

Symmetric form for the hyperbolic-parabolic system of fourth-gradient fluid model

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Abstract The fourth-gradient model for fluids - associated with an extended molecular mean-field theory of capillarity - is considered. By producing fluctuations of density near the critical point like in computational molecular dynamics, the model is more realistic and richer than van der Waals' one and other models associated with a second order expansion.

The aim of the paper is to prove - with a fourth-gradient internal energy already obtained by the mean field theory - that the quasi-linear system of conservation laws can be written in an Hermitian symmetric form implying the stability of constant solutions. The result extends the symmetric hyperbolicity property of governing-equations' systems when an equation of energy associated with high order deformation of a continuum medium is taken into account.

Keywords: Fourth-gradient model; Hyperbolic-parabolic systems; Extended van der Waals' model; Fluid energy equation.

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1 Introduction

Many physical models are represented by quasi-linear first order systems of N balance laws (in particular conservation laws),

$$\frac{\partial \mathbf{F}^0(\mathbf{u})}{\partial t} + \frac{\partial \mathbf{F}^j(\mathbf{u})}{\partial x^j} = \mathbf{f}(\mathbf{u}), \quad (1)$$

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with an additional scalar balance equation (typically the energy equation in pure mechanical case or the entropy equation in thermodynamics):

$$\frac{\partial h^0(\mathbf{u})}{\partial t} + \frac{\partial h^j(\mathbf{u})}{\partial x^j} = \Sigma(\mathbf{u}),$$

where $\mathbf{F}^0, \mathbf{F}^j$ ($j = 1, 2, \dots, n$), \mathbf{f}, \mathbf{u} are column vectors of R^N and h^0, h^j , ($j = 1, 2, \dots, n$), Σ are scalar functions; $t, \mathbf{x} \equiv (x^1, \dots, x^n)$ are the time and space coordinates, respectively; we adopt sum convention on the repeated indices.

Function h^0 is assumed convex with respect to field $\mathbf{F}^0(\mathbf{u}) \equiv \mathbf{u}$, [1–5]. Boillat [3] introduces dual-vector field \mathbf{u}' , associated with Legendre transform h'^0 and potentials h'^j , such that

$$\mathbf{u}' = \left(\frac{\partial h^0}{\partial \mathbf{u}} \right)^*, \quad h'^0 = \mathbf{u}'^* \mathbf{u} - h^0, \quad h'^j = \mathbf{u}'^* \mathbf{F}^j(\mathbf{u}) - h^j, \quad (2)$$

where superscript “ $*$ ” denotes the transposition. Therefore by convexity argument, it is possible to take \mathbf{u}' as field and we obtain from (2):

$$\mathbf{u} = \left(\frac{\partial h'^0}{\partial \mathbf{u}'} \right)^*, \quad \mathbf{F}^j(\mathbf{u}) = \left(\frac{\partial h'^j}{\partial \mathbf{u}'} \right)^*. \quad (3)$$

Inserting (3) into (1), system (1) becomes symmetric :

$$\frac{\partial}{\partial t} \left(\frac{\partial h'^0}{\partial \mathbf{u}'} \right) + \frac{\partial}{\partial x^j} \left(\frac{\partial h'^j}{\partial \mathbf{u}'} \right) = \mathbf{f}(\mathbf{u}'), \quad (4)$$

which is equivalent to

$$\mathbf{A}^0 \frac{\partial \mathbf{u}'}{\partial t} + \mathbf{A}^j \frac{\partial \mathbf{u}'}{\partial x^j} = \mathbf{f}(\mathbf{u}'), \quad (5)$$

where matrix $\mathbf{A}^0 \equiv (\mathbf{A}^0)^*$ is symmetric positive definite and matrices $\mathbf{A}^j = (\mathbf{A}^j)^*$ are symmetric :

$$\mathbf{A}^0 \equiv (\mathbf{A}^0)^* = \frac{\partial^2 h'^0}{\partial \mathbf{u}' \partial \mathbf{u}'}, \quad \mathbf{A}^j \equiv (\mathbf{A}^j)^* = \frac{\partial^2 h'^j}{\partial \mathbf{u}' \partial \mathbf{u}'}, \quad (j = 1, 2, \dots, n). \quad (6)$$

The symmetric form of governing equations implies hyperbolicity. For conservation laws with vanishing productions, the hyperbolicity is equivalent to the stability of constant solutions with respect to perturbations in form $e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}$, where $i^2 = -1$, $\mathbf{k}^* = [k_1, \dots, k_n] \in (R^n)^*$ and ω is a real scalar. Indeed, the symmetric form of governing equations for an unknown vector \mathbf{u} , ($\mathbf{u}^* = [u_1, \dots, u_n]$) implies the *dispersion relation* :

$$\det(\mathbf{A}_{(k)} - \omega \mathbf{A}^0) = 0 \quad \text{with} \quad \mathbf{A}_{(k)} = \mathbf{A}^j k_j,$$

which determines real values of ω for any *real wave vector* \mathbf{k} . In this case, phase velocities are real and coincide with the characteristic velocities of hyperbolic system [6,7]. Moreover right eigenvectors of $\mathbf{A}_{(k)}$ with respect to \mathbf{A}^0 are linearly independent and any symmetric system is also automatically hyperbolic.

The previous technique was generalized in covariant relativistic formulation by Ruggeri and Strumia [4] that recognized the importance of field that symmetrizes

the original system and they proposed to call \mathbf{u}' *main field*. Boillat called symmetric form (5) with relations (6), *Godunov systems*. This kind of systems are the typical ones of Rational Extended Thermodynamics [8].

In the case of systems with parabolic structure (*hyperbolic-parabolic systems*), the following generalization of symmetric system (5) was considered :

$$\mathbf{A}^0 \frac{\partial \mathbf{u}'}{\partial t} + \mathbf{A}^j \frac{\partial \mathbf{u}'}{\partial x^j} - \frac{\partial}{\partial x^j} \left(\mathbf{B}^{jl} \frac{\partial \mathbf{u}'}{\partial x^l} \right) = 0, \quad (7)$$

where matrices $\mathbf{B}^{jl} = (\mathbf{B}^{jl})^*$ are symmetric and $\mathbf{B}_{(k)} = \mathbf{B}^{jl} k_j k_l$ are non-negative definite.

The compatibility of system (7) with entropy principle and the corresponding determination of main field was given by Ruggeri in [9] for Navier-Stokes-Fourier fluids and in general case by Kawashima and Shizuta [10]. The same authors in [11] considered linearized version of system (7) proving that the constant solutions are stable. For capillarity fluids, symmetric form (7) was studied in the simplest case by Gavriluk and Gouin [12].

