

Thermodynamics of a continuous medium with electric and magnetic dipoles

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Abstract. The thermodynamics of an electrically charged, multicomponent fluid with spontaneous electric and magnetic dipoles is analysed in the presence of electromagnetic fields. Taking into account the chemical composition of the current densities and stress tensors leads to three types of irreversible terms: scalars, vectors and pseudo-vectors. The scalar terms account for chemical reactivities, the vectorial terms account for transport and the pseudo-vectorial terms account for relaxation. The linear phenomenological relations, derived from the irreversible evolution, describe notably the Lehmann and electric Lehmann effects, the Debye relaxation of polar molecules and the Landau-Lifshitz relaxation of the magnetisation. This formalism accounts for the thermal and electric magnetisation accumulations and magnetisation waves. It also predicts that a temperature gradient affects the dynamics of magnetic vortices and drives magnetisation waves.

1 Introduction

Spin caloritronics is mainly focused on studying the effects of a temperature gradient on the time evolution of the distribution of the local spin average of a physical system [1]. In many experimental situations, the system can be treated as a classical continuum with magnetisation on the scale of interest where the quantum fluctuations average out and the underlying microscopic structure is smoothed out. In such a case, the local system is sufficiently large from a microscopic perspective to be treated classically, but it is sufficiently small from a macroscopic perspective to be considered as infinitesimal. In order to understand the interplay between the magnetisation and a temperature gradient, a consistent classical thermodynamic theory of continuous media with magnetisation needs to be established. It is of interest to include also the electric counterpart of the magnetisation, that is the electric polarisation. The thermodynamics of continuous media is presented in details in textbooks by Gurtin et al. [2], Wilmanski [3] and Silhavy [4]. In order to account for the intrinsic rotation of the matter, we need to introduce explicitly an intrinsic angular velocity. The introduction of intrinsic rotations into the thermodynamics of continuous media was discussed notably by Müller [5] and Muschik et al. [6,7]. The electrodynamics of an electrically charged continuous medium interacting with electromagnetic fields is detailed in textbooks by Eringen and Maugin [8] and O'Dell [9]. Some pioneering work on the non-equilibrium thermodynamics of a continuous medium

with electromagnetic fields was carried out by Liu and Müller [10], Hutter [11] and de Groot and Mazur [12]. In the present work, we follow essentially the approach of Stueckelberg detailed in reference [13] and extend his formalism to describe locally the thermodynamics of an electrically charged continuous medium with spontaneous electric and magnetic dipoles in the presence of electromagnetic fields.

In order to be able to define intensive fields such as the temperature, the chemical potential, the electrostatic potential, the electric field and the magnetic induction field, we require the local infinitesimal system to be homogeneous, uniform and at equilibrium.

We introduce phenomenological relations that express, in terms of the chemical constituents of the continuous medium, the mass density, the electric charge density, the intrinsic angular mass density, the electric polarisation and magnetisation, the momentum and the intrinsic angular momentum. Using these phenomenological relations, we express the electric polarisation and magnetisation current tensors as well as the stress and angular stress tensors in terms of the chemical current densities. We show that by expressing the state fields, such as the electric polarisation and the magnetisation, and the dynamical fields, such as the momentum density and the intrinsic angular momentum density, in terms of the different elementary constituents of the medium, three types of irreversible terms appear. First, there are scalar terms that account for the chemical reactivities and are expressed as the product between the chemical reaction rate densities and the chemical affinities. Second, there are vectorial terms that account for the transport and are expressed as the

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dot product between the current densities and the forces. Third, there are pseudo-vectorial terms that account for the relaxation and are expressed as the dot product between the intrinsic rotation rate densities and the intrinsic torques.

To illustrate our formalism, we derive explicit expressions for the irreversible relations characterising the Lehmann and electric Lehmann effects, and the relaxation of the electric and magnetic dipoles. Furthermore, we show how this formalism accounts for the thermal and electric magnetisation accumulations and magnetisation waves. We also predict how a temperature gradient affects the dynamics of magnetic vortices and drives magnetisation waves.

The structure of this publication is the following. In Section 2, we define the continuity equations, which describe the local time evolution for the material state variables. Then, we express the time evolution equations in terms of the different chemical constituents. In Section 3, using the fact that the energy is a state function, we combine the time evolution equations for the internal energy, the entropy and the other extensive material state variables. This ensures the local compatibility of the first and second laws of thermodynamics with the laws of mechanics. Through symmetry arguments, we establish the thermostatic, the reversible and irreversible thermodynamic equations. Finally, in Section 4, we establish the irreversible thermodynamic phenomenology and describe some physical phenomena to illustrate our formalism, with a particular emphasis on spintronics.

2 Thermodynamic description

2.1 Continuity equations

The state of a continuous medium is defined by a set of matter state variables. The local state of a continuous medium is defined by a set of matter state fields that are function of the space and time coordinates. To keep the notation concise, we do not explicitly write the space and time dependence of the state fields. The local thermodynamic state of a classical electrically charged continuous medium consisting of N chemical substances with electric polarisation and magnetisation interacting with electromagnetic fields is determined by the following state fields:

- the entropy density field s ;
- the number density fields n_A of N chemical substances, where $A \in \{1, \dots, N\}$;
- the electric charge density field q ;
- the spontaneous electric polarisation field \mathbf{P} ;
- the spontaneous magnetisation field \mathbf{M} ;
- the velocity field \mathbf{v} ;
- the intrinsic angular velocity field $\boldsymbol{\omega}$.

It is important to stress that the system does not include the electromagnetic fields in the vacuum, but only their interaction with the matter fields. Thus, the state fields q , \mathbf{P} and \mathbf{M} , accounting for the electromagnetic properties of the matter, are purely matter fields, in contrast to our

previous work [14,15] where the electric displacement field \mathbf{D} and the magnetic induction field \mathbf{B} were accounting for the electromagnetic properties of the matter and of the electromagnetic fields in the vacuum.

Moreover, in this publication, we restrict our analysis to a system with spontaneous electric polarisation \mathbf{P} and magnetisation \mathbf{M} , whereas in our former work [14,15], we considered only a system with induced electric polarisation and magnetisation.

The classical time evolution of every extensive physical property in a local infinitesimal system is expressed by a continuity equation, which is a local detailed balance equation. In this publication, we shall refer to the continuity equations expressed in terms of the material time derivative as the material continuity equations.

The material continuity equation for an extensive scalar property F is given by reference [14],

$$\dot{f} + (\nabla \cdot \mathbf{v}) f + \nabla \cdot \mathbf{j}_f = \rho_f, \quad (1)$$

where \dot{f} is the material time derivative of the scalar density f , \mathbf{j}_f is the vectorial diffusive current density and ρ_f is the scalar source density of F . Note that the frame-independent material time derivative operator is related to the partial time derivative operator by,

$$\dot{} = \partial_t + (\mathbf{v} \cdot \nabla).$$

The material continuity equation for an extensive vectorial property \mathbf{F} is given by [14],

$$\dot{\mathbf{f}} + (\nabla \cdot \mathbf{v}) \mathbf{f} + \nabla \cdot \mathbf{j}_f = \boldsymbol{\rho}_f, \quad (2)$$

where $\dot{\mathbf{f}}$ is the material time derivative of the vectorial density \mathbf{f} , \mathbf{j}_f is the rank-2 tensorial diffusive current density and $\boldsymbol{\rho}_f$ is the vectorial source density of \mathbf{F} .

The material continuity equation (1) for the entropy yields,

$$\dot{s} + (\nabla \cdot \mathbf{v}) s + \nabla \cdot \mathbf{j}_s = \rho_s \geq 0, \quad (3)$$

where s is the entropy density, \mathbf{j}_s is the diffusive entropy current density and ρ_s is the entropy source density that is positive-definite in order to satisfy locally the second law of thermodynamics. Note that the strictly positive part, i.e. $\rho_s > 0$, accounts for the irreversibility by breaking the symmetry of the dynamics under time inversion, which defines a time arrow.

The material continuity equation (1) for the number of elementary units of chemical substance A is given by,

$$\dot{n}_A + (\nabla \cdot \mathbf{v}) n_A + \nabla \cdot \mathbf{j}_A = \sum_a \nu_{aA} \omega_a, \quad (4)$$

where n_A is the number density of the chemical substance A , \mathbf{j}_A is the diffusive current density and the source density ρ_A is expressed explicitly as the sum over all the chemical reactions a of the product of the stoichiometric coefficients ν_{aA} and the reaction rate densities ω_a .

The material continuity equation (1) for the electric charge is given by,

$$\dot{q} + (\nabla \cdot \mathbf{v}) q + \nabla \cdot \mathbf{j}_q = 0, \quad (5)$$

where q is the electric charge density, \mathbf{j}_q is the diffusive electric current density and there is no electric charge source density ρ_q since the electric charge is an invariant. Note that the electric current density \mathbf{j} is the sum of the convective and diffusive electric current densities, i.e.

$$\mathbf{j} = q \mathbf{v} + \mathbf{j}_q. \quad (6)$$

The material continuity equation (2) for the spontaneous electric dipoles is given by,

$$\dot{\mathbf{P}} + (\nabla \cdot \mathbf{v}) \mathbf{P} + \nabla \cdot \mathbf{j}_P = \sum_A \Omega_A^m \times \mathbf{p}_A, \quad (7)$$

where \mathbf{P} is the spontaneous electric polarisation, \mathbf{j}_P is the diffusive electric polarisation current tensor. The source density ρ_P of the spontaneous electric dipoles \mathbf{p}_A of the chemical substances A is an irreversible term accounting for the relaxation of the spontaneous electric dipoles \mathbf{p}_A . It is due to the rotational motion of the chemical substances A , that is expressed in terms of the intrinsic rotation rate densities Ω_A^m of the matter where the super-script m stands for ‘‘matter’’.

The uniformity of the local infinitesimal system implies that there is a unique local rotation rate density Ω_A^m for each chemical substance A , that is defined with respect to the local frame where the electric polarisation \mathbf{P} does not rotate. This frame corresponds to the local frame where the intrinsic rotation of the matter vanishes, i.e. $\boldsymbol{\omega} = \mathbf{0}$, since the spontaneous electric polarisation is rigidly attached to the matter.

The material continuity equation (2) for the spontaneous magnetic dipoles yields,

$$\dot{\mathbf{M}} + (\nabla \cdot \mathbf{v}) \mathbf{M} + \nabla \cdot \mathbf{j}_M = \sum_A \left(\gamma_A n_A (\mathbf{m}_A \times \mathbf{B}) + \Omega_A^M \times \mathbf{m}_A \right), \quad (8)$$

where \mathbf{M} is the spontaneous magnetisation, \mathbf{j}_M is the diffusive magnetisation current tensor. The source density ρ_M of the spontaneous magnetic dipoles \mathbf{m}_A of the chemical substances A consists of two terms. The first term is a reversible term accounting for the precession of the magnetic dipoles \mathbf{m}_A around the magnetic induction field \mathbf{B} that is expressed in terms of their gyromagnetic ratios γ_A . The second term is an irreversible term accounting for the relaxation of the magnetic dipoles \mathbf{m}_A with respect to the magnetic induction field \mathbf{B} that is expressed in terms of the intrinsic rotation rate densities Ω_A^M where the super-script M stands for ‘‘magnetisation’’. These rotation rate densities consist of two contributions. The first contribution corresponds to the micromagnetic hypothesis [16], that accounts for the fact that electrons are point-like particles that have no known substructure and thus no intrinsic angular mass. The second contribution accounts for the rotational motion of the chemical substances A . The second contribution is generally much smaller than the first.

The uniformity of the local infinitesimal system implies that there is a unique local rotation rate density Ω_A^M for

each chemical substance A , that is defined with respect to the local frame where the magnetisation \mathbf{M} does not rotate. In contrast to the case of the electric polarisation, this frame does not correspond to the local frame where the intrinsic rotation of the matter vanishes, since the spontaneous magnetisation is not rigidly attached to the matter. Note that by symmetry, only axial vectors such as the magnetic dipoles \mathbf{m}_A are allowed to precess. Thus, no precession of the electric dipoles \mathbf{p}_A is to be expected.

The material continuity equation (2) for the momentum is given by,

$$\dot{\mathbf{p}} + (\nabla \cdot \mathbf{v}) \mathbf{p} - \nabla \cdot \boldsymbol{\sigma} = \mathbf{f}^{\text{ext}}, \quad (9)$$

where \mathbf{p} is the momentum density, $\boldsymbol{\sigma}$ is the stress tensor and \mathbf{f}^{ext} is the external force density. The momentum density $\mathbf{p}(n_A, \mathbf{v})$ of the matter is proportional to the velocity \mathbf{v} , i.e.

$$\mathbf{p}(n_A, \mathbf{v}) = m(n_A) \mathbf{v}, \quad (10)$$

where $m(n_A)$ is the mass density.

The material continuity equation (1) for the mass yields,

$$\dot{m} + (\nabla \cdot \mathbf{v}) m = 0. \quad (11)$$

There is no diffusive mass current density \mathbf{j}_m by definition of the local centre of mass and there is no mass source density ρ_m since the mass is a non-relativistic invariant. Substituting the relation (10) and the continuity equation (11) for the mass into the continuity equation (9) for the momentum, the latter yields Newton’s second law of motion, i.e.

$$m \dot{\mathbf{v}} = \mathbf{f}^{\text{ext}} + \nabla \cdot \boldsymbol{\sigma}. \quad (12)$$

The material continuity equation (2) for the intrinsic angular momentum yields,

$$\dot{\mathbf{s}} + (\nabla \cdot \mathbf{v}) \mathbf{s} - \nabla \cdot \boldsymbol{\Theta} = \boldsymbol{\tau}^{\text{ext}}, \quad (13)$$

where \mathbf{s} is the intrinsic angular momentum density, $\boldsymbol{\Theta}$ is the intrinsic angular stress tensor and $\boldsymbol{\tau}^{\text{ext}}$ is the intrinsic torque density. The local infinitesimal system is considered as a homogeneous sphere. Thus, the intrinsic angular momentum density $\mathbf{s}(n_A, \boldsymbol{\omega})$ of the matter is proportional to the intrinsic angular velocity $\boldsymbol{\omega}$, i.e.

$$\mathbf{s}(n_A, \boldsymbol{\omega}) = I(n_A) \boldsymbol{\omega}, \quad (14)$$

where $I(n_A)$ is the intrinsic angular mass density, which is a scalar density. For a homogeneous sphere of constant infinitesimal radius dr , the intrinsic angular mass density I is related to the mass density m by,

$$I = \frac{2}{5} m dr^2. \quad (15)$$

Thus, the material continuity equation (11) for the mass and the relation (15) imply that the material continuity equation (1) for the intrinsic angular mass is given by:

$$\dot{I} + (\nabla \cdot \mathbf{v}) I = 0. \quad (16)$$

There is no diffusive intrinsic angular mass current density \mathbf{j}_I and no intrinsic angular mass source density ρ_I

since for a homogeneous local system the intrinsic angular mass density I is totally determined by the mass density m .

Substituting the relation (14) and the continuity equation (16) for the intrinsic angular mass into the continuity equation (13) for the intrinsic angular momentum, the latter yields Newton's second law in intrinsic rotation, i.e.

$$I \dot{\boldsymbol{\omega}} = \boldsymbol{\tau}^{\text{ext}} + \nabla \cdot \boldsymbol{\Theta}. \quad (17)$$

The material continuity equation (1) for the energy is given by,

$$\dot{e} + (\nabla \cdot \mathbf{v}) e + \nabla \cdot \mathbf{j}_e = \mathbf{f}^{\text{ext}} \cdot \mathbf{v} + \boldsymbol{\tau}^{\text{ext}} \cdot \boldsymbol{\omega}, \quad (18)$$

where e is the energy density, \mathbf{j}_e is the diffusive energy current density and the energy source density ρ_e is the power density due to the external force densities \mathbf{f}^{ext} and external intrinsic torque densities $\boldsymbol{\tau}^{\text{ext}}$. The energy density $e(s, n_A, q, \mathbf{P}, \mathbf{M}, \mathbf{v}, \boldsymbol{\omega})$ is the sum of the translational kinetic energy density, i.e. $\frac{1}{2} m \mathbf{v}^2$, the intrinsic rotational kinetic energy density, i.e. $\frac{1}{2} I \boldsymbol{\omega}^2$, and the internal energy density, i.e. $u(s, n_A, q, \mathbf{P}, \mathbf{M})$,

$$e(s, n_A, q, \mathbf{P}, \mathbf{M}, \mathbf{v}, \boldsymbol{\omega}) = \frac{1}{2} m \mathbf{v}^2 + \frac{1}{2} I \boldsymbol{\omega}^2 + u(s, n_A, q, \mathbf{P}, \mathbf{M}), \quad (19)$$

where the internal energy density is the energy density in the local rest frame where $\mathbf{v} = \mathbf{0}$ and $\boldsymbol{\omega} = \mathbf{0}$. Using the continuity equations for the mass (11) and the intrinsic angular mass (16), Newton's second law in translation (12) and in intrinsic rotation (17), the time derivative of the relation (19) yields,

$$\dot{e} = \dot{u} + (\nabla \cdot \mathbf{v})(u - e) + (\nabla \cdot \boldsymbol{\sigma}) \cdot \mathbf{v} + (\nabla \cdot \boldsymbol{\Theta}) \cdot \boldsymbol{\omega} + \mathbf{f}^{\text{ext}} \cdot \mathbf{v} + \boldsymbol{\tau}^{\text{ext}} \cdot \boldsymbol{\omega}. \quad (20)$$

Substituting the relation (20) into the continuity equation (18) for the energy and using the vectorial identities

$$\begin{aligned} (\nabla \cdot \boldsymbol{\sigma}) \cdot \mathbf{v} &= \nabla_{\boldsymbol{\sigma}} \cdot (\boldsymbol{\sigma} \cdot \mathbf{v}) - \boldsymbol{\sigma} \cdot (\nabla \odot \mathbf{v}), \\ (\nabla \cdot \boldsymbol{\Theta}) \cdot \boldsymbol{\omega} &= \nabla_{\boldsymbol{\Theta}} \cdot (\boldsymbol{\Theta} \cdot \boldsymbol{\omega}) - \boldsymbol{\Theta} \cdot (\nabla \odot \boldsymbol{\omega}), \end{aligned}$$

yields the material continuity equation (1) for the internal energy density, i.e.

$$\dot{u} + (\nabla \cdot \mathbf{v}) u + \nabla \cdot \mathbf{j}_u = \boldsymbol{\sigma} \cdot (\nabla \odot \mathbf{v}) + \boldsymbol{\Theta} \cdot (\nabla \odot \boldsymbol{\omega}), \quad (21)$$

where the symbol \odot denotes a symmetrised tensorial product \otimes and the indices $\boldsymbol{\sigma}$ or $\boldsymbol{\Theta}$ denote that there is a dot product between the covariant differential operator ∇ and the contravariant components of the stress tensors $\boldsymbol{\sigma}$ and $\boldsymbol{\Theta}$, respectively.

The internal energy current density \mathbf{j}_u is related to the energy current density \mathbf{j}_e by:

$$\mathbf{j}_e = \mathbf{j}_u - \boldsymbol{\sigma} \cdot \mathbf{v} - \boldsymbol{\Theta} \cdot \boldsymbol{\omega}. \quad (22)$$

Note that the source density terms $\boldsymbol{\sigma} \cdot (\nabla \odot \mathbf{v})$ and $\boldsymbol{\Theta} \cdot (\nabla \odot \boldsymbol{\omega})$ in the continuity equation (21) for the internal energy account for the elastic expansion or contraction of the matter and for the dissipative translational and rotational viscous friction. The dissipation is defined as the irreversible part of the internal energy source density ρ_u .

2.2 Dynamics in terms of the chemical composition

2.2.1 Time evolution of the electric charge density

The electric charge density q is defined as the product of the number density n_A and the electric charge q_A carried by the elementary units of chemical substances A [14], i.e.

$$q = \sum_A n_A q_A. \quad (23)$$

In order to characterise physically the time evolution of the electric polarisation, we substitute the expression (23) for the chemical composition of the electric charge density and the continuity equation for the chemical substance (4) into the continuity equation (5) for the electric charge. The latter then becomes,

$$\sum_A \left(-(\nabla \cdot \mathbf{j}_A) q_A + \sum_a \nu_{aA} \omega_a q_A \right) + \nabla \cdot \mathbf{j}_q = 0. \quad (24)$$

Using the fact that the electric charge q_A is an invariant, i.e.

$$(\nabla \cdot \mathbf{j}_A) q_A = \nabla \cdot (q_A \mathbf{j}_A),$$

the continuity equation (24) for the electric charge can be recast as:

$$\sum_{a,A} \nu_{aA} \omega_a q_A + \nabla \cdot \left(\mathbf{j}_q - \sum_A q_A \mathbf{j}_A \right) = 0. \quad (25)$$

The continuity equation (25) has to hold for any current flow, which yields an explicit expression for the diffusive electric current density, i.e.

$$\mathbf{j}_q = \sum_A q_A \mathbf{j}_A. \quad (26)$$

Moreover, it has to hold for every chemical reaction a , which implies that

$$\sum_A \nu_{aA} q_A = 0, \quad (27)$$

and means that the chemical reaction a preserves the total electric charge in the local infinitesimal system.

2.2.2 Time evolution of the electric dipoles

The electric polarisation \mathbf{P} is defined as the product of the number density n_A and the electric dipoles \mathbf{p}_A carried by the elementary units of chemical substances A , i.e.

$$\mathbf{P} = \sum_A n_A \mathbf{p}_A. \quad (28)$$

In order to characterise physically the dynamics of the electric polarisation, we substitute the expression (28) for the chemical composition of the electric polarisation and the continuity equation for the chemical substance (4) into

the continuity equation (7) for the electric dipoles. The latter then becomes,

$$\sum_A \left(n_A \dot{\mathbf{p}}_A - (\nabla \cdot \mathbf{j}_A) \mathbf{p}_A + \sum_a \nu_{aA} \omega_a \mathbf{p}_A \right) + \nabla \cdot \mathbf{j}_P = \sum_A \boldsymbol{\Omega}_A^m \times \mathbf{p}_A. \quad (29)$$

Using the vectorial identity,

$$(\nabla \cdot \mathbf{j}_A) \mathbf{p}_A = \nabla_{\mathbf{j}} \cdot (\mathbf{p}_A \odot \mathbf{j}_A) - (\mathbf{j}_A \cdot \nabla) \mathbf{p}_A,$$

where the index \mathbf{j} denotes that there is a dot product between the covariant differential operator ∇ and the contravariant current density \mathbf{j}_A , the continuity equation (29) for the electric dipoles can be recast as,

$$\sum_A \left(n_A \dot{\mathbf{p}}_A - \boldsymbol{\Omega}_A^m \times \mathbf{p}_A + (\mathbf{j}_A \cdot \nabla) \mathbf{p}_A \right) + \sum_{a,A} \nu_{aA} \omega_a \mathbf{p}_A + \nabla_{\mathbf{j}} \cdot \left(\mathbf{j}_P - \sum_A \mathbf{p}_A \odot \mathbf{j}_A \right) = \mathbf{0}. \quad (30)$$

The continuity equation (30) has to hold for any current flow, which yields an explicit expression for the electric polarisation current tensor, i.e.

$$\mathbf{j}_P = \sum_A \mathbf{p}_A \odot \mathbf{j}_A. \quad (31)$$

The condition (31) implies that the continuity equation (30) reduces to:

$$\sum_A \left(n_A \dot{\mathbf{p}}_A - \boldsymbol{\Omega}_A^m \times \mathbf{p}_A + (\mathbf{j}_A \cdot \nabla) \mathbf{p}_A + \sum_a \nu_{aA} \omega_a \mathbf{p}_A \right) = \mathbf{0}, \quad (32)$$

which describes the time evolution of the electric dipoles \mathbf{p}_A of all the chemical substances.

The local time evolution of the electric dipoles \mathbf{p}_A of a specific chemical substance A is given by:

$$n_A \dot{\mathbf{p}}_A = -\mathbf{p}_A \times \boldsymbol{\Omega}_A^m - (\mathbf{j}_A \cdot \nabla) \mathbf{p}_A - \sum_a \nu_{aA} \omega_a \mathbf{p}_A, \quad (33)$$

where the first, second and third terms on the RHS describe, respectively, the relaxation, the transport and the chemistry of the electric dipoles \mathbf{p}_A . Note the intrinsic rotation rate densities $\boldsymbol{\Omega}_A^m$ are functions of the electric dipoles of all the chemical substances. They account for the dissipative couplings between the electric dipoles of the different chemical substances.

2.2.3 Time evolution of the magnetic dipoles

The magnetisation \mathbf{M} is defined as the product of the number density n_A and the magnetic dipoles \mathbf{m}_A carried by the elementary units of chemical substances A , i.e.

$$\mathbf{M} = \sum_A n_A \mathbf{m}_A. \quad (34)$$

In order to characterise physically the time evolution of the magnetisation, we substitute the expression (34) for the chemical composition of the magnetisation and the continuity equation for the chemical substance (4) into the continuity equation (8) for the magnetic dipoles. The latter then becomes,

$$\sum_A \left(n_A \dot{\mathbf{m}}_A - (\nabla \cdot \mathbf{j}_A) \mathbf{m}_A + \sum_a \nu_{aA} \omega_a \mathbf{m}_A \right) + \nabla \cdot \mathbf{j}_M = \sum_A \left(\gamma_A n_A \mathbf{m}_A \times \mathbf{B} + \boldsymbol{\Omega}_A^M \times \mathbf{m}_A \right). \quad (35)$$

Using the vectorial identity,

$$(\nabla \cdot \mathbf{j}_A) \mathbf{m}_A = \nabla_{\mathbf{j}} \cdot (\mathbf{m}_A \odot \mathbf{j}_A) - (\mathbf{j}_A \cdot \nabla) \mathbf{m}_A,$$

the continuity equation (35) for the magnetic dipoles can be recast as,

$$\sum_A \left(n_A (\dot{\mathbf{m}}_A - \gamma_A \mathbf{m}_A \times \mathbf{B}) - \boldsymbol{\Omega}_A^M \times \mathbf{m}_A + (\mathbf{j}_A \cdot \nabla) \mathbf{m}_A \right) + \sum_{a,A} \nu_{aA} \omega_a \mathbf{m}_A + \nabla_{\mathbf{j}} \cdot \left(\mathbf{j}_M - \sum_A \mathbf{m}_A \odot \mathbf{j}_A \right) = \mathbf{0}. \quad (36)$$

The continuity equation (36) has to hold for any current flow, which yields an explicit expression for the magnetisation current tensor, i.e.

$$\mathbf{j}_M = \sum_A \mathbf{m}_A \odot \mathbf{j}_A. \quad (37)$$

Note that this expression for the magnetisation current tensor is the classical counterpart of the tensorial ‘‘spin-current’’ used in spintronics [17]. The condition (37) implies that the continuity equation (36) reduces to,

$$\sum_A \left(n_A (\dot{\mathbf{m}}_A - \gamma_A \mathbf{m}_A \times \mathbf{B}) - \boldsymbol{\Omega}_A^M \times \mathbf{m}_A + (\mathbf{j}_A \cdot \nabla) \mathbf{m}_A + \sum_a \nu_{aA} \omega_a \mathbf{m}_A \right) = \mathbf{0}, \quad (38)$$

which describes the local time evolution of the magnetic dipoles of all the chemical substances.

In order to establish the local time evolution of the magnetic dipoles \mathbf{m}_A of a specific chemical substance A in the presence of other chemical substances B carrying magnetic dipoles \mathbf{m}_B , we need to take explicitly into account the local magnetic torque that couples \mathbf{m}_A and \mathbf{m}_B . This torque is antisymmetric under the permutation of different chemical substances A and B , which implies that the torque density vanishes after summation over all the chemical substances, i.e.

$$\sum_{A,B} \gamma_{AB} n_A n_B (\mathbf{m}_A \times \mathbf{m}_B) = \mathbf{0}, \quad (39)$$

where γ_{AB} is the symmetric coupling coefficient. Note that the magnetic torques do not affect the magnetisation \mathbf{M} ,

which is defined as the density of magnetic dipoles of all the chemical substances. Thus, they do not affect the Larmor energy of the local system. Note that by symmetry, the interaction torques are allowed only for axial vectors such as the magnetic dipoles \mathbf{m}_A and \mathbf{m}_B . Thus, no interaction torque of the electric dipoles \mathbf{p}_A and \mathbf{p}_B is to be expected.

Equations (38) and (39) imply that the local time evolution of the magnetic dipoles \mathbf{m}_A of a specific chemical substance A is given by,

$$n_A \dot{\mathbf{m}}_A = \gamma_A n_A (\mathbf{m}_A \times \mathbf{B}) - \mathbf{m}_A \times \boldsymbol{\Omega}_A^M - (\mathbf{j}_A \cdot \nabla) \mathbf{m}_A - \sum_a \nu_{aA} \omega_a \mathbf{m}_A + \sum_B \gamma_{AB} n_A (\mathbf{m}_A \times n_B \mathbf{m}_B), \quad (40)$$

where the first, second, third and fourth terms on the RHS describe, respectively, the precession, the relaxation, the transport and the chemistry of the magnetic dipoles \mathbf{m}_A , and the last term describes the local interaction of the magnetic dipoles \mathbf{m}_A with the magnetic dipoles \mathbf{m}_B of the other chemical substances B . Note the intrinsic rotation rate densities $\boldsymbol{\Omega}_A^M$ are functions of the magnetic dipoles of all the chemical substances. They account for the dissipative couplings between the magnetic dipoles of the different chemical substances.

2.2.4 Time evolution of the mass density

The mass density m is defined as the product of the number density n_A and the mass m_A carried by the elementary units of chemical substances A [14], i.e.

$$m = \sum_A n_A m_A. \quad (41)$$

In order to characterise physically the time evolution of the mass density, we substitute the expression (41) for the chemical composition of the mass density and the continuity equation for the chemical substance (4) into the continuity equation (11) for the mass. The latter then becomes,

$$\sum_A \left(-(\nabla \cdot \mathbf{j}_A) m_A + \sum_a \nu_{aA} \omega_a m_A \right) = 0. \quad (42)$$

Using the fact that the mass m_A is a non-relativistic invariant, i.e.

$$(\nabla \cdot \mathbf{j}_A) m_A = \nabla \cdot (m_A \mathbf{j}_A),$$

the continuity equation (42) for the mass can be recast as,

$$\sum_{a,A} \nu_{aA} \omega_a m_A - \nabla \cdot \left(\sum_A m_A \mathbf{j}_A \right) = 0. \quad (43)$$

The continuity equation (43) has to hold for any current flow, which defines a set of frames where the local centre of mass of the matter element is at rest, i.e.

$$\sum_A m_A \mathbf{j}_A = \mathbf{0}. \quad (44)$$

Moreover, it has to hold for every chemical reaction a , which implies that

$$\sum_A \nu_{aA} m_A = 0, \quad (45)$$

and means that the chemical reaction a preserves the total mass in the local infinitesimal system.

2.2.5 Time evolution of the momentum

The momentum density $\mathbf{p} = m \mathbf{v}$ is defined as the density of momentum carried by the elementary units of chemical substances A of velocities \mathbf{v}_A , i.e.

$$\mathbf{p} = \sum_A n_A m_A \mathbf{v}_A. \quad (46)$$

It consists of convective part and a diffusive parts according to,

$$\sum_A n_A m_A \mathbf{v}_A = \sum_A n_A m_A \mathbf{v} + \sum_A m_A \mathbf{j}_A. \quad (47)$$

Thus, using the relation (44), the velocity of the matter element is found to be,

$$\mathbf{v} = \left(\sum_A n_A m_A \right)^{-1} \left(\sum_A n_A m_A \mathbf{v}_A \right), \quad (48)$$

and represents the velocity of the local centre of mass.

In order to characterise physically the time evolution of the momentum, we substitute the expression (46) for the chemical composition of the momentum density and the continuity equation for the chemical substance (4) into the continuity equation (9) for the momentum. The latter then becomes,

$$\sum_A \left(n_A m_A \dot{\mathbf{v}}_A - (\nabla \cdot \mathbf{j}_A) m_A \mathbf{v}_A + \sum_a \nu_{aA} \omega_a m_A \mathbf{v}_A \right) - \nabla \cdot \boldsymbol{\sigma} = \mathbf{f}^{\text{ext}}. \quad (49)$$

Using the vectorial identity,

$$(\nabla \cdot \mathbf{j}_A) \mathbf{v}_A = \nabla_{\mathbf{j}} \cdot (\mathbf{v}_A \odot \mathbf{j}_A) - (\mathbf{j}_A \cdot \nabla) \mathbf{v}_A,$$

and the extensivity of the force, i.e.

$$\mathbf{f}^{\text{ext}} = \sum_A \mathbf{f}_A^{\text{ext}},$$

where $\mathbf{f}_A^{\text{ext}}$ represents the external force density acting on the chemical substance A , the continuity equation (49) for the momentum can be recast as,

$$\sum_A \left(n_A m_A \dot{\mathbf{v}}_A + m_A (\mathbf{j}_A \cdot \nabla) \mathbf{v}_A - \mathbf{f}_A^{\text{ext}} \right) + \sum_{a,A} \nu_{aA} \omega_a m_A \mathbf{v}_A - \nabla_{\mathbf{j}} \cdot \left(\boldsymbol{\sigma} + \sum_A m_A \mathbf{v}_A \odot \mathbf{j}_A \right) = \mathbf{0}. \quad (50)$$

The matter stress tensor σ is split into a reversible part due to the pressure P and an irreversible part $\tilde{\sigma}$ according to,

$$\sigma = -P\mathbb{1} + \tilde{\sigma}, \quad (51)$$

which implies that the continuity equation (50) is recast as:

$$\begin{aligned} & \sum_A \left(n_A m_A \dot{\mathbf{v}}_A + m_A (\mathbf{j}_A \cdot \nabla) \mathbf{v}_A - \mathbf{f}_A^{\text{ext}} \right) + \nabla P \\ & + \sum_{a,A} \nu_{aA} \omega_a m_A \mathbf{v}_A - \nabla \mathbf{j} \cdot \left(\tilde{\sigma} + \sum_A m_A \mathbf{v}_A \odot \mathbf{j}_A \right) = \mathbf{0}. \end{aligned} \quad (52)$$

The continuity equation (52) has to hold for any current flow, which yields an explicit expression for the irreversible part of the stress tensor, i.e.

$$\tilde{\sigma} = - \sum_A m_A \mathbf{v}_A \odot \mathbf{j}_A. \quad (53)$$

Moreover, every chemical reaction preserves the total momentum in the local infinitesimal system, i.e.

$$\sum_A \nu_{aA} m_A \mathbf{v}_A = \mathbf{0}. \quad (54)$$

Taking into account the fact that the pressure P of the continuous medium is the sum of the partial pressures P_A of the different chemical substances A multiplied by the dimensionless fugacity coefficients φ_A [18], i.e.

$$P = \sum_A \varphi_A P_A, \quad (55)$$

and using the conditions (53) and (54), the continuity equation (52) reduces to,

$$\sum_A \left(n_A m_A \dot{\mathbf{v}}_A + m_A (\mathbf{j}_A \cdot \nabla) \mathbf{v}_A - \mathbf{f}_A^{\text{ext}} + \nabla (\varphi_A P_A) \right) = \mathbf{0}, \quad (56)$$

which describes the local time evolution of all the chemical substances. Note that the dimensionless fugacity coefficient φ_A in the relation (55) accounts for the local interactions between the constituents of the chemical substance A .

In order to establish the local time evolution of a specific chemical substance A in the presence of other chemical substances B , we need to take explicitly into account the local internal force densities $\mathbf{f}_{B \rightarrow A}^{\text{int}}$ exerted by the chemical substances B on the chemical substance A . These force densities do not change the local internal energy of the system, which means that they are the densities of conservative forces accounting for elastic collisions or scattering. Newton's third law implies that the sum of the internal force densities over all the chemical substances vanishes, i.e.

$$\sum_{A,B} \mathbf{f}_{B \rightarrow A}^{\text{int}} = \mathbf{0}. \quad (57)$$

Equations (56) and (57) imply that the local time evolution of a specific chemical substance A is given by,

$$n_A m_A \dot{\mathbf{v}}_A = \mathbf{f}_A^{\text{ext}} - \nabla (\varphi_A P_A) - m_A (\mathbf{j}_A \cdot \nabla) \mathbf{v}_A + \sum_B \mathbf{f}_{B \rightarrow A}^{\text{int}}, \quad (58)$$

where the first, second and third terms on the RHS describe, respectively, the action of the external forces, of the fugacity gradient and of the transport, and the last term describes the action of the internal forces due to the local interaction of the chemical substance A with the other chemical substances B .

2.2.6 Time evolution of the intrinsic angular momentum

The intrinsic angular momentum density $\mathbf{s} = I\boldsymbol{\omega}$ is defined as the density of intrinsic angular momentum carried by the elementary units of chemical substances A of intrinsic angular velocities $\boldsymbol{\omega}_A$, i.e.

$$\mathbf{s} = \sum_A n_A I_A \boldsymbol{\omega}_A, \quad (59)$$

where $n_A I_A$ represent the intrinsic angular mass density of the elementary units of chemical substance A and the relations (15) and (41) imply that

$$n_A I_A = \frac{2}{5} n_A m_A dr^2. \quad (60)$$

The intrinsic angular momentum density \mathbf{s} consists of convective and diffusive parts according to,

$$\sum_A n_A I_A \boldsymbol{\omega}_A = \sum_A n_A I_A \boldsymbol{\omega} + \sum_A I_A \boldsymbol{\Omega}_A^{\text{m}}, \quad (61)$$

where the sum of the diffusive intrinsic angular momenta of all the chemical substances A vanish, i.e.

$$\sum_A I_A \boldsymbol{\Omega}_A^{\text{m}} = \mathbf{0}, \quad (62)$$

which defines a set of frames where the matter has no average intrinsic rotational motion. Thus, the intrinsic angular velocity of the matter element is found to be,

$$\boldsymbol{\omega} = \left(\sum_A n_A I_A \right)^{-1} \left(\sum_A n_A I_A \boldsymbol{\omega}_A \right), \quad (63)$$

and represents the average intrinsic angular velocity around the local centre of mass.

In order to characterise physically the time evolution of the intrinsic angular momentum, we substitute the expression (59) for the chemical composition of the intrinsic angular momentum density and the continuity equation for the chemical substance (4) into the continuity equation (13) for the intrinsic angular momentum. The latter then becomes,

$$\begin{aligned} & \sum_A \left(n_A I_A \dot{\boldsymbol{\omega}}_A - (\nabla \cdot \mathbf{j}_A) I_A \boldsymbol{\omega}_A + \sum_a \nu_{aA} \omega_a I_A \boldsymbol{\omega}_A \right) \\ & - \nabla \cdot \boldsymbol{\Theta} = \boldsymbol{\tau}^{\text{ext}}. \end{aligned} \quad (64)$$

Using the vectorial identity,

$$(\nabla \cdot \mathbf{j}_A) \boldsymbol{\omega}_A = \nabla_{\mathbf{j}} \cdot (\boldsymbol{\omega}_A \odot \mathbf{j}_A) - (\mathbf{j}_A \cdot \nabla) \boldsymbol{\omega}_A,$$

and the extensivity of the torque, i.e.

$$\boldsymbol{\tau}^{\text{ext}} = \sum_A \boldsymbol{\tau}_A^{\text{ext}},$$

where $\boldsymbol{\tau}_A^{\text{ext}}$ represents the external torque density acting on the chemical substance A , the continuity equation (64) for the intrinsic angular momentum is recast as:

$$\begin{aligned} & \sum_A \left(n_A I_A \dot{\boldsymbol{\omega}}_A + I_A (\mathbf{j}_A \cdot \nabla) \boldsymbol{\omega}_A - \boldsymbol{\tau}_A^{\text{ext}} \right) \\ & + \sum_{a,A} \nu_{aA} \omega_a I_A \boldsymbol{\omega}_A - \nabla_{\mathbf{j}} \cdot \left(\boldsymbol{\Theta} + \sum_A I_A \boldsymbol{\omega}_A \odot \mathbf{j}_A \right) = \mathbf{0}. \end{aligned} \quad (65)$$

The continuity equation (65) has to hold for any current flow, which yields an explicit expression for the irreversible part of the intrinsic angular stress tensor, i.e.

$$\boldsymbol{\Theta} = - \sum_A I_A \boldsymbol{\omega}_A \odot \mathbf{j}_A. \quad (66)$$

Moreover, every chemical reaction preserves the total intrinsic angular momentum in the local infinitesimal system, i.e.

$$\sum_A \nu_{aA} I_A \boldsymbol{\omega}_A = \mathbf{0}. \quad (67)$$

The conditions (66) and (67) imply that the continuity equation (65) reduces to,

$$\sum_A \left(n_A I_A \dot{\boldsymbol{\omega}}_A + I_A (\mathbf{j}_A \cdot \nabla) \boldsymbol{\omega}_A - \boldsymbol{\tau}_A^{\text{ext}} \right) = \mathbf{0}, \quad (68)$$

which describes the local intrinsic time evolution of all the chemical substances.

In order to establish the local intrinsic time evolution of a specific chemical substance A in the presence of other chemical substances B , we need to take explicitly into account the local internal intrinsic torque densities $\boldsymbol{\tau}_{B \rightarrow A}^{\text{int}}$ exerted by the chemical substances B on the chemical substance A . These torque densities do not change the local internal energy of the system, which means that they are the densities of conservative torques accounting for elastic collisions or scattering. Newton's third law in intrinsic rotation implies that the sum of the internal torque densities over all the chemical substances vanishes, i.e.

$$\sum_{A,B} \boldsymbol{\tau}_{B \rightarrow A}^{\text{int}} = \mathbf{0}. \quad (69)$$

Equations (68) and (69) imply that the local intrinsic time evolution of a specific chemical substance A is given by,

$$n_A I_A \dot{\boldsymbol{\omega}}_A = \boldsymbol{\tau}_A^{\text{ext}} - I_A (\mathbf{j}_A \cdot \nabla) \boldsymbol{\omega}_A + \sum_B \boldsymbol{\tau}_{B \rightarrow A}^{\text{int}}, \quad (70)$$

where the first and second terms on the RHS describe, respectively, the action of the external torques and of the transport, and the last term describes the action of the internal torques due to the local interaction of the chemical substance A with the other chemical substances B .

3 Thermostatistics and thermodynamics

3.1 Thermostatic equation, reversible and irreversible thermodynamic equations

The thermostatistics and thermodynamics of the continuous medium are contained within the internal energy balance (21). Since the internal energy density $u(s, n_A, q, \mathbf{P}, \mathbf{M})$ is a state function in the local rest frame, the time derivative of the internal energy density field is given by, i.e.

$$\dot{u} = T \dot{s} + \sum_A \mu_A \dot{n}_A + V \dot{q} - \mathbf{E} \cdot \dot{\mathbf{P}} - \mathbf{B} \cdot \dot{\mathbf{M}}, \quad (71)$$

where the temperature T , the chemical potential μ_A of the substance A , the electric potential V , the opposite of the electric field $-\mathbf{E}$ and the opposite of the magnetic induction field $-\mathbf{B}$ are defined as the intensive conjugate fields of the extensive state fields s , n_A , q , \mathbf{P} and \mathbf{M} , respectively [19,20], i.e.

$$\begin{aligned} T &\equiv \frac{\partial u}{\partial s}, & \mu_A &\equiv \frac{\partial u}{\partial n_A}, & V &\equiv \frac{\partial u}{\partial q}, \\ -\mathbf{E} &\equiv \frac{\partial u}{\partial \mathbf{P}}, & -\mathbf{B} &\equiv \frac{\partial u}{\partial \mathbf{M}}. \end{aligned} \quad (72)$$

Note that these intensive fields can only be defined locally if the system satisfies the local equilibrium hypothesis.

Using the relation (62), the continuity equations for the entropy (3), the density of the chemical substance A (4), the electric charge density (5), the electric polarisation (7) and the magnetisation (8), the continuity equation for the internal energy (21) is recast as,

$$\begin{aligned} & T \left(\rho_s - (\nabla \cdot \mathbf{v}) s - \nabla \cdot \mathbf{j}_s \right) \\ & + \sum_A \mu_A \left(\sum_a \nu_{aA} \omega_a - (\nabla \cdot \mathbf{v}) n_A - \nabla \cdot \mathbf{j}_A \right) \\ & + V \left(-(\nabla \cdot \mathbf{v}) q - \nabla \cdot \mathbf{j}_q \right) \\ & - \mathbf{E} \cdot \left(\sum_A \boldsymbol{\Omega}_A^{\text{m}} \times \mathbf{p}_A - (\nabla \cdot \mathbf{v}) \mathbf{P} - \nabla \cdot \mathbf{j}_P \right) \\ & - \mathbf{B} \cdot \left(\sum_A (\gamma_A n_A (\mathbf{m}_A \times \mathbf{B}) + \boldsymbol{\Omega}_A^{\text{M}} \times \mathbf{m}_A) \right. \\ & \quad \left. - (\nabla \cdot \mathbf{v}) \mathbf{M} - \nabla \cdot \mathbf{j}_M \right) \\ & + (\nabla \cdot \mathbf{v}) u + \nabla \cdot \mathbf{j}_u = \boldsymbol{\sigma} \cdot (\nabla \odot \mathbf{v}) + \boldsymbol{\Theta} \cdot (\nabla \odot \boldsymbol{\omega}). \end{aligned} \quad (73)$$

Using the vectorial identities,

$$\begin{aligned}
T(\nabla \cdot \mathbf{j}_s) &= \nabla \cdot (T \mathbf{j}_s) - \mathbf{j}_s \cdot \nabla T, \\
\mu_A(\nabla \cdot \mathbf{j}_A) &= \nabla \cdot (\mu_A \mathbf{j}_A) - \mathbf{j}_A \cdot \nabla \mu_A, \\
V(\nabla \cdot \mathbf{j}_q) &= \nabla \cdot (\mathbf{j}_q V) - \mathbf{j}_q \cdot \nabla V, \\
\mathbf{E} \cdot (\nabla \cdot \mathbf{j}_P) &= \nabla \cdot (\mathbf{j}_P \cdot \mathbf{E}) - \mathbf{j}_P \cdot (\nabla \odot \mathbf{E}), \\
\mathbf{B} \cdot (\nabla \cdot \mathbf{j}_M) &= \nabla \cdot (\mathbf{j}_M \cdot \mathbf{B}) - \mathbf{j}_M \cdot (\nabla \odot \mathbf{B}), \\
\mathbf{E} \cdot (\boldsymbol{\Omega}_A^m \times \mathbf{p}_A) &= \boldsymbol{\Omega}_A^m \cdot (\mathbf{p}_A \times \mathbf{E}), \\
\mathbf{B} \cdot (\gamma_A n_A (\mathbf{m}_A \times \mathbf{B})) &= 0, \\
\mathbf{B} \cdot (\boldsymbol{\Omega}_A^M \times \mathbf{m}_A) &= \boldsymbol{\Omega}_A^M \cdot (\mathbf{m}_A \times \mathbf{B}),
\end{aligned}$$

the splitting (51) of the stress tensor into a reversible and an irreversible part, and expressing the chemical irreversibility in terms of the chemical affinities \mathcal{A}_a , i.e.

$$\mathcal{A}_a = \sum_A \nu_{aA} (-\mu_A), \quad (74)$$

the internal energy balance equation (73) is recast as,

$$\begin{aligned}
&(u - Ts + P - \sum_A \mu_A n_A - qV + \mathbf{P} \cdot \mathbf{E} + \mathbf{M} \cdot \mathbf{B})(\nabla \cdot \mathbf{v}) \\
&+ \nabla \cdot (\mathbf{j}_u - T \mathbf{j}_s - \sum_A \mu_A \mathbf{j}_A - \mathbf{j}_q V + \mathbf{j}_P \cdot \mathbf{E} + \mathbf{j}_M \cdot \mathbf{B}) \\
&+ T \rho_s - \sum_a \omega_a \mathcal{A}_a + \mathbf{j}_s \cdot \nabla T + \sum_A \mathbf{j}_A \cdot \nabla \mu_A - \mathbf{j}_q \cdot \nabla V \\
&- \tilde{\boldsymbol{\sigma}} \cdot (\nabla \odot \mathbf{v}) - \boldsymbol{\Theta} \cdot (\nabla \odot \boldsymbol{\omega}) \\
&- \mathbf{j}_P \cdot (\nabla \odot \mathbf{E}) - \mathbf{j}_M \cdot (\nabla \odot \mathbf{B}) \\
&- \sum_A \boldsymbol{\Omega}_A^m \cdot (\mathbf{p}_A \times \mathbf{E}) - \sum_A \boldsymbol{\Omega}_A^M \cdot (\mathbf{m}_A \times \mathbf{B}) = 0.
\end{aligned} \quad (75)$$

Using the expressions (23), (28) and (34) for the chemical composition of the electric charge density, the electric polarisation and magnetisation, we deduce the identities,

$$\begin{aligned}
qV &= \sum_A q_A V n_A, \\
\mathbf{P} \cdot \mathbf{E} &= \sum_A (\mathbf{p}_A \cdot \mathbf{E}) n_A, \\
\mathbf{M} \cdot \mathbf{B} &= \sum_A (\mathbf{m}_A \cdot \mathbf{B}) n_A.
\end{aligned} \quad (76)$$

Moreover, using the expressions (26), (31) and (37) for the diffusive electric current density vector, the electric polarisation and the magnetisation current tensors, respectively, we obtain the identities,

$$\begin{aligned}
\mathbf{j}_q V &= \sum_A q_A V \mathbf{j}_A, \\
\mathbf{j}_P \cdot \mathbf{E} &= \sum_A (\mathbf{p}_A \cdot \mathbf{E}) \mathbf{j}_A, \\
\mathbf{j}_M \cdot \mathbf{B} &= \sum_A (\mathbf{m}_A \cdot \mathbf{B}) \mathbf{j}_A.
\end{aligned} \quad (77)$$

Furthermore, using the expressions (53), (66), (31) and (37), for the momentum and intrinsic angular momentum stress tensors as well as for the electric polarisation and the magnetisation current tensors, respectively, we obtain the identities,

$$\begin{aligned}
\tilde{\boldsymbol{\sigma}} \cdot (\nabla \odot \mathbf{v}) &= \sum_A \mathbf{j}_A \cdot \left(-m_A \mathbf{v}_A \nabla \mathbf{v} \right), \\
\boldsymbol{\Theta} \cdot (\nabla \odot \boldsymbol{\omega}) &= \sum_A \mathbf{j}_A \cdot \left(-I_A \omega_A \nabla \boldsymbol{\omega} \right), \\
\mathbf{j}_P \cdot (\nabla \odot \mathbf{E}) &= \sum_A \mathbf{j}_A \cdot \left(\mathbf{p}_A \nabla \mathbf{E} \right), \\
\mathbf{j}_M \cdot (\nabla \odot \mathbf{B}) &= \sum_A \mathbf{j}_A \cdot \left(\mathbf{m}_A \nabla \mathbf{B} \right).
\end{aligned} \quad (78)$$

At this point it is useful to introduce the generalised chemical potential $\bar{\mu}_A$ that is defined as:

$$\bar{\mu}_A = \mu_A + q_A V - \mathbf{p}_A \cdot \mathbf{E} - \mathbf{m}_A \cdot \mathbf{B}. \quad (79)$$

Using the identities (76)–(78) and the definition (79), the internal energy balance equation (75) is recast explicitly in terms of the physical properties of the chemical components, i.e.

$$\begin{aligned}
&(u - Ts + P - \sum_A \bar{\mu}_A n_A)(\nabla \cdot \mathbf{v}) \\
&+ \nabla \cdot (\mathbf{j}_u - T \mathbf{j}_s - \sum_A \bar{\mu}_A \mathbf{j}_A) \\
&+ T \rho_s - \sum_a \omega_a \mathcal{A}_a - \mathbf{j}_s \cdot (-\nabla T) \\
&- \sum_A \mathbf{j}_A \cdot \left(-\nabla \mu_A - q_A \nabla V \right) \\
&- \sum_A \mathbf{j}_A \cdot \left(-m_A \mathbf{v}_A \nabla \mathbf{v} - I_A \omega_A \nabla \boldsymbol{\omega} \right) \\
&- \sum_A \mathbf{j}_A \cdot \left(\mathbf{p}_A \nabla \mathbf{E} + \mathbf{m}_A \nabla \mathbf{B} \right) \\
&- \sum_A \boldsymbol{\Omega}_A^m \cdot (\mathbf{p}_A \times \mathbf{E}) - \sum_A \boldsymbol{\Omega}_A^M \cdot (\mathbf{m}_A \times \mathbf{B}) = 0.
\end{aligned} \quad (80)$$

Firstly, the internal energy balance equation (80) has to hold locally for all flows. This implies that the terms in the first brackets have to vanish, which yields the thermostatic equilibrium equation for the continuous medium, i.e.

$$u = Ts - P + \sum_A \left(\mu_A + q_A V - \mathbf{p}_A \cdot \mathbf{E} - \mathbf{m}_A \cdot \mathbf{B} \right) n_A. \quad (81)$$

Secondly, the internal energy balance equation (80) has to hold locally for all currents. This implies that the terms in the second brackets have to vanish, which yields the reversible thermodynamic evolution equation for the continuous medium, i.e.

$$\mathbf{j}_u = T \mathbf{j}_s + \sum_A \left(\mu_A + q_A V - \mathbf{p}_A \cdot \mathbf{E} - \mathbf{m}_A \cdot \mathbf{B} \right) \mathbf{j}_A. \quad (82)$$

Thirdly, the thermostatic equilibrium equation (81) and the reversible thermodynamic evolution equation (82) imply that the internal energy balance equation (80) yields the irreversible thermodynamic evolution equation for the continuous medium, i.e.

$$\begin{aligned} \rho_s = \frac{1}{T} & \left\{ \sum_a \omega_a \mathcal{A}_a + \mathbf{j}_s \cdot (-\nabla T) \right. \\ & + \sum_A \mathbf{j}_A \cdot (-\nabla \mu_A - q_A \nabla V) \\ & + \sum_A \mathbf{j}_A \cdot (-m_A \mathbf{v}_A \nabla \mathbf{v} - I_A \omega_A \nabla \omega) \\ & + \sum_A \mathbf{j}_A \cdot (\mathbf{p}_A \nabla \mathbf{E} + \mathbf{m}_A \nabla \mathbf{B}) \\ & \left. + \sum_A \boldsymbol{\Omega}_A^m \cdot (\mathbf{p}_A \times \mathbf{E}) + \sum_A \boldsymbol{\Omega}_A^M \cdot (\mathbf{m}_A \times \mathbf{B}) \right\}. \end{aligned} \quad (83)$$

Finally, note that the time derivative of the thermostatic equation (81) and the expression (71) for the time derivative of the internal energy determines a generalised Gibbs-Duhem relation that yields the time evolution of the intensive fields (72), i.e.

$$s\dot{T} - \dot{P} + \sum_A n_A \dot{\mu}_A + q\dot{V} - \mathbf{P} \cdot \dot{\mathbf{E}} - \mathbf{M} \cdot \dot{\mathbf{B}} = 0. \quad (84)$$

4 Thermodynamical phenomenology

4.1 Linear phenomenological relations and Onsager matrix elements

In order to deduce the linear phenomenological relations, we extend the approach developed by Onsager [21,22] to include intrinsic rotations. The expression (83) can be formally split into a scalar, a vectorial and a pseudo-vectorial part, which are irreducible representations of the Euclidean group and have different symmetries. Thus, the entropy source density (83) is expressed formally as,

$$\rho_s = \frac{1}{T} \left\{ \sum_a \omega_a \mathcal{A}_a + \sum_\alpha \mathbf{j}_\alpha \cdot \mathbf{F}_\alpha + \sum_{A,i} \boldsymbol{\Omega}_A^i \cdot \mathbf{T}_A^i \right\}, \quad (85)$$

where ω_a is a scalar chemical reaction rate density, \mathcal{A}_a is a scalar chemical affinity, \mathbf{j}_α is a vectorial current density, \mathbf{F}_α is a vectorial force, $\boldsymbol{\Omega}_A^i$ is a pseudo-vectorial intrinsic rotation rate density and \mathbf{T}_A^i is a pseudo-vectorial intrinsic torque.

In the relation (85), there are two types of vectorial current densities and forces (i.e. $\alpha \in \{s, A\}$). First, there is a thermal current density \mathbf{j}_s and an irreversible thermal force $\mathbf{F}_s = -\nabla T$. Second, there are current densities \mathbf{j}_A for the chemical substances A and irreversible forces \mathbf{F}_A

acting on these substances. The expression for the forces \mathbf{F}_A is given by the irreversible relation (83), i.e.

$$\begin{aligned} \mathbf{F}_A = & -\nabla \mu_A - q_A \nabla V - m_A \mathbf{v}_A \nabla \mathbf{v} - I_A \omega_A \nabla \omega \\ & + \mathbf{p}_A \nabla \mathbf{E} + \mathbf{m}_A \nabla \mathbf{B}, \end{aligned} \quad (86)$$

where the first term on the RHS is the chemical force, the second term is the Coulomb force, the third and the fourth terms are the viscous forces, the fifth term is the electric polarisation force and the sixth term is the magnetisation force. Note that in a stationary regime, the viscous forces, the electric polarisation force and the magnetisation force generate dielectrophoresis and magnetophoresis.

In the relation (85), there are also two types of pseudo-vectorial intrinsic rotation rate densities and torques (i.e. $\boldsymbol{\Omega}_A^i \in \{A, A\}^m, M$) associated to the chemical substances A . First, there are intrinsic rotation rate densities $\boldsymbol{\Omega}_A^m$ and irreversible torques \mathbf{T}_A^m associated with the intrinsic rotational motion of the matter. Second, there are intrinsic rotation rate densities $\boldsymbol{\Omega}_A^M$ and irreversible torques \mathbf{T}_A^M associated with micro-magnetism and matter. The expression for the torques \mathbf{T}_A^i is given by the irreversible relation (83), i.e.

$$\begin{aligned} \mathbf{T}_A^m & = \mathbf{p}_A \times \mathbf{E}, \\ \mathbf{T}_A^M & = \mathbf{m}_A \times \mathbf{B}. \end{aligned} \quad (87)$$

The local expression of the second law (3) requires the entropy source density to be locally positive definite, i.e. $\rho_s \geq 0$. Extending Onsager's approach, in the neighbourhood of a local thermodynamic state where the scalar affinities \mathcal{A}_a , the vectorial forces \mathbf{F}_α and the pseudo-vectorial intrinsic torques \mathbf{T}_A^i are sufficiently small, the entropy source density can be expressed formally as the sum of quadratic forms of \mathcal{A}_a , \mathbf{F}_α and \mathbf{T}_A^i , which ensures that it is positive definite, i.e.

$$\begin{aligned} \rho_s = \frac{1}{T} & \left(\sum_{a,b} L_{ab} \mathcal{A}_a \mathcal{A}_b + \sum_{\alpha,\beta} \mathbf{L}_{\alpha\beta} \cdot (\mathbf{F}_\alpha \odot \mathbf{F}_\beta) \right. \\ & \left. + \sum_{\substack{A,B \\ i,j}} \mathbf{L}_{AB}^{ij} \cdot (\mathbf{T}_A^i \odot \mathbf{T}_B^j) \right) \geq 0, \end{aligned} \quad (88)$$

where the phenomenological components are the Onsager matrix elements, which are of two different types: scalars $L_{\alpha\beta}$ and rank-2 tensors $\mathbf{L}_{\alpha\beta}$ and \mathbf{L}_{AB}^{ij} . The symmetries of the Onsager matrices are given by the Onsager reciprocity relations, i.e.

$$\begin{aligned} L_{ab}(s, n_A, q, \mathbf{P}, \mathbf{M}) & = L_{ba}(s, n_A, q, \mathbf{P}, -\mathbf{M}), \\ \mathbf{L}_{\alpha\beta}(s, n_A, q, \mathbf{P}, \mathbf{M}) & = \mathbf{L}_{\beta\alpha}(s, n_A, q, \mathbf{P}, -\mathbf{M}), \\ \mathbf{L}_{AB}^{ij}(s, n_A, q, \mathbf{P}, \mathbf{M}) & = \mathbf{L}_{AB}^{ji}(s, n_A, q, \mathbf{P}, -\mathbf{M}), \end{aligned} \quad (89)$$

which cannot be derived within a thermodynamic approach but require a statistical treatment since they are a consequence of the time reversibility of the microscopic dynamics [21]. The inequality (88) has to hold for each part, which implies that each quadratic form has to be positive definite.

Thus, the chemical reaction rate densities ω_a are related to the chemical affinities \mathcal{A}_b through scalar linear phenomenological relations, i.e.

$$\omega_a = \sum_b L_{ab} \mathcal{A}_b, \quad (90)$$

where the Onsager matrix has to satisfy,

$$\frac{1}{T} \{L_{ab}\} \geq 0, \quad (91)$$

in order for the scalar quadratic form in the relation (88) to be positive definite. The scalar linear phenomenological relations (90) account for the irreversibility due to the chemistry. Note that these linear relations are only accurate provided that the chemical affinities \mathcal{A}_b are sufficiently small compared to the thermal excitation, i.e. $\mathcal{A}_b \ll k_B T$ where k_B is Boltzmann's constant, as pointed out by Glansdorf and Prigogine [23] and de Groot and Mazur [12].

Similarly, the vectorial current densities \mathbf{j}_α are related to the vectorial forces \mathbf{F}_β through vectorial linear phenomenological relations, i.e.

$$\mathbf{j}_\alpha = \sum_\beta L_{\alpha\beta} \cdot \mathbf{F}_\beta, \quad (92)$$

where the Onsager matrix has to satisfy,

$$\frac{1}{T} \{L_{\alpha\beta}\} \geq 0, \quad (93)$$

in order for the vectorial quadratic form in the relation (88) to be positive definite. The vectorial linear phenomenological relations (92) are expressed explicitly in terms of the currents densities \mathbf{j}_s and \mathbf{j}_A and forces \mathbf{F}_s and \mathbf{F}_A as,

$$\begin{cases} \mathbf{j}_s = L_{ss} \cdot (-\nabla T) + \sum_B L_{sB} \cdot \mathbf{F}_B \\ \mathbf{j}_A = L_{As} \cdot (-\nabla T) + \sum_B L_{AB} \cdot \mathbf{F}_B \end{cases} \quad (94)$$

where the Onsager matrix (93) is positive definite, i.e.

$$\frac{1}{T} \begin{pmatrix} L_{ss} & L_{sB} \\ L_{As} & L_{AB} \end{pmatrix} \geq 0. \quad (95)$$

The vectorial linear phenomenological relations (94) account for the irreversibility due to the transport.

Likewise, the pseudo-vectorial intrinsic rotation rate densities Ω_A^i are related to the pseudo-vectorial torques \mathbf{T}_B^j through pseudo-vectorial linear phenomenological relations, i.e.

$$\Omega_A^i = \sum_{B,j} L_{AB}^{ij} \cdot \mathbf{T}_B^j, \quad (96)$$

where the Onsager matrix has to satisfy,

$$\frac{1}{T} \{L_{AB}^{ij}\} \geq 0, \quad (97)$$

in order for the pseudo-vectorial quadratic form in the relation (88) to be positive definite. The pseudo-vectorial linear phenomenological relations (96) are expressed explicitly in terms of the intrinsic rotation rate densities Ω_A^i and torques \mathbf{T}_B^j as,

$$\begin{cases} \Omega_A^m = \sum_B \left(L_{AB}^{mm} \cdot (\mathbf{p}_B \times \mathbf{E}) + L_{AB}^{mM} \cdot (\mathbf{m}_B \times \mathbf{B}) \right) \\ \Omega_A^M = \sum_B \left(L_{AB}^{Mm} \cdot (\mathbf{p}_B \times \mathbf{E}) + L_{AB}^{MM} \cdot (\mathbf{m}_B \times \mathbf{B}) \right) \end{cases} \quad (98)$$

where the Onsager matrix (97) is positive definite, i.e.

$$\frac{1}{T} \begin{pmatrix} L_{AB}^{mm} & L_{AB}^{mM} \\ L_{AB}^{Mm} & L_{AB}^{MM} \end{pmatrix} \geq 0. \quad (99)$$

The pseudo-vectorial linear phenomenological relations (98) account for the irreversibility due to the relaxation. They also represent the classical counterpart of the spin-orbit coupling, since they couple the rotation rate density Ω_A^M of the magnetisation with the rotation rate density of the matter Ω_A^m .

It is worth emphasising that although the scalar (90), the vectorial (94) and the pseudo-vectorial (98) linear phenomenological relations are structurally independent, they are coupled through the time evolution equations. The chemical affinities \mathcal{A}_a defined in relation (74) couple the scalar (90) and vectorial (94) linear phenomenological relations. This coupling has interesting applications for spintronics, some of which were investigated in reference [24].

In the particular case of a continuous medium made of a single chemical substance A , the scalar linear relations (90) vanish, the vectorial linear relations (94) become,

$$\begin{cases} \mathbf{j}_s = L_{ss} \cdot (-\nabla T) + L_{sA} \cdot \mathbf{F}_A \\ \mathbf{j}_A = L_{As} \cdot (-\nabla T) + L_{AA} \cdot \mathbf{F}_A \end{cases} \quad (100)$$

and the pseudo-vectorial linear relations (98) reduce to,

$$\begin{cases} \Omega_A^m = L_{AA}^{mm} \cdot (\mathbf{p}_A \times \mathbf{E}) + L_{AA}^{mM} \cdot (\mathbf{m}_A \times \mathbf{B}) \\ \Omega_A^M = L_{AA}^{Mm} \cdot (\mathbf{p}_A \times \mathbf{E}) + L_{AA}^{MM} \cdot (\mathbf{m}_A \times \mathbf{B}). \end{cases} \quad (101)$$

4.2 Physical applications

4.2.1 Lehmann effect

In 1900, Lehmann [25] observed that droplets of liquid crystals, that have a chiral cholesteric structure, were set in rotation by a temperature gradient. The explanation given by Leslie [26] is now questioned [27] by recent observations. Here, we show how our formalism accounts for the Lehmann effect.

It is worth mentioning that the irreversible thermodynamics of a continuum of nematic liquid crystals has

been examined by Müller [5], Blenk and Muschik [28], and Muschik et al. [29], even though these authors did not explicitly deduce the Lehmann effect from their formalism. The “director” vector field they introduced corresponds to the preferred axis $\hat{\mathbf{n}}$ of the continuum of liquid crystals. In contrast to them, we do not enlarge the spatial manifold to account for $\hat{\mathbf{n}}$. Instead, we require the net electric polarisation \mathbf{P} , which is a state field, to be collinear to $\hat{\mathbf{n}}$.

We consider a uniform continuum made of identical liquid crystals that are oriented in the same direction. The continuum of liquid crystals is gyrotropic since the chirality of the liquid crystals defines a preferred axis of unit vector $\hat{\mathbf{n}}$. Each liquid crystal is helicoidal and made of identical elements A that are dielectrics, i.e. $q_A = 0$, and have electric dipoles \mathbf{p}_A orthogonal to the helix axis [30]. Thus, in the absence of an external interaction, the liquid crystals have no net electric polarisation. On a macroscopic scale, the continuum is homogeneous, i.e. $\nabla \mu_A = \mathbf{0}$. We assume that the viscosity can be neglected, i.e. $\nabla \mathbf{v}_A = \mathbf{0}$ and $\nabla \boldsymbol{\omega}_A = \mathbf{0}$. The liquid crystals are trapped in an experimental set-up such that they have no translational motion, i.e. $\mathbf{j}_A = \mathbf{0}$. Moreover, a temperature gradient ∇T is applied along a direction that is different from the preferred axis of the crystals.

In such a case, the linear phenomenological relation (100) reduces to,

$$\mathbf{p}_A \nabla \mathbf{E} = (\mathbf{L}_{AA}^{-1} \cdot \mathbf{L}_{As}) \cdot \nabla T, \quad (102)$$

where the temperature gradient ∇T induces an electric field \mathbf{E} and the electric dipoles \mathbf{p}_A rotate in an asymmetric manner in order to lower the Debye energy $-\mathbf{p}_A \cdot \mathbf{E}$. Thus, a net electric polarisation \mathbf{P} is generated along the preferred axis $\hat{\mathbf{n}}$, i.e.

$$\mathbf{P} = n_A \mathbf{p}_A = n_A (\mathbf{p}_A \cdot \hat{\mathbf{n}}) \hat{\mathbf{n}}, \quad (103)$$

and the linear relation (102) is recast as,

$$\mathbf{P} \nabla \mathbf{E} = (n_A \mathbf{L}_{AA}^{-1} \cdot \mathbf{L}_{As}) \cdot \nabla T. \quad (104)$$

The spatial symmetry requires the electric polarisation force $\mathbf{P} \nabla \mathbf{E}$ and the thermal force $-\nabla T$ to be collinear, i.e.

$$\mathbf{P} \nabla \mathbf{E} = n_A L_{AA}^{-1} L_{As} \nabla T, \quad (105)$$

where $\mathbf{L}_{AA}^{-1} = L_{AA}^{-1} \mathbb{1}$ and $\mathbf{L}_{As} = L_{As} \mathbb{1}$. Thus, the phenomenological relation (105) can be recast as:

$$\mathbf{P} \nabla \mathbf{E} = \lambda n_A k_B \nabla T, \quad (106)$$

where λ is a dimensionless parameter given by:

$$\lambda = \frac{L_{AA}^{-1} L_{As}}{k_B}. \quad (107)$$

In the absence of an applied magnetic field, i.e. $\mathbf{B} = \mathbf{0}$, the electric field \mathbf{E} is expressed in terms of the electric potential V as,

$$\mathbf{E} = -\nabla V. \quad (108)$$

The electric polarisation force density $\mathbf{P} \nabla \mathbf{E}$ satisfies the vectorial identity,

$$\begin{aligned} \mathbf{P} \nabla \mathbf{E} &= (\mathbf{P} \cdot \nabla) \mathbf{E} + \mathbf{P} \times (\nabla \times \mathbf{E}) = (\mathbf{P} \cdot \nabla) \mathbf{E} \\ &= \nabla_{\mathbf{P}} \cdot (\mathbf{P} \odot \mathbf{E}) + (-\nabla \cdot \mathbf{P}) \mathbf{E}, \end{aligned} \quad (109)$$

where we used Faraday’s law (108) and the index \mathbf{P} denotes that there is a dot product between the covariant differential operator ∇ and the electric polarisation \mathbf{P} . The term $\nabla_{\mathbf{P}} \cdot (\mathbf{P} \odot \mathbf{E})$ in the identity (109) corresponds to a surface contribution after integration over the volume of the liquid crystal continuum [31]. Thus, it can be neglected in the bulk of the continuum where the electric polarisation force density $\mathbf{P} \nabla \mathbf{E}$ is expressed as,

$$\mathbf{P} \nabla \mathbf{E} = (-\nabla \cdot \mathbf{P}) \mathbf{E}. \quad (110)$$

Moreover, using the definition of the bound electric charge density $q_{\mathbf{P}}$, i.e.

$$q_{\mathbf{P}} \equiv -\nabla \cdot \mathbf{P}, \quad (111)$$

the electric polarisation force density (110) is recast as [31],

$$\mathbf{P} \nabla \mathