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# Properties of thermocapillary fluids and symmetrization of motion equations

Henri Gouin

Aix-Marseille Univ, CNRS, Centrale Marseille, M2P2 UMR 7340, 13451 Marseille, France

## Abstract

The equations of fluid motions are considered in the case of internal energy depending on mass density, volume entropy and their spatial derivatives. The model corresponds to domains with large density gradients in which the temperature is not necessary uniform. In this new general representation writes in symmetric form with respect to the mass and entropy densities. For conservative motions of perfect *thermocapillary fluids*, Kelvin's circulation theorems are always valid. Dissipative cases are also considered; we obtain the balance of energy and we prove that equations are compatible with the second law of thermodynamics. The internal energy form allows to obtain a Legendre transformation inducing a quasi-linear system of conservation laws which can be written in a divergence form and the stability near equilibrium positions can be deduced. The result extends classical hyperbolicity theory for governing-equations' systems in hydrodynamics, but symmetric matrices are replaced by Hermitian matrices.

*Key words:* Thermocapillarity; second gradient models; fluid interfaces; hyperbolicity

# 1 Introduction

Theoretical and experimental studies show that, when working far from critical conditions, the liquid-vapour capillary layer has a few molecular-beams' thickness [1–3]. Consequently, liquid-vapour interfaces are generally represented by

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*Email address:* henri.gouin@univ-amu.fr;henri.gouin@yahoo.fr (Henri Gouin).

material surfaces endowed with surface energy related to Laplace's surfacetension [4]. The surfaces have their own characteristic behaviours and energy properties [5]. In interfacial layers, molecular models - as used in kinetic theory of gases - express behaviours associated with non-convex internal energies [6– 9]. These models appear advantageous as they provide a more precise verification of Maxwell's rule applied to isothermal phase-transitions [8,10]. Nonetheless, they present two disadvantages. First, for densities that lie between bulk densities, the pressure may become negative. However, simple physical experiments can be used to cause traction that leads to negative pressure values [11]. Second, in the field between bulks, internal energies cannot be represented by convex surfaces associated with the variation of densities. The fact seems to contradict the existence of equilibrium states. To overcome the disadvantages, the thermodynamics replaces the non-convex portions corresponding to internal energies by planar domains [12]; the fluid can no longer be considered as a continuous medium.

At equilibrium, an appropriate modification of the layer stress-tensor, expressed in an anisotropic form, can eliminate the previous disadvantages; then, the continuous-medium energies change [6,9] and near the critical point, allow to study interfaces of non-molecular size [13,14]. The approach is not new and dates back to van der Walls [15,16] and Korteweg [17]; it corresponds to what is known as a Landau-Ginzburg theory [7]. The contradiction between Korteweg's classical stress theory and the Clausius-Duhem inequality [18] is corrected by Eglit [19], Dunn and Serrin [20], Casal and Gouin [21].

To study capillary layers and bulks, the second gradient theory [22,23] - conceptually simpler than Laplace's theory - led to a capillary model for isothermal liquid-vapour interfaces. Fluids endowed with internal capillarity yield equations of motion and energy including additive terms. The internal energy of such fluids is a function of the entropy, the mass density and the gradient of mass density [24-26]. Gradient theory can be extended to solid mechanics, materials, nanofluidics, fluid mixtures [27-32] and developed at *n*-order (n > 2) [33].

The simplest model in continuum mechanics considers a volume internal energy  $\varepsilon$  as the sum of two terms: a first one  $\varepsilon_0$  corresponding to a medium with uniform composition equal to the local one and a second one associated with the non-uniformity of the fluid and is approximated by a gradient expansion, typically truncated to the second order [6,15]:

$$\varepsilon = \varepsilon_0(\rho, \eta) + \frac{1}{2} m (\operatorname{grad} \rho)^2,$$

where  $\rho$  is the mass density (or volume mass),  $\eta$  the volume entropy,  $\varepsilon_0$  the volume internal-energy of the fluid assumed to be homogeneous and m is a coefficient independent of  $\eta$ , grad  $\rho$  and of any higher derivatives [9]. In such a model,  $\eta$  varies with  $\rho$  through isothermal interface in the same way as in

bulks and at given temperature T satisfies

$$\frac{\partial \varepsilon_0}{\partial \eta}(\rho, \eta) = T,\tag{1}$$

so,  $\varepsilon_0 = \varepsilon_0(\rho, T)$ . At given temperature T, the points representing phase states in the  $(\rho, \eta, \varepsilon_0)$  space lie on a curve instead of surface  $\varepsilon_0 = \varepsilon_0(\rho, \eta)$ . In fact, the assumption is not exact for realistic potentials; in practice the potential for the two-density form of van der Waals' theory is not constructed by prescription (1) but by other means [9] (Ch. 8). Aside from the question of accuracy, there are qualitative features of some interfaces, especially in systems of more than one component, that require two or more independently varying densities - entropy included - for their description; in fact, when we have non-monotonic behaviours, one-density models inevitably lead to monotonic variations of densities [8]. In our case, the model must be extended by taking account of not only the strong variations of matter density through interfacial layers but also the strong variations of entropy. For this purpose, Rowlinson and Widom in [9] (Ch. 3 and Ch. 9) introduced an energy arising from the mean-field theory and depending on the deviations of densities  $\rho$  and  $\eta$  from their values at the critical point and on the gradients of densities. Consequently, we can also imagine non-isothermal steady motions in zones with large density gradients [34].

The paper is presented as follows :

In Section 2, we consider different forms of equation of motions in the most general case. The Hamilton principle yields the equation of conservative motions in a symmetric form with respect to mass and entropy volumes. In [35] we considered thermocapillary fluids as fluids with a specific internal energy in the form  $\alpha = \alpha(\rho, s, \text{grad } \rho, \text{grad } s)$  where s is the specific entropy. But, it is more convenient to consider the volume entropy in place of the specific entropy to obtain a simpler system of equations.

In Section 3, we extend the balance equations to viscous fluids. The equation of energy is completed with a heat flux and a heat supply. We get an additive *interstitial-working term* similar to a heat-flux vector and the processes' equations are compatible with the second law of thermodynamics.

In Section 4, we revisit Kelvin's circulation-theorem and analyse the surface tension of planar interfaces at equilibrium. The Maxwell rule is extended for thermocapillary fluids.

Section 5 is a completely new study. A Legendre transformation yields a system of equations in a divergence form when conjugated variables - with respect to the mass density, volume entropy and their gradients - are used. The hyperbolicity of the system of governing equations can be studied. Small perturbations near an equilibrium position are analysed. Eigenvalues associated with Hermitian matrices conclude to the stability of equilibrium positions by extending Godunov and Lax-Friedrichs analyses [36,37].

A conclusion ends the paper.

For any vectors  $\boldsymbol{a}, \boldsymbol{b}$  we use the notation  $\boldsymbol{a}^*\boldsymbol{b}$  for the scalar product (the line vector is multiplied by the column vector) and  $\boldsymbol{a} \ \boldsymbol{b}^*$  for the tensor product (or  $\boldsymbol{a} \otimes \boldsymbol{b}$  the column vector is multiplied by the line vector), where superscript \* denotes the transposition. Divergence of a linear transformation  $\boldsymbol{D}$  is the covector div  $\boldsymbol{D}$  such that, for any constant vector  $\boldsymbol{c}$ ,  $(\operatorname{div} \boldsymbol{D}) \boldsymbol{c} = \operatorname{div}(\boldsymbol{D} \boldsymbol{c})$ . The identical transformation is denoted by  $\boldsymbol{I}$ .

# 2 Equation of motions

## 2.1 Preliminaries

The volume internal energy of a thermocapillary fluid is represented by a development in *gradients* with respect to  $\rho$  and  $\eta$ :

$$\varepsilon = \varepsilon(\rho, \eta, \operatorname{grad} \rho, \operatorname{grad} \eta).$$
 (2)

A particular case of volume internal energy can be

$$\varepsilon = \varepsilon_0(\rho, \eta) + \frac{1}{2} \left( C \left( \operatorname{grad} \rho \right)^2 + 2 D \left( \operatorname{grad} \rho \right)^* \operatorname{grad} \eta + E \left( \operatorname{grad} \eta \right)^2 \right), \quad (3)$$

where C, D, E are assumed to be constant; in special case D = 0 and E = 0, we get Cahn and Hilliard's fluids [6].