Continuum models of capillarity can be interpreted by using *gradient theories* [13–15]. The models are useful to study interactions between fluids and solid walls [16, 17] and they can be obtained thanks to molecular methods [18–20]. In fact, the fourth-gradient model for fluids corresponds to development in continuum mechanics when the principle of virtual powers needs to obtain a separated form in the sense of distributions' theory on the physical domain and its boundaries, edges and end points where only vector forces are applied at end points [21, 22].

The study of models containing higher-order derivatives of the density has a clear interpretation in the framework of the mean-field molecular model. In the mean-field theory of hard-sphere molecules, the van der Waals forces exert stresses on fluid molecules producing surface tension effects [23, 24]. The second-gradient theory provides a construction of the energy density such that capillarity effects appear as a consequence of the molecular model in domains where the change of mass density is important [25, 26].

The fourth-gradient model for fluids is the background of the paper: the volume energy can be extended to obtain a fourth-gradient expansion of Cahn and Hilliard's equation [27] near the critical point [28]. The model is richer than the expansion of second order by van der Waals and others [29]. Such extension obtained via the request of molecular range turns out to be effective in the construction of a new interpolating model compatible with fluctuations of density near the critical point; the effects are not negligible and it is possible to deduce a Fisher-Kolmogorov equation [30] generating observable hydrodynamics fluctuations [31]. The differences in pulse-wave oscillations between second- and fourth-gradient models allow to revisit papers introducing kinks versus pulses as in [32]. We believe that this result is remarkable and will hopefully stimulate further and deeper investigations on both theoretical and phenomenological nature. It is interesting to note – and it is not the case for the second-gradient model – that the fourth gradient model is able to take the range of London intermolecular forces into account [28].

Using a statistical model in mean-field molecular theory, specific internal energy ε and volume free energy F of the fourth-gradient fluid are in the form,

$$\varepsilon = \alpha(\rho, s) - \frac{\lambda}{2} \Delta \rho - \frac{\gamma}{2} \Delta^2 \rho, \quad (8)$$

and

$$F = f(\rho, T) - \frac{\lambda}{2} \rho \Delta \rho - \frac{\gamma}{2} \rho \Delta^2 \rho,$$

with $\Delta \equiv \text{div grad}$ and $\Delta^2 \equiv \text{div}\{\text{grad}(\text{div grad})\}$ denote the harmonic and biharmonic operators, where div and grad denote the divergence and gradient operators, respectively ; ρ is the fluid density, s the specific entropy, T the Kelvin temperature and λ, γ are two scalar functions of ρ and s (or ρ and T). Term $\alpha(\rho, s)$ is the specific internal energy and $f(\rho, T)$ is the volume free energy of the homogeneous fluid bulk of densities ρ and s at temperature T . In the mean-field simplest model, near the critical point of the fluid, λ and γ can be considered as constant, conditions assumed along the paper.

In case $\gamma = 0$,

We get the internal energy expression given in [27]. However, authors used $\lambda/2 (\text{grad } \rho)^2$ in place of $-(\lambda/2) \rho \Delta \rho$.

But, $\rho \Delta \rho = \text{div}(\rho \text{ grad } \rho) - (\text{grad } \rho)^2$; consequently, $\lambda/2 \text{div}(\rho \text{ grad } \rho)$ can be integrated on the fluid boundary and is null when the fluid is homogeneous (as in the bulks).

In case $\gamma \neq 0$,

$$\rho \Delta^2 \rho = \text{div} [\rho \text{ grad}(\text{div grad } \rho) - (\text{div grad } \rho) \text{ grad } \rho] + [\text{div grad } \rho]^2.$$

Term $\text{div} [\rho \text{ grad}(\text{div grad } \rho) - (\text{div grad } \rho) \text{ grad } \rho]$ can be integrated on the boundary domain and is null when the fluid is homogeneous (as in the bulks); then, $-(\gamma/2) \rho \Delta^2 \rho$ can be replaced with $-\gamma/2 (\Delta \rho)^2$.

Consequently, for fourth-gradient fluids, the specific internal energy and the free volume energy can be respectively replaced by:

$$\varepsilon = \alpha(\rho, s) + \frac{1}{\rho} \left(\frac{\lambda}{2} (\text{grad } \rho)^2 - \frac{\gamma}{2} (\Delta \rho)^2 \right), \quad (9)$$

and

$$F = f(\rho, T) + \frac{\lambda}{2} (\text{grad } \rho)^2 - \frac{\gamma}{2} (\Delta \rho)^2,$$

We note that the equation of motion is the same for the two energy representations (8) and (9) but the boundary conditions, corresponding to the integrated terms, are different as it is pointed out in [20].

Here and later, for any vectors \mathbf{a}, \mathbf{b} we use the notation $\mathbf{a}^* \mathbf{b}$ for the scalar product (the line is multiplied by the column vector) and $\mathbf{a} \mathbf{b}^*$ for the tensor product (or $\mathbf{a} \otimes \mathbf{b}$ the column vector is multiplied by the line vector). Divergence of a linear transformation \mathbf{D} is the covector $\text{div}(\mathbf{D})$ such that, for any constant vector \mathbf{d} , $\text{div}(\mathbf{D}) \mathbf{d} = \text{div}(\mathbf{D} \mathbf{d})$. The identical transformation is denoted by \mathbf{I} .

The paper is organized as follows. In Section 2, thanks to the principle of virtual powers, we obtain the equation of conservative motions. In Section 3, we get the equation of energy and extends the *interstitial-working* notion obtained in second-gradient model [33]. In Section 4, we propose a system of quasi-linear equations in divergence form. Using a convenient change of variables associated with a Legendre transformation of the total fluid energy, near an equilibrium position we obtain an *Hermitian symmetric form* for the equations of perturbations. For the equations of *fourth-gradient capillary fluids* that belong to the class of dispersive systems, we get an analog of symmetric form (7) with main field given by (2)¹. The system is proved to be stable. A conclusion and two appendices end the paper.

