Unconditionally gradient-stable computational schemes in problems of fast phase transitions

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(Received 24 September 2010; published 14 February 2011)

Equations of fast phase transitions, in which the phase boundaries move with velocities comparable with the atomic diffusion speed or with the speed of local structural relaxation, are analyzed. These equations have a singular perturbation due to the second derivative of the order parameter with respect to time, which appears due to phenomenologically introduced local nonequilibrium. To develop unconditionally stable computational schemes, the Eyre theorem [D. J. Eyre, unpublished] proved for the classical equations, based on hypotheses of local equilibrium, is used. An extension of the Eyre theorem for the case of equations for fast phase transitions is given. It is shown that the expansion of the free energy on contractive and expansive parts, suggested by Eyre for the classical equations of Cahn-Hilliard and Allen-Cahn, is also true for the equations of fast phase transitions. Grid approximations of these equations lead to gradient-stable algorithms with an arbitrary time step for numerical modeling, ensuring monotonic nonincrease of the free energy. Special examples demonstrating the extended Eyre theorem for fast phase transitions are considered.

DOI: 10.1103/PhysRevE.83.026705 PACS number(s): 02.70.−c, 05.70.Fh, 05.70.Ln, 64.60.−i

I. INTRODUCTION

Freezing of a system into the disordered state leads to relaxation of frozen degrees of freedom during some characteristic time [1]. Sizes of ordered regions of new phases, forming under degeneration of the initial disordered state (symmetrical phase), change with the velocity controlled by thermodynamic driving forces. A difference in growth velocities leads to competition of various phases with broken symmetry and to the appearance of one of the ordered states. In accordance with this scheme, equations for phase transitions of local equilibrium systems are described by partial differential equations that are of first order with respect to time. Well-known equations, based on the hypothesis of local equilibrium, are the equations of Allen and Cahn (a model of antiphase domain motion [2]), Cahn and Hilliard (a model of spinodal decomposition [3]), Swift and Hohenberg (a model for hydrodynamic instability [4]), and Elder and Grant (a model of phase-field crystals [5,6]).

Numerical modeling of equations for phase transitions is an important tool for the investigation of pattern formation evolving in real systems and materials under nonequilibrium conditions [6,7]. Usually, existing computational algorithms are inefficient due to their limitation by the time of numeric modeling of phase transitions with small numeric time step. For example, numeric "checkerboard instability" appears in the integration of the Cahn-Hilliard equation by the Euler method due to chosen time step for a length scale larger than some characteristic distance δx [8]. This leads to a fixed time step δt, which limits computation of a whole stage of the system dynamics. Therefore, for usual numeric schemes and algorithms following straightforwardly from them, there is an essential restriction of an opportunity of modeling, because numerical modeling with constant and limited time step is superfluously exact and demands excessive computer memory. In an ideal situation, a stable algorithm of integration is necessary which defines the time step from the requirement of accuracy, instead of restriction on the stability of the numerical algorithm. Rather recently, in this direction a certain progress has been reached with the development of the gradient-stable methods [9–13]. These methods were applied to the phase transformations, the description of which rests on the local equilibrium hypothesis. This description is well justified for systems not too far from equilibrium, that is, for infinitesimal deviation from local thermodynamic equilibrium [14]. It can be applied essentially to phase transitions having a linear response against small perturbation.

For many classes of materials—such as non-Newtonian fluids, viscoelastic bodies, rapidly solidifying alloys and systems, materials deeply quenched into the spinodal region, and, more generally, systems with memory—the hypothesis of local equilibrium does not apply (for an overview, see literature in Ref. [15]). This hypothesis also fails for fast phase transformations and during transient periods from an unstable state to a metastable or even stable state [16,17]. Therefore, a wide class of materials and processes cannot be coherently described by the formalism based on the hypothesis of local equilibrium, which is avoided by the introduction of fast independent thermodynamic variables such as thermodynamic fluxes or some internal variables [18,19].

The use of methods of extended irreversible thermodynamics [19] leads to singular perturbation in equations of fast phase transitions due to the appearance of the second derivative of the order parameter with respect to time [16,17]. Complex-valued dispersion relations for equations of phase transitions [20,21] testify that the systems described by these equations may have a behavior associated with transition from an oscillatory regime to a regime with monotonic relaxation [21,22]. This behavior adds complexity to numerical modeling of problems

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of fast phase transitions due to the instability of the known algorithms. In this sense, because the success of gradient-stable methods [11–13] in a modeling of the local equilibrium tasks is based on the Eyre theorem [9–11], the main goal of this paper is to extend the Eyre theorem for description of fast phase transitions under local nonequilibrium conditions.

The paper is organized as follows. In Sec. II, equations for fast phase transitions are analyzed. Using the Eyre theorem, Sec. III presents main results of the work in the form of gradient-stable numeric schemes for systems undergoing fast phase transitions. Section IV illustrates the Eyre theorem in application to the problems of fast transitions with conserved and nonconserved order parameters. In Sec. V, the condition of nonincrease of the free energy for developed numeric schemes is proved. Section VI briefly summarizes the main results of the present work. Finally, the Appendix gives inequalities necessary for proofs of the work.

II. EQUATIONS FOR SYSTEMS UNDERGOING FAST PHASE TRANSITIONS

Extension of equations of Cahn-Hilliard (CH) equation and Allen-Cahn (AC) equation as well as the phase-field crystals (PFC) equation and Swift-Hohenberg (SH) equation to the case of fast phase transitions leads to the extension of a set of independent thermodynamic variables due to introducing dynamical fluxes (for the conserved order parameter) and the rate of change of the order parameter (for the nonconserved order parameter) [16,17,21].

Assuming isothermal approximation, first, the relaxation dynamics is defined by the free energy density, which is chosen as the sum of the local equilibrium contribution $f(\varphi)$ and gradient terms. As a result, the local equilibrium free energy is defined by the functional of the Ginzburg-Landau form [23,24] for AC and CH equations,

$$F_1[\varphi] = \int_V \left[\frac{1}{2}\varepsilon^2(\vec{\nabla}\varphi)^2 + f(\varphi)\right] dV, \tag{1}$$

and it is defined by the functional of the Brazovskii and Swift-Hohenberg form [4,25] for SH and PFC equations,

$$F_2[\varphi] = \int_V \left[\frac{1}{2}\varepsilon_1^2(\nabla^2\varphi)^2 - \frac{1}{2}\varepsilon_2^2(\vec{\nabla}\varphi)^2 + f(\varphi)\right] dV. \tag{2}$$

Second, extension of the space of thermodynamical variables to the case of phase transitions leads to the pure nonequilibrium contributions to the free energy in the following form [17]: for AC and SH equations (with nonconserved order parameter),

$$F_n[u] = \frac{1}{2}\alpha_n \int_V u^2 dV, \quad \alpha_n > 0, \tag{3}$$

where u is the rate of change of the order parameter given by

$$\dot{\varphi} = u, \tag{4}$$

and for CH and PFC equations (with conserved order parameter),

$$F_c[\vec{J}] = \frac{1}{2}\alpha_c \int_V \vec{J}\cdot\vec{J} dV, \quad \alpha_c > 0, \tag{5}$$

where $\vec{J}$ is the dynamical flux which guarantees the balance law

$$\dot{\varphi} + \vec{\nabla}\cdot\vec{J} = 0. \tag{6}$$

Here (and in the text that follows), a dot over the functions denotes, as usual, the partial derivative $\partial/\partial t$ with respect to time t.

Nonequilibrium contributions (3) and (5) have a meaning of kinetic energy as has been shown in examples of spinodal decomposition [26]. The kinetic coefficients α_n and α_c in Eqs. (3) and (5) are assumed to be positive, because the local nonequilibrium contribution should lead to an increase of the free energy and limit a range of accessible states in a phase space of the system [20]. Combining Eqs. (1) and (2) with Eqs. (3) and (5) results in four functionals which decrease monotonically during relaxation of the system in time.

