsigma \eta[\, \boldsymbol{x}^\varsigma \,] \right)^{-1} (\boldsymbol{r}) = \int \frac{\partial_{\theta(\boldsymbol{r}')} \varepsilon(\tilde{\boldsymbol{x}}^\theta(\boldsymbol{r}'))}{\partial_{\theta(\boldsymbol{r}')} \eta(\tilde{\boldsymbol{x}}^\theta(\boldsymbol{r}'))} \, \delta(\boldsymbol{r}' - \boldsymbol{r}) \, dv(\boldsymbol{r}')
$$

$$
= \frac{\partial_\theta \varepsilon(\tilde{\boldsymbol{x}}^\theta)}{\partial_\theta \eta(\tilde{\boldsymbol{x}}^\theta)} \, (\boldsymbol{r}) \, .
$$

(61)

Comparing this with $(51)_1$, one sees that (56) with $\varsigma = \theta$ and this latter relation agree when

$$
\theta = \frac{\partial_\theta \varepsilon(\tilde{\boldsymbol{x}}^\theta)}{\partial_\theta \eta(\tilde{\boldsymbol{x}}^\theta)}
$$

(62)

holds. Note that this condition is automatically satisfied if both ε and η are derived from a Helmholtz free energy density ψ, $(55)_1$, with $\eta = -\partial_\theta \psi(\tilde{\boldsymbol{x}}^\theta)$. In the sequel, it is assumed that ε and η indeed do originate from a Helmholtz free energy density ψ in the way just mentioned, i.e., that the consistency relation (62) is satisfied, and θ will be referred to as the absolute temperature in the sequel.

In the following, all model components summarized in the "Complete model: combination of reversible and irreversible dynamics" section are reduced to the weakly non-local case. While $(51)_1$ for the absolute temperature θ has already been dealt with above (in terms of (62)), one obtains for the coldness $(51)_2$, with a calculation analogous to (61),

$$
\vartheta = \theta^{-1} \, ,
$$

(63)

i.e., the coldness is the inverse of the absolute temperature. Furthermore, with the help of (59), one obtains for the functional derivatives (50)

$$
\begin{aligned}
\mathcal{D}_\chi E[\, \boldsymbol{x}^\eta \,] &= \delta_\chi \psi\left(\tilde{\boldsymbol{x}}^\theta\right) = -\nabla \cdot \partial_{\nabla \chi} \psi\left(\tilde{\boldsymbol{x}}^\theta\right) \, , \\
\mathcal{D}_\varrho S[\, \boldsymbol{x}^\varepsilon \,] &= \delta_\varrho \hat{\psi}\left(\tilde{\boldsymbol{x}}^\theta\right) \, , \\
\mathcal{D}_\phi S[\, \boldsymbol{x}^\varepsilon \,] &= \delta_\phi \hat{\psi}\left(\tilde{\boldsymbol{x}}^\theta\right) \, ,
\end{aligned}
$$

(64)

which is a realization of (54) with the densities of the Helmholtz free energy and the free entropy defined in (55). Using these reduced forms for the functional derivatives, the evolution equations specified in (47) are modified in terms of the reduced momentum balance for the weakly non-local case,

$$
\dot{\boldsymbol{m}} = -\mathcal{D}_\chi E[\, \boldsymbol{x}^\eta \,] = \nabla \cdot \partial_{\nabla \chi} \psi\left(\tilde{\boldsymbol{x}}^\theta\right) \, .
$$

(65)

The term in brackets is naturally identified as the stress tensor that agrees with earlier results (Hütter and Svendsen 2011; Gladkov et al. 2016). The driving forces for the irreversible dynamics, (48), assume the form

$$
\boldsymbol{f}_\varepsilon = \nabla \vartheta \, , \quad \boldsymbol{f}_\varrho = \nabla \delta_\varrho \hat{\psi}\left(\tilde{\boldsymbol{x}}^\theta\right) \, , \quad \boldsymbol{f}_\phi = \delta_\phi \hat{\psi}\left(\tilde{\boldsymbol{x}}^\theta\right) \, ,
$$

(66)

in agreement with the non-isothermal weakly non-local formulation developed in Gladkov et al. (2016). As evidenced by (65) and (66), the reversible dynamics and irreversible dynamics are driven by derivatives of the Helmholtz free energy density ψ and derivatives of the free entropy density $\hat{\psi}$, respectively (Mielke 2011).