2 Equation of conservative motions

2.1 The principle of virtual powers

The principle of virtual powers is a convenient way to obtain the equation of motions [34, 35]. A particle is identified in Lagrange's representation by a reference position \mathbf{X} of coordinates (X, Y, Z) belonging to reference configuration \mathcal{D}_0 ; its position is given in physical space \mathcal{D} by Euler's representation \mathbf{x} of coordinates (x, y, z) . The variations of particle motions are deduced from families of virtual motions of the fluid written as

$$\mathbf{X} = \psi(\mathbf{x}, t; \beta),$$

where β denotes a real parameter defined in the vicinity of 0, and the real motion corresponds to $\beta = 0$. Virtual displacements in reference configuration are associated with any variation of the real motion written as in [14],

$$\tilde{\delta}\mathbf{X} = \left. \frac{\partial \psi}{\partial \beta}(\mathbf{x}, t; \beta) \right|_{\beta=0}.$$

Variation $\tilde{\delta}$ is *dual* and mathematically equivalent to Serrin's variation denoted δ ([36], p. 145). It is important to note that - due to virtual displacement $\tilde{\delta}\mathbf{X}$ - the variation commutes with the derivative with respect to physical-space variable \mathbf{x} ($\tilde{\delta} \text{grad}^p \rho = \text{grad}^p \tilde{\delta} \rho$, $p \in \mathbb{N}$). Consequently, for complex fluids, $\tilde{\delta}$ -variation is straightforward and a lot simpler than δ -variation [14, 37].

Neglecting the body forces, the Lagrangian of the fluid writes,

$$L = \frac{1}{2} \rho \mathbf{v}^* \mathbf{v} - \rho \varepsilon,$$

where \mathbf{v} denotes the particle velocity. Conservative motions stationarize the Hamilton action

$$\mathcal{G} = \int_{\mathcal{D}} L \, dx, \quad (10)$$

where dx denotes the volume element in \mathcal{D} . The density satisfies the mass conservation

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho \mathbf{v}) = 0 \quad \Longleftrightarrow \quad \rho \det \mathbf{F} = \rho_0(\mathbf{X}) \quad (11)$$

with $\mathbf{F} \equiv \partial \mathbf{x} / \partial \mathbf{X}$, where ρ_0 is the reference density defined on \mathcal{D}_0 . The specific entropy verifies

$$\dot{s} = 0 \quad \Longleftrightarrow \quad s = s_0(\mathbf{X}), \quad (12)$$

where s_0 is defined on \mathcal{D}_0 and superposed dot denotes the material derivative.

Classical methods of variation calculus yield the variation of \mathcal{G} . Virtual displacements can be assumed to be null in the vicinity of the boundary of \mathcal{D}_0 and consequently, variations of integrated terms are null on the boundary \mathcal{D} . By using Stokes' formula, we can integrate by parts the variations of integral (10); from $\tilde{\delta}\mathcal{G} = (\partial \mathcal{G} / \partial \beta)|_{\beta=0}$, we get (see Appendix A for details)

$$\tilde{\delta}\mathcal{G} = \int_{\mathcal{D}} \left\{ \left[\frac{1}{2} \mathbf{v}^* \mathbf{v} - \rho \frac{\partial \alpha}{\partial \rho} - \alpha + \lambda \Delta \rho + \gamma \Delta^2 \rho \right] \tilde{\delta} \rho - \rho \frac{\partial \alpha}{\partial s} \tilde{\delta} s + \rho \mathbf{v}^* \tilde{\delta} \mathbf{v} \right\} dx.$$

Moreover:

Equation (11) implies

$$\tilde{\delta}\rho = \rho \operatorname{div}_0 \tilde{\delta}\mathbf{X} + \frac{1}{\det \mathbf{F}} \frac{\partial \rho_0}{\partial \mathbf{X}} \tilde{\delta}\mathbf{X}, \quad \text{where} \quad \operatorname{div}_0 \tilde{\delta}\mathbf{X} = \operatorname{tr} \left(\frac{\partial \tilde{\delta}\mathbf{X}}{\partial \mathbf{x}} \frac{\partial \mathbf{x}}{\partial \mathbf{X}} \right) \equiv \operatorname{tr} \left(\frac{\partial \tilde{\delta}\mathbf{X}}{\partial \mathbf{X}} \right).$$

Operator div_0 denotes the divergence operator in \mathcal{D}_0 .

The definition of the velocity implies

$$\frac{\partial \mathbf{X}(\mathbf{x}, t)}{\partial \mathbf{x}} \mathbf{v} + \frac{\partial \mathbf{X}(\mathbf{x}, t)}{\partial t} = 0,$$

and consequently,

$$\frac{\partial \tilde{\delta}\mathbf{X}}{\partial \mathbf{x}} \mathbf{v} + \frac{\partial \mathbf{X}}{\partial \mathbf{x}} \tilde{\delta}\mathbf{v} + \frac{\partial \tilde{\delta}\mathbf{X}}{\partial t} = 0 \quad \Longleftrightarrow \quad \tilde{\delta}\mathbf{v} = -F \dot{\tilde{\delta}\mathbf{X}}.$$

By denoting

$$H = \alpha + \frac{\mathcal{P}}{\rho}, \quad K = H - \lambda \Delta \rho - \gamma \Delta^2 \rho \quad \text{and} \quad m = \frac{1}{2} \mathbf{v}^* \mathbf{v} - K,$$

where \mathcal{P} is the thermodynamical pressure, we obtain

$$\tilde{\delta}\mathcal{G} = \int_{\mathcal{D}} \left[m \tilde{\delta}\rho - \rho (\mathbf{v}^* \mathbf{F}) \dot{\tilde{\delta}\mathbf{X}} - \rho T (\operatorname{grad}_0^* s_0) \tilde{\delta}\mathbf{X} \right] dx \quad \text{where} \quad \operatorname{grad}_0 s = \frac{\partial s_0(\mathbf{X})}{\partial \mathbf{X}}$$

and by integration by part on \mathcal{D}_0 ,

$$\tilde{\delta}\mathcal{G} = \int_{\mathcal{D}_0} \rho_0 \left[(\dot{\mathbf{v}^* \mathbf{F}}) - \operatorname{grad}_0^* m - T \operatorname{grad}_0^* s_0 \right] \tilde{\delta}\mathbf{X} dX.$$

Terms grad_0 and dX denote the gradient and the volume element in \mathcal{D}_0 , respectively.