A. Dynamics of nonconserved order parameter

Consider the dynamics with a nonconserved variable in a system with volume V and outer surface S. After differentiation of the free energy $F = F_1[\varphi] + F_n[u]$ with respect to time and using Eqs. (1), (3), and (4), one obtains

$$\frac{d}{dt}F = \int_V u\left[\alpha_n\dot{u} + \frac{\delta F_1}{\delta\varphi}\right] dV + \varepsilon^2 \oint_S u(\vec{\nabla}\varphi)\cdot(d\vec{S}) \leqslant 0, \tag{7}$$

where $d\vec{S}$ is the vectorial element of the surface S. To exclude free energy exchange on S which is described by the second integral in Eq. (7), we consider an isolated system with the following natural boundary condition:

$$\nabla_n\varphi|_S = 0, \tag{8}$$

where ∇_n is the projection of the nabla operator $\vec{\nabla}$ on the vector $\vec{n}$ normal to the outer surface S. Then a monotonic decrease of F is guaranteed in the simplest case by the following choice:

$$\alpha_n\dot{u} + \frac{\delta F_1}{\delta\varphi} = -\frac{1}{M_n}u, \tag{9}$$

where $M_n(\varphi) > 0$ is the mobility for the nonconserved order parameter. Equations (9) and (4) lead to the following system of equations:

$$\begin{cases} \tau_n\dot{u} + u = -M_n\dfrac{\delta F_1}{\delta\varphi}, \\ \dot{\varphi} = u, \end{cases} \tag{10}$$

where the coefficient

$$\tau_n = \alpha_n M_n \tag{11}$$

is the relaxation time of the rate $\dot{\varphi} = u$ of change of the order parameter φ. For many materials, the time τ_n has a small value (for metals, $\tau_n \approx 10^{-9}$–10^{-12} s); therefore, we assume $\tau_n \equiv$ const, although the mobility M_n may depend on the order parameter φ. The obtained system of equations (10) is equivalent to the single equation of second order in time

(which is the hyperbolic AC equation) [17]:

$$\tau_n\ddot{\varphi}+\dot{\varphi}=-M_n\frac{\delta F_1}{\delta\varphi} \equiv M_n\left(\varepsilon^2\nabla^2\varphi-\frac{\partial f(\varphi)}{\partial\varphi}\right). \quad (12)$$

Substituting $F_1\to F_2$ into Eq. (12) leads to the hyperbolic SH equation [21]

$$\tau_n\ddot{\varphi}+\dot{\varphi}=-M_n\frac{\delta F_2}{\delta\varphi} \equiv -M_n\left(\varepsilon_1^2\Delta^2\varphi+\varepsilon_2^2\Delta\varphi+\frac{\partial f(\varphi)}{\partial\varphi}\right). \quad (13)$$

For the isolated system, described by the hyperbolic SH equation (13), we assume again an absence of the free energy transfer through the outer surface S. This leads to

$$\oint_S\left[\varepsilon_1^2(\Delta\varphi\vec{\nabla}u-u\vec{\nabla}(\Delta\varphi))-\varepsilon_2^2u\vec{\nabla}\varphi\right]\cdot d\vec{S}=0, \quad (14)$$

with $u=\dot{\varphi}$. Obviously, Eq. (14) holds if

$$\nabla_n\varphi|_S=0 \quad \text{and} \quad \nabla_n(\Delta\varphi)|_S=0. \quad (15)$$

B. Dynamics of conserved order parameter

For the conserved variable with the balance law (6), differentiation of the free energy $F=F_1[\varphi]+F_c[\vec{J}]$ with respect to time leads to the following expression:

$$\frac{d}{dt}F=\int_V\vec{J}\cdot\left[\alpha_c\dot{\vec{J}}+\vec{\nabla}\left(\frac{\delta F_1}{\delta\varphi}\right)\right]dV +\varepsilon^2\oint_S\dot{\varphi}(\vec{\nabla}\varphi)\cdot(d\vec{S})-\oint_S\left(\frac{\delta F_1}{\delta\varphi}\right)\vec{J}\cdot d\vec{S}\leqslant 0. \quad (16)$$

The natural boundary conditions for Eq. (16) are

$$\nabla_n\varphi|_S=0 \quad \text{and} \quad J_n|_S=0, \quad (17)$$

where J_n is the projection of the vector $\vec{J}$ on the vector $\vec{n}$ normal to the outer surface S. Taking conditions (17) into account (monotonic nonincrease of the free energy F), Eq. (16) is guaranteed by the linear relation between the flux and conjugated thermodynamic force:

$$\alpha_c\dot{\vec{J}}+\vec{\nabla}\left(\frac{\delta F_1}{\delta\varphi}\right)=-\frac{1}{M_c}\vec{J}, \quad (18)$$

In Eq. (18), the driving force for flux $\vec{J}$ consists of a contribution from the gradient of $\delta F_1/\delta\varphi$ and from the time relaxation $\propto\dot{\vec{J}}$. Note, especially, that nonincreasing of the free energy in isolated systems occurs with the positive value of mobility, $M_c(\varphi)>0$, which may depend on the spatial distribution of φ. Introducing the relaxation time

$$\tau_c=\alpha_cM_c \quad (19)$$

of the atomic flux $\vec{J}$ to a steady state, assuming it is constant [independently of choice of $M_c(\varphi)$], and using Eq. (18), one can obtain the following system of equations:

$$\begin{cases}\tau_c\dot{\vec{J}}+\vec{J}=-M_c\vec{\nabla}\left(\dfrac{\delta F_1}{\delta\varphi}\right),\\ \dot{\varphi}+\vec{\nabla}\cdot\vec{J}=0.\end{cases} \quad (20)$$

Because the second of Eqs. (20) has only $\vec{\nabla}\cdot\vec{J}$, and defining

$$\vec{\nabla}\cdot\vec{J}\equiv u, \quad (21)$$

one can obtain the following system of equations:

$$\begin{cases}\tau_c\dot{u}+u=-\vec{\nabla}\cdot\left[M_c\vec{\nabla}\left(\dfrac{\delta F_1}{\delta\varphi}\right)\right],\\ \dot{\varphi}+u=0.\end{cases} \quad (22)$$

Equation (22) shows that, due to the identity $\vec{\nabla}\cdot(\vec{\nabla}\times)=\mathrm{div}(\mathrm{rot})=0$, the curl component of the vector field $\vec{J}$ does not influence the dynamics of the order parameter. Thus, the physics of the process is defined by the divergence of fluxes completely. The fluxes themselves just have a role of additional variables. In this sense, the system of equations in its form (22) is preferred to system (20) because it does not require obtaining vector fields and it represents two equations equivalent to the hyperbolic CH equation [16,17]:

$$\tau_c\ddot{\varphi}+\dot{\varphi}=\vec{\nabla}\cdot\left[M_c\vec{\nabla}\left(\frac{\delta F_1}{\delta\varphi}\right)\right] \equiv\vec{\nabla}\cdot\left[M_c\vec{\nabla}\left(\frac{\partial f(\varphi)}{\partial\varphi}-\varepsilon^2\nabla^2\varphi\right)\right]. \quad (23)$$

Finally, substituting $F_1\to F_2$ into Eq. (23) leads to the hyperbolic PFC equation

$$\tau_c\ddot{\varphi}+\dot{\varphi}=\vec{\nabla}\cdot\left[M_c\vec{\nabla}\left(\frac{\delta F_2}{\delta\varphi}\right)\right] \equiv\vec{\nabla}\cdot\left[M_c\vec{\nabla}\left(\varepsilon_1^2\Delta^2\varphi+\varepsilon_2^2\Delta\varphi+\frac{\partial f(\varphi)}{\partial\varphi}\right)\right]. \quad (24)$$

Physically reasonable argumentation for Eq. (24) originates from the model of fast transitions [17,21] and also from the self-consistent incorporation of both fast elastic relaxation and multiple crystal orientations in the behavior of nanocrystals [27].

