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Author for correspondence:

P. Ván

e-mail: van.peter@wigner.mta.hu

Variational principles and nonequilibrium thermodynamics

P. Ván^{1,2,3} and R. Kovács^{1,2,3}

¹Department of Theoretical Physics, Wigner Research Centre for Physics, Konkoly Thege Miklós u. 29-33, 1525 Budapest, Hungary

²Department of Energy Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Műegyetem rkp. 3, 1111 Budapest, Hungary

³Montavid Thermodynamic Research Group, 1112 Budapest, Hungary

PV, 0000-0002-9396-4073; RK, 0000-0001-5822-6035

Variational principles play a fundamental role in deriving the evolution equations of physics. They work well in the case of non-dissipative evolution, but for dissipative systems, the variational principles are not unique and not constructive. With the methods of modern nonequilibrium thermodynamics, one can derive evolution equations for dissipative phenomena and, surprisingly, in several cases, one can also reproduce the Euler–Lagrange form and symplectic structure of the evolution equations for non-dissipative processes. In this work, we examine some demonstrative examples and compare thermodynamic and variational techniques. Then, we argue that, instead of searching for variational principles for dissipative systems, there is another viable programme: the second law alone can be an effective tool to construct evolution equations for both dissipative and non-dissipative processes.

This article is part of the theme issue 'Fundamental aspects of nonequilibrium thermodynamics'.

1. Variational principles for dissipative processes

Variational principles are the ultimate tools for deriving the evolution equations of physics, in particular the equations of motion in mechanics. They work well without dissipation when the evolution is based on symmetric differential operators [1,2]. However, for dissipative systems, most of the differential equations

are not symmetric; therefore, special modifications are necessary to construct a variational principle in order to circumvent the strict mathematical conditions [3–5].

The applied tricks can be very different: for instance, changing the original equations, increasing the number of variables, reducing the corresponding function spaces, turning to statistical interpretations, and so on [6–13]. The obtained variational principles are special, different and not equivalent [14]. A general construction methodology of dissipative evolution equations in physics is still a challenge. A particularly strong requirement for any candidate principle, variational or not, is that it should deal with both dissipative and non-dissipative evolution equations. For the non-dissipative case, it must be compatible with Hamiltonian principles, and in the dissipative case, with the laws of thermodynamics. Moreover, it must be universal and simple.

In this paper, we reverse the usual logic. Instead of looking for variational principles of dissipative systems, we argue that the second law can be used to derive evolution equations for both dissipative and non-dissipative evolution. First, recent developments of nonequilibrium thermodynamics are outlined (§2). Then the evolution equation of a single point mass is treated from a thermodynamic point of view (§3), in the spirit of dual thermodynamic variables. Then a more complicated example, a two-component compressible fluid, is considered, where constraints and weak non-locality lead to the Fourier–Navier–Stokes–Cahn–Hilliard–Korteweg equations (§4). There, a weakly non-local extension of the entropy principle is used, and one obtains evolution equations where the time-independent part has an Euler–Lagrange form. In the next section (§5), using a combination of the previous methods, dual variables and weak non-locality, an inertial and dissipative extension of the classical gravity for self-gravitating heat-conducting fluids is derived. Finally, we summarize, compare and discuss the results.

2. Gradients and internal variables in nonequilibrium thermodynamics

In classical non-relativistic irreversible thermodynamics, one calculates the entropy balance in order to obtain constitutive functions that are compatible with the second law [15]. There, the basic assumption is the hypothesis of local equilibrium; memory and non-local effects are not considered. In modern theories of nonequilibrium thermodynamics, memory and non-local effects are considered in a weak sense, with the help of internal variables and gradients, respectively. Both aspects are reflected in methods exploiting the entropy inequality, and must consider frame indifference and objectivity as a fundamental background requirement [16,17].

The second law, the entropy inequality, is conditional. An absolute, i.e. frame-independent, treatment requires a four-dimensional space–time representation of the physical quantities also in a non-relativistic case. In this work, we consider the following aspects of a more rigorous non-relativistic formalism [18,19]:

- The entropy density and the entropy flux are both constitutive quantities; they are the time-like and space-like components of the entropy four-vector. Therefore, the entropy flux is to be determined according to the second law.
- Gradients are objective, reference-frame-independent quantities, because a gradient is a non-relativistic space-like co-vector. Therefore, spatial gradients can appear in constitutive functions with a good chance of being frame-independent. However, time derivatives depend on the frame.

Using these simple rules, objectivity and frame independence are respected, using three-dimensional abstract index-based calculations, and classical second law analysis without formal, transformation rule-based analysis or a rigorous space–time formalism [19,20].

In extended thermodynamics, the dissipative fluxes are not constitutive functions. They are basic state variables [21,22], and the evolution equations of these new fields are to be constructed according to the requirements of the second law [23]. In general, the idea of extending the thermodynamic state space with a new field, that of internal variables, has

a long history [24–27]. There are many interesting applications in their thermodynamically consistent use with particular physical examples [28–30]. Their combined use with classical fields in nonequilibrium thermodynamics verifies the second-law-based evolution equations by introducing thermodynamically consistent inertial, memory effects due to internal structural changes.

There are many different methods to introduce weak non-locality in space, that is, extending the constitutive functions with the gradients of the thermodynamic state variables, for both classical and internal ones. Without being complete, one needs to mention the method of virtual power [31–33], GENERIC [34–36], configurational forces [37,38] and phase-field theories [39]. All of these approaches can produce successful theories, with various levels of thermodynamic consistency and objectivity. For example, phase-field theories introduce functional derivatives for the new fields and then the variational techniques are combined with thermodynamic methods.

There are some particular aspects for the correct application of weak time and space non-locality in a thermodynamic framework.