Due to the principle of virtual work :

$$\text{For any displacement } \tilde{\delta}\mathbf{X} \text{ null on the edge of } \mathcal{D}_0, \tilde{\delta}\mathcal{G} = 0,$$

$$\text{we get } (\dot{\mathbf{v}^* \mathbf{F}}) = \operatorname{grad}_0^* m + T \operatorname{grad}_0^* s_0.$$

Noticing that $(\mathbf{a}^* + \mathbf{v}^* \frac{\partial \mathbf{v}}{\partial \mathbf{x}}) \mathbf{F} = (\dot{\mathbf{v}^* \mathbf{F}})$, where \mathbf{a} is the acceleration vector, we get

$$\mathbf{a} + \operatorname{grad} K - T \operatorname{grad} s = 0.$$

But,

$$dH = \frac{d\mathcal{P}}{\rho} + T ds$$

and consequently, the equation of motion writes

$$\rho \mathbf{a} + \operatorname{grad} \mathcal{P} - \lambda \rho \operatorname{grad} \Delta \rho - \gamma \rho \operatorname{grad} \Delta^2 \rho = 0. \quad (13)$$

2.2 Divergence form of the equation of motion

On one hand, we note

$$\sigma_1 \equiv \lambda \left[\frac{1}{2} (\text{grad } \rho)^2 + \rho \Delta \rho \right] \mathbf{I} - \lambda (\text{grad } \rho) (\text{grad}^* \rho)$$

Then,

$$\text{div } \sigma_1 = \lambda \left[\text{grad}^* \rho \frac{\partial \text{grad } \rho}{\partial \mathbf{x}} + \Delta \rho \text{ grad}^* \rho + \rho \text{ grad}^* \Delta \rho - \Delta \rho \text{ grad}^* \rho - \text{grad}^* \rho \frac{\partial \text{grad } \rho}{\partial \mathbf{x}} \right],$$

and consequently,

$$\text{div } \sigma_1 = \lambda \rho \text{ grad } \Delta \rho.$$

On the other hand,

$$\rho \text{ grad } \Delta^2 \rho = \text{grad}(\rho \Delta^2 \rho) - \text{div}^*[(\text{grad } \Delta \rho) \text{ grad}^* \rho] + \frac{\partial \text{grad } \rho}{\partial \mathbf{x}} \text{ grad } \Delta \rho \quad (14)$$

and after some calculations (See Appendix B),

$$\text{div } \sigma_2 = \gamma \rho \text{ grad } \Delta^2 \rho$$

with

$$\sigma_2 \equiv \gamma \left\{ \left[\rho \Delta^2 \rho - \frac{1}{2} \text{tr} \left(\frac{\partial \text{grad } \rho}{\partial \mathbf{x}} \right)^2 \right] \mathbf{I} + \left(\frac{\partial \text{grad } \rho}{\partial \mathbf{x}} \right)^2 - (\text{grad } \Delta \rho) \text{ grad}^* \rho \right\}.$$

The equation of motion can be written in divergence form :

$$\frac{\partial \rho \mathbf{v}^*}{\partial t} + \text{div} [\rho \mathbf{v} \mathbf{v}^* + \mathcal{P} \mathbf{I} - \sigma] = 0,$$

where

$$\begin{aligned} \sigma &\equiv \sigma_1 + \sigma_2 \\ &= \left[\frac{\lambda}{2} (\text{grad } \rho)^2 - \frac{\gamma}{2} \text{tr} \left(\frac{\partial \text{grad } \rho}{\partial \mathbf{x}} \right)^2 + \lambda \rho \Delta \rho + \gamma \rho \Delta^2 \rho \right] \mathbf{I} - (\lambda \text{ grad } \rho + \gamma \text{ grad } \Delta \rho) \text{ grad}^* \rho + \gamma \left(\frac{\partial \text{grad } \rho}{\partial \mathbf{x}} \right)^2 \end{aligned}$$

In fact, σ has the physical dimension of a stress tensor but is not a Cauchy stress tensor as we will notice in section 3.

3 Equation of energy

Multiplying Eq. (13) by \mathbf{v} , we get

$$\rho \mathbf{a}^* \mathbf{v} + (\text{grad } \mathcal{P})^* \mathbf{v} - \lambda \rho (\text{grad } \Delta \rho)^* \mathbf{v} - \gamma \rho (\text{grad } \Delta^2 \rho)^* \mathbf{v} = 0.$$

Due to Gibbs' identity, the volume energy of the homogeneous fluid yields

$$\rho T \dot{s} = \rho \frac{d\alpha}{dt} - \frac{\mathcal{P}}{\rho} \dot{\rho}.$$

Taking eqs (11) and (12) into account, we obtain

$$\rho \frac{d}{dt} \left[\frac{1}{2} \mathbf{v}^2 + \alpha - \lambda \Delta \rho - \gamma \Delta^2 \rho \right] + \text{div} (\mathcal{P} \mathbf{v}) + \rho \Delta \left[\lambda \frac{\partial \rho}{\partial t} + \gamma \Delta \left(\frac{\partial \rho}{\partial t} \right) \right] = 0,$$

with $\mathbf{v}^2 \equiv \mathbf{v}^* \mathbf{v} \equiv |\mathbf{v}|^2$, and

$$\begin{aligned} & \frac{\partial}{\partial t} \left[\rho \left(\frac{1}{2} \mathbf{v}^2 + \alpha - \lambda \Delta \rho - \gamma \Delta^2 \rho \right) \right] + \\ & \text{div} \left[\rho \left(\frac{1}{2} \mathbf{v}^2 + \alpha - \lambda \Delta \rho - \gamma \Delta^2 \rho \right) \mathbf{v} + \mathcal{P} \mathbf{v} \right] + \rho \Delta \left[\lambda \frac{\partial \rho}{\partial t} + \gamma \Delta \left(\frac{\partial \rho}{\partial t} \right) \right] = 0. \end{aligned}$$

Taking account of relations

$$-\lambda \Delta \rho \frac{\partial \rho}{\partial t} - \text{div} [\lambda \Delta \rho \rho \mathbf{v}] = \frac{\partial}{\partial t} \left[\frac{\lambda}{2} (\text{grad } \rho)^2 \right] - \text{div} \left(\lambda \Delta \rho \rho \mathbf{v} + \lambda \frac{\partial \rho}{\partial t} \text{grad } \rho \right)$$

and

$$-\gamma \Delta^2 \rho \frac{\partial \rho}{\partial t} - \text{div} [\gamma \Delta^2 \rho \rho \mathbf{v}] = -\frac{\partial}{\partial t} \left(\frac{\gamma}{2} (\Delta \rho)^2 \right) + \text{div} \left[\gamma \Delta \rho \text{grad } \frac{\partial \rho}{\partial t} - \gamma \left(\frac{\partial \rho}{\partial t} \right) \text{grad } \Delta \rho - \gamma \Delta^2 \rho \rho \mathbf{v} \right],$$

we obtain

$$\begin{aligned} & \frac{\partial}{\partial t} \left[\rho \left(\frac{1}{2} \mathbf{v}^2 + \alpha \right) + \frac{\lambda}{2} (\text{grad } \rho)^2 - \frac{\gamma}{2} (\Delta \rho)^2 \right] + \text{div} \left[\rho \left(\frac{1}{2} \mathbf{v}^2 + \alpha - \lambda \Delta \rho - \gamma \Delta^2 \rho \right) \mathbf{v} + \mathcal{P} \mathbf{v} \right] \\ & - \text{div} \left[\frac{\partial \rho}{\partial t} (\lambda \text{grad } \rho + \gamma \text{grad } \Delta \rho) - \gamma \Delta \rho \text{grad} \left(\frac{\partial \rho}{\partial t} \right) \right] = 0. \end{aligned} \quad (15)$$