• Thermodynamical potential  $\varepsilon_0(\rho, \eta) = \rho \alpha(\rho, s)$  is the volume internal energy of the fluid bulk with volume mass  $\rho$  and volume entropy  $\eta$  (the same potential expression as for compressible fluids). Consequently,

$$d\alpha(\rho, s) = \frac{P}{\rho^2} d\rho + T \, ds \,,$$

where P is the thermodynamical pressure and T the Kelvin temperature. Then,

$$d\varepsilon_0 = \mu_0 \, d\rho + T \, d\eta \,,$$

where  $\mu_0 = \frac{\partial \varepsilon_0(\rho, \eta)}{\partial \rho}$  is the bulk chemical-potential. We get

$$P = \rho \,\mu_{_{0}} + \eta \,T - \varepsilon_{_{0}}$$

and

$$\eta \, dT = dP - \rho \, d\mu_{\rm o}.$$

• For thermocapillary fluids associated with Eq. (2) we denote,

$$d\varepsilon = \mu \, d\rho + \mathcal{T} \, d\eta + \Phi^* d(\operatorname{grad} \rho) + \Psi^* d(\operatorname{grad} \eta)$$
  
with  $\mu = \frac{\partial \varepsilon}{\partial \rho}, \quad \mathcal{T} = \frac{\partial \varepsilon}{\partial \eta}, \quad \Phi^* = \frac{\partial \varepsilon}{\partial \operatorname{grad} \rho}, \quad \Psi^* = \frac{\partial \varepsilon}{\partial \operatorname{grad} \eta}.$  (4)

We always denote

 $\mathcal{P} = \rho \, \mu + \eta \, \mathcal{T} - \varepsilon,$ 

where  $\mathcal{P}$  is called the *thermocapillary pressure*,  $\mu$  and  $\mathcal{T}$  are extended by Eq. (4) as the *thermocapillary chemical-potential* and the *thermocapillary temperature*, respectively.

In the particular case of Eq. (3) we obtain,

$$\begin{split} \mu &= \frac{\partial \varepsilon_0}{\partial \rho} \,, \quad \mathcal{T} = \frac{\partial \varepsilon_0}{\partial \eta} \,, \\ \Phi^\star &= C(\operatorname{grad} \rho)^\star + D(\operatorname{grad} \eta)^\star \quad \text{and} \quad \Psi^\star = D(\operatorname{grad} \rho)^\star + E(\operatorname{grad} \eta)^\star \,, \end{split}$$

where  $\mu \equiv \mu_0$  and  $\mathcal{T} \equiv T$  are also the chemical potential and the temperature of bulks.

# 2.2 The Hamilton principle [38]

The mass conservation writes :

$$\frac{\partial \rho}{\partial t} + \operatorname{div} \rho \boldsymbol{u} = 0.$$
 (5)

For isentropic motions, the volume entropy conservation writes :

$$\frac{\partial \eta}{\partial t} + \operatorname{div} \eta \boldsymbol{u} = 0. \tag{6}$$

The Hamilton action between time  $t_1$  and time  $t_2$  is [39,40],

$$S = \int_{t_1}^{t_2} \int_{D_t} L \, dv \, dt \quad \text{with} \quad L = \frac{1}{2} \, \rho \, \boldsymbol{u}^* \boldsymbol{u} - \varepsilon - \rho \, \Omega$$

where L is the Lagrangian, dv is the volume element of physical space  $D_t$ at time t, dt is the time differential,  $\boldsymbol{u}$  is the fluid velocity-vector and  $\Omega$  the external-force potential. We have the properties associated with the variations of  $\boldsymbol{u}, \rho$  and  $\eta$ 

$$\delta \boldsymbol{u} = \frac{d\boldsymbol{\zeta}}{dt}, \quad \delta \rho = -\rho \operatorname{div} \boldsymbol{\zeta}, \quad \delta \eta = -\eta \operatorname{div} \boldsymbol{\zeta}, \tag{7}$$

where  $\boldsymbol{\zeta} = \delta \boldsymbol{x}$  notes the variation of Euler position  $\boldsymbol{x}$  as defined by Serrin in [41]. Equation (7<sup>3</sup>) corresponds to an isentropic variation when the motion is conservative and isentropic.

Thanks to Eqs. (4-7), the variation of Hamilton's action is [42],

$$\delta S = \int_{t_1}^{t_2} \int_{D_t} \left[ \rho \, \boldsymbol{u}^* \frac{d\boldsymbol{\zeta}}{dt} + (\rho \, \boldsymbol{\mu} - \varepsilon + \eta \, \mathcal{T}) \operatorname{div} \boldsymbol{\zeta} \right. \\ \left. - \delta(\operatorname{grad} \rho)^* \, \boldsymbol{\Phi} - \delta(\operatorname{grad} \eta)^* \, \boldsymbol{\Psi} - \rho \, \frac{\partial \Omega}{\partial \boldsymbol{x}} \, \boldsymbol{\zeta} \right] dv \, dt.$$

Relations :

$$(\rho \mu - \varepsilon + \eta \mathcal{T}) \operatorname{div} \boldsymbol{\zeta} = \operatorname{div}[(\rho \mu - \varepsilon + \eta \mathcal{T}) \boldsymbol{\zeta}] \\ + \left[ \boldsymbol{\Phi}^{\star} \frac{\partial \operatorname{grad} \rho}{\partial \boldsymbol{x}} + \boldsymbol{\Psi}^{\star} \frac{\partial \operatorname{grad} \eta}{\partial \boldsymbol{x}} - \rho \frac{\partial \mu}{\partial \boldsymbol{x}} - \eta \frac{\partial \mathcal{T}}{\partial \boldsymbol{x}} \right],$$

and

$$\delta(\operatorname{grad} \rho)^{\star} = \frac{\partial \delta \rho}{\partial \boldsymbol{x}} - \frac{\partial \rho}{\partial \boldsymbol{x}} \frac{\partial \boldsymbol{\zeta}}{\partial \boldsymbol{x}} \quad \text{and} \quad \delta(\operatorname{grad} \eta)^{\star} = \frac{\partial \delta \eta}{\partial \boldsymbol{x}} - \frac{\partial \eta}{\partial \boldsymbol{x}} \frac{\partial \boldsymbol{\zeta}}{\partial \boldsymbol{x}},$$

imply

$$-\delta(\operatorname{grad} \rho)^{\star} \Phi = \operatorname{div} \left[ \Phi \, \frac{\partial \rho}{\partial x} \, \zeta - \Phi \, \delta \rho + (\rho \operatorname{div} \, \Phi) \zeta \right] \\ + \left[ \frac{\partial(\rho \operatorname{div} \Phi)}{\partial x} - \operatorname{div} \left( \Phi \, \frac{\partial \rho}{\partial x} \right) \right] \zeta \, .$$

and

$$-\delta(\operatorname{grad} \eta)^{\star} \Psi = \operatorname{div} \left[ \Psi \, \frac{\partial \eta}{\partial \boldsymbol{x}} \, \boldsymbol{\zeta} - \Psi \, \delta \eta + (\eta \operatorname{div} \, \Psi) \boldsymbol{\zeta} \right] \\ + \left[ \frac{\partial(\eta \operatorname{div} \Psi)}{\partial \boldsymbol{x}} - \operatorname{div} \left( \Psi \, \frac{\partial \eta}{\partial \boldsymbol{x}} \right) \right] \boldsymbol{\zeta} \; .$$