- Internal variables are incorporated in the thermodynamic state space, i.e. the entropy density may depend on them. The related Gibbs relation is a convenient tool in calculations.
- The entropy density may depend on the gradients of the state variables, including internal variables, contrary to [40].
- A field theoretical approach is preferable, that is, the densities or specific quantities are primary in a continuum theory. That holds for the entropy, too. Consequently, the first-order Euler homogeneity of the entropy applies for internal variables and gradient extensions.

In the following, we demonstrate that the second law provides a constructive method to obtain evolution equations, for both dissipative and non-dissipative situations, for internal variables and with weakly non-local, gradient-dependent state spaces. Moreover, nonequilibrium thermodynamics leads to surprising information for evolution in classical theories, for pure mechanics, as well. We present three examples. First, the emergence of a Hamiltonian, that is, a symplectic structure, with dual thermodynamic state variables is demonstrated. Then, the case of gradient-dependent state spaces is presented with the example of two-component compressible and diffusive Fourier–Navier–Stokes–Cahn–Hilliard–Korteweg fluids. This is a complicated, but typical, example of the problems of phase-field methodology with variational derivatives. Our final illustration is the derivation of equations of classical gravitation with inertia. This is a combination of internal variables and classical fields with both gradient and memory effects.

In general, the conceptual foundation is the universality of a thermodynamic approach. We do not intend to look for the microscopic or mesoscopic origin of the continuum fields. They may be considered as emergent, but here it is not important. Whenever the second law is valid, the thermodynamic treatment is uniformly applicable, also if we do not expect a statistical background, e.g. for point masses or gravitation.

From now on, we use the abstract index notation, with i, j, k being spatial indices, denoting the tensorial character of a physical quantity in a three-dimensional vector space without particular coordinates [41]. Identical upper and lower indices denote contraction, according to the summation rule.

3. Example I: thermodynamic approach to point mass mechanics

In this section, the Newtonian, Lagrangian and Hamiltonian mechanics of a single point mass [42] is connected to nonequilibrium thermodynamics. This aspect is hidden in the traditional textbook treatment where dissipation is a secondary aspect. However, a simple change of attitude, a thermodynamic approach, leads to the complete dissipative version of Hamiltonian mechanics.

(a) Pure mechanics

The Hamilton equations of a point mass are generated from a phase-space potential, called Hamiltonian, $H(x^i, p^i)$, and are given as

$$\dot{x}^i = \partial_{p^i} H \quad (3.1)$$

and

$$\dot{p}^i = -\partial_{x^i} H, \quad (3.2)$$

with the upper dot denoting the time derivative.

A partial Legendre transformation gives the Lagrangian $L(x^i, \dot{x}^i)$ as

$$L(x^i, \dot{x}^i) + H(x^i, p^i) = p^i \dot{x}^i. \quad (3.3)$$

The derivatives of this formula yield

$$\partial_{\dot{x}^i} L = p^i, \quad (3.4)$$

$$\partial_{p^i} H = \dot{x}^i \quad (3.5)$$

and

$$\partial_{x^i} L = -\partial_{x^i} H. \quad (3.6)$$

The equation of motion, the Euler–Lagrange equation, follows by eliminating p^i , the momentum, from the system in order to obtain a differential equation for the position x^i :

$$\frac{d}{dt} \partial_{\dot{x}^i} L - \partial_{x^i} L = p^i + \partial_{x^i} H = 0. \quad (3.7)$$

This formula is obtained with the usual interpretation that (3.1) (and (3.5)) is the definition of the momentum, and (3.2) is the equation of motion. A Lagrangian is convenient to perform the elimination through Legendre transformation (3.3), without directly inverting the Hamiltonian.

Equation (3.7) is an evolution equation without dissipative terms. Dissipative terms are secondary in the usual approach and are given by separate, additional assumptions, such as imitating the non-dissipative mathematical structure by Rayleigh dissipation potentials. The addition of dissipative terms requires additional concepts. Without introducing thermodynamics, one cannot be sure whether a modification of (3.7) is dissipative or not. For instance, in an additive damping term like $\beta \dot{x}^i$, the coefficient β has a definite sign because of the second law.

(b) Pure thermodynamics

The thermodynamic construction of an evolution equation for state variables is also simple. One assumes the existence of a concave potential, which is increasing in time. Let us observe the direction of argumentation: One does not start from the evolution equations, but from the potential that determines the evolution according to the thermodynamic requirements. It is important to note that *these two conditions define a thermodynamic system* hereafter. In mechanics, starting from the Hamiltonian is exceptional; in thermodynamics, starting from entropy is the standard method. Let us denote the state variable by p^i , and the potential by S ; then

$$\dot{S}(p^i) = \partial_{p^i} S \dot{p}^i \geq 0. \quad (3.8)$$

Therefore, in the simplest case, when $S = -p^2/2m$ with $m > 0$, a linear isotropic solution of the above inequality leads to

$$\dot{p}^i = -\frac{l}{m} p^i, \quad (3.9)$$

where $l > 0$. This is a relaxation dynamics; the concavity of the potential ($m > 0$) together with the condition (3.8) guarantee the asymptotic stability of the equilibrium, $p^i = 0$. This is what one expects from a thermodynamic evolution: a hot coffee cools to the temperature of the environment; the asymptotic stability of the equilibrium is ensured. Indeed, the time-dependent classical thermodynamics of homogeneous systems rigorously and generally satisfies

that expectation, as proven by Matolcsi [43]. One may think that thermodynamics excludes inertial effects, and for mechanics, or for analogous evolutions with inertia, a variational principle is necessary, as argued e.g. in [28,44]. However, that is not so.