Equation (15) is the balance equation of energy of the fourth-gradient fluid. Let us consider the specific energy in form (9), then the total volume energy of the fluid is,

$$e = \frac{1}{2} \rho \mathbf{v}^2 + \rho \alpha(\rho, s) + \frac{\lambda}{2} (\text{grad } \rho)^2 - \frac{\gamma}{2} (\Delta \rho)^2. \quad (16)$$

Term $\alpha + \mathcal{P}/\rho$ is the enthalpy of the homogeneous bulk, and

$$\mathcal{H} \equiv \rho \alpha + \frac{\mathcal{P}}{\rho} - \lambda \Delta \rho - \gamma \Delta^2 \rho$$

is the enthalpy of the fourth-gradient fluid. Let us note

$$\Xi \equiv \frac{\partial \rho}{\partial t} (\lambda \text{grad } \rho + \gamma \text{grad } \Delta \rho) - \gamma \Delta \rho \text{grad} \left(\frac{\partial \rho}{\partial t} \right),$$

then, balance equation of energy (15) becomes

$$\frac{\partial e}{\partial t} + \operatorname{div} \left[\left(\frac{1}{2} \mathbf{v}^2 + \mathcal{H} \right) \rho \mathbf{v} \right] - \operatorname{div} \Xi = 0. \quad (17)$$

In the special case of capillary fluids, Eq. (17) reduces to

$$\frac{\partial e_0}{\partial t} + \operatorname{div} [(e_0 - \sigma_1) \mathbf{v}] - \operatorname{div} (\lambda \dot{\rho} \operatorname{grad} \rho) = 0$$

where $\sigma_1 = -p \mathbf{I} - \lambda \operatorname{grad} \rho \operatorname{grad}^* \rho$ with $p = \mathcal{P} - \lambda (\operatorname{grad} \rho)^2 / 2 - \lambda \rho \Delta \rho$, corresponds to the *stress tensor*, $\lambda \dot{\rho} \operatorname{grad} \rho$ is the *interstitial working* vector and $e_0 = \frac{1}{2} \rho \mathbf{v}^2 + \rho \alpha(\rho, s) + \frac{\lambda}{2} (\operatorname{grad} \rho)^2$ is the total volume energy of the capillary fluid, respectively. Or, with

$$\mathcal{H}_0 \equiv \rho \alpha + \frac{\mathcal{P}}{\rho} - \lambda \Delta \rho, \quad \text{and} \quad \Xi_0 \equiv \lambda \frac{\partial \rho}{\partial t} \operatorname{grad} \rho,$$

$$\frac{\partial e_0}{\partial t} + \operatorname{div} \left[\left(\frac{1}{2} \mathbf{v}^2 + \mathcal{H}_0 \right) \rho \mathbf{v} \right] - \operatorname{div} \Xi_0 = 0,$$

which is specific to gradient fluids because σ_1 is not associated with a Cauchy stress tensor of an elastic medium.

4 Governing equations in symmetric form

The internal energy per unit volume of the fourth-gradient fluid is taken in the form

$$\rho \varepsilon(\rho, \eta, \mathbf{w}) = \epsilon(\rho, \eta) + \frac{\lambda |\mathbf{w}|^2}{2} - \frac{\gamma}{2} (\Delta \rho)^2,$$

where $\epsilon = \rho \alpha$, $\mathbf{w} = \operatorname{grad} \rho$ and $\eta = \rho s$ is the entropy per unit volume. *Homogeneous* internal energy per unit volume ϵ satisfies the Gibbs identity,

$$T d\eta = d\epsilon - \mu d\rho$$

where $\mu = (\epsilon + \mathcal{P} - T\eta)/\rho$ is the chemical potential of the fluid bulk. The governing equations of the fourth-gradient fluid write in the form

$$\left\{ \begin{array}{l} \frac{\partial \rho}{\partial t} + \operatorname{div} \mathbf{j} = 0 \\ \frac{\partial \eta}{\partial t} + \operatorname{div} \left(\frac{\eta}{\rho} \mathbf{j} \right) = 0 \\ \frac{\partial \mathbf{j}^*}{\partial t} + \operatorname{div} \left(\frac{\mathbf{j} \mathbf{j}^*}{\rho} + \mathcal{P} \mathbf{I} \right) - \lambda \rho \operatorname{grad}^* (\operatorname{div} \mathbf{w}) - \gamma \rho \operatorname{grad}^* \Delta^2 \rho = \mathbf{0}^* \end{array} \right. \quad (18)$$

where $\mathbf{j} \equiv \rho \mathbf{v}$. The gradient of the mass conservation law verifies another conservation law,

$$\frac{\partial \mathbf{w}}{\partial t} + \operatorname{grad} \operatorname{div} \mathbf{j} = 0. \quad (19)$$

Conversely, if we consider \mathbf{w} as an independent variable, and if we add the initial condition

$$\mathbf{w} |_{t=0} = \operatorname{grad} \rho |_{t=0},$$

$\mathbf{w} = \text{grad } \rho$ is a consequence of the governing equations.

Similarly, we denote $a = \Delta \rho$ the Laplace operator, the mass conservation equation yields,

$$\frac{\partial a}{\partial t} + \Delta(\text{div } \mathbf{j}) = 0.$$

Conversely, if we add the initial condition

$$a|_{t=0} = \Delta \rho|_{t=0},$$

we can consider a as an independent variable.

Finally, we obtain the system of equations (18) in the following equivalent non-divergence form

$$\left\{ \begin{array}{l} \frac{\partial \rho}{\partial t} + \text{div } \mathbf{j} = 0 \\ \frac{\partial \eta}{\partial t} + \text{div} \left(\frac{\eta}{\rho} \mathbf{j} \right) = 0 \\ \frac{\partial \mathbf{j}^*}{\partial t} + \text{div} \left(\frac{\mathbf{j} \mathbf{j}^*}{\rho} + \mathcal{P} \mathbf{I} \right) - \lambda \rho \text{grad}^* (\text{div } \mathbf{w}) - \gamma \rho \text{grad}^* \Delta^2 \rho = \mathbf{0}^* \\ \frac{\partial \mathbf{w}}{\partial t} + \text{grad } \text{div } \mathbf{j} = 0 \\ \frac{\partial a}{\partial t} + \Delta(\text{div } \mathbf{j}) = 0 \end{array} \right. \quad (20)$$

Remark: We choose energy equation (15) as supplementary equation. In usual thermodynamical theories the energy equation is a part of the system and the entropy balance equation is taken as a supplementary equation (entropy principle). In the case of weak solutions, the fact is very important; in particular, for shock waves, the entropy is growing across the shock. But when we consider classical solutions, we can, without losing generality, switch roles of entropy and energy.