Finally, according to (53), the evolution equation for the absolute temperature θ is given by

$$
\begin{aligned}
\left(\partial_\theta \varepsilon(\tilde{x}^\theta)\right) \dot{\theta} = {} & (\nabla \dot{\chi}) : \partial_{\nabla_\chi} \left(-\theta \eta(\tilde{x}^\theta)\right) \\
& -\nabla \cdot j_\varepsilon + \sum_{k=1}^m \left(d_{\varrho_k} \varepsilon(\tilde{x}^\theta)\right) \diamond \left(\nabla \cdot j_{\varrho_k}\right) - \sum_{l=1}^s \left(d_{\phi_l} \varepsilon(\tilde{x}^\theta)\right) \diamond j_{\phi_l},
\end{aligned}
\tag{67}
$$

where we have used $\partial_\eta \varepsilon(\tilde{x}^\eta) = \theta$, which follows from (62). Furthermore, the operator notation,

$$
(d_s a) \diamond b := (\partial_s a)\, b + (\partial_{\nabla_s} a) \cdot \nabla b,
\tag{68}
$$

has been introduced in terms of scalar s and b. The temperature evolution (67) is in complete agreement with the weakly non-local formulation developed in Gladkov et al. (2016).

Furthermore, (67) includes the irreversible contributions to the temperature evolution equation derived in Bollada et al. (2017) (see Eq. (111) therein), which is based on the bracket formalism (Beris and Edwards 1994).

Non-isothermal generalization of a phase-field crystal model for binary alloys

As a second example application of the current model formulation summarized in the "Complete model: combination of reversible and irreversible dynamics" section, the phase-field crystal (PFC) model for binary alloys from Elder et al. (2007) is generalized to non-isothermal conditions. Since deformation χ, momentum m, and non-conservative phase fields ϕ play no role in this model, they are neglected here. In this case, the reduced set

$$
x_{\text{PFC}}^\varsigma = (\varsigma, \varrho_A, \varrho_B)
\tag{69}
$$

of GENERIC variables and corresponding evolution relations

$$
\begin{aligned}
\dot{\varrho}_A &= -\nabla \cdot j_{\varrho_A}, \\
\dot{\varrho}_B &= -\nabla \cdot j_{\varrho_B}, \\
\dot{\varsigma} * \mathcal{D}_\varsigma \varepsilon[x^\varsigma] &= -\nabla \cdot j_\varepsilon + \left(\nabla \cdot j_{\varrho_A}\right) * \mathcal{D}_{\varrho_A} \varepsilon[x^\varsigma] + \left(\nabla \cdot j_{\varrho_B}\right) * \mathcal{D}_{\varrho_B} \varepsilon[x^\varsigma],
\end{aligned}
\tag{70}
$$

from (47) and (53), respectively, are relevant. Here, ϱ_A and ϱ_B represent the (number) densities of alloy species A and B, respectively. Consistent with the model formulation in Elder et al. (2007) based on a free energy functional, the current non-isothermal formulation is based on the choice

$$
\varsigma = T
\tag{71}
$$

in (69), adopting now the notation T for the absolute temperature. Likewise, the forms

$$
\begin{aligned}
\varepsilon\left[x_{\text{PFC}}^T\right] = {} & -\tfrac{1}{2} \left\{ \delta\varrho_A \left(\Gamma_{AA}^E * \delta\varrho_A\right) + \delta\varrho_B \left(\Gamma_{BB}^E * \delta\varrho_B\right) + 2\,\delta\varrho_A \left(\Gamma_{AB}^E * \delta\varrho_B\right) \right\}, \\
\eta\left[x_{\text{PFC}}^T\right] = {} & -k_{\text{B}} \left\{ \varrho_A \ln(\varrho_A/\varrho_{A,\ell}) - \delta\varrho_A + \varrho_B \ln(\varrho_B/\varrho_{B,\ell}) - \delta\varrho_B \right\} \\
& -\tfrac{1}{2} \left\{ \delta\varrho_A \left(\Gamma_{AA}^S * \delta\varrho_A\right) + \delta\varrho_B \left(\Gamma_{BB}^S * \delta\varrho_B\right) + 2\,\delta\varrho_A \left(\Gamma_{AB}^S * \delta\varrho_B\right) \right\},
\end{aligned}
\tag{72}
$$