(c) Thermodynamics and mechanics: the embedded evolution

The entropy of the simplest thermodynamic body with a constant heat capacity, C , and internal energy, U , is $S(U) = C \ln(U)$. Then the caloric equation of state, the relation of the internal energy and the temperature, is $U = CT$. However, the internal energy is the difference between the total energy, E , and the Hamiltonian, $U = E - H(x^i, p^i)$; therefore, the entropy is a function of E , x^i and p^i . These variables are the state variables of the system and the system is isolated; the total energy is constant, and the entropy cannot decrease, independently of the dynamics prescribed for \dot{x}^i and \dot{p}^i . Hence

$$\dot{S}(E - H(x^i, p^i)) = \partial_{x^i} S \dot{x}^i + \partial_{p^i} S \dot{p}^i \geq 0. \quad (3.10)$$

If H is a twice differentiable function with continuous second derivative on a simply connected domain, then a quasi-linear solution of this inequality can be written, in general, as

$$\dot{x}^i = -l_{11} \partial_{x^i} H - l_{12} \partial_{p^i} H \quad (3.11)$$

and

$$\dot{p}^i = -l_{21} \partial_{x^i} H - l_{22} \partial_{p^i} H. \quad (3.12)$$

Here, l_{AB} is proportional to the reciprocal temperature, where $CT = U = E - H(x^i, p^i)$. This is not constant in this adiabatic situation; however, in several cases, when that particular equation of state is valid, considering the relation of thermal and mechanical energies, a constant temperature is a very good approximation. Regarding more precise arguments and conditions for the quasi-linear solution of (3.10), see e.g. the appendix B in [45]. Regarding some interesting consequences of the general dynamics with varying temperature, see [46]. Then, with these assumptions, the conductivity tensor,

$$L = \begin{pmatrix} l_{11} & l_{12} \\ l_{21} & l_{22} \end{pmatrix}, \quad (3.13)$$

is not necessarily symmetric, but its symmetric part is positive definite, according to the inequality (3.10), and therefore

$$l_{11} > 0, \quad l_{22} > 0, \quad l_{11}l_{22} - \frac{(l_{11} + l_{12})^2}{2} > 0. \quad (3.14)$$

It is convenient to split L into symmetric and antisymmetric parts, L_D and L_I , as

$$L = L_I + L_D = \begin{pmatrix} 0 & -k \\ k & 0 \end{pmatrix} + \begin{pmatrix} l_{11} & l \\ l & l_{22} \end{pmatrix}, \quad (3.15)$$

where the notation $l = (l_{12} + l_{21})/2$ and $k = (l_{21} - l_{12})/2$ is introduced. Then the third condition in (3.14) is $l_{11}l_{22} - l^2 > 0$. If the dissipative (symmetric) part is zero, the conservation of H and the conservation of S follow. The antisymmetric part generates non-dissipative evolution, a symplectic dynamics. It is identical with (3.1)–(3.2) if $k = 1$. That choice is linked to the definition of inertial mass.

This is a gradient dynamics, a particular case of the GENERIC framework, where the compatibility conditions are trivial [47–49], and also like metriplectic dynamics [50]. In both cases, the dynamics may be more general: in the case of GENERIC, with the help of two generators [35,36]; in the case of metriplectic theory, where a metric structure is important, with the help of a single generator, which can be the entropy alone [51].

It has been argued several times that the symplectic structure is the core of mechanics. A Lagrangian can be calculated from a Hamiltonian, and then the classical variational principles can be constructed. However, the Lagrangian and Hamiltonian approaches are not equivalent in the non-dissipative case and the situation is worse for dissipative systems.

(d) Mechanothermodynamics

Normally, in a textbook treatment, mechanics starts from the Newton equation without dissipation. However, in a dissipative system, this start can be misleading. As we have already mentioned, equations (3.11)–(3.12) are pure mechanical, with generalized dissipation. In order to get an impression about the physics of this generalization, it is worth considering a specific example, a usual Hamiltonian with kinetic and potential energies, i.e. $H(x^i, p^i) = p^2/2m + V(x^i)$, where V is a convex function. Therefore, the complete Hamiltonian is convex, too, and the entropy is concave; thermodynamic stability is fulfilled. In this case, (3.11)–(3.12) can be written as

$$\dot{x}^i = \frac{k}{m} p^i - l_{11} \partial_{x^i} V - \frac{l}{m} p^i \quad (3.16)$$

and

$$\dot{p}^i = -k \partial_{x^i} V - l \partial_i V - \frac{l_{22}}{m} p^i. \quad (3.17)$$

Then p^i can be expressed explicitly, and one obtains

$$p^i = m \frac{\dot{x}^i + l_{11} \partial_{x^i} V}{k - l}. \quad (3.18)$$

This is a strange, potential-dependent ‘momentum’. Substituting this expression into (3.17), one derives the corresponding dissipative equation of motion:

$$\hat{m} \ddot{x}^i + \hat{m} \left(l_{11} \partial_{x^i \dot{x}^j} V + \frac{l_{22}}{m} \delta_{ij} \right) \dot{x}^j + \partial_{x^i} V = 0, \quad (3.19)$$

where $\hat{m} = m/(k^2 + l_{11}l_{22} - l^2)$, $\hat{m} > m > 0$. All coefficients are non-negative according to the second law and the convexity of the Hamiltonian. We can see that, following the usual reasoning, i.e. starting from the equation of motion, the separation of the dissipative and non-dissipative parts is not straightforward. The mass increases due to dissipation and the damping is affected by the potential. However, when non-dissipative motion is what we have in mind, then the simple damping term in the middle of (3.19) seems to be completely satisfactory. Also, normally $k \neq 1$ is invisible, because inertial mass is defined by the Newton equation. With the interpretation of dissipative terms in (3.16), the ‘phase dissipation’ is peculiar from a mechanical point of view. However, it can be interpreted in a stochastic framework; for example, it may appear when the position is fluctuating [52], like in the case of wave function collapse [53,54].