The theory of capillary usually applied for van der Waals-like fluids can be extended to fourth-gradient fluids. For such fluids the energy $\epsilon(\rho, \eta)$ is not convex for all values of ρ and η . We assume that we are in the vicinity of an equilibrium state (ρ_e, η_e) where the energy function is locally convex.

With $\mathbf{u} \equiv (\rho, \eta, \mathbf{j}^*, \mathbf{w}^*, a)^*$ and $h^0 \equiv e$ (given by Eq. (16)), from Eq. (2)¹ we deduce the main field

$$\mathbf{u}' \equiv (q, \theta, \mathbf{v}', \mathbf{r}^*, b)^*$$

coming from

$$\begin{aligned} de &= \left(\mu - \frac{|\mathbf{v}|^2}{2} \right) d\rho + T d\eta + \mathbf{v}^* d\mathbf{j} + \lambda \mathbf{w}^* d\mathbf{w} - \gamma a da \\ &= q d\rho + \theta d\eta + \mathbf{v}'^* d\mathbf{j} + \mathbf{r}^* d\mathbf{w} + b da \end{aligned}$$

and therefore

$$q = \mu - \frac{|\mathbf{v}|^2}{2}, \quad \theta = T, \quad \mathbf{v}' = \mathbf{v}, \quad \mathbf{r}^* = \lambda \text{grad}^* \rho \quad \text{and} \quad b = -\gamma \Delta \rho.$$

Legendre transformation $h'^0 = \Pi$ of total energy $h^0 = e$ given by Eq. (2)² is

$$\Pi = \rho q + \eta T + \mathbf{j}^* \mathbf{v} + \mathbf{w}^* \mathbf{r} + a b - E = \mathcal{P} + \frac{|\mathbf{r}|^2}{2\lambda} - \frac{b^2}{2\gamma},$$

where thermodynamic pressure \mathcal{P} is considered as a function of q , θ and \mathbf{v} . Therefore, from Eq. (3)¹ we get

$$\frac{\partial \Pi}{\partial q} = \rho, \quad \frac{\partial \Pi}{\partial T} = \eta, \quad \frac{\partial \Pi}{\partial \mathbf{u}} = \mathbf{j}^*, \quad \frac{\partial \Pi}{\partial \mathbf{r}} = \mathbf{w}^*, \quad \frac{\partial \Pi}{\partial b} = a,$$

If we introduce matrix $\mathbf{B} \equiv -\gamma \frac{\partial \text{grad} \rho}{\partial \mathbf{x}}$, System (20) can be rewritten as a symmetric form (7) in which the hyperbolic part is in the form (4), (5) :

$$\left\{ \begin{array}{l} \frac{\partial}{\partial t} \left(\frac{\partial \Pi}{\partial q} \right) + \text{div} \left[\frac{\partial(\Pi \mathbf{v})}{\partial q} \right] = 0 \\ \frac{\partial}{\partial t} \left(\frac{\partial \Pi}{\partial T} \right) + \text{div} \left[\frac{\partial(\Pi \mathbf{v})}{\partial T} \right] = 0 \\ \frac{\partial}{\partial t} \left(\frac{\partial \Pi}{\partial \mathbf{v}} \right) + \text{div} \left[\frac{\partial(\Pi \mathbf{v})}{\partial \mathbf{v}} - \frac{\partial \Pi}{\partial q} \frac{\partial \mathbf{r}}{\partial \mathbf{x}} + \left\{ \frac{1}{2} \frac{\partial \Pi}{\partial b} b - \frac{1}{2} \text{tr} \left(\mathbf{B} \frac{\partial}{\partial \mathbf{x}} \left(\frac{\partial}{\partial \mathbf{x}} \frac{\partial \Pi}{\partial q} \right)^* \right) \right. \right. \\ \left. \left. + \frac{\partial \Pi}{\partial q} \text{tr} \left(\frac{\partial}{\partial \mathbf{x}} \left(\frac{\partial}{\partial \mathbf{x}} \frac{\partial \Pi}{\partial b} \right)^* \right) \right\} \mathbf{I} - (\text{grad} b) \text{grad}^* \left(\frac{\partial \Pi}{\partial q} \right) + \mathbf{B} \frac{\partial}{\partial \mathbf{x}} \left(\frac{\partial(\frac{\partial \Pi}{\partial q})}{\partial \mathbf{x}} \right)^* \right] = 0 \\ \frac{\partial}{\partial t} \left(\frac{\partial \Pi}{\partial \mathbf{r}} \right) + \text{div} \left[\frac{\partial(\Pi \mathbf{v})}{\partial \mathbf{r}} + \frac{\partial \Pi}{\partial q} \frac{\partial \mathbf{v}}{\partial \mathbf{x}} \right] = 0 \\ \frac{\partial}{\partial t} \left(\frac{\partial \Pi}{\partial b} \right) + \text{div} \left[\frac{\partial}{\partial \mathbf{x}} \left(\text{tr} \left\{ \frac{\partial}{\partial \mathbf{x}} \left(\frac{\partial(\Pi)}{\partial \mathbf{v}} \right) \right\} \right)^* \right] = 0, \end{array} \right. \quad (21)$$

Therefore, the system has a Cauchy problem well posed according with the general results proved in [10,11] for hyperbolic-parabolic systems in form (7). If the capillary coefficients λ and γ are zero, $\Pi = \mathcal{P}$ and we get gas-dynamics' equation and the symmetric hyperbolic form of Godunov [1].

5 Stability of constant states

System (21) admits constant solutions $(\rho_e, \eta_e, \mathbf{v}_e, \mathbf{w}_e = \mathbf{0}, a_e = 0)$. Since the governing equations are invariant under Galilean transformation, we can assume that $\mathbf{v}_e = \mathbf{0}$.

Near equilibrium, we look for the solutions of the linearized system proportional to $e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}$, ($i^2 = -1$, $\mathbf{k}^* \mathbf{k} = 1$) :

$$\mathbf{u} = \mathbf{u}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)} \quad \text{with} \quad \mathbf{u}^* = [q, T, \mathbf{v}, \mathbf{r}, b] \quad \text{and} \quad \mathbf{u}_0^* = [q_0, T_0, \mathbf{u}_0, \mathbf{r}_0, b_0].$$

We obtain

$$\frac{\partial}{\partial t} \left(\frac{\partial \Pi}{\partial \mathbf{u}} \right)_e = \frac{\partial}{\partial \mathbf{u}} \left(\frac{\partial \Pi}{\partial \mathbf{u}} \right)_e \frac{\partial \mathbf{u}}{\partial t} = -i \omega \frac{\partial}{\partial \mathbf{u}} \left(\frac{\partial \Pi}{\partial \mathbf{u}} \right)_e \mathbf{u}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}$$

where subscript e means at equilibrium and we note $\mathbf{m}^* = [q, T, \mathbf{v}, \mathbf{r}]$ and $\mathbf{m}_0^* = [q_0, T_0, \mathbf{v}_0, \mathbf{r}_0]$ such that $\mathbf{u}^* = [\mathbf{m}^*, b]$ and $\mathbf{u}_0^* = [\mathbf{m}_0^*, b_0]$.

$$\text{div} \left(\frac{\partial \Pi \mathbf{v}}{\partial \mathbf{m}} \right) = \sum_{j=1}^3 \left[\left(\frac{\partial \Pi v^j}{\partial \mathbf{m}} \right)_{,x^j} \right]^* = \sum_{j=1}^3 \frac{\partial}{\partial \mathbf{m}} \left(\frac{\partial \Pi v^j}{\partial \mathbf{m}} \right)^* \frac{\partial \mathbf{m}}{\partial x^j},$$

and at equilibrium,

$$\operatorname{div} \left(\frac{\partial \Pi \mathbf{v}}{\partial \mathbf{m}} \right)_e = \sum_{j=1}^3 i F^j k_j \mathbf{m}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)},$$

where

$$F^j \equiv \frac{\partial}{\partial \mathbf{m}} \left(\frac{\partial \Pi v^j}{\partial \mathbf{m}} \right)_e^* \quad \text{and we note} \quad F \equiv \sum_{j=1}^3 F^j k_j.$$

• To Eq. (20)³ (or equivalently Eq. (21)³), we must add two terms with respect to classical fluids' equations :

First term,

$$-\lambda \rho \operatorname{grad}^* (\operatorname{div} \mathbf{w}) = -\rho \operatorname{div} \left(\frac{\partial \mathbf{r}}{\partial \mathbf{x}} \right) \quad \text{with} \quad \frac{\partial \mathbf{r}}{\partial \mathbf{x}} = i \mathbf{r}_0 \mathbf{k}^* e^{i(\mathbf{k}^* \mathbf{x} - \omega t)},$$

where $\mathbf{r} = \mathbf{r}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}$; consequently,

$$[-\lambda \rho \operatorname{grad}^* (\operatorname{div} \mathbf{w})]_e = \rho_e \mathbf{r}_0^* \mathbf{k} \mathbf{k}^* e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}.$$

Second term,

$$-\gamma \rho \operatorname{grad}^* \Delta^2 \rho = \rho \operatorname{grad}^* (\operatorname{div} \operatorname{grad} b) \quad \text{with} \quad b = b_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}.$$

But

$$-\gamma \Delta^2 \rho = \operatorname{div} \operatorname{grad} b = b_0 i^2 \mathbf{k}^* \mathbf{k} e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}.$$

Then, at equilibrium,

$$-\gamma \rho \operatorname{grad}^* \Delta^2 \rho = -i \rho_e b_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)} \mathbf{k}^*.$$

• To Eq. (21)⁴ at equilibrium, we must add the term,

$$\operatorname{div} \left[\frac{\partial \Pi}{\partial q} \frac{\partial \mathbf{v}}{\partial \mathbf{x}} \right]_e = -\rho_e \mathbf{v}_0^* \mathbf{k} \mathbf{k}^* e^{i(\mathbf{k}^* \mathbf{x} - \omega t)} \quad \text{with} \quad \mathbf{v} = \mathbf{v}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}.$$

• To Eq. (20)⁵ (or equivalently Eq. (21)⁵), we must add the term,

$$\begin{aligned} \Delta(\operatorname{div} \mathbf{j}) &= \operatorname{divgrad} [(\operatorname{grad} \rho)^* \mathbf{v} + \rho \operatorname{div} \mathbf{v}] = \\ \operatorname{div} \left[\left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^* \right] \mathbf{v} &+ \operatorname{tr} \left[\left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^* \frac{\partial \mathbf{v}}{\partial \mathbf{x}} + \left(\frac{\partial \mathbf{v}}{\partial \mathbf{x}} \right)^* \frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right] + \operatorname{div} \left[\left(\frac{\partial \mathbf{v}}{\partial \mathbf{x}} \right)^* \right] \operatorname{grad} \rho \\ &+ (\operatorname{div} \mathbf{v}) (\operatorname{divgrad} \rho) + \frac{\partial \operatorname{div} \mathbf{v}}{\partial \mathbf{x}} \operatorname{grad} \rho + \rho \Delta(\operatorname{div} \mathbf{v}) + \operatorname{grad}^* \rho \operatorname{grad}(\operatorname{div} \mathbf{v}). \end{aligned}$$

At equilibrium, near $\rho = \rho_e$, the only remaining term is $\rho_e \Delta(\operatorname{div} \mathbf{v})$, and taking $\mathbf{v} = \mathbf{v}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}$ into account, we obtain

$$\rho_e \Delta(\operatorname{div} \mathbf{v}) = -i \rho_e \mathbf{k}^* \mathbf{v}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)}.$$

Let us denote

$$\mathbf{A} = \frac{\partial}{\partial \mathbf{u}} \left[\left(\frac{\partial \Pi}{\partial \mathbf{u}} \right)^* \right]_e, \quad \mathbf{G} = \begin{bmatrix} F & \mathbf{0} \\ \mathbf{0}^* & 0 \end{bmatrix},$$

where $\underline{\mathbf{0}}$ and $\underline{\mathbf{0}}^*$ are column and row matrices with nine zeros: $\underline{\mathbf{0}}^* = [0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0]$,

$$\mathbf{H} = \rho_e \begin{bmatrix} 0 & 0 & \mathbf{0}^* & \mathbf{0}^* & 0 \\ 0 & 0 & \mathbf{0}^* & \mathbf{0}^* & 0 \\ \mathbf{0} & \mathbf{0} & \mathbf{0}_3 & -i\mathbf{k}\mathbf{k}^* & -\mathbf{k} \\ \mathbf{0} & \mathbf{0} & i\mathbf{k}\mathbf{k}^* & \mathbf{0}_3 & \mathbf{0} \\ 0 & 0 & -\mathbf{k}^* & \mathbf{0}^* & 0 \end{bmatrix} \quad \text{with} \quad \mathbf{0}_3 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad \text{and} \quad \mathbf{0}^* = [0\ 0\ 0].$$