In addition to condition (14), the surface contribution from the fluxes appears:

$$\oint_S\frac{\delta F_2}{\delta\varphi}\vec{J}\cdot d\vec{S} \quad (25)$$

for Eq. (24). For the isolated system, the surface contribution (25) disappears if the flux through the outer surface is absent:

$$J_n|_S=0. \quad (26)$$

III. GRADIENT STABILITY IN THE DYNAMICS OF FAST TRANSITIONS

The theorem of Eyre [9–11] defines conditions at which the unconditionally stable numeric computations for the parabolic AC and CH equations are guaranteed. These conditions were also used in numerical solutions of the PFC equation [13,28] and in various numerical experiments described by the AC equation [29].

The main quantity of the Eyre theorem is the Hessian matrix

$$H_{ij} = \frac{\partial^2 F[\varphi]}{\partial\varphi_i \partial\varphi_j}, \tag{27}$$

where $F[\varphi]$ is the free energy that depends on the order parameter, and φ_i represents the field of the order parameter on the ith grid node (for simplicity, only a one-component scalar field is considered). For the free energy used to describe pattern formation [see Eqs. (1), (2), (3), and (5)], the Hessian matrix (27) has both positive and negative eigenvalues.

Eyre found a numeric scheme of first order in time for the AC and SH parabolic equations by separating the free energy $F[\varphi]$ into a *contractive* part $F_C[\varphi]$ and an *expansive* part $F_E[\varphi]$ [9,10]:

$$F[\varphi] = F_C[\varphi] + F_E[\varphi]. \tag{28}$$

In Eq. (28), the contractive part $F_C[\varphi]$ is a convex function; that is, for any interval $[\varphi_a,\varphi_b]$ and $\forall\varphi \in [\varphi_a,\varphi_b]$ the value of the function $F_C[\varphi]$ is bounded above by the linear function

$$F_C[\varphi] \leqslant \frac{\varphi-\varphi_a}{\varphi_b-\varphi_a} F_C[\varphi_b] + \frac{\varphi_b-\varphi}{\varphi_b-\varphi_a} F_C[\varphi_a],$$

having the boundary points φ_a and φ_b of the interval. Analogously, the expansive part $F_E[\varphi]$ in Eq. (28) is a concave function; that is, $\forall\varphi \in [\varphi_a,\varphi_b]$ the value of φ is bounded from below by the value of the linear function

$$F_E[\varphi] \geqslant \frac{\varphi-\varphi_a}{\varphi_b-\varphi_a} F_E[\varphi_b] + \frac{\varphi_b-\varphi}{\varphi_b-\varphi_a} F_E[\varphi_a].$$

In this case, the eigenvalues λ^C of the Hessian H^C_{ij} for the convex function $F_C[\varphi]$ are strictly not negative, but the eigenvalues λ^E of the Hessian H^E_{ij} for the concave function $F_E[\varphi]$ are strictly not positive for every configuration of the φ field.

Now we formulate the main results which present spectral relations, obtained on the basis of the Eyre theorem due to expansion (28), which hold for the local nonequilibrium system described by Eqs. (22), (23), (13), and (24) for the fast phase transitions. It allows the construction of computational algorithms which are gradient stable.

Note one important feature for the Hessian (27) in application to the considered case of a local nonequilibrium system. Due to the quadratic structure of expressions for the local nonequilibrium contributions (3) and (5) into the free energy, Hessian (27) has a block-diagonal structure by the flux $\vec{J}$ (or by the rate of change $\dot{\varphi}$ for the nonconserved order parameter) and by the order parameter φ. In other words, the Hessian has the following form:

$$\begin{pmatrix} \alpha_{c(n)} & 0 \\ 0 & H_{ij}(\varphi) \end{pmatrix}. \tag{29}$$

Therefore, in what follows, we save the notation H_{ij} for the contribution from the order parameter into Hessian, explicitly writing respective contribution from the flux $\vec{J}$ (or from the rate of change $\dot{\varphi}$ for the nonconserved order parameter).

Let $\lambda_{\min} < 0$ represent a lower boundary for eigenvalues of the matrix H_{ij} for any field φ. This boundary exists due to the Eyre theorem [9] and the quadratic structure of nonequilibrium contributions (3) and (5). Furthermore, let $\lambda^E_{\max} \leqslant 0$ be the upper boundary for eigenvalues of the Hessian H^E_{ij}. The first main result consists in that if the condition

$$\lambda^E_{\max} \leqslant \tfrac{1}{2}\lambda_{\min} \tag{30}$$

holds, then the difference scheme for the nonconserved order parameter may be written as

$$\begin{cases} \varphi^{(k+1)} + \dfrac{M_n \delta t^2}{\delta t + 2\tau_n} \dfrac{\delta F_C}{\delta\varphi}\Big|^{(k+1)} = \varphi^{(k)} - \dfrac{M_n \delta t^2}{\delta t + 2\tau_n} \dfrac{\delta F_E}{\delta\varphi}\Big|^{(k)} + \dfrac{2\tau_n}{\delta t + 2\tau_n} u^{(k)}, \\ u^{(k+1)} = -u^{(k)} + \dfrac{2}{\delta t}\left(\varphi^{(k+1)} - \varphi^{(k)}\right). \end{cases} \tag{31}$$

Assuming that nonconstant mobility $M_n = M_n(\varphi(x))$ should be taken from the previous time step, $M_n \equiv M_n^{(k)}$, then the difference scheme (31) leads to a monotonic nonincrease of the free energy in time:

$$F[\varphi^{(k+1)},u^{(k+1)}] \leqslant F[\varphi^{(k)},u^{(k)}]. \tag{32}$$

In the limit $\tau_n \to 0$, the rate of the order parameter splits and the difference scheme (31) transforms into Eq. (A3) of Ref. [11] obtained for the parabolic AC equation.

Analogously, the second main result for the conserved order parameter consists of the formulation of the following numerical scheme:

$$\begin{cases} \varphi^{(k+1)} - \dfrac{\delta t^2}{\delta t + 2\tau_c} \vec{\nabla}\cdot\left[M_c \vec{\nabla}\left(\dfrac{\delta F_C}{\delta\varphi}\right)\right]\Big|^{(k+1)} = \varphi^{(k)} + \dfrac{\delta t^2}{\delta t + 2\tau_c} \vec{\nabla}\cdot\left[M_c \vec{\nabla}\left(\dfrac{\delta F_E}{\delta\varphi}\right)\right]\Big|^{(k)} - \dfrac{2\tau_c}{\delta t + 2\tau_c} u^{(k)}, \\ u^{(k+1)} = -u^{(k)} - \dfrac{2}{\delta t}\left(\varphi^{(k+1)} - \varphi^{(k)}\right), \\ \Delta\vec{J}^{(k+1)} = \vec{\nabla}\cdot u^{(k+1)}, \end{cases} \tag{33}$$

where the mobility has to be taken from the previous numerical time step: $M_c \equiv M_c^{(k)}$. With condition (30), numerical scheme (33) leads to a monotonic nonincrease of the free energy in time for the system with conserved order parameter:

$$F[\varphi^{(k+1)}, \vec{J}^{(k+1)}] \leqslant F[\varphi^{(k)}, \vec{J}^{(k)}]. \tag{34}$$

Within the limit $\tau_c \to 0$, Eqs. (33) simply transform into Eq. (A4) of Ref. [11], which is obtained for the parabolic CH equation. As in the case of the nonconserved order parameter, the limit $\tau_c \to 0$ splits equations of the system (33) in two independent expressions in such a way that the nonstationarity on the flux does not play a role in the dynamics of the order parameter.