We have seen that inertial effects can be introduced by pure thermodynamics, and this generalization leads to a fundamental reinterpretation of the concepts and expectations for the simplest mechanical systems, such as a point mass in an external potential. But aside from the thermodynamics of a point mass, one thing is clear. In order to get Hamiltonian dynamics, one does not need a variational principle; the second law with dual variables will do the job, and if entropy is conserved, then the emergent dynamics is not dissipative.

4. Example II: phase fields, Fourier–Navier–Stokes–Cahn–Hilliard–Korteweg equations

Classical phase-field models, the Allen–Cahn (or Ginzburg–Landau, model A) and the Cahn–Hilliard (model B) equations were not obtained from a thermodynamic treatment [39,45]. As we have already mentioned, there are several different methods to derive them, with or without variational derivatives [9,45,55–58]. The Cahn–Hilliard equation is exceptionally challenging for rigorous derivation, because of the higher space derivatives [59]. In this section, we investigate a two-component compressible heat-conducting fluid with diffusion. Using variational principles, it is not easy to realize the balances as thermodynamic constraints, because of the higher-order derivatives in the state space, and the non-trivial contributions to the pressure [60]. Here, we give

a simple, but complete, treatment, using the Gibbs relation as a starting point. We identify the entropy flux with the method of separation of divergences [15,59,61,62].

(a) Basic balances

A two-component fluid with component densities ρ_1, ρ_2 and velocities v_1^i, v_2^i will be characterized by the density ρ , the barycentric velocity v^i , the concentration c and the diffusion flux j^i . They are defined as

$$\rho = \rho_1 + \rho_2, \quad v^i = \frac{\rho_1 v_1^i + \rho_2 v_2^i}{\rho_1 + \rho_2}, \quad c = \frac{\rho_1}{\rho}, \quad j^i = \rho_1(v_1^i - v^i). \quad (4.1)$$

The balances of mass and concentration are

$$\dot{\rho} + \rho \partial_i v^i = 0 \quad (4.2)$$

and

$$\rho \dot{c} + \partial_i j^i = 0, \quad (4.3)$$

where the dot denotes the substantial time derivative and ∂_i is the nabla operator. The balance of momentum, the Cauchy equation, and the balance of internal energy are

$$\rho \dot{v}^i + \partial_j P^{ij} = 0 \quad (4.4)$$

and

$$\rho \dot{e} + \partial_i q^i = -P^{ij} \partial_i v_j, \quad (4.5)$$

where P^{ij} is the pressure tensor and q^i is the heat flux.

The basic fields of the system are the internal energy, the density, the velocity and the concentration. In order to get a closed system of equations, one must derive the constitutive functions for the diffusion flux j^i , the pressure P^{ij} and the heat flux q^i . These relations can be determined with the help of the entropy inequality.

Our thermodynamic state variables are the internal energy, the density and the concentration. The specific entropy is a function of these fields and also depends on the concentration gradient, i.e. $s(e, \rho, c, \partial_i c)$, which is required to obtain the necessary couplings. The Gibbs relation introduces the intensive thermodynamic quantities

$$de = T ds - p dv + \mu dc + \frac{Z^i}{\rho} d\partial_i c. \quad (4.6)$$

That is, the partial derivatives of the specific entropy are

$$\partial_e s = \frac{1}{T}, \quad \partial_\rho s = -\frac{p}{\rho^2 T}, \quad \partial_c s = -\frac{\mu}{T} \quad \text{and} \quad \partial_{\partial_i c} s = -\frac{Z^i}{\rho T}, \quad (4.7)$$

where T is the temperature, $v = 1/\rho$ is the specific volume, p is the thermostatic pressure, μ stands for the chemical potential and Z^i represents the derivative of the entropy by the concentration gradient. Z^i is introduced respecting the extensivity of the entropy. Now the entropy production can be calculated from the time derivative of the entropy considering the balances (4.2), (4.3) and (4.5) as constraints, then separating the surface and volumetric effects. This is a short, simple and straightforward calculation

$$\begin{aligned} \rho \dot{s}(e, \rho, c, \partial_i c) &= \frac{1}{T} \rho \dot{e} - \frac{p}{\rho T} \dot{\rho} - \frac{\rho \mu}{T} \dot{c} - \frac{Z^i}{T} (\partial_i \dot{c}) \\ &= -\partial_i \left[\frac{q^i - \mu j^i - Z^i \partial_k j^k / \rho}{T} + \frac{j^i}{\rho} \partial_k \left(\frac{Z^k}{T} \right) \right] \\ &\quad + q^i \partial_i \frac{1}{T} - j^i \partial_i \left[\frac{\mu}{T} - \frac{1}{\rho} \partial_k \left(\frac{Z^k}{T} \right) \right] - \frac{\partial_i v_j}{T} [P^{ij} - p \delta^{ij} - Z^i \partial^j c]. \end{aligned} \quad (4.8)$$

Table 1. Entropic representation of the thermodynamic fluxes and forces of Fourier–Navier–Stokes–Cahn–Hilliard–Korteweg fluids.

	thermal	diffusive	mechanical
fluxes	q^i	j^i	$p^{ij} - p\delta^{ij} - Z^i\partial^j c$
forces	$\partial_i \left(\frac{1}{T} \right)$	$-\partial_i \left(\frac{\mu}{T} - \frac{1}{\rho} \partial_k \left(\frac{Z^k}{T} \right) \right)$	$-\frac{\partial_i v_j}{T}$