Consequently,

$$\rho \, \boldsymbol{u}^{\star} \frac{d\boldsymbol{\zeta}}{dt} = \frac{\partial}{\partial t} (\rho \, \boldsymbol{u}^{\star} \boldsymbol{\zeta}) + \operatorname{div}[\rho(\boldsymbol{u}^{\star} \boldsymbol{\zeta}) \boldsymbol{u}] - \rho \, \boldsymbol{a}^{\star} \boldsymbol{\zeta},$$

where a denotes the acceleration vector of the fluid. We get,

$$\delta S = \int_{t_1}^{t_2} \int_{D_t} \left\{ -\rho \, \boldsymbol{a}^* - \rho \, \frac{\partial \mu}{\partial \boldsymbol{x}} - \eta \, \frac{\partial \mathcal{T}}{\partial \boldsymbol{x}} + \boldsymbol{\Phi}^* \, \frac{\partial \operatorname{grad} \rho}{\partial \boldsymbol{x}} + \boldsymbol{\Psi}^* \, \frac{\partial \operatorname{grad} \eta}{\partial \boldsymbol{x}} \right. \\ \left. + \frac{\partial}{\partial \boldsymbol{x}} \left( \rho \operatorname{div} \boldsymbol{\Phi} + \eta \operatorname{div} \boldsymbol{\Psi} \right) - \operatorname{div} \left( \boldsymbol{\Phi} \, \frac{\partial \rho}{\partial \boldsymbol{x}} + \boldsymbol{\Psi} \, \frac{\partial \eta}{\partial \boldsymbol{x}} \right) - \rho \, \frac{\partial \Omega}{\partial \boldsymbol{x}} \right\} \boldsymbol{\zeta} \, dv \, dt \\ \left. + \int_{t_1}^{t_2} \int_{D_t} \left\{ \operatorname{div} \left[ \boldsymbol{\Phi} \, \frac{\partial \rho}{\partial \boldsymbol{x}} \, \boldsymbol{\zeta} - \boldsymbol{\Phi} \, \delta\rho + (\rho \operatorname{div} \boldsymbol{\Phi}) \, \boldsymbol{\zeta} + \boldsymbol{\Psi} \, \frac{\partial \eta}{\partial \boldsymbol{x}} \, \boldsymbol{\zeta} - \boldsymbol{\Psi} \, \delta\eta + (\eta \operatorname{div} \boldsymbol{\Psi}) \, \boldsymbol{\zeta} \right. \\ \left. + (\rho \, \mu - \varepsilon + \eta \, \mathcal{T}) \, \boldsymbol{\zeta} \right] \left. + \frac{\partial}{\partial t} (\rho \, \boldsymbol{u}^* \boldsymbol{\zeta}) + \operatorname{div} [\rho (\boldsymbol{u}^* \boldsymbol{\zeta}) \boldsymbol{u}] \right\} dv \, dt.$$

By integration, the second integral vanishes when the virtual displacement is null on the boundary of  $[t_1, t_2] \times D_t$ .

From Hamilton principle,

$$\forall \ \boldsymbol{x} \in D_t \rightarrow \boldsymbol{\zeta}(\boldsymbol{x}), \text{with } \boldsymbol{\zeta}(\boldsymbol{x}) \text{ null on the boundary of } D_t, \ \delta S = 0,$$

we can deduce the motion equation of conservative and isentropic fluids.

# 2.3 First form of motion equation

From Hamilton principle, we deduce

$$\rho \boldsymbol{a} + \rho \operatorname{grad} \mu + \eta \operatorname{grad} \mathcal{T} - \frac{\partial \operatorname{grad} \rho}{\partial \boldsymbol{x}} \boldsymbol{\Phi} - \frac{\partial \operatorname{grad} \eta}{\partial \boldsymbol{x}} \boldsymbol{\Psi}$$
(8)  
- grad ( $\rho \operatorname{div} \boldsymbol{\Phi} + \eta \operatorname{div} \boldsymbol{\Psi}$ ) + div<sup>\*</sup> ( $\boldsymbol{\Phi} \operatorname{grad}^* \rho + \boldsymbol{\Psi} \operatorname{grad}^* \eta$ ) +  $\rho \operatorname{grad} \Omega = 0$ .

From relations

$$\begin{aligned} \frac{\partial(\rho \operatorname{div} \boldsymbol{\Phi})}{\partial \boldsymbol{x}} &= (\operatorname{div} \boldsymbol{\Phi}) \frac{\partial \rho}{\partial \boldsymbol{x}} + \rho \frac{\partial \operatorname{div} \boldsymbol{\Phi}}{\partial \boldsymbol{x}} \quad \text{and} \\ \frac{\partial(\eta \operatorname{div} \boldsymbol{\Psi})}{\partial \boldsymbol{x}} &= (\operatorname{div} \boldsymbol{\Psi}) \frac{\partial \eta}{\partial \boldsymbol{x}} + \eta \frac{\partial \operatorname{div} \boldsymbol{\Psi}}{\partial \boldsymbol{x}}, \\ \operatorname{div} \left( \boldsymbol{\Phi} \frac{\partial \rho}{\partial \boldsymbol{x}} \right) &= (\operatorname{div} \boldsymbol{\Phi}) \frac{\partial \rho}{\partial \boldsymbol{x}} + \boldsymbol{\Phi}^{\star} \frac{\partial \operatorname{grad} \rho}{\partial \boldsymbol{x}} \quad \text{and} \\ \operatorname{div} \left( \boldsymbol{\Psi} \frac{\partial \eta}{\partial \boldsymbol{x}} \right) &= (\operatorname{div} \boldsymbol{\Psi}) \frac{\partial \eta}{\partial \boldsymbol{x}} + \boldsymbol{\Psi}^{\star} \frac{\partial \operatorname{grad} \eta}{\partial \boldsymbol{x}}, \end{aligned}$$

we obtain,

$$\rho \boldsymbol{a} + \rho \operatorname{grad}(\mu - \operatorname{div} \boldsymbol{\Phi} + \Omega) + \eta \operatorname{grad}(\mathcal{T} - \operatorname{div} \boldsymbol{\Psi}) = 0, \quad (9)$$

or,

$$\boldsymbol{a} + \operatorname{grad}(\mu - \operatorname{div} \boldsymbol{\Phi} + \Omega) + s \operatorname{grad}(\boldsymbol{\mathcal{T}} - \operatorname{div} \boldsymbol{\Psi}) = 0, \quad (10)$$

where  $s = \eta/\rho$ . From Eq. (9) we deduce,

$$\boldsymbol{a} + \operatorname{grad} \boldsymbol{\Xi} - \boldsymbol{\theta} \operatorname{grad} \boldsymbol{s} = 0 \tag{11}$$

with

$$\theta = \mathcal{T} - \operatorname{div} \Psi$$
 and  $\Xi = \mu - \operatorname{div} \Phi + \Omega + (\mathcal{T} - \operatorname{div} \Psi) s.$ 

In case of internal energy (3) we get

$$\theta = \mathcal{T} - D\,\Delta\rho - E\,\Delta\eta \qquad \text{and} \qquad \Xi = \mu - C\,\Delta\rho - D\,\Delta\eta + \Omega + (\mathcal{T} - D\,\Delta\rho - E\,\Delta\eta)\,s$$
  
where  $\Delta$  is the Laplace operator. From  $d\mu = \frac{d\mathcal{P}}{\rho} - s\,d\mathcal{T}$ , Eq. (9) can be written  
 $\rho\,\boldsymbol{a} + \operatorname{grad}\mathcal{P} + \rho\,\operatorname{grad}(\Omega - \operatorname{div}\boldsymbol{\Phi}) - \eta\,\operatorname{grad}\operatorname{div}\boldsymbol{\Psi} = 0.$  (12)

# 2.4 Second form of the motion equation

If we denote

$$p = \mathcal{P} - \rho \operatorname{div} \Phi - \eta \operatorname{div} \Psi$$
 and  $\sigma = -p I - \Phi \frac{\partial \rho}{\partial x} - \Psi \frac{\partial \eta}{\partial x}$ 

equation (8) yields

$$\rho \boldsymbol{a} - \operatorname{div}^{\star} \boldsymbol{\sigma} + \rho \operatorname{grad} \Omega = 0.$$
(13)

In the case of internal energy (3), we obtain the value of  $\sigma$ ,

$$\boldsymbol{\sigma} = -p \, \boldsymbol{I} - (C \operatorname{grad} \rho + D \operatorname{grad} \eta) \, \frac{\partial \rho}{\partial \boldsymbol{x}} - (D \operatorname{grad} \rho + E \operatorname{grad} \eta) \, \frac{\partial \eta}{\partial \boldsymbol{x}}.$$
 (14)

# 2.5 Adiabatic motions

If the total entropy of the fluid in domain  $D_t$  is constant, its variation is null,

$$\delta \int_{D_t} \eta \, dv \equiv \delta \int_{D_t} \rho s \, dv = 0,$$

and it exists a constant Lagrange multiplier  ${\cal T}_0$  such that the variation of Hamilton's action

$$\delta S \equiv \int_{t_1}^{t_2} \int_{D_t} \rho \,\delta\left(\frac{1}{2} \,\boldsymbol{u}^{\star} \boldsymbol{u} - \frac{\varepsilon}{\rho} - \Omega + T_0 \,s\right) dv \,dt = 0$$

is null, with always

$$\delta \boldsymbol{u} = \frac{d\boldsymbol{\zeta}}{dt}$$
 and  $\delta \rho = -\rho \operatorname{div} \boldsymbol{\zeta}.$ 