Equations (31) and (33) are true for every configuration of the $\varphi^{(k+1)}$ field and corresponding fast variables (u and $\vec{J}$) as well as for any time step $\delta t > 0$. The convexity of F_C guarantees that the implicit equation for $\varphi^{(k+1)}$ has a unique solution. Note that spatial indices are omitted in Eqs. (31) and (33) and temporal values of fields are denoted as $\varphi^{(k+1)} = \varphi(t + \delta t)$ and $\varphi^{(k)} = \varphi(t)$.

The feature of energy dissipation in Eqs. (32) and (34) (together with other natural requirements, e.g., with the requirement of positive definite F) is referred to by Eyre as *gradient stability* [9]. As far as the gradient stability can be obtained for many algorithms (also for an explicit Euler scheme with a small enough time step δt), algorithms, defined by Eqs. (31) and (33), guarantee stability for *arbitrarily large* values of δt limited only by the accuracy requirement.

A useful consequence of the Eyre theorem lies in the fact that, if the condition on eigenvalues (30) is held for some limited set of fields φ, then the monotonic behavior in Eqs. (32) is always held for any values of δt. This can be useful when the order parameter φ is physically limited by its own dynamics. For example, it is true for a concentration field or a phase field when φ is the field with $\varphi_i^2 < \varphi_0^2$ for any i and some constant φ_0. In particular, it should be noted that separation of the free energy into the convex part F_C and concave part F_E, leading to numeric schemes (31) and (33), can be a complicated task and is not uniquely defined.

Proof of the nonincreasing free energy in Eqs. (32) and (34) for schemes (31) and (33) is given in Sec. V. Before this proof, two examples of the application of the Eyre theorem to the problems of fast transitions with conserved and nonconserved order parameters are presented.

IV. APPLICATION OF THE EYRE THEOREM TO FAST PHASE TRANSITIONS

A. Example for hyperbolic AC and CH equations

To illustrate the presently extended Eyre theorem, we choose an equilibrium part of the free energy in a form of double-well potential

$$f(\varphi) = \tfrac{1}{4}(\varphi^2 - 1)^2, \tag{35}$$

which has equilibrium values $\varphi = \pm 1$. Dimensionless Eq. (12) for the nonconserved order parameter is given by

$$\tau_n \ddot{\varphi} + \dot{\varphi} = \Delta\varphi - \varphi^3 + \varphi, \tag{36}$$

and dimensionless Eq. (23) for the conserved order parameter is presented as

$$\tau_c \ddot{\varphi} + \dot{\varphi} = \Delta(\varphi^3 - \varphi - \Delta\varphi). \tag{37}$$

The total free energy, dependent only on the order parameter

$$F[\varphi] = \int_V \left[\frac{1}{2}(\vec{\nabla}\varphi)^2 + \frac{1}{4}(\varphi^2 - 1)^2\right] dV, \tag{38}$$

can be treated as a sum of three contributions (with accuracy up to additive constant):

$$F[\varphi] = F_1 + F_2 + F_3,$$

where

$$\begin{cases} F_1 = -\frac{1}{2}\int \varphi^2\, dV, \\ F_2 = \frac{1}{2}\int (\vec{\nabla}\varphi)^2\, dV, \\ F_3 = \frac{1}{4}\int \varphi^4\, dV. \end{cases}$$

For each of these contributions, grid Hessians (27) are equal to

$$(M_1)_{ij} = -\delta_{ij}, \quad (M_2)_{ij} = -\Delta_{ij}, \quad (M_3)_{ij} = 3\varphi_i^2\delta_{ij},$$

where δ_{ij} is Kronecker's delta and Δ_{ij} is the grid Laplace operator. For estimation of the lower-bound eigenvalues of the whole Hessian we take into account the nonpositive spectrum of the grid Laplace operator with homogeneous boundary conditions of second order,

$$\Delta_{ij} f_j = -\mu^2 f_i.$$

Then one obtains the matrix sums as

$$\lambda(M) = \lambda(M_1 + M_2 + M_3) \geqslant -1 + \mu^2 + 3\varphi^2.$$

This expression reaches its minimum at the eigenvalue $\lambda_{\min} = -1$. As a result, from Eq. (30) one finds $\lambda^E_{\max} \leqslant -1/2$.

For the contractive and expansive parts of the free energy, we choose the following interpolation:

$$F_E(\varphi) = \sum_{i=1}^{3} a_i F_i, \quad F_C(\varphi) = \sum_{i=1}^{3} (1 - a_i) F_i,$$

from which it follows that, for the upper-bound eigenvalues of the matrix M^E, one can write

$$\lambda(M^E) = -a_1 + a_2\mu^2 + 3a_3\varphi^2.$$

To obtain the lower-bound limit for the latter expression, we skip the contribution from eigenvalues of the Laplacian by assuming $a_2 \leqslant 0$. Taking for simplicity $a_2 = 0$ and using the maximal contribution of the arbitrary configuration of the field for order parameter $\varphi^2 = 1$, one can get the following condition:

$$\lambda^E_{\max} = -a_1 + 3a_3 \leqslant -1/2.$$

To avoid iterations at every time step, it is convenient to calculate the nonlinear contribution in equations explicitly. It is equivalent to the equality $a_3 = 1$. As a result, we obtain a direct linear method for iteration by time. For the coefficient a_1, one

then obtains the condition $a_1 \geqslant 7/2$, which is in agreement with the previous result [11].

In a general case of d measurable Cartesian space, the maximal eigenvalue of the grid Laplace operator on the grid with a space δx_i $(i = 1, \ldots, d)$ can be evaluated as

$$\mu^2 \leqslant \pi^2 \sum_{i=1}^{d} \frac{1}{\delta x_i^2} \leqslant \frac{\pi^2 d}{\min(\delta x_i)^2}. \tag{39}$$

In this case, the region of values of the task parameters, satisfying the inequality

$$a_1 - a_2 \frac{\pi^2 d}{\min(\delta x_i)^2} - 3a_3 \geqslant 1/2, \tag{40}$$

defines possible values of the coefficients a_1, a_2, and a_3. With these coefficients, the difference scheme (31) for the hyperbolic AC equation is described by

$$\begin{cases} \varphi^{(k+1)} - \dfrac{\delta t^2}{\delta t + 2\tau_n}[(1-a_1)\varphi + (1-a_2)\Delta\varphi - (1-a_3)\varphi^3]|^{(k+1)} = \varphi^{(k)} + \dfrac{\delta t^2}{\delta t + 2\tau_n}[a_1\varphi + a_2\Delta\varphi - a_3\varphi^3]|^{(k)} + \dfrac{2\tau_n}{\delta t + 2\tau_n}u^{(k)}, \\ u^{(k+1)} = -u^{(k)} + \dfrac{2}{\delta t}(\varphi^{(k+1)} - \varphi^{(k)}), \end{cases} \tag{41}$$

and the difference scheme (33) for the hyperbolic CH equation is given by

$$\begin{cases} \varphi^{(k+1)} + \dfrac{\delta t^2}{\delta t + 2\tau_c}\Delta[(1-a_1)\varphi \quad + (1-a_2)\Delta\varphi - (1-a_3)\varphi^3]|^{(k+1)} \\ \quad = \varphi^{(k)} - \dfrac{\delta t^2}{\delta t + 2\tau_c}\Delta[a_1\varphi + a_2\Delta\varphi - a_3\varphi^3]|^{(k)} - \dfrac{2\tau_c}{\delta t + 2\tau_c}u^{(k)}, \\ u^{(k+1)} = -u^{(k)} - \dfrac{2}{\delta t}(\varphi^{(k+1)} - \varphi^{(k)}), \\ \Delta\vec{J}^{(k+1)} = \nabla u^{(k+1)}. \end{cases} \tag{42}$$

Numerical schemes (41) and (42) provide monotonic nonincreasing of the free energy in time (see Sec. V A) and are gradient stable.