On the right-hand side of equation (4.8), the first term is identified as the divergence of the entropy flux and the second one is the entropy production. One can identify the thermodynamic fluxes and forces according to table 1. That can be considered as an entropic representation, because the gradients of the entropic intensives ($1/T$, $-\mu/T$, $-Z^i/T$) appear everywhere in the expression. It is notable that the flux of thermal interaction is the heat flux, in spite of the modified entropy flux expression. The linear relation between the fluxes and forces in isotropic materials leads to the following constitutive equations:

$$\left. \begin{aligned} q^i &= \lambda \partial_i \left(\frac{1}{T} \right) - \chi \partial_i \left(\frac{\mu}{T} - \frac{1}{\rho} \partial_k \left(\frac{Z^k}{T} \right) \right), \\ j^i &= \chi \partial_i \left(\frac{1}{T} \right) - \zeta \partial_i \left(\frac{\mu}{T} - \frac{1}{\rho} \partial_k \left(\frac{Z^k}{T} \right) \right), \\ P^{ij} &= p\delta^{ij} + Z^i\partial^j c - 2\eta\partial^{(i}v^{j)} - \eta_b\partial_k v^k\delta^{ij}. \end{aligned} \right\} \quad (4.9)$$

and

Here, the conductivity coefficients λ , χ and ζ in the entropy representation are the thermal conductivity, the cross-coupling coefficient and the thermodynamic diffusion coefficient, respectively. The cross-coupling coefficients are equal, according to Onsager reciprocity. In (4.9), $\partial^{(i}v^{j)}$ denotes the symmetric (deviatoric) part of the velocity gradient, and η and η_b are the shear and bulk viscosities, respectively. Let us remark that in isotropic materials the pressure tensor is symmetric since the entropy depends only on the length of the concentration gradient vector, that is, on $\sqrt{\partial_i c \partial^i c}$; therefore Z^i is proportional to $\partial_i c$. The non-negativity of the entropy production requires the following inequalities:

$$\lambda > 0, \quad \chi > 0, \quad \zeta > 0, \quad \lambda\zeta - \chi^2 \geq 0, \quad \eta > 0, \quad \eta_b > 0. \quad (4.10)$$

It is remarkable that, for single-temperature mixtures, a different, reasonable flux–force representation is obtained writing the entropy balance in the following, equivalent form:

$$\begin{aligned} \rho\dot{s} + \partial_i \left[\frac{q^i - \mu j^i - (Z^i \partial_k j^k - j^i \partial_k Z^k)/\rho}{T} \right] \\ = \left(q^i - \mu j^i - \frac{Z^i \partial_k j^k - j^i \partial_k Z^k}{\rho} \right) \partial_i \frac{1}{T} - \frac{j^i}{T} \partial_i \left[\mu - \frac{\partial_k Z^k}{\rho} \right] \\ - \frac{\partial_i v_j}{T} \left[P^{ij} - p\delta^{ij} - Z^i \partial^j c \right] \geq 0. \end{aligned} \quad (4.11)$$

Based on the last line of the above expression, one can identify the thermodynamic fluxes and forces again. This choice is called the *energy representation*. It is notable that the entropy flux equals the thermal flux over the temperature. Recognizing that the entropy production is proportional to the reciprocal temperature, a system of fluxes and forces is given in table 2. These are identical

Table 2. Thermal representation of the thermodynamic fluxes and forces of Fourier–Navier–Stokes–Cahn–Hilliard–Korteweg fluids.

	thermal	diffusive	mechanical
fluxes	$q^i - \mu j^i - \frac{Z^i \partial_k j^k - j^i \partial_k Z^k}{\rho}$	j^i	$p^{ij} - p \delta^{ij} - Z^i \partial^j c$
forces	$-\frac{\partial_i T}{T}$	$\partial_i \left(\mu - \frac{\partial_k Z^k}{\rho} \right)$	$-\partial_i v_j$

with that from [63]. In isotropic materials, the linear relation between the fluxes and forces results in the following constitutive equations for q^i , j^i and P^{ij} , respectively:

$$\left. \begin{aligned} q^i - \mu j^i - \frac{Z^i \partial_k j^k - j^i \partial_k Z^k}{\rho} &= -\hat{\lambda} \frac{\partial_i T}{T} - \hat{\chi} \partial_i \left(\mu - \frac{\partial_k Z^k}{\rho} \right), \\ j^i &= \hat{\lambda} \frac{\partial_i T}{T} - \hat{\zeta} \partial_i \left(\mu - \frac{\partial_k Z^k}{\rho} \right) \\ \text{and} \quad P^{ij} &= p \delta^{ij} + Z^i \partial^j c - 2\eta \partial^{(i} v^{j)} - \eta_b \partial_k v^k \delta^{ij}, \end{aligned} \right\} \quad (4.12)$$

where the coefficients $\hat{\lambda}$, $\hat{\chi}$ and $\hat{\zeta}$ are seemingly different from the previous ones.

(b) Equivalence of representations

It is frequently argued that the various flux–force representations are not equivalent [64]. In our case, the viscosities are the same in the two representations, but the fluxes and forces, and also the coefficients of thermodiffusion, look rather different. However, the (4.8) and (4.11) entropy balances are the same, and with the general quasi-linear solution of the inequality—as in our previous example—the coefficients are state-dependent. Then, if the entropy flux does not change, a straightforward calculation gives a unique relation between the representations. For example, in the case of simple diffusion, without the Cahn–Hilliard extension, $Z^i \equiv 0$, and one obtains

$$\hat{\lambda} = \lambda + \zeta \mu^2, \quad \hat{\chi} = \chi - \zeta \mu \quad \text{and} \quad \hat{\zeta} = \zeta. \quad (4.13)$$

Therefore, the representations are equivalent, and the reciprocity is preserved. Moreover, if the inequalities in (4.10) of the entropic representation are fulfilled, then similar inequalities are valid in the energy representation, too. However, the coefficient inequalities that guarantee the non-negative entropy production in (4.11) are not sufficient to get (4.10), in particular, the first inequality.