From variation field  $\boldsymbol{\zeta}$ , we get the same equation of motions (Eq. (9)). When  $\boldsymbol{\zeta} = 0$ , independent variation of  $\eta ~(\delta \eta = \rho \, \delta s)$  yields

$$\delta S = \int_{t_1}^{t_2} \int_{D_t} \left( -\delta \varepsilon + \rho T_0 \,\delta s \right) \, dv \, dt = 0.$$

Due to Eq. (4),  $\delta \varepsilon = \mathcal{T} \, \delta \eta - \Psi^* \, \delta \operatorname{grad} \eta$  and  $\boldsymbol{\zeta} = 0$  implies  $\delta \operatorname{grad} \eta = \left(\frac{\partial \delta \eta}{\partial \boldsymbol{x}}\right)^*$ . Consequently,

$$\delta S = \int_{t_1}^{t_2} \int_{D_t} \left[ (T_0 - \mathcal{T}) \,\delta\eta - \Psi^* \,\delta \operatorname{grad} \,\eta \right] \, dv \, dt$$
$$= \int_{t_1}^{t_2} \int_{D_t} \left( T_0 - \mathcal{T} + \operatorname{div} \Psi \right) \,\,\delta\eta \,\, dv \, dt - \int_{t_1}^{t_2} \int_{D_t} \operatorname{div} \left( \Psi \,\delta\eta \right) dv \, dt.$$

We consider that  $\delta \eta = 0$  on the boundary of  $D_t$ . By integration on the  $D_t$ -boundary, the second integral is null and the Hamilton principle yields :

$$\mathcal{T} = T_0 + \operatorname{div} \boldsymbol{\Psi} \,,$$

and in the special case of a volume energy in form (3),

$$\mathcal{T} = T_0 + D\,\Delta\rho + E\,\Delta\eta\,. \tag{15}$$

We note that  $\theta = \mathcal{T} - \text{div } \Psi$  is constant equal to  $T_0$  which is the temperature in the homogeneous parts of thermocapillary fluids (corresponding to the bulks).

#### 3 Equation of energy and second law of thermodynamics [43,44]

# 3.1 Equation of motions of viscous thermocapillary fluids

For a viscous fluid, we add a stress tensor in the Newtonian form

$$\boldsymbol{\sigma}_{v} = \tau \left( \operatorname{tr} \mathcal{D} \right) \boldsymbol{I} + 2 \,\kappa \,\mathcal{D},$$

where  $\mathcal{D}$  is the velocity deformation tensor;  $\tau, \kappa$  are constant. We are in first gradient model for the viscosity but experiments prove that such a model is always correct for capillary layers [45]. The Hamilton principle becomes the

principle of virtual powers (or virtual works) [10] and Eq. (13) allows to obtain

$$\rho \boldsymbol{a} - \operatorname{div}^{\star} (\boldsymbol{\sigma} + \boldsymbol{\sigma}_{v}) + \rho \operatorname{grad} \Omega = 0,$$

where  $\boldsymbol{\sigma}$  verifies Eq. (14).

# 3.2 Equation of energy

We extend the results proposed in [19–21,46]. Let us note

$$\begin{cases} \boldsymbol{M} = \rho \, \boldsymbol{a} - \operatorname{div}^{\star} \left( \boldsymbol{\sigma} + \boldsymbol{\sigma}_{v} \right) + \rho \operatorname{grad} \Omega \\ B = \frac{\partial \rho}{\partial t} + \operatorname{div} \rho \, \boldsymbol{u} \\ N = \rho \left( \boldsymbol{\mathcal{T}} - \operatorname{div} \boldsymbol{\Psi} \right) \dot{\boldsymbol{s}} + \operatorname{div} \boldsymbol{q} - r - tr \left( \boldsymbol{\sigma}_{v} \boldsymbol{\mathcal{D}} \right) \\ F = \frac{\partial e}{\partial t} + \operatorname{div} \left[ \left( e \boldsymbol{I} - \boldsymbol{\sigma} - \boldsymbol{\sigma}_{v} \right) \boldsymbol{u} \right] - \operatorname{div} \left( \dot{\rho} \, \boldsymbol{\Phi} + \dot{\eta} \, \boldsymbol{\Psi} \right) + \operatorname{div} \boldsymbol{q} - r - \rho \frac{\partial \Omega}{\partial t} \\ (16)$$

where  $e = \frac{1}{2} \rho \mathbf{u}^* \mathbf{u} + \varepsilon + \rho \Omega$  is the total volume energy of the fluid,  $\mathbf{q}$  and r are the heat flux vector and the heat supply, respectively; superscript  $\cdot$  denotes the material derivative and the free enthalpy is  $h \equiv \frac{\varepsilon + p}{\rho}$ . We get :

**Theorem 1** For an internal energy in form (2) and for any motion of thermocapillary fluids,

$$F - \mathbf{M}^{\star} \mathbf{u} - \left(\frac{1}{2} \, \mathbf{u}^{\star} \mathbf{u} + h + \Omega\right) B - N \equiv 0.$$
(17)

The proof is proposed in Appendix 1.

**Corollary 2** For any motion of conservative thermocapillary fluids, the conservation of specific entropy  $\dot{s} = 0$  (or  $\partial \eta / \partial t + \operatorname{div} \eta \, \boldsymbol{u} = 0$ ) is equivalent to

$$\frac{\partial e}{\partial t} + \operatorname{div}\left[\left(e\boldsymbol{I} - \boldsymbol{\sigma}\right)\boldsymbol{u}\right] - \operatorname{div}\left(\dot{\rho}\,\boldsymbol{\Phi} + \dot{\eta}\,\boldsymbol{\Psi}\right) - \rho\,\frac{\partial\Omega}{\partial t} = 0.$$

**Corollary 3** For any motions of dissipative thermocapillary fluids, equation of energy

$$\frac{\partial e}{\partial t} + \operatorname{div}\left[\left(e\boldsymbol{I} - \boldsymbol{\sigma} - \boldsymbol{\sigma}_{v}\right)\boldsymbol{u}\right] - \operatorname{div}\left(\dot{\rho}\,\boldsymbol{\Phi} + \dot{\eta}\,\boldsymbol{\Psi}\right) + \operatorname{div}\boldsymbol{q} - r - \rho\frac{\partial\Omega}{\partial t} = 0$$

is equivalent to "equation of entropy"

$$\rho \left( \mathcal{T} - \operatorname{div} \Psi \right) \dot{s} + \operatorname{div} \boldsymbol{q} - r - tr \left( \boldsymbol{\sigma}_{v} \mathcal{D} \right) = 0.$$
(18)

Term  $\dot{\rho} \Phi + \dot{\eta} \Psi$  has the physical dimension of a heat flux vector; it corresponds to the *interstitial working term* [20] and reveals the existence of an additional term to the heat flux even if the motion is conservative. The result extends the ones obtained for capillary fluids when terms associated with grad  $\eta$  are not taken into account.

#### 3.3 Planck and Clausius-Duhem inequalities

For any motion of thermocapillary fluids,  $tr(\boldsymbol{\sigma}_{v}\mathcal{D}) \geq 0$  [43]. Equation (18) implies *Planck's inequality* [47]

$$\rho \left( \mathcal{T} - \operatorname{div} \boldsymbol{\Psi} \right) \dot{s} + \operatorname{div} \boldsymbol{q} - r \ge 0.$$

We assume the *Fourier law* in the general form,

$$q^* \operatorname{grad} \theta \leq 0$$
, with  $\theta = \mathcal{T} - \operatorname{div} \Psi$ 

and we obtain

$$p\dot{s} + \operatorname{div}\left(\frac{\boldsymbol{q}}{\theta}\right) - \frac{r}{\theta} \ge 0,$$

which is the extended form for thermocapillary fluids of *Clausius-Duhem's* inequality. We note that temperature  $\theta$  corresponds to the temperature value in homogeneous parts of thermocapillary fluids.

## 4 Some properties of thermocapillary fluids

1

#### 4.1 First integrals and Kelvin's circulation-theorems [48]

**Theorem 4** The velocity circulation on a closed, isentropic fluid-curve is constant.

The circulation of velocity vector  $\boldsymbol{u}$  on a closed fluid-curve  $\mathcal{C}$  is  $\mathcal{J} = \oint_{\mathcal{C}} \boldsymbol{u}^* d\boldsymbol{x}$ . From [41] p. 162,

$$\frac{d}{dt} \oint_{\mathcal{C}} \boldsymbol{u}^{\star} \, d\boldsymbol{x} = \oint_{\mathcal{C}} \boldsymbol{a}^{\star} \, d\boldsymbol{x}$$

and thanks to Eq. (11), we deduce

$$\oint_{\mathcal{C}} \boldsymbol{a}^{\star} d\boldsymbol{x} = \oint_{\mathcal{C}} \operatorname{grad}^{\star} \Xi \ d\boldsymbol{x} = 0,$$

which proves the theorem.