B. Example for hyperbolic SH and PFC equations

For the SH and PFC equations, we choose the potential of the form

$$f(\varphi) = \tfrac{1}{4}\varphi^4 + \tfrac{1}{2}(1-\varepsilon)\varphi^2, \tag{43}$$

where ε is the undercooling into the metastable (unstable) region of the phase diagram. In the dimensionless form, Eq. (13) for the nonconserved order parameter is described by

$$\tau_n\ddot{\varphi} + \dot{\varphi} = [\varepsilon - (1+\Delta)^2]\varphi - \varphi^3, \tag{44}$$

and the dimensionless equation (24) for the conserved order parameter is

$$\tau_c\ddot{\varphi} + \dot{\varphi} = \Delta\{[-\varepsilon + (1+\Delta)^2]\varphi + \varphi^3\}. \tag{45}$$

The whole free energy of the system, excluding the pure nonequilibrium contribution, is given by

$$F[\varphi] = \int_V \left[\frac{1}{2}(\Delta\varphi)^2 - (\vec{\nabla}\varphi)^2 + \frac{1}{4}\varphi^4 + \frac{1}{2}(1-\varepsilon)\varphi^2\right]dV. \tag{46}$$

This free energy can be represented by four terms,

$$F(\varphi) = F_1 + F_2 + F_3 + F_4,$$

which are defined as

$$\begin{cases} F_1 = -\int(\vec{\nabla}\varphi)^2\,dV, \\ F_2 = \frac{1}{2}\int(\Delta\varphi)^2\,dV, \\ F_3 = \frac{1}{4}\int\varphi^4\,dV, \\ F_4 = \frac{1}{2}(1-\varepsilon)\int\varphi^2\,dV. \end{cases}$$

Each of these terms has the following grid Hessians (27):

$$(M_4)_{ij} = (1-\varepsilon)\delta_{ij}, \quad (M_1)_{ij} = \Delta_{ij},$$
$$(M_2)_{ij} = \Delta_{ij}^2, \quad (M_3)_{ij} = 3\varphi_i^2\delta_{ij}.$$

Lower-bound eigenvalues of the whole Hessian are obtained by

$$\lambda(M) \geqslant 1 - \varepsilon - \mu^2 + \mu^4 + 3\varphi^2.$$

Expression $\mu^4 - \mu^2$ has a minimum equal to $-1/4$ at $\mu^2 = 1/2$. Therefore, the minimal value of $\lambda(M)$ is reached at the eigenvalue $\lambda_{\min} = 3/4 - \varepsilon$. Consistent with relation (30), it leads to the following limitation:

$$\lambda^E_{\max} \leqslant \tfrac{3}{8} - \tfrac{1}{2}\varepsilon.$$

Taking contractive and expansive parts of the free energy in the form

$$F_E(\varphi) = \sum_{i=1}^{4} a_i F_i, \quad F_C(\varphi) = \sum_{i=1}^{4}(1-a_i)F_i,$$

we find the expression

$$\lambda(M^E) = a_1(1-\varepsilon) - a_2\mu^2 + a_3\mu^4 + 3a_4\varphi_0^2,$$

which is limited at $a_2 \geqslant 0$ and $a_3 \leqslant 0$. To evaluate the upper-bound eigenvalues of the matrix M^E, one may choose, for simplicity, $a_2 = 0$ and $a_3 = 0$. Then, taking into account the maximal contribution of arbitrary configuration of the field of order parameter φ_0, we find the following condition:

$$\lambda^E_{\max} = a_1(1-\varepsilon) + 3a_4\varphi_0^2.$$

Using an explicit interpolation for nonlinear terms by explicit form ($a_4 = 1$), one obtains a direct linear method for time iteration. For the coefficient a_1 we obtain the following condition:

$$a_1(1-\varepsilon) + 3a_4\varphi^2 \leqslant \tfrac{3}{8} - \tfrac{1}{2}\varepsilon.$$

In the general case, from estimation of spectrum (39), one obtains possible values of the coefficients a_1, a_2, a_3, and a_4:

$$a_1(1-\varepsilon) - a_2\frac{\pi^2 d}{\min(\delta x_i)^2} + a_3\frac{\pi^4 d^2}{\min(\delta x_i)^4} + 3a_4\varphi_0^2 \leqslant \frac{3}{8} - \frac{1}{2}\varepsilon. \tag{47}$$

For these coefficients, numeric scheme (31) for the hyperbolic SH equation is described by

$$\begin{cases} \varphi^{(k+1)} + \dfrac{\delta t^2}{\delta t + 2\tau_n}[(1-a_1)(1-\varepsilon)\varphi + 2(1-a_2)\Delta\varphi + (1-a_3)\Delta^2\varphi + (1-a_4)\varphi^3]|^{(k+1)} \\ \quad = \varphi^{(k)} - \dfrac{\delta t^2}{\delta t + 2\tau_n}[a_1(1-\varepsilon)\varphi + 2a_2\Delta\varphi + a_3\Delta^2\varphi + a_4\varphi^3]|^{(k)} + \dfrac{2\tau_n}{\delta t + 2\tau_n}u^{(k)}, \\ u^{(k+1)} = -u^{(k)} + \dfrac{2}{\delta t}(\varphi^{(k+1)} - \varphi^{(k)}), \end{cases} \tag{48}$$

and difference scheme (31) for the hyperbolic PFC equation is given by

$$\begin{cases} \varphi^{(k+1)} + \dfrac{\delta t^2}{\delta t + 2\tau_c}\Delta[(1-a_1)(1-\varepsilon)\varphi + 2(1-a_2)\Delta\varphi + (1-a_3)\Delta^2\varphi + (1-a_4)\varphi^3]|^{(k+1)} \\ \quad = \varphi^{(k)} - \dfrac{\delta t^2}{\delta t + 2\tau_c}\Delta[a_1(1-\varepsilon)\varphi + 2a_2\Delta\varphi + a_3\Delta^2\varphi + a_4\varphi^3]|^{(k)} + \dfrac{2\tau_c}{\delta t + 2\tau_c}u^{(k)}, \\ u^{(k+1)} = -u^{(k)} - \dfrac{2}{\delta t}(\varphi^{(k+1)} - \varphi^{(k)}), \\ \Delta\vec{J}^{(k+1)} = \vec{\nabla}u^{(k+1)}. \end{cases} \tag{49}$$

Numeric schemes (48) and (49) provide monotonic nonincreasing of the free energy in time (see Sec. V B) and are gradient stable.

V. MONOTONIC NONINCREASE IN TIME OF THE FREE ENERGY

As is shown in the extended irreversible thermodynamics [15], hyperbolic equations of transport or motion provide monotonic relaxation of thermodynamic functions to their extremal values in the evolution of the system to global equilibrium. It has been clearly demonstrated, for example, for monotonic increasing of the entropy to its maximum in a thermally nonequilibrium system (see Fig. 2.1 in Ref. [19]). Here we also assume that hyperbolic equations (12), (13), (23), and (24) provide a monotonic nonincrease of the free energy even though highly nonequilibrium fast transitions may result in both oscillating behavior and monotonic relaxation of the order parameter (as described in Refs. [21,22]).