(c) Cahn–Hilliard equation

The Cahn–Hilliard equation is obtained if the density, the internal energy and the velocity fields are homogeneous and there is a linear relation between the diffusion flux and force:

$$j^i = -\zeta \partial^i \left[\frac{\mu}{T} - \frac{1}{\rho} \partial_k \left(\frac{Z^k}{T} \right) \right] = -\zeta \partial^i \left[\partial_c s - \frac{1}{\rho} \partial_k \partial_{\partial_k c} (\rho s) \right]. \quad (4.14)$$

It is clear that the right-hand side of the equation is proportional to the gradient of the functional derivative of the specific entropy only if the density is constant. Then, substituting this equation into the concentration balance, one obtains the Cahn–Hilliard equation. This is a simple explanation of the mentioned differences between the variational derivation, the Lowengrub–Truskinovsky model [60] and the thermodynamic methods, analysed carefully in [63].

For phase transition models, the concavity of the entropy and the proper convexity relations for the free energy are important requirements. This property ensures the stability of equilibria, and the basin of attraction is related to simply connected concave regions. With additional considerations for boundary conditions, the total entropy is a good candidate to be a Lyapunov functional of equilibria, as indicated in [65].

5. Example III: inertial gravitation

Recently, it was shown that a single scalar internal variable, when additively separated from the internal energy, leads to a dissipative extension of classical gravitation [66]. Now, in our third example, we extend that calculation and introduce dual internal variables in order to explore a theory of gravitation with inertia.

We consider a single-component fluid; therefore the balances of mass and energy, (4.2) and (4.5), are the constraints for the entropy balance. Two scalar fields are introduced, φ and ψ . Our basic assumption is that the internal energy u is the difference between the total energy and the energies of the gravitating matter and field:

$$u = e - \varphi - \frac{\partial_i \varphi \partial^i \varphi}{8\pi G \rho} - \frac{\psi^2}{2K}. \quad (5.1)$$

Here, G is the gravitational constant and φ is the gravitational potential. It is subtracted from the specific internal energy, and the square gradient of φ represents the energy of the gravitational field [67]. Additive separation of the internal variables from the energy leads to energy interpretation. The last quadratic term is responsible for inertial effects; K is an inertial coefficient, as usual in variational principles. Then the specific entropy is a function also of u and the specific volume $v = 1/\rho$. The Gibbs relation is

$$de = T ds + \left(\frac{p}{\rho^2} - \frac{\partial_i \varphi \partial^i \varphi}{8\pi G \rho^2} \right) d\rho + d\varphi + \left(\frac{\partial_i \varphi}{8\pi G \rho} \right) d\partial^i \varphi + \frac{\psi}{K} d\psi. \quad (5.2)$$

Similarly as before, the calculation of the entropy balance is

$$\begin{aligned} \rho \dot{s} + \partial_i \left[\frac{1}{T} \left(q^i + \frac{1}{4\pi G} \dot{\varphi} \partial^i \varphi \right) \right] \\ = \left(q^i + \frac{\dot{\varphi}}{4\pi G} \partial^i \varphi \right) \partial_i \left(\frac{1}{T} \right) + \frac{\dot{\varphi}}{4\pi G T} \left(\partial_k \partial^k \varphi - 4\pi G \rho \right) \\ - \frac{\rho}{KT} \psi \dot{\psi} - \left[P^{ij} - p \delta^{ij} - \frac{1}{4\pi G} \left(\partial^i \varphi \partial^j \varphi - \frac{1}{2} \partial_k \varphi \partial^k \varphi \delta^{ij} \right) \right] \frac{\partial_i v_j}{T} \geq 0. \end{aligned} \quad (5.3)$$

The corresponding fluxes and forces are shown in table 3. Both the mechanical and thermal thermodynamic forces have changed due to the presence of gravitation. There is a contribution to the pressure, $P_{\text{grav}}^{ij} = (1/4\pi G)(\partial^i \varphi \partial^j \varphi - \frac{1}{2} \partial_k \varphi \partial^k \varphi \delta^{ij})$, too. The first term, the thermal interaction, is a product of vectors; the last term, the mechanical one, is a product of second-order tensors. The scalar second and the third terms determine the evolution equations for the gravitational potential and for the second scalar field, φ and ψ , respectively. Therefore, in isotropic materials, cross-effects are possible between the scalar part of the pressure and the scalar fields. Hence the linear constitutive equations are

$$q^i + \frac{\dot{\varphi}}{4\pi G} \partial^i \varphi = -\lambda \frac{\partial_i T}{T}, \quad (5.4)$$

$$\dot{\varphi} = l_1 \left(\frac{\partial_i \partial^i \varphi}{4\pi G} - \rho \right) - l_{12} \rho \frac{\psi}{K} - l_{13} \partial_k v^k, \quad (5.5)$$

$$\dot{\psi} = l_{21} \left(\frac{\partial_i \partial^i \varphi}{4\pi G} - \rho \right) - l_{2\rho} \frac{\psi}{K} - l_{23} \partial_k v^k, \quad (5.6)$$

Table 3. Thermodynamic fluxes and forces of self-gravitating fluids with gravitational inertia. P_{grav}^{ij} is the gravitational pressure.