**Corollary 5** In a homentropic motion (the entropy is uniform in the fluid), the velocity circulation on a fluid-curve is constant.

**Theorem 6** The velocity circulation on a closed fluid-curve such that  $\mathcal{T} - \text{div } \Psi = T_0$  is constant.

From Eq. (10) we get,

$$\boldsymbol{a} + \operatorname{grad} \boldsymbol{\Xi} - (\boldsymbol{\mathcal{T}} - \operatorname{div} \boldsymbol{\Psi}) \operatorname{grad} \boldsymbol{s} = 0.$$

But,

$$\boldsymbol{a} - \frac{1}{2} \operatorname{grad} \boldsymbol{u}^2 = \frac{\partial \boldsymbol{u}}{\partial t} + \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} \boldsymbol{u} - \left(\frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}}\right)^* \boldsymbol{u} = \frac{\partial \boldsymbol{u}}{\partial t} + \operatorname{rot} \boldsymbol{u} \times \boldsymbol{u}.$$

For a stationary motion,

$$\operatorname{rot} \boldsymbol{u} \times \boldsymbol{u} = (\mathcal{T} - \operatorname{div} \boldsymbol{\Psi}) \operatorname{grad} s - \operatorname{grad} \left( \Xi + \frac{\boldsymbol{u}^2}{2} + \Omega \right).$$
(19)

Equation(19) is the generalized Crocco-Vazsonyi relation for thermocapillary fluids.

# 4.2 Superficial tension of thermocapillary fluids

We consider a planar interface between liquid and vapour bulks of a thermocapillary fluid. In the interfacial layer, density gradients are important. With internal energy (3), the stress tensor is

$$\boldsymbol{\sigma} = -\left(\mathcal{P} - \frac{C}{2}\operatorname{grad}^{\star}\rho \operatorname{grad}\rho - D\operatorname{grad}^{\star}\rho \operatorname{grad}\eta - \frac{E}{2}\operatorname{grad}^{\star}\eta \operatorname{grad}\eta\right)\boldsymbol{I} - (C\operatorname{grad}\rho + D\operatorname{grad}\eta)\frac{\partial\rho}{\partial\boldsymbol{x}} - (D\operatorname{grad}\rho + E\operatorname{grad}\eta)\frac{\partial\eta}{\partial\boldsymbol{x}}.$$

When the extraneous force potential is neglected, the equation of the equilibrium is

div 
$$\boldsymbol{\sigma} = 0.$$

For a flat interface, normal to  $\operatorname{grad} \rho$  and  $\operatorname{grad} \eta$ , the coordinate normal to the interface being denoted z, the eigenvalues of stress tensor  $\sigma$  are

$$\lambda_1 = -\mathcal{P} + \frac{C}{2} \left(\frac{d\rho}{dz}\right)^2 + D \frac{d\rho}{dz} \frac{d\eta}{dz} + \frac{E}{2} \left(\frac{d\eta}{dz}\right)^2$$

(associated with the plane of interface), and

$$\lambda_2 = -\mathcal{P} - \frac{C}{2} \left(\frac{d\rho}{dz}\right)^2 - D \frac{d\rho}{dz} \frac{d\eta}{dz} - \frac{E}{2} \left(\frac{d\eta}{dz}\right)^2$$

(associated with direction normal to the plane of interface).

In an orthonormal system with third coordinate z, the stress tensor writes

$$oldsymbol{\sigma} = egin{bmatrix} \lambda_1 & 0 & 0 \ 0 & \lambda_1 & 0 \ 0 & 0 & \lambda_2 \end{bmatrix}.$$

The equation of balance momentum in the planar interface implies

$$\lambda_2 = -P_0 \,,$$

where  $P_0$  is the common pressure in the bulks. The force per unit of length on the edge of the interface is (see Fig. 1) :

$$\mathcal{F} = \int_{z_1}^{z_2} \lambda_1 \, dz =$$
$$-P_0(z_2 - z_1) + \int_{z_1}^{z_2} \left[ C \left( \frac{d\rho}{dz} \right)^2 + 2 D \frac{d\rho}{dz} \frac{d\eta}{dz} + E \left( \frac{d\eta}{dz} \right)^2 \right] \, dz,$$

where  $z_2 - z_1$  corresponds to the physical interface thickness. Due to the small thickness of the interface,  $-P_0(z_2 - z_1)$  is negligible. Let us note

$$H_1 = \int_{z_1}^{z_2} C \left(\frac{d\rho}{dz}\right)^2 dz, \quad H_2 = \int_{z_1}^{z_2} 2D \frac{d\rho}{dz} \frac{d\eta}{dz} dz, \quad H_3 = \int_{z_1}^{z_2} E \left(\frac{d\eta}{dz}\right)^2 dz.$$

The line force per unit of length on the interface edge is

$$H = H_1 + H_2 + H_3,$$

where H represents the surface tension of the planar interface at equilibrium. If we consider the approximation

$$\frac{\partial \varepsilon_0}{\partial \eta}(\rho, \eta) = T_1, \tag{20}$$

where  $T_1$  is the temperature value in the liquid and vapour bulks, then  $\eta$  is a function of  $\rho$ . Due to the variation principle, the surface tension calculated for capillary fluids (corresponding to D = 0 and E = 0) with approximation (20) is necessary greater than the surface tension when

$$\frac{\partial \varepsilon_0}{\partial \eta}(\rho,\eta) = T_1 + D\,\Delta\,\rho + E\,\Delta\,\eta\,.$$

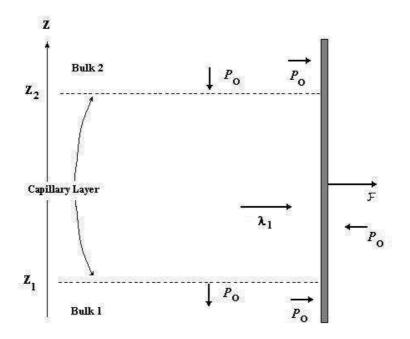


Fig. 1. Interpretation of the surface tension

In fact, experiments prove that the entropy effects are small enough on surface tension value and when the critical point is approached, the one - and twodensity theories become equivalent as a general property of critical point ([9], Ch. 3), [14].

# 4.3 Maxwell's rule for thermocapillary fluids

We consider the case when the volume internal energy is in form (3). In the case of capillary fluids (corresponding to D = 0 and E = 0), the Maxwell rule of planar liquid-vapour interface at equilibrium can be written in equivalent form

$$\int_{\rho_v}^{\rho_l} \left( \mu_0 - \mu_1 \right) d\rho = 0,$$

where  $\rho_l$  and  $\rho_v$  are the mass density in the liquid and vapour bulks;  $\mu_1$  is the common value of the chemical potential in the bulks [11]. We denote  $\eta_l$  and  $\eta_v$  the volume entropies in the liquid and vapour bulks, respectively.

Equation of temperature (15) of thermocapillary fluids yields

$$\mathcal{T} - T_1 = D \frac{d^2 \rho}{dz^2} + E \frac{d^2 \eta}{dz^2}.$$

Without body forces, equation of equilibrium (10) of thermocapillary fluids yields

$$\operatorname{grad}(\mu - \operatorname{div} \Phi) = 0$$

or by integration,

$$\mu - \mu_1 = C \frac{d^2 \rho}{dz^2} + D \frac{d^2 \eta}{dz^2} \,.$$

Consequently,

$$\int_{\rho_v}^{\rho_l} (\mu - \mu_1) \, d\rho + \int_{\eta_v}^{\eta_l} (\mathcal{T} - T_1) \, d\eta$$
  
= 
$$\int_{z_1}^{z_2} \left[ C \, \frac{d^2 \rho}{dz^2} \, \frac{d\rho}{dz} + D \left( \frac{d^2 \eta}{dz^2} \, \frac{d\rho}{dz} + \frac{d^2 \rho}{dz^2} \, \frac{d\eta}{dz} \right) + E \frac{d^2 \eta}{dz^2} \, \frac{d\eta}{dz} \right] \, dz$$
  
= 
$$\left[ \frac{C}{2} \left( \frac{d\rho}{dz} \right)^2 + D \left( \frac{d\rho}{dz} \right) \left( \frac{d\eta}{dz} \right) + \frac{E}{2} \left( \frac{d\eta}{dz} \right)^2 \right]_{z_1}^{z_2} \equiv 0 \, .$$

The generalisation of Maxwell's rule for thermocapillary fluids writes in the form :  $c_{i}$ 

$$\int_{\rho_v}^{\rho_l} (\mu - \mu_1) \, d\rho + \int_{\eta_v}^{\eta_l} (\mathcal{T} - T_1) \, d\eta = 0 \, .$$

## 5 Governing equations in divergence form and hyperbolicity

Conservative motions with balance equation of energy lead to an interesting class of quasilinear systems previously pointed out by Godunov [36], Friedrichs and Lax [37]. In classical mechanics and relativity, many studies on hyperbolic systems were developed in the literature for hydrodynamics, elasticity and classical materials [49–52]. The section extends results presented in [53] for the capillary-fluids' simplest case. The small motions near an equilibrium position are studied thanks to a convenient system of governing equations associated with a Legendre transformation of the internal energy.