Due to $\overline{\mathbf{H}}^* = \mathbf{H}$, matrices \mathbf{G} and \mathbf{H} are Hermitian and the perturbations of system (21) verify

$$i[\mathbf{C} - \omega\mathbf{A}] \mathbf{u}_0 e^{i(\mathbf{k}^* \mathbf{x} - \omega t)} = \mathbf{0},$$

where $\mathbf{C} \equiv \mathbf{G} + \mathbf{H}$ and \mathbf{A} are Hermitian and symmetric matrices, respectively. Consequently, ω -values are the roots of the characteristic equation

$$\det[\mathbf{C} - \omega\mathbf{A}] = 0,$$

where ω are the eigenvalues of \mathbf{C} with respect to \mathbf{A} and \mathbf{u}_0 are the corresponding eigenvectors. Hence, ω is real if \mathbf{A} is positive definite.

6 Conclusion

The fourth-gradient model of capillarity yields a conservation energy equation. By a Legendre transformation of energy variables, its quasi-linear system of conservation laws can be symmetrized in the sense of Hermitian matrices.

This result extends the simplest case of capillarity with second-gradient model [12] and the problem of stability of fluids in gradient theories for mass density.

A Useful formulae

$$\rho \operatorname{div}(\operatorname{grad} \rho) = \operatorname{div}(\rho \operatorname{grad} \rho) - (\operatorname{grad} \rho)^2.$$

Term $\operatorname{div}(\rho \operatorname{grad} \rho)$ can be integrated on the boundary of \mathcal{D} and consequently $-\tilde{\delta} \left(\frac{\lambda}{2} (\operatorname{grad} \rho)^2 \right)$ corresponds in \mathcal{D} to

$$-\lambda \operatorname{grad}^* \rho \operatorname{grad} \tilde{\delta} \rho = -\lambda \operatorname{div}(\tilde{\delta} \rho \operatorname{grad} \rho) + \lambda \operatorname{div}(\operatorname{grad} \rho) \tilde{\delta} \rho.$$

Term $\operatorname{div}(\tilde{\delta} \rho \operatorname{grad} \rho)$ can be integrated on the boundary of \mathcal{D} and the variation of $\frac{\lambda}{2} \rho \Delta \rho$ is $\lambda \Delta \rho \tilde{\delta} \rho$.

In a similar way,

$$\rho \operatorname{div}[\operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)] = \operatorname{div}[\rho \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)] - \operatorname{grad}^* \rho \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho).$$

but, $\operatorname{div}[\rho \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)]$ can be integrated on the boundary of \mathcal{D} and

$$-\operatorname{grad}^* \rho \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho) = -\operatorname{div}[(\operatorname{div} \operatorname{grad} \rho) \operatorname{grad} \rho] + (\operatorname{div} \operatorname{grad} \rho)^2.$$

Integrating on the boundary of \mathcal{D} term $-\operatorname{div}[(\operatorname{div} \operatorname{grad} \rho) \operatorname{grad} \rho]$, and considering that variation of $\rho \operatorname{div}[\operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)]$ is the same as variation of $(\operatorname{div} \operatorname{grad} \rho)^2$, we obtain

$$2 (\operatorname{div} \operatorname{grad} \rho) (\operatorname{div} \operatorname{grad} \tilde{\delta} \rho) = 2 \operatorname{div}[(\operatorname{div} \operatorname{grad} \rho) \operatorname{grad} \tilde{\delta} \rho] - 2 \operatorname{grad}^*(\operatorname{div} \operatorname{grad} \rho) \operatorname{grad} \tilde{\delta} \rho.$$

Term $2 \operatorname{div}[(\operatorname{div} \operatorname{grad} \rho) \operatorname{grad} \tilde{\delta} \rho]$ can be integrated on the boundary of \mathcal{D} and

$$-2 \operatorname{grad}^*(\operatorname{div} \operatorname{grad} \rho) \operatorname{grad} \tilde{\delta} \rho = -2 \operatorname{div}[\tilde{\delta} \rho \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)] + 2 [\operatorname{div} \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)] \tilde{\delta} \rho.$$

Term $-2 \operatorname{div}[\tilde{\delta} \rho \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)]$ can be integrated on the boundary of \mathcal{D} and variation of $\frac{\gamma}{2} \rho \Delta^2 \rho$ is

$$\gamma [\operatorname{div} \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)] \tilde{\delta} \rho = \gamma (\Delta^2 \rho) \tilde{\delta} \rho.$$

B Additive calculations to Subsection 2.2

In Rel. (14) we have to study term $\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho)$. Due to

$$\operatorname{grad}(\operatorname{div} \operatorname{grad} \rho) = \operatorname{div}^* \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right) \quad \text{and} \quad \frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} = \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^*,$$

$$\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho) = \left[\operatorname{div} \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right) \frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right]^*$$

Each term of covector $\operatorname{div} \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right) \frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}}$ is in the form $\{\rho_{,kkj} \rho_{,jl}\}$.

From

$$\rho_{,kkj} \rho_{,jl} = (\rho_{,lj} \rho_{,jk})_{,k} - \rho_{,ljk} \rho_{,jk}$$

and

$$(\rho_{,jk} \rho_{,jk})_{,l} = \rho_{,jkl} \rho_{,jk} + \rho_{,jlk} \rho_{,jk},$$

together with the Schwarz theorem we get

$$\rho_{,ljk} \rho_{,jk} = \frac{1}{2} (\rho_{,jk} \rho_{,jk})_{,l}$$

which are the elements of $\frac{1}{2} \frac{\partial}{\partial \mathbf{x}} \left[\operatorname{tr} \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^2 \right]$. But $(\rho_{,lj} \rho_{,jk})_{,k}$ are the elements of $\operatorname{div} \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^2$. Consequently,

$$\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \operatorname{grad}(\operatorname{div} \operatorname{grad} \rho) = \operatorname{div}^* \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^2 - \frac{1}{2} \operatorname{grad} \left[\operatorname{tr} \left(\frac{\partial \operatorname{grad} \rho}{\partial \mathbf{x}} \right)^2 \right].$$

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