	thermal	gravitational	mechanical	gravoinertial
fluxes	$q^i + \frac{\dot{\varphi}}{4\pi G} \partial^i \varphi$	$\dot{\varphi}$	$P^{ij} - \rho \delta^{ij} - P_{\text{grav}}^{ij}$	$\dot{\psi}$
forces	$\partial_i \left(\frac{1}{T} \right)$	$\frac{1}{T} \left(\frac{\partial_k \partial^k \varphi}{4\pi G} - \rho \right)$	$-\frac{\partial_i v_j}{T}$	$-K\rho\psi$

$$P_i^i - 3p + \frac{\partial_i \varphi \partial^i \varphi}{8\pi G} = l_{31} \left(\frac{\partial_i \partial^i \varphi}{4\pi G} - \rho \right) - l_{32} \rho \frac{\psi}{K} - l_3 \partial_k v^k \quad (5.7)$$

and

$$P^{ij} - P_k^k \frac{\delta^{ij}}{3} - \frac{1}{4\pi G} \left(\partial^i \varphi \partial^j \varphi - \frac{1}{3} \partial_k \varphi \partial^k \varphi \delta^{ij} \right) = -\eta \left(\partial^i v^j + \partial^j v^i - \frac{2}{3} \partial_k v^k \delta^{ij} \right). \quad (5.8)$$

Here, $l_3/3 = \eta_b$ is the bulk viscosity and η is the shear viscosity. The classical Newtonian gravitation is obtained if there is no gravitation-related dissipation, that is, the thermodynamic force of gravitation is zero, that is, the Poisson equation is valid and l_{12}, l_{13}, l_{31} and l_{32} are zero. In this case, the gravitational pressure has the following remarkable property: $\partial_j P_{\text{grav}}^{ij} = \rho \partial^i \varphi$. Let us observe the contribution of the dissipative terms in (5.6) and (5.7), but without the coupling with divergence of velocity, that is

$$\dot{\varphi} = l_1 \left(\frac{\partial_i \partial^i \varphi}{4\pi G} - \rho \right) - l_{12} \rho \frac{\psi}{K} \quad (5.9)$$

and

$$\dot{\psi} = l_{21} \left(\frac{\partial_i \partial^i \varphi}{4\pi G} - \rho \right) - l_2 \rho \frac{\psi}{K}, \quad (5.10)$$

where $l_1 > 0$, $l_2 > 0$ and $l_1 l_2 - (l_{12} + l_{21})^2/4 > 0$, according to the second law. Eliminating the second field, ψ , one obtains the following differential equation:

$$\begin{aligned} K\rho\ddot{\varphi} - K\rho l_1 \frac{d}{dt} \left(\frac{\partial_i^i \varphi}{4\pi G} - \rho \right) - L \left(\frac{\partial_i^i \varphi}{4\pi G} - \rho \right) + l_2 \dot{\varphi} \\ = K\rho l_1 \frac{d}{dt} \left(\frac{\partial_i^i \varphi}{4\pi G} - \frac{\dot{\rho}}{l_1} - \rho \right) - LK \left(\frac{\partial_i^i \varphi}{4\pi G} - \frac{l_2}{L} \dot{\varphi} - \rho \right) = 0. \end{aligned} \quad (5.11)$$

This is the governing equation for the dissipative, massive Newtonian gravitation. The non-dissipative case is obtained if the transport matrix is antisymmetric, i.e. $l_1 = l_2 = 0$ and $l_{12} = -l_{21} = a$. In this case, (5.11) reduces to the wave equation, as expected:

$$\frac{K4\pi G}{a^2} \rho \ddot{\varphi} - \partial_i^i \varphi - 4\pi G \rho = 0. \quad (5.12)$$

6. Summary and conclusion

In this paper, we have surveyed the capabilities of modern thermodynamics to construct evolution equations reflecting memory and non-local phenomena. The focus was on the evolution equations of ideal processes, which are typically constructed from variational principles. The demonstrative examples of the previous sections considered pure memory, pure non-local and a mixed memory and non-local situations. In every case, the second law was applied to obtain the evolution equations and non-dissipative processes emerged in the case of zero entropy production.

In §3, we have shown that Hamiltonian dynamics of a point mass can be reproduced in a thermodynamic framework: instead of starting from a Lagrangian, one can get the symplectic form of Hamiltonian mechanics and also the dissipative extension, both with direct

thermodynamic arguments. Some dissipative effects are unexpected and surprising from a pure mechanical point of view. In §4, the example of a two-component Fourier–Navier–Stokes–Cahn–Hilliard–Korteweg fluid demonstrated thermodynamic methods in the case of a weakly non-local continuum. The simplest classical method of divergence separation was applied, with a straightforward generalization of the Gibbs relation for deriving the Cahn–Hilliard equation. This equation is typically constructed with a combination of variational and thermodynamic principles. Here, the balances of mass and energy are constraints for the entropy inequality. The traditional method of nonequilibrium thermodynamics reproduced the same set of equations that have corrected the variational approach of Lowengrub & Truskinovsky [60] and were obtained by rigorous arguments of rational thermodynamics [63].

In §5, we have shown a combined utilization of dual variables and divergence separation to get both memory and non-local equations by constructing a dissipative theory of Newtonian gravitation with inertial effects. It is remarkable that the wave equation of the gravitational potential was only a part of the non-dissipative limit of the theory: the scalar evolution equation of the gravitational potential can be coupled to the scalar part of the mechanical interaction, to the spherical part of the stress field.

In every example we have given some particular predictions, some new aspects due to the construction method and to the thermodynamic approach. These examples demonstrate that nonequilibrium thermodynamics can substitute variational principles and construct both non-dissipative and dissipative parts of the evolution equations without any further ado. The Hamilton principle emerges for the non-dissipative part. For phase fields, where only the spatial part of a variational principle is necessary, the substitution is complete.