## 5.1 Governing equations in divergence form

Let us denote  $\boldsymbol{\beta} \equiv \text{grad } \rho$ ,  $\boldsymbol{\chi} \equiv \text{grad } \eta$  and  $\boldsymbol{j} \equiv \rho \boldsymbol{u}$ . The gradient of the mass-conservation balance verifies another conservation equation,

$$\frac{\partial \boldsymbol{\beta}}{\partial t} + \operatorname{grad}\operatorname{div}\boldsymbol{j} = 0, \qquad (21)$$

Conversely, if we consider  $\beta$  as an independent vector verifying Eq. (21), and if we add initial condition

$$\boldsymbol{\beta}|_{t=0} = \operatorname{grad} \rho|_{t=0} ,$$

then  $\beta \equiv \operatorname{grad} \rho$  becomes a consequence of governing equation (21). Similarly, the gradient of the balance of entropy verifies another conservation equation,

$$\frac{\partial \boldsymbol{\chi}}{\partial t} + \operatorname{grad}\operatorname{div}(\eta \,\boldsymbol{u}) = 0. \tag{22}$$

In the same way, if we add initial condition

$$\boldsymbol{\chi}|_{t=0} = \operatorname{grad} \eta|_{t=0} ,$$

 $\chi \equiv \operatorname{grad} \eta$  becomes a consequence of governing equation (22) and we can consider  $\chi$  as an independent vector verifying Eq. (22).

Without body forces, with the new notations, Eqs. (5, 6, 12, 21, 22) immediately yield the system of governing equations in the form

$$\begin{cases} \frac{\partial \rho}{\partial t} + \operatorname{div} \boldsymbol{j} = 0 \\ \frac{\partial \eta}{\partial t} + \operatorname{div} \left(\frac{\eta}{\rho} \boldsymbol{j}\right) = 0 \\ \frac{\partial \boldsymbol{j}^{\star}}{\partial t} + \operatorname{div} \left(\frac{\boldsymbol{j}\boldsymbol{j}^{\star}}{\rho} + \mathcal{P} \boldsymbol{I}\right) - \rho \operatorname{grad}^{\star} (\operatorname{div} \boldsymbol{\Phi}) - \eta \operatorname{grad}^{\star} (\operatorname{div} \boldsymbol{\Psi}) = 0 \quad (23) \\ \frac{\partial \boldsymbol{\beta}}{\partial t} + \operatorname{grad} (\operatorname{div} \boldsymbol{j}) = 0 \\ \frac{\partial \boldsymbol{\chi}}{\partial t} + \operatorname{grad} \operatorname{div} \left(\frac{\eta}{\rho} \boldsymbol{j}\right) = 0. \end{cases}$$

With the new notations, the total volume energy of the fluid is

$$\mathcal{E} = \frac{\boldsymbol{j}^* \boldsymbol{j}}{2 \rho} + \varepsilon \,.$$

We denote  $q = \mu - \frac{\boldsymbol{u}^* \boldsymbol{u}}{2}$ . Consequently,

$$d\mathcal{E} = q \, d\rho + \mathcal{T} \, d\eta + \boldsymbol{u}^* d\boldsymbol{j} + \boldsymbol{\Phi}^* d\boldsymbol{\beta} + \boldsymbol{\Psi}^* d\boldsymbol{\chi},$$

The Legendre transform of  $\mathcal{E}$  with respect to  $\rho, \eta, j, \beta, \chi$  is

$$\Pi = \rho q + \eta \mathcal{T} + \boldsymbol{j}^* \boldsymbol{u} + \boldsymbol{\Phi}^* \boldsymbol{\beta} + \boldsymbol{\Psi}^* \boldsymbol{\chi} - \mathcal{E}.$$
(24)

Conjugate variables  $q, \mathcal{T}, \boldsymbol{u}, \boldsymbol{\Phi}, \boldsymbol{\Psi}$  verify

$$\frac{\partial \Pi}{\partial q} = \rho, \quad \frac{\partial \Pi}{\partial \mathcal{T}} = \eta, \quad \frac{\partial \Pi}{\partial \boldsymbol{u}} = \boldsymbol{j}^{\star}, \quad \frac{\partial \Pi}{\partial \boldsymbol{\Phi}} = \boldsymbol{\beta}^{\star}, \quad \frac{\partial \Pi}{\partial \boldsymbol{\Psi}} = \boldsymbol{\chi}^{\star}$$

System (23) can be written in divergence form as (See Appendix B) :

$$\begin{cases}
\frac{\partial}{\partial t} \left( \frac{\partial \Pi}{\partial q} \right) + \operatorname{div} \left[ \frac{\partial (\Pi u)}{\partial q} \right] = 0 \\
\frac{\partial}{\partial t} \left( \frac{\partial \Pi}{\partial T} \right) + \operatorname{div} \left[ \frac{\partial (\Pi u)}{\partial T} \right] = 0 \\
\frac{\partial}{\partial t} \left( \frac{\partial \Pi}{\partial u} \right) + \operatorname{div} \left[ \frac{\partial (\Pi u)}{\partial u} - \frac{\partial \Pi}{\partial q} \frac{\partial \Phi}{\partial x} - \frac{\partial \Pi}{\partial T} \frac{\partial \Psi}{\partial x} \right] = 0 \quad (25) \\
\frac{\partial}{\partial t} \left( \frac{\partial \Pi}{\partial \Phi} \right) + \operatorname{div} \left[ \frac{\partial (\Pi u)}{\partial \Phi} + \frac{\partial \Pi}{\partial q} \frac{\partial u}{\partial x} \right] = 0 \\
\frac{\partial}{\partial t} \left( \frac{\partial \Pi}{\partial \Psi} \right) + \operatorname{div} \left[ \frac{\partial (\Pi u)}{\partial \Psi} + \frac{\partial \Pi}{\partial T} \frac{\partial u}{\partial x} \right] = 0.
\end{cases}$$

When  $\varepsilon = \varepsilon(\rho, \eta)$ , we get the classical gas dynamics equations and the conservative form of Godunov [36]. In the simplest special case, when  $\varepsilon = \varepsilon_0(\rho, \eta) + \frac{C}{2}(\operatorname{grad} \rho)^2$ , we obtain the results [53].

# 5.2 Hyperbolicity of governing equations

The system of governing equations generates dispersive relations with multiple eigenvalues near an equilibrium position. In this subsection we extend the results presented in [54–56]. System (25) yields constant solutions

$$(\rho_e, \eta_e, \boldsymbol{j}_e, \boldsymbol{\beta}_e = 0, \boldsymbol{\chi}_e = 0),$$

where subscript e means at equilibrium. Since the governing equations are invariant under Galilean transformation, we can assume that  $u_e = 0$  which implies  $j_e = 0$ .