A. Proof for the nonconserved order parameter

A proof of Eq. (32) for the discrete equation of the dynamics with nonconserved order parameter (31) is based on inequalities (A5) and (A7) from the Appendix. These inequalities express the properties of a function of many variables and transform to the following form:

$$F[\varphi^{(k+1)},u^{(k+1)}] - F[\varphi^{(k)},u^{(k)}] \leqslant \left\langle \delta\varphi\frac{\partial F}{\partial\varphi}\bigg|^{(k+1)} + \delta u\frac{\partial F}{\partial u}\bigg|^{(k+1)} \right\rangle - \frac{1}{2}\lambda_{\min}\langle\delta\varphi^2\rangle - \frac{1}{2}\alpha_n\langle\delta u^2\rangle, \tag{50}$$

and

$$\left\langle \delta\varphi\left(\frac{\partial F_E}{\partial\varphi}\bigg|^{(k+1)} - \frac{\partial F_E}{\partial\varphi}\bigg|^{(k)}\right)\right\rangle \leqslant \lambda^E_{\max}\langle\delta\varphi^2\rangle. \tag{51}$$

In Eqs. (50) and (51), brackets $\langle\cdots\rangle$ represent the summation over grid nodes $\langle A\rangle \equiv \sum_i A_i$; notations $\delta\varphi$ and δu give the variations of fields φ and u at each grid node: $\delta\varphi_i \equiv \varphi_i^{(k+1)} - \varphi_i^{(k)}$ and $\delta u_i \equiv u_i^{(k+1)} - u_i^{(k)}$, respectively.

Considering the numeric approximation of the first of Eqs. (10), one may write

$$\tau_n\dot{u} + \dot{\varphi} = -M_n\frac{\delta F}{\delta\varphi}. \tag{52}$$

Replacing time derivatives by finite differences and using functions on the grid, one finds

$$\tau_n\frac{\delta u_i}{\delta t} + \frac{\delta\varphi_i}{\delta t} + M_n\frac{\delta F_C}{\delta\varphi_i}\bigg|^{(k+1)} = -M_n\frac{\delta F_E}{\delta\varphi_i}\bigg|^{(k)}, \tag{53}$$

where $M_n \equiv M_n^{(k)}$. Adding $M_n \delta F_E/\delta\varphi_i|^{(k+1)}$ to both sides of the equation of motion (53), one gets

$$\left.\frac{\delta F}{\delta\varphi_i}\right|^{(k+1)} = -\frac{1}{M_n}\left(\tau_n\frac{\delta u_i}{\delta t}+\frac{\delta\varphi_i}{\delta t}\right) + \left(\left.\frac{\delta F_E}{\delta\varphi_i}\right|^{(k+1)} - \left.\frac{\delta F_E}{\delta\varphi_i}\right|^{(k)}\right). \tag{54}$$

Substituting Eq. (54) into inequality (50) and calculating $\delta F/\delta u$ from Eq. (3) leads to

$$\begin{aligned} &F[\varphi^{(k+1)},u^{(k+1)}] - F[\varphi^{(k)},u^{(k)}] \\ &\leqslant \left\langle \delta\varphi\left(\left.\frac{\delta F_E}{\delta\varphi}\right|^{(k+1)} - \left.\frac{\delta F_E}{\delta\varphi}\right|^{(k)}\right)\right\rangle + \alpha_n\langle u^{(k+1)}\delta u\rangle \\ &\quad -\frac{1}{2}\alpha_n\langle\delta u^2\rangle - \frac{1}{2}\lambda_{\min}\langle\delta\varphi^2\rangle \\ &\quad -\frac{1}{\delta t}\left\langle M_n^{-1}\delta\varphi^2\right\rangle - \frac{1}{\delta t}\left\langle\delta\varphi\tau_n M_n^{-1}\delta u\right\rangle. \end{aligned} \tag{55}$$

Equation (55), together with inequality (51) and expression (12), gives

$$\begin{aligned} &F[\varphi^{(k+1)},u^{(k+1)}] - F[\varphi^{(k)},u^{(k)}] \\ &\leqslant \left(\lambda_{\max}^E - \frac{1}{2}\lambda_{\min}\right)\langle\delta\varphi^2\rangle - \frac{1}{\delta t}\left\langle M_n^{-1}\delta\varphi^2\right\rangle \\ &\quad +\alpha_n\left\langle\delta u\left(\frac{1}{2}(u^{(k+1)}+u^{(k)}) - \frac{\delta\varphi}{\delta t}\right)\right\rangle. \end{aligned} \tag{56}$$

With $M_n > 0$, the term $\langle M_n^{-1}\delta\varphi^2\rangle/\delta t$ can be omitted. Choosing

$$\delta\varphi = \tfrac{1}{2}\delta t(u^{(k+1)}+u^{(k)}),$$

the free energy nonincreases in time by Eqs. (32) if

$$\lambda_{\max}^E \leqslant \tfrac{1}{2}\lambda_{\min}.$$

The latter semi-inequality agrees with the Eyre theorem condition (30) for the parabolic AC equation. Finally, we find the following difference scheme in time:

$$\begin{cases} \tau_n\dfrac{\delta u_i}{\delta t} + \dfrac{\delta\varphi_i}{\delta t} + M_n\left.\dfrac{\delta F_C}{\delta\varphi_i}\right|^{(k+1)} = -M_n\left.\dfrac{\delta F_E}{\delta\varphi_i}\right|^{(k)}, \\ \delta\varphi_i = \dfrac{1}{2}\delta t\left(u_i^{(k+1)}+u_i^{(k)}\right), \end{cases} \tag{57}$$

from which Eq. (31) directly follows.

B. Proof for the conserved order parameter

Proof of Eq. (34) for the discrete equation with conserved order parameter (33) is based on inequalities (A5) and (A7) from the Appendix, which are reduced to the following expressions:

$$\begin{aligned} &F[\varphi^{(k+1)},\vec{J}^{(k+1)}] - F[\varphi^{(k)},\vec{J}^{(k)}] \\ &\leqslant \left\langle\delta\varphi\left.\frac{\delta F}{\delta\varphi}\right|^{(k+1)} + \delta\vec{J}\cdot\left.\frac{\delta F}{\delta\vec{J}}\right|^{(k+1)}\right\rangle \\ &\quad -\frac{1}{2}\lambda_{\min}\langle\delta\varphi^2\rangle - \frac{1}{2}\alpha_c\langle\delta\vec{J}^{\,2}\rangle, \end{aligned} \tag{58}$$

and

$$\left\langle\delta\varphi\left(\left.\frac{\delta F_E}{\delta\varphi}\right|^{(k+1)} - \left.\frac{\delta F_E}{\delta\varphi}\right|^{(k)}\right)\right\rangle \leqslant \lambda_{\max}^E\langle\delta\varphi^2\rangle. \tag{59}$$

As before, brackets $\langle\cdots\rangle$ in Eq. (59) represent the summation over the grid nodes $\langle A\rangle \equiv \sum_i A_i$, and $\delta\varphi$ and $\delta\vec{J}$ give the variations of fields φ and u, respectively, at each grid node: $\delta\varphi_i \equiv \varphi_i{}^{(k+1)} - \varphi_i{}^{(k)}$ and $\delta\vec{J}_i \equiv \vec{J}_i^{(k+1)} - \vec{J}_i^{(k)}$.