At the end, let us outline the Hamilton variational principle for a scalar field ϕ in non-relativistic space–time with a first-order weakly non-local density. This is constructed from a Lagrangian, $L(\phi, \partial_t\phi, \partial_i\phi)$, and based on the next formal procedure, the ‘variation’ of the action functional, the integral of the Lagrangian at a compact $V \times [t_1, t_2]$ space–time domain:

$$\begin{aligned} \delta S(\phi) &= \delta \int_t \int_V L(\phi, \partial_t\phi, \partial_i\phi) dt dV = \int_t \int_V \delta L(\phi, \partial_t\phi, \partial_i\phi) dt dV \\ &= \int_t \int_V [\partial_\phi L - \partial_t(\partial_{\partial_t\phi} L) - \partial_i(\partial_{\partial_i\phi} L)] \delta\phi dt dV \\ &\quad + \int_V \partial_{\partial_t\phi} L \delta\phi dV \Big|_{t_1}^{t_2} + \int_t \oint_{\partial V} \partial_{\partial_i\phi} L \delta\phi dt dA_i = 0. \end{aligned} \quad (6.1)$$

Then one concludes that the following Euler–Lagrange equation is valid:

$$\partial_\phi L - \partial_t(\partial_{\partial_t\phi} L) - \partial_i(\partial_{\partial_i\phi} L) = 0. \quad (6.2)$$

When properly formulated it is a differentiation of S on a Banach space of functions that disappear at $\partial V \times \{t_1, t_2\}$, at a boundary of the space–time domain. There is an affine space behind, with natural boundary conditions fixed with the last two integrals of (6.1). The Banach space derivative is zero, because of the assumed extremum property, the Euler–Lagrange equation (6.2) is a necessary condition.

As a physical principle, it is a disaster: the Lagrangian is coming out of the blue, the action is not an extremum in several physical theories, initial value problems are not included, most of the fields in physics are measures and not functions, etc., considering only some aspects of the mathematical formulation. Moreover, in most of the classical, everyday physics, dissipative processes are excluded; evolution equations for dissipative processes cannot be generated using this variational principle without any further ado.

One can remedy several of the mentioned problems: there are more general function spaces than Banach, initial value problems can be considered, some dissipative processes can be added. As a tool, it is useful; however, as a principle, it is compromised. The limited validity of the approach cannot be denied.

Up to now, attempts to find a principal approach for constructing evolution equations for dissipative processes have been based on well-established methods for non-dissipative processes. We were looking for extensions and modifications of the variational principle (6.1) preserving some elements of the formal method and the idea of the extremum. In this paper, we argue that one can start from the other end and assume an inequality instead of an equality. Interestingly and importantly, several methods based on the analysis of this inequality result in solutions in an Euler–Lagrange form. This observation enables us to formulate a research programme. One can postpone the decision whether the fields are emergent or not and what kind of microscopic and submicroscopic composition of the material can lead to the inequality. Instead, we may assume that entropy, like a Lagrangian, is the generating potential of the evolution of the fields and deal with the consequences.

This programme is already running, mostly unconsciously, in the various branches of nonequilibrium thermodynamics. Any methods that construct evolution equations with thermodynamic methods are contributing. We have already mentioned extended thermodynamics, GENERIC, conservation–dissipation formalism, steepest entropy descent, virtual power and internal variables [21,22,35,36,68–71]. It is very successful in classical, non-relativistic situations where dissipation is apparent.

There are also some conceptual consequences. First of all, it is a unification. Our evolution generator extremum principle, the selection rule for the laws of Nature, is not independent any more; it is of thermodynamic origin. Thermodynamic, but not statistical; the axioms are related to the stability structure, the existence of a concave and increasing potential. Is the second law elevated to a first principle level, or are the first principles better understood as emergent? We prefer the first interpretation: the rule is to select the stable materials with the entropy as a Lyapunov functional of the equilibrium. Stability is clearly a selection rule; unstable materials survive only under special conditions. The second interpretation denies the existence of first principles, e.g. for theories like gravitation [72,73]. If everything is emergent, then processes without dissipation are exceptional. Then energy does not leak towards the ever existing microlevel, it is somehow insulated. The fact that the two interpretations are practically the same is a disturbing consequence. It may be reasonable to postpone the decision until more is known about the performance of this possible unification, e.g. until electrodynamics is derived from nonequilibrium thermodynamics.

If everything were to be dissipative, non-dissipative evolution arises as a particular limit. However, in most of the fundamental theories, it is definitely not an approximation. We believe that it is an inherent property of physical phenomena; we consider the corresponding equations identically fulfilled. Let us recall, for example, gravitation. As far as we know, gravitation is not dissipative. That is true, but that is not a contradiction.

- First, this is always an experimental question. There are more and more precise experiments that measure the validity of equivalence principles of inertial and gravitational masses and other experiments that check the validity of Lorentz invariance [74]. In these cases, most of our theories assume equivalence; violation of Lorentz covariance is not expected. Still, it is always necessary to validate our most fundamental assumptions, too.
- Second, what is the reason for the existence of ideal, non-dissipative evolution at various levels of length scales? Atomic, hadronic and quark levels are seemingly independent and apparently non-dissipative. Then a theory in which stability of the material is the guiding idea provides a practical and deep explanation: the various ideal levels are stabilized due to physical processes. We are far from understanding the details here [75], but renormalization is definitely a dissipative method [76,77].
- There is also a third, very practical, point here: it is easier to solve dissipative equations than non-dissipative ones. Therefore, a dissipative extension can be a tool to design powerful numerical procedures [75,78].

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