Near equilibrium, we look for the solutions proportional to  $e^{i(\mathbf{k}^* \mathbf{x} - \lambda t)}$ , where  $\mathbf{k}^* = [k_1, k_2, k_3]$  is a constant covector,  $\lambda$  a constant scalar and  $i^2 = -1$ ,

$$oldsymbol{v} = oldsymbol{v}_{_0} e^{i(oldsymbol{k}^\staroldsymbol{x}-\lambda t)} ext{ with } oldsymbol{v}^\star = [q,\mathcal{T},oldsymbol{u}, \Phi,\Psi] ext{ and } oldsymbol{v}^\star_{_0} = [q_{_0},\mathcal{T}_{_0},oldsymbol{u}_{_0},\Phi_{_0},\Psi_{_0}] ext{ .}$$

We obtain

$$\frac{\partial}{\partial t} \left( \frac{\partial \Pi}{\partial \boldsymbol{v}} \right)_{e}^{\star} \equiv \frac{\partial}{\partial \boldsymbol{v}} \left( \frac{\partial \Pi}{\partial \boldsymbol{v}} \right)_{e}^{\star} \frac{\partial \boldsymbol{v}}{\partial t} \equiv -i \lambda \frac{\partial}{\partial \boldsymbol{v}} \left( \frac{\partial \Pi}{\partial \boldsymbol{v}} \right)_{e}^{\star} \boldsymbol{v}_{0} e^{i(\boldsymbol{k}^{\star}\boldsymbol{x} - \lambda t)}.$$

$$\operatorname{div}\left(\frac{\partial \Pi \boldsymbol{u}}{\partial \boldsymbol{v}}\right)^{\star} = \sum_{j=1}^{3} \frac{\partial}{\partial x^{j}} \left(\frac{\partial \Pi u^{j}}{\partial \boldsymbol{v}}\right)^{\star} = \sum_{j=1}^{3} \frac{\partial}{\partial \boldsymbol{v}} \left(\frac{\partial \Pi u^{j}}{\partial \boldsymbol{v}}\right)^{\star} \frac{\partial \boldsymbol{v}}{\partial x^{j}},$$

with  $\boldsymbol{x}^{\star} = [x^1, x^2, x^3]$  and

div 
$$\left(\frac{\partial \Pi \boldsymbol{u}}{\partial \boldsymbol{v}}\right)_{e}^{\star} = \sum_{j=1}^{3} i \, \boldsymbol{F}^{j} \, k_{j} \, \boldsymbol{v}_{0} \, e^{i(\boldsymbol{k}^{\star}\boldsymbol{x}-\lambda t)},$$

where

$$\boldsymbol{F}^{j} \equiv \frac{\partial}{\partial \boldsymbol{v}} \left( \frac{\partial \Pi u^{j}}{\partial \boldsymbol{v}} \right)_{e}^{\star}; \text{ we denote } \boldsymbol{F} \equiv \sum_{j=1}^{3} \boldsymbol{F}^{j} k_{j}.$$

# At equilibrium,

 $\bullet$  For Eq. (23)^3 (or equivalently Eq. (25)^3), we add two additive terms to classical-fluids' equations :

First term,

$$\operatorname{div}\left(\frac{\partial\Pi}{\partial q}\frac{\partial\Phi}{\partial \boldsymbol{x}}\right) = (\operatorname{grad}^{\star}\rho) \ \frac{\partial\Phi}{\partial \boldsymbol{x}} + \rho \ \operatorname{div}\left(\frac{\partial\Phi}{\partial \boldsymbol{x}}\right) \ .$$

At equilibrium,  $\operatorname{grad}_e \rho = 0$  . Then, from  $\mathbf{\Phi} = \mathbf{\Phi}_{\scriptscriptstyle 0} e^{i(\mathbf{k}^\star \mathbf{x} - \lambda t)},$ 

$$\operatorname{div}\left(\frac{\partial\Pi}{\partial q}\frac{\partial \Phi}{\partial \boldsymbol{x}}\right)_{e} = \rho_{e} \operatorname{div}\left(\frac{\partial \Phi}{\partial \boldsymbol{x}}\right) = i^{2} \rho_{e} \boldsymbol{\Phi}_{0}^{\star} \boldsymbol{k} \, \boldsymbol{k}^{\star} e^{i(\boldsymbol{k}^{\star} \boldsymbol{x} - \lambda t)}.$$

Second term,

$$\operatorname{div}\left(\frac{\partial\Pi}{\partial\mathcal{T}}\frac{\partial\Psi}{\partial\boldsymbol{x}}\right) = (\operatorname{grad}^{\star}\eta) \ \frac{\partial\Psi}{\partial\boldsymbol{x}} + \eta \ \operatorname{div}\left(\frac{\partial\Psi}{\partial\boldsymbol{x}}\right) \ .$$

At equilibrium,  $\operatorname{grad}_e\eta=0$  . Then, from  $\pmb{\Psi}=\pmb{\Psi}_{\scriptscriptstyle 0}e^{i(\pmb{k}^\star\pmb{x}-\lambda t)},$ 

$$\operatorname{div}\left(\frac{\partial \Pi}{\partial \mathcal{T}}\frac{\partial \Psi}{\partial \boldsymbol{x}}\right)_{e} = \rho_{e} \operatorname{div}\left(\frac{\partial \Psi}{\partial \boldsymbol{x}}\right) = i^{2} \eta_{e} \boldsymbol{\Psi}_{0}^{\star} \boldsymbol{k} \, \boldsymbol{k}^{\star} e^{i(\boldsymbol{k}^{\star} \boldsymbol{x} - \lambda t)}.$$

Taking account of  $\boldsymbol{u} = \boldsymbol{u}_0 e^{i(\boldsymbol{k}^*\boldsymbol{x} - \lambda t)}$ ,

• For Eq.  $(25)^4$  at equilibrium, we add term

$$\operatorname{div}\left[\frac{\partial \Pi}{\partial q}\frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}}\right]_{e} = i^{2} \rho_{e} \boldsymbol{u}_{0}^{\star} \boldsymbol{k} \, \boldsymbol{k}^{\star} e^{i(\boldsymbol{k}^{\star}\boldsymbol{x}-\lambda t)}.$$

• For Eq.  $(25)^5$  at equilibrium, we add term

div 
$$\left[\frac{\partial \Pi}{\partial \mathcal{T}} \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}}\right]_e = i^2 \eta_e \, \boldsymbol{u}_0^* \boldsymbol{k} \, \boldsymbol{k}^* e^{i(\boldsymbol{k}^* \boldsymbol{x} - \lambda t)}.$$

Let us define  $\boldsymbol{A}, \boldsymbol{C}$  such that

$$\boldsymbol{A} = \frac{\partial}{\partial \boldsymbol{v}} \left[ \left( \frac{\partial \Pi}{\partial \boldsymbol{v}} \right)^{\star} \right]_{e},$$

$$\boldsymbol{C} = -\boldsymbol{C}^{\star} = \begin{bmatrix} 0 & 0 & \boldsymbol{0}^{\star} & \boldsymbol{0}^{\star} & \boldsymbol{0}^{\star} \\ 0 & 0 & \boldsymbol{0}^{\star} & \boldsymbol{0}^{\star} & \boldsymbol{0}^{\star} \\ \boldsymbol{0} & \boldsymbol{0} & \boldsymbol{O}^{\star} & -\rho_{e} \boldsymbol{k} \boldsymbol{k}^{\star} & -\eta_{e} \boldsymbol{k} \boldsymbol{k}^{\star} \\ \boldsymbol{0} & \boldsymbol{0} & \rho_{e} \boldsymbol{k} \boldsymbol{k}^{\star} & \boldsymbol{O} & \boldsymbol{O} \\ \boldsymbol{0} & \boldsymbol{0} & \eta_{e} \boldsymbol{k} \boldsymbol{k}^{\star} & \boldsymbol{O} & \boldsymbol{O} \end{bmatrix}$$

with

$$\boldsymbol{O} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad \text{and} \quad \boldsymbol{O}^* = \begin{bmatrix} 0 & 0 & 0 \end{bmatrix}.$$

Due to  $\overline{iC}^* \equiv iC$ , where overline denotes the complex conjugation; matrix iC is hermitian.

The solutions corresponding to the perturbations of system (25) verify :

$$i \left[ \mathbf{F} + i \mathbf{C} - \lambda \mathbf{A} \right] \mathbf{v}_0 e^{i(\mathbf{k}^* \mathbf{x} - \lambda t)} = 0,$$

where  $D = \overline{D}^* \equiv F + i C$  is Hermitian matrix and A is symmetric matrix; so,  $\lambda$  are the roots of the characteristic equation :

$$\det\left[\boldsymbol{D}-\lambda\,\boldsymbol{A}\right]=0,$$

and  $\lambda$  is eigenvalue of D with respect to A and  $v_0$  is its eigenvector. Near an equilibrium state where the local internal energy is locally convex, A is positive definite; eigenvalues are real and the small perturbations are stable with respect to equilibrium positions.