Note that the analysis of conserved dynamics is complicated due to the appearance of the elliptical Laplace operator in Eq. (33). We consider a grid of arbitrary dimensionality with n nodes, on which the grid elliptical Laplace operator $(\vec{\nabla}\cdot M_c\vec{\nabla})_{ij} \equiv A_{ij}$ is defined. Here we again assume that $M_c \equiv M_c^{(k)}$. The Laplacian is defined by the symmetric $n\times n$ matrix with eigenvalues $\lambda_1 = 0$ and $\lambda_m < 0$ for all $m > 1$. Let $v_i^{(m)}$ represent the ith component of the mth eigenvector of the matrix A. Then, regarding the condition of completeness, one has

$$\delta_{ik} = \sum_{m=1}^{n} v_i^{(m)}v_k^{(m)} = \sum_{j=1}^{n}\widetilde{A}_{ij}A_{jk} + v_i^{(1)}v_k^{(1)}, \tag{60}$$

where the pseudoinverse matrix $\widetilde{A}$ is defined by

$$\widetilde{A}_{ij} = \sum_{m\neq 1}^{n}\frac{1}{\lambda_m}v_i^{(m)}v_j^{(m)}. \tag{61}$$

The eigenvalue $\lambda_1 = 0$ corresponds to a uniform field $v_i^{(1)} = 1/\sqrt{n}$ for all ith indices. Substituting the identity operator (60) into the sum for expression (58) and summing over the grid, one finds

$$\begin{aligned} &F[\varphi^{(k+1)},\vec{J}^{(k+1)}] - F[\varphi^{(k)},\vec{J}^{(k)}] \\ &\leqslant \left\langle\delta\varphi\widetilde{A}A\left.\frac{\delta F}{\delta\varphi}\right|^{(k+1)}\right\rangle + \alpha_c\langle\vec{J}^{(k+1)}\delta\vec{J}\rangle - \frac{1}{2}\lambda_{\min}\langle\delta\varphi^2\rangle \\ &\quad -\frac{1}{2}\alpha_c\langle\delta\vec{J}^{\,2}\rangle. \end{aligned} \tag{62}$$

Here the contribution from the zero mode has dropped out due to the existence of the conservation law $\langle\delta\varphi\rangle = 0$.

By analogy with the dynamics of the nonconserved order parameter (see Sec. V A), the first of Eqs. (22) can be written as

$$\tau_c\dot{u} - \dot{\varphi} = -\vec{\nabla}\cdot\left[M_c\vec{\nabla}\left(\frac{\delta F}{\delta\varphi}\right)\right]. \tag{63}$$

Then the corresponding difference scheme in time is given by

$$\tau_c\frac{\delta u_i}{\delta t} - \frac{\delta\varphi_i}{\delta t} + A_{ij}\left.\left(\frac{\delta F_C}{\delta\varphi_j}\right)\right|^{(k+1)} = -A_{ij}\left.\left(\frac{\delta F_E}{\delta\varphi_j}\right)\right|^{(k)}. \tag{64}$$

Adding $A_{ij}[\partial F_E/\partial\varphi_j]^{(k+1)}$ to both sides of Eq. (64) gives

$$\begin{aligned} &\sum_k A_{jk}\left.\frac{\delta F}{\delta\varphi_k}\right|^{(k+1)} \\ &\quad = -\tau_c\frac{\delta u_i}{\delta t} + \frac{\delta\varphi_i}{\delta t} + \sum_k A_{jk}\left(\left.\frac{\delta F_E}{\delta\varphi_k}\right|^{(k+1)} - \left.\frac{\delta F_E}{\delta\varphi_k}\right|^{(k)}\right). \end{aligned} \tag{65}$$

Substituting Eq. (65) into expression (62) leads to

$$\begin{aligned} F[\varphi^{(k+1)},\vec{J}^{(k+1)}] - F\left[\varphi^{(k)},\vec{J}^{(k)}\right] \\ \leqslant \left\langle \delta\varphi\left(\frac{\delta F_E}{\delta\varphi_k}\Big|^{(k+1)} - \frac{\delta F_E}{\delta\varphi_k}\Big|^{(k)}\right)\right\rangle + \alpha_c\langle\vec{J}^{(k+1)}\delta\vec{J}\rangle \\ -\frac{1}{2}\lambda_{\min}\langle\delta\varphi^2\rangle + \frac{1}{\delta t}\langle\delta\varphi\widetilde{A}\delta\varphi\rangle \\ -\frac{1}{\delta t}\langle\delta\varphi\tau_c\widetilde{A}\delta u\rangle - \frac{1}{2}\alpha_c\langle\delta\vec{J}^{\,2}\rangle. \end{aligned} \tag{66}$$

Using Eq. (59) and combining the fluxes in the last two terms on the right-hand side of Eq. (66), we arrive at the following expression:

$$\begin{aligned} F[\varphi^{(k+1)},\vec{J}^{(k+1)}] - F[\varphi^{(k)},\vec{J}^{(k)}] \\ \leqslant \left(\lambda^E_{\max} - \frac{1}{2}\lambda_{\min}\right)\langle\delta\varphi^2\rangle + \frac{1}{\delta t}\langle\delta\varphi\widetilde{A}\delta\varphi\rangle \\ + \frac{1}{2}\alpha_c\langle(\vec{J}^{(k+1)} + \vec{J}^{(k)})\delta\vec{J}\rangle - \frac{1}{\delta t}\langle\delta\varphi\tau_c\widetilde{A}\delta u\rangle. \end{aligned} \tag{67}$$

Regarding the nonpositivity of the spectrum of the grid Laplacian and by definition (61), the following quadratic form is negatively defined:

$$\frac{1}{\delta t}\langle\delta\varphi\widetilde{A}\delta\varphi\rangle \leqslant 0. \tag{68}$$

Therefore, the corresponding term in Eq. (67) can be omitted.

To evaluate the contribution from the last two terms in Eq. (67), one can take into account that the field dynamics of the conserved order parameter is defined only up to a divergence of the vector field $\vec{J}$ that follows from Eqs. (22). As is known [30], applying the Helmholtz theorem for the three-dimensional Euclidean space, any vector field is unambiguously defined through the expansion of gradient and curl contributions:

$$\vec{J} = \vec{\nabla}\psi + \vec{\nabla}\times\vec{A}.$$

Note that only the gradient contribution gives the divergence term:

$$\vec{\nabla}\cdot\vec{J} = \vec{\nabla}\cdot(\vec{\nabla}\psi + \vec{\nabla}\times\vec{A}) = \Delta\psi,$$

where ψ is some differentiable function.

Taking into account the form of the elliptic operator in the right-hand side of Eq. (22), the flux can be presented in a more general form (in comparison with the standard gradient operator):

$$\vec{J} = M_c\vec{\nabla}\psi. \tag{69}$$

Obviously, the curl of such a substitution is not zero:

$$\vec{\nabla}\times(M_c\vec{\nabla}\psi) = \vec{\nabla}M_c\times\vec{\nabla}\psi \neq 0,$$

but it is defined only by inhomogeneity of mobility. Consequently, the gradient part of vector field $\vec{J}$ is contained in $\vec{J} = M_c\vec{\nabla}\psi$, which gives the expression for estimation of terms in inequality (67).