for the internal energy, and entropy, density functionals, respectively, represent direct generalizations of the free energy density functional in Eq. (15) of Elder et al. (2007). Here, k_{B} is the Boltzmann constant, and $\delta\varrho_k = \varrho_k - \varrho_{k,\ell}$, where $\varrho_{k,\ell}$ denotes the liquidus density of the respective phase (Elder et al. 2007). In (72), $\Gamma_{kk'}^{E|S} = \Gamma_{kk'}^{E|S}(r, r')$ represent energetic and entropic two-point correlation kernels. For the special case

that T is constant in space and time, i.e., the case usually considered in phase-field crystal modeling, one recovers with $\int (\varepsilon - T\eta)\, dv$ the free energy used in Elder et al. (2007), with $\left(\Gamma_{kk'}^{E} - T\Gamma_{kk'}^{S} \right)/(k_\mathrm{B}T)$ equal to the two-point correlation kernels $C^{kk'}$ of Elder et al. (2007).

Note that the dependence of the internal energy and entropy density functionals in (72) on T is contained in the correlation kernels $\Gamma_{kk'}^{E|S}$. Once that dependence is known, the heat capacity (kernel) on the l.h.s. of $(70)_2$ and particularly the driving forces $(48)_{1,2}$, using $(50)_2$ and $(51)_2$, can be calculated. For the special case that the T-dependence in (72) is such that $\vartheta = 1/T$ by way of $(51)_2$, the simplified relation $(54)_2$ can be used for the driving forces, in terms of the free entropy.

The non-isothermal extension of phase-field crystal modeling is completed by specifying the relation between the fluxes $\boldsymbol{j}_\varepsilon$ and $\boldsymbol{j}_\varrho$ on the one hand and the forces $\boldsymbol{f}_\varepsilon$ and $\boldsymbol{f}_\varrho$ on the other hand. For example, one may choose $\boldsymbol{D}$ in $(31)_2$ to be diagonal, which includes the diffusive dynamics studied in Elder et al. (2007) for the densities ϱ_A and ϱ_B.

Summary and discussion

In this paper, the GENERIC approach (Grmela and Öttinger 1997; Öttinger and Grmela 1997; Öttinger 2005) has been used to formulate a class of spatially strongly non-local and non-isothermal thermodynamic models for multiphase, multicomponent solids, with conservative and non-conservative dynamics. Particularly, phase transitions as well as mass and heat transport can be described with these models. For the model formulation, the reversible and irreversible dynamics have been distinguished carefully. For the present class of models, the reversible dynamics is of a purely kinematic origin, supplemented by the key ingredient that the referential entropy density is constant in reversible dynamics. With respect to the formulation of the irreversible dynamics, the operator split (30) was employed in order to relate to the constitutive model formulation in terms of thermodynamic forces and fluxes (Edwards 1998). As shown in the last part of the work, the current strongly non-local model formulation reduces consistently to the weakly non-local one of Gladkov et al. (2016). As shown in that work, the weakly non-local model formulation is a direct non-isothermal generalization of the well-known phase-field models of Cahn and Hilliard (1958) for conservative dynamics and of Allen and Cahn (1979) for non-conservative dynamics in the spirit of Penrose and Fife (e.g., Penrose and Fife 1990, 1993). As well, the application of the current GENERIC-based approach to the non-isothermal generalization of a phase-field crystal model for binary alloys from Elder et al. (2007) demonstrates its potential as a framework for the formulation of strongly non-local models for long-range dynamics in chemically and structurally inhomogeneous materials and systems.

In view of the goal of formulating models that are spatially strongly non-local in nature, the weakly non-local relation (the "Decomposition of the friction operator" section) between the thermodynamic fluxes ($\boldsymbol{j}_\varepsilon$ and $\boldsymbol{j}_\varrho$) and the corresponding irreversible dynamics (of ε and $\boldsymbol{\varrho}$) may seem inappropriate. Since both ε and the components of $\boldsymbol{\varrho}$ are densities of conserved quantities, however, the weakly non-local ansatzes are justified if the corresponding densities at a certain position evolve by virtue of exchange with the immediate neighborhood. This gives rise to the spatial gradient operator ∇ in the model formulation. The following two generalizations of this ansatz can be incorporated in a straightforward manner in the treatment presented in this paper. The first

generalization concerns the exchange of internal energy over (finite) distance, i.e., by radiation and absorption, making a weakly non-local ansatz inappropriate. This could be incorporated by including in the friction matrix element $M_{\varepsilon\varepsilon}$ a strongly non-local contribution that satisfies the condition $(29)_1$ for the conservation of energy, with $M_{\varepsilon\varepsilon}$ being strongly non-local in the sense of not making use of any spatial derivative operator. The second generalization of the weakly non-local ansatz concerns the account of chemical reactions of the components of ϱ (e.g., Bazant 2013). While the thermodynamic potential generating the dynamics (in our case $S[x^{\varepsilon}]$) can be of a strongly non-local character, the relation between the derivatives of the thermodynamic potential ($\mathcal{D}_{\varrho}S[x^{\varepsilon}]$) and the rate of change of the species ϱ is usually assumed to be a local one. In this sense, the reader is referred to Öttinger and Grmela (1997) for the implementation of chemical reactions as an amendment to the model formulation in this paper.

The isothermal special case of the current GENERIC-based formulation is in some ways reminiscent of phase-field crystal (e.g., Elder and Grant 2004; Elder et al. 2007; Provatas and Elder 2010), which itself is based on the classic density functional theory (of freezing). In particular, all of these are based on spatial strong non-locality. Also relevant to the modeling of complex multiscale dynamics in general is temporal non-locality. This leads to models that use a memory kernel that does not decay infinitesimally rapidly. In many cases, e.g., in modeling the rheology of complex fluids, it has been found that the memory kernels can be avoided by including additional, microstructural dynamic variables in the description (Bird et al. 1987a, b). However, problems can arise if appropriate auxiliary dynamic variables can not be found or if the system does not obey a clear separation of characteristic time scales (e.g., Öttinger 2005, Chapter 6), i.e., if a clear separation into "slow" and "fast" dynamics is not possible. In these cases, explicit temporally truly non-local model formulations seem unavoidable. This is a topic beyond the scope of this paper and is therefore left as a subject to further research.

Endnotes

[1] Account of such non-locality in coarse-grained form is possible for example with the help of projection-operator methods (e.g., Öttinger 2005, Chapter 6), something well-beyond the scope of the current work.

[2] For notational simplicity, the time argument in $x(r, t)$ is suppressed in the notation in what follows.

[3] A number of concepts from the theory of functionals and their derivatives (see, e.g., Parr and Yang 1989; Davis 1996; Engel and Dreizler 2011 for more details) required for this purpose are briefly summarized in Appendix A: brief summary of concepts from the theory of functionals and illustrated in Appendix B: functional derivatives based on delta function.

[4] In this work, the short-hand notation $\mathcal{D}_{x_i}A$ is used to denote the partial functional derivative of A with respect to x_i in the set of variables x, i.e., keeping all other "components" $x \backslash x_i$ of x fixed. When necessary for clarity (e.g., to emphasize which variables $x \backslash x_i$ are being kept fixed), the more explicit notation $\mathcal{D}_{x_i}A[x]$ for this derivative will also be employed.

[5] Recall that $\mathcal{D}_x E \cdot \delta x$ represents an energy *density* (see Appendix A: brief summary of concepts from the theory of functionals). In particular, then, $\mathcal{D}_{\eta}E$ has units of energy density divided by entropy density, i.e., temperature.

[6] This implies a weakly non-local connection of fluxes to irreversible dynamics. Possible generalizations of that are discussed in the "Summary and discussion" section.

[7] In this work, the same symbol "δ" signifies two things: (i) the variation δf of any field f and (ii) the spatial Dirac delta function $\delta(\boldsymbol{r})$. From the context, it should be clear which is intended.

Appendix

Appendix A: brief summary of concepts from the theory of functionals

The following synopsis is based on the treatment of Engel and Dreizler (2011), Appendix A. As the name implies, a functional A maps one or more functions or fields $\boldsymbol{x}(\boldsymbol{r})$ to the value $A[\boldsymbol{x}]$ of A. This value could be a number or, more generally, a function or field. Following common practice, both A and its value $A[\boldsymbol{x}]$ are denoted by the notation $A[\boldsymbol{x}]$ in what follows. If $A[\boldsymbol{x}]$ is a functional, then $A(\epsilon) := A[\boldsymbol{x} + \epsilon \boldsymbol{v}]$ is a function of ϵ for any fields $\boldsymbol{x}, \boldsymbol{v}$. Assuming sufficient differentiability, the Taylor series expansion

$$A[\boldsymbol{x} + \epsilon \boldsymbol{v}] = A[\boldsymbol{x}] + \sum_{k=1}^{n} \frac{\epsilon^k}{k!} D_\epsilon^k A[\boldsymbol{x} + \epsilon \boldsymbol{v}]\,|_{\epsilon=0} + o\left(\epsilon^{n+1}\right) \tag{73}$$

about $\epsilon = 0$ makes sense. The kth-order functional derivative $\mathcal{D}_{\boldsymbol{x}_1} \otimes \cdots \otimes \mathcal{D}_{\boldsymbol{x}_k} A$ of A is a multifield of order k defined via

$$\langle \mathcal{D}_{\boldsymbol{x}_1} \otimes \cdots \otimes \mathcal{D}_{\boldsymbol{x}_k} A, \boldsymbol{v}_1 \otimes \cdots \otimes \boldsymbol{v}_k \rangle := D_\epsilon^k A[\boldsymbol{x} + \epsilon \boldsymbol{v}]\,|_{\epsilon=0}\,, \tag{74}$$

with

$$\langle \mathcal{D}_{\boldsymbol{x}_1} \otimes \cdots \otimes \mathcal{D}_{\boldsymbol{x}_k} A, \boldsymbol{v}_1 \otimes \cdots \otimes \boldsymbol{v}_k \rangle$$
$$:= \int dv_1\, \boldsymbol{v}_1 \otimes \cdots \otimes \int dv_k\, \boldsymbol{v}_k \cdot \mathcal{D}_{\boldsymbol{x}_1} \otimes \cdots \otimes \mathcal{D}_{\boldsymbol{x}_k} A\,, \tag{75}$$

and

$$f_k := f(\boldsymbol{r}_k)\,, \quad \int f_k\, dv_k := \int f(\boldsymbol{r}_k)\, dv(\boldsymbol{r}_k)\,. \tag{76}$$

Interpreting $\boldsymbol{v}$ in (74) as a variation[7] $\delta \boldsymbol{x}$ of $\boldsymbol{x}$, let

$$\delta^k A[\boldsymbol{x}] := \langle \mathcal{D}_{\boldsymbol{x}_1} \otimes \cdots \otimes \mathcal{D}_{\boldsymbol{x}_k} A, \delta \boldsymbol{x}_1 \otimes \cdots \otimes \delta \boldsymbol{x}_k \rangle \tag{77}$$

represent the kth-order variation of $A[\boldsymbol{x}]$ induced by $\delta \boldsymbol{x}$. A particular kind of variation of the field $\boldsymbol{x}$ arises of course when these are time-dependent. For example, the induced variation

$$\dot{A} = \langle \mathcal{D}_{\boldsymbol{x}} A, \dot{\boldsymbol{x}} \rangle \tag{78}$$

of any $A = A[\boldsymbol{x}]$ with respect to $\dot{\boldsymbol{x}}$ represents its time derivative in agreement with (5). Finally, let a represents the volume density of A, i.e.,

$$A = \int a(\boldsymbol{r})\, dv(\boldsymbol{r})\,. \tag{79}$$

By definition, then, a has units of A per unit volume. Clearly, for A to be finite, $a(\boldsymbol{r})$ must be bounded, i.e., reduce to zero as $|\boldsymbol{r}| \to \infty$. In general, a itself is a functional. Via the chain rule for functional differentiation (e.g., Engel and Dreizler 2011, (A.38)), we have

$$\mathcal{D}_{\boldsymbol{x}(\boldsymbol{r})} A = \int \mathcal{D}_{a(\boldsymbol{r}')} A\, \mathcal{D}_{\boldsymbol{x}(\boldsymbol{r})} a(\boldsymbol{r}')\, dv(\boldsymbol{r}') = \int \mathcal{D}_{\boldsymbol{x}(\boldsymbol{r})} a(\boldsymbol{r}')\, dv(\boldsymbol{r}') \tag{80}$$

via the linearity of A in a.

Appendix B: functional derivatives based on delta function

Consider the specific choice

$$\boldsymbol{v}(\boldsymbol{r}') = \delta_{\boldsymbol{r}}(\boldsymbol{r}')\,\boldsymbol{e}_i \tag{81}$$

for $\boldsymbol{v}$ in (74) based on the delta function $\delta_{\boldsymbol{r}}(\boldsymbol{r}') := \delta(\boldsymbol{r}' - \boldsymbol{r})$ at fixed $\boldsymbol{r}$. Here,

$$\boldsymbol{e}_i = (\delta_{i1}, \ldots, \delta_{ij}, \ldots, \delta_{ig}) \tag{82}$$

is the Cartesian "basis vector" in space of the GENERIC variables. Then,

$$D_{\epsilon}^1 A[\,\boldsymbol{x} + \epsilon\,\boldsymbol{v}\,]\,|_{\epsilon=0} = \int dv'\,\boldsymbol{v}' \cdot \mathcal{D}_{\boldsymbol{x}'} A = \int dv'\,\delta_{\boldsymbol{r}}'\,\boldsymbol{e}_i \cdot \mathcal{D}_{\boldsymbol{x}'} A = \mathcal{D}_{x_i(\boldsymbol{r})} A \tag{83}$$

follows from (74) for the first-order functional derivative of A with respect to $x_i := \boldsymbol{e}_i \cdot \boldsymbol{x}$. This δ-function-based form of the functional derivative is common in physics (e.g., Parr and Yang 1989; Davis 1996).

As an illustrative example, consider a functional of the form

$$A[\,\theta, \boldsymbol{\chi}\,] = \int a(\theta, \nabla\boldsymbol{\chi})\,dv, \tag{84}$$

with a a *function* of θ and $\nabla\boldsymbol{\chi}$ explicitly independent of $\boldsymbol{r}$. In the context of (83), we have

$$\mathcal{D}_{\theta(\boldsymbol{r})} A = \int \frac{\partial a(\theta', \nabla'\boldsymbol{\chi}')}{\partial \theta'}\,\delta_{\boldsymbol{r}}'\,dv' = \frac{\partial a(\theta(\boldsymbol{r}), \nabla\boldsymbol{\chi}(\boldsymbol{r}))}{\partial \theta(\boldsymbol{r})}\,. \tag{85}$$

Recall that a and its derivatives are implicit fields via the field arguments of a. Analogously,

$$\begin{aligned}
\mathcal{D}_{\chi_k(\boldsymbol{r})} A &= \int \frac{\partial a(\theta', \nabla'\boldsymbol{\chi}')}{\partial \nabla'\chi_k'} \cdot \nabla'\delta_{\boldsymbol{r}}'\,dv' \\
&= \int \nabla' \cdot \delta_{\boldsymbol{r}}' \frac{\partial a(\theta', \nabla'\boldsymbol{\chi}')}{\partial \nabla'\chi_k'}\,dv' - \int \delta_{\boldsymbol{r}}'\,\nabla' \cdot \frac{\partial a(\theta', \nabla'\boldsymbol{\chi}')}{\partial \nabla'\chi_k'}\,dv' \\
&= -\nabla \cdot \frac{\partial a(\theta(\boldsymbol{r}), \nabla\boldsymbol{\chi}(\boldsymbol{r}))}{\partial \nabla\chi_k(\boldsymbol{r})}\,.
\end{aligned} \tag{86}$$

This specific example, (84), is relevant for the reduction of the strongly non-local model formulation to the weakly non-local case in the "Special case: weakly non-local model formulation" section.

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