#### 6 Conclusion

For conservative processes associated with system (23), Legendre transformation (24) of the internal energy yields a system of governing equations which extends the classical models of hyperbolicity to non-local behaviour. The Lax-Friedrichs method [57] is a numerical method we can consider as an alternative to Godunov's scheme [58] in which one avoids solving a Riemann problem at each cell interface, at the expense of adding artificial viscosity. The stability of quasi-linear perturbations allows to forecast an extention of the Lax-Friedrichs method for thermocapillary fluids.

## 7 Appendix A: Proof of relation (17)

By using System (16) in the first member of Eq. (17), dissipative terms  $\boldsymbol{q}, r, \boldsymbol{\sigma}_v$  can be algebraically simplified. Also are terms associated with inertia and  $\Omega$ . The remaining terms are

$$\begin{cases} \boldsymbol{M}_{0} = -\operatorname{div}^{\star}(\boldsymbol{\sigma}) \\ B = \frac{\partial \rho}{\partial t} + \operatorname{div} \rho \, \boldsymbol{u} \\ N_{0} = \rho \left( \boldsymbol{\mathcal{T}} - \operatorname{div} \boldsymbol{\Psi} \right) \dot{s} \\ F_{0} = \frac{\partial \varepsilon}{\partial t} + \operatorname{div} \left[ \left( \varepsilon \boldsymbol{I} - \boldsymbol{\sigma} \right) \boldsymbol{u} \right] - \operatorname{div} \left( \dot{\rho} \, \boldsymbol{\Phi} + \dot{\eta} \, \boldsymbol{\Psi} \right) \,, \end{cases}$$

and we have to prove

$$F_0 - \boldsymbol{M}_0^{\star} \boldsymbol{u} - h B - N_0 \equiv 0.$$
<sup>(26)</sup>

From

$$\begin{aligned} \frac{\partial \varepsilon}{\partial t} + \operatorname{div}(\varepsilon \, \boldsymbol{u}) - (\operatorname{div} \boldsymbol{\sigma}) \, \boldsymbol{u} - \operatorname{div} \left( \dot{\rho} \, \boldsymbol{\Phi} + \dot{\eta} \, \boldsymbol{\Psi} \right) &= \\ \frac{\varepsilon + p}{\rho} \, B + \mathcal{P} \, \dot{\rho} + \rho \, \mathcal{T} \, \dot{s} + \boldsymbol{\Phi}^* \frac{d \operatorname{grad} \rho}{dt} + \boldsymbol{\Psi}^* \frac{d \operatorname{grad} \eta}{dt} - p \, \frac{\dot{\rho}}{\rho} \\ + \boldsymbol{\Phi}^* \left( \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} \right)^* \operatorname{grad} \rho + \boldsymbol{\Psi}^* \left( \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} \right)^* \operatorname{grad} \eta - \operatorname{div} \left( \dot{\rho} \, \boldsymbol{\Phi} + \dot{\eta} \, \boldsymbol{\Psi} \right) = \\ \frac{\varepsilon + p}{\rho} \, B + \rho \left( \mathcal{T} - \operatorname{div} \, \boldsymbol{\Psi} \right) \dot{s} + \boldsymbol{\Phi}^* \operatorname{grad} \frac{\partial \rho}{\partial t} + \boldsymbol{\Phi}^* \, \frac{\partial \operatorname{grad} \rho}{\partial \boldsymbol{x}} \, \boldsymbol{u} + \boldsymbol{\Psi}^* \operatorname{grad} \frac{\partial \eta}{\partial t} \\ + \boldsymbol{\Psi}^* \frac{\partial \operatorname{grad} \eta}{\partial \boldsymbol{x}} \, \boldsymbol{u} + \, \boldsymbol{\Phi}^* \left( \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} \right)^* \operatorname{grad} \rho + \boldsymbol{\Psi}^* \left( \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} \right)^* \operatorname{grad} \eta \\ - \left( \operatorname{grad}^* \frac{\partial \rho}{\partial t} \right) \, \boldsymbol{\Phi} \, - \boldsymbol{u}^* \, \frac{\partial \operatorname{grad} \rho}{\partial \boldsymbol{x}} \, \boldsymbol{\Psi} - \left( \operatorname{grad}^* \rho \right) \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} \, \boldsymbol{\Psi}, \end{aligned}$$

and

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

we get,

$$\frac{\partial \varepsilon}{\partial t} + \operatorname{div}(\varepsilon \, \boldsymbol{u}) - (\operatorname{div} \, \boldsymbol{\sigma}) \, \boldsymbol{u} - \operatorname{div}(\dot{\rho} \, \boldsymbol{\Phi} + \dot{\eta} \, \boldsymbol{\Psi}) = \frac{\varepsilon + p}{\rho} \, B + \rho \left( \mathcal{T} - \operatorname{div} \boldsymbol{\Psi} \right) \dot{s}.$$

Relation

$$\operatorname{div}(\boldsymbol{\sigma}\,\boldsymbol{u}) = (\operatorname{div}\boldsymbol{\sigma})\,\boldsymbol{u} + tr\left(\boldsymbol{\sigma}\,\frac{\partial\boldsymbol{u}}{\partial\boldsymbol{x}}\right)$$

yields relation (26).

# 8 Appendix B: Proof of relation (25)

Relations  $\frac{\partial \Pi}{\partial q} = \rho$  and  $\frac{\partial (\Pi \boldsymbol{u})}{\partial q} = \rho \boldsymbol{u}$  imply Eq. (25<sup>1</sup>).

Relations  $\frac{\partial \Pi}{\partial \mathcal{T}} = \eta$  and  $\frac{\partial (\Pi \boldsymbol{u})}{\partial \mathcal{T}} = \eta \, \boldsymbol{u}$  imply Eq. (25<sup>2</sup>).

From relation

$$\frac{\partial \Pi}{\partial \boldsymbol{u}} = \boldsymbol{j}^{\star} \implies \frac{\partial (\Pi \boldsymbol{u})}{\partial \boldsymbol{u}} = \boldsymbol{u} \boldsymbol{j}^{\star} + \Pi \boldsymbol{I},$$

we obtain,

$$div \left( \frac{\partial (\Pi \boldsymbol{u})}{\partial \boldsymbol{u}} - \frac{\partial \Pi}{\partial q} \frac{\partial \boldsymbol{\Phi}}{\partial \boldsymbol{x}} - \frac{\partial \Pi}{\partial \mathcal{T}} \frac{\partial \boldsymbol{\Psi}}{\partial \boldsymbol{x}} \right) = div \left( \rho \, \boldsymbol{u} \, \boldsymbol{u}^{\star} \right) + \frac{\partial \Pi}{\partial \boldsymbol{x}}$$
$$- div \left( \rho \, \frac{\partial \boldsymbol{\Phi}}{\partial \boldsymbol{x}} + \eta \, \frac{\partial \boldsymbol{\Psi}}{\partial \boldsymbol{x}} \right)$$
$$= div \left( \rho \, \boldsymbol{u} \, \boldsymbol{u}^{\star} \right) + \rho \, \frac{\partial \boldsymbol{\mu}}{\partial \boldsymbol{x}} + \eta \, \frac{\partial \mathcal{T}}{\partial \boldsymbol{x}} - \rho \, \frac{\partial div \, \boldsymbol{\Phi}}{\partial \boldsymbol{x}} - \eta \, \frac{\partial div \, \boldsymbol{\Psi}}{\partial \boldsymbol{x}}$$

and consequently, the motion equation writes on form  $(25^3)$ .

From relation

$$\frac{\partial \Pi}{\partial \Phi} = \boldsymbol{\beta}^{\star} \quad \text{and} \quad \frac{\partial (\Pi \boldsymbol{u})}{\partial \Phi} + \frac{\partial \Pi}{\partial q} \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} = \boldsymbol{u} \, \boldsymbol{\beta}^{\star} + \rho \frac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} = \frac{\partial (\rho \boldsymbol{u})}{\partial \boldsymbol{x}},$$

we deduce Eq.  $(25^4)$ .

From relation

$$rac{\partial \Pi}{\partial \boldsymbol{\Psi}} = \boldsymbol{\chi}^{\star} \quad ext{and} \quad rac{\partial (\Pi \boldsymbol{u})}{\partial \boldsymbol{\Psi}} + rac{\partial \Pi}{\partial \mathcal{T}} \, rac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} = \boldsymbol{u} \, \boldsymbol{\chi}^{\star} + \eta \, rac{\partial \boldsymbol{u}}{\partial \boldsymbol{x}} = rac{\partial (\eta \boldsymbol{u})}{\partial \boldsymbol{x}} \,,$$

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