Using the above statements, the divergence of the flux is $u = \vec{\nabla}\cdot(M_c\vec{\nabla}\psi)$. Because the difference operator is taken from the previous time step in Eq. (64), we assume for the difference approximation that $\vec{J}^{(k+1)} = M_c\vec{\nabla}\psi^{(k+1)}$, $\vec{J}^{(k)} = M_c\vec{\nabla}\psi^{(k)}$, $u^{(k+1)} = \vec{\nabla}\cdot(M_c\vec{\nabla}\psi^{(k+1)})$, and $u^{(k)} = \vec{\nabla}\cdot(M_c\vec{\nabla}\psi^{(k)})$. Accordingly, one obtains

$$\delta\vec{J} = M_c\vec{\nabla}\delta\psi, \quad \delta u = \vec{\nabla}\cdot(M_c\vec{\nabla}\delta\psi).$$

Then, substituting δu_i into Eq. (67) and using $\delta u_i = A_{ij}\delta\psi_j$, one finds

$$\frac{1}{\delta t}\langle\delta\varphi\tau_c\widetilde{A}\delta u\rangle = \frac{1}{\delta t}\langle\tau_c\delta\varphi\delta\psi\rangle. \tag{70}$$

Similarly, substituting $\delta\vec{J}_i = M_c\vec{\nabla}_{ij}\delta\psi_j$ (where ∇_{ij} is the grid approximation of the operator $\vec{\nabla}$) into Eq. (67) allows us to write

$$\begin{aligned} \tfrac{1}{2}\alpha_c\langle(\vec{J}^{(k+1)} + \vec{J}^{(k)})\delta\vec{J}\rangle &= \tfrac{1}{2}\alpha_c\langle(\vec{J}^{(k+1)} + \vec{J}^{(k)})M_c\vec{\nabla}\delta\psi\rangle \\ &= \tfrac{1}{2}\tau_c\langle(\vec{J}^{(k+1)} + \vec{J}^{(k)})\vec{\nabla}\delta\psi\rangle. \end{aligned} \tag{71}$$

Using partial summation and boundary conditions for fluxes (17) and (26) and taking into account the difference approximation of definition (21) in the form of

$$u_i = \vec{\nabla}_{ij}\vec{J}_j,$$

one finds

$$\begin{aligned} \tfrac{1}{2}\alpha_c\langle(\vec{J}^{(k+1)} + \vec{J}^{(k)})\delta\vec{J}\rangle &= -\tfrac{1}{2}\tau_c\langle\vec{\nabla}(\vec{J}^{(k+1)} + \vec{J}^{(k)})\delta\psi\rangle \\ &= -\tfrac{1}{2}\tau_c\langle(u^{(k+1)} + u^{(k)})\delta\psi\rangle. \end{aligned} \tag{72}$$

Equation (72) asserts that the last two terms in Eq. (67) mutually annihilate if the condition

$$\frac{\delta\varphi_i}{\delta t} = -\frac{1}{2}\left(u_i^{(k+1)} + u_i^{(k)}\right)$$

is satisfied. In this case, the Eyre condition (30) arises again and the respective unconditionally stable numeric scheme takes the following form:

$$\begin{cases} \tau_c\dfrac{\delta u_i}{\delta t} - \dfrac{\delta\varphi_i}{\delta t} + A_{ij}\left(\dfrac{\delta F_C}{\delta\varphi_j}\right)\Big|^{(k+1)} = -A_{ij}\left(\dfrac{\delta F_E}{\delta\varphi_j}\right)\Big|^{(k)}, \\ \dfrac{\delta\varphi_i}{\delta t} = -\dfrac{1}{2}\left(u_i^{(k+1)} + u_i^{(k)}\right). \end{cases} \tag{73}$$

From Eq. (73), obviously, it follows the system of equations (33).

VI. CONCLUSIONS

An approach to the numerical solution of equations of fast phase transitions, describing the local nonequilibrium evolution of spatially inhomogeneous systems, is presented. Using the Eyre expansion for free energy on contractive and expansive parts for equations of fast transitions, the gradient-stable computational algorithms can be developed. These algorithms are defined by numerical schemes (31) and (33), which (1) guarantee computational stability for arbitrary time steps limited only by the requirement of computational accuracy, and (2) provide a monotonic nonincrease of the free energy with arbitrary time step in numerical modeling. In examples of dynamics with conserved and nonconserved order parameters, an application of the Eyre theorem to the problems of fast phase transitions is shown.

Finally, the second temporal derivatives in Eqs. (12), (13), (23), and (24) describe the inertia in local nonequilibrium

systems [17]. In this sense, local nonequilibrium phenomena in Eqs. (12), (13), (23), and (24) are introduced phenomenologically by the temporal nonlocality. In more general case, a local nonequilibrium contribution is introduced into the system dynamics by the temporal memory function and spatial nonlocality [17,21]. For the latter case, gradient-stable numeric schemes might be developed additionally.

ACKNOWLEDGMENTS

V.L. acknowledges support from the Russian Foundation of Basic Research under Project No. 08-02-91957NNIO_a and from ROSNAUKA (the Russian Scientific Foundation) under Project No. 2009-1.5-507-007-002. P.G. acknowledges support from DFG (the German Research Foundation) under Project No. HE 160/19 and DLR Space Management under Contract No. 50WM1140.

APPENDIX: NECESSARY INEQUALITIES

To complete the analysis, we summarize here the inequalities [11] which are necessary to prove the gradient stability. Consider an arbitrary function $f(x)$ which can be differentiated twice in Euclidian space $\mathbf{E}^{(n)}$. From the fundamental theorem of calculus, one gets

$$f(\mathbf{x}+\mathbf{y}) - f(\mathbf{x}) = \sum_i y_i \int_0^1 ds_1 \partial_i f\Big|_{\mathbf{x}+s_1\mathbf{y}}, \tag{A1}$$

where $\partial_i f \equiv \partial f/\partial x_i$ and the parameter s_1 changes from 0 to 1. Analogously, for the first derivatives, one gets

$$\partial_i f\big|_{\mathbf{x}+\mathbf{s_1}\mathbf{y}} - \partial_i f\big|_{\mathbf{x}} = \sum_j y_j \int_0^{s_1} ds_2 H_{ij}\Big|_{\mathbf{x}+s_2\mathbf{y}}, \tag{A2}$$

where Hessian $H_{ij} \equiv \partial^2 f/\partial x_i \partial x_j$ is the matrix of the second derivatives. Combination of Eqs. (A1) and (A2) leads to the following obvious identity:

$$f(\mathbf{x}+\mathbf{y}) - f(\mathbf{x}) = \sum_i y_i \partial_i f\Big|_{\mathbf{x}} + \int_0^1 ds_1 \int_0^{s_1} ds_2 \sum_{i,j} y_i y_j H_{ij}\Big|_{\mathbf{x}+s_2\mathbf{y}}. \tag{A3}$$

Let the eigenvalues of the Hessian H_{ij} be limited from below by some value $\lambda_{\min}$ for any value of $\mathbf{x}$. Then, for the quadratic form with matrix H_{ij}, the following estimation holds:

$$\sum_{i,j} y_i y_j H_{ij}\Big|_{\mathbf{x}+s_2\mathbf{y}} \geqslant \lambda_{\min}\langle \mathbf{y}^2\rangle, \tag{A4}$$

where $\langle \mathbf{y}^2\rangle = \sum_i y_i^2$. Combination of Eqs. (A3) and (A4) leads to

$$f(\mathbf{x}+\mathbf{y}) - f(\mathbf{x}) \geqslant \sum_i y_i \partial_i f\Big|_{\mathbf{x}} + \frac{1}{2}\lambda_{\min}\langle \mathbf{y}^2\rangle, \tag{A5}$$

where the factor $1/2$ appears in Eq. (A5) after integration by s_i.

Also, at $s_1 = 1$ in Eq. (A2), one can get the second inequality from Eq. (A2) by multiplying it by y_i. After summation, one has

$$\sum_i y_i(\partial_i f|_{\mathbf{x}+\mathbf{y}} - \partial_i f|_{\mathbf{x}}) = \sum_{i,j} y_i y_j \int_0^1 ds\, H_{ij}|_{\mathbf{x}+s\mathbf{y}}. \tag{A6}$$

If the spectrum of the Hessian H_{ij} is limited from above by the number $\lambda_{\max}$, then we arrive, finally, at the following estimation:

$$\sum_i y_i(\partial_i f|_{\mathbf{x}+\mathbf{y}} - \partial_i f|_{\mathbf{x}}) \leqslant \lambda_{\max}\langle \mathbf{y}^2\rangle. \tag{A7}$$

Conditions for the lower boundary (A5) and upper boundary (A7) define the initial inequalities (50) and (51), as well as inequalities (58) and (59). These inequalities are used for the proof of the monotonic nonincrease of the free energy in the system described by the difference schemes (57) and (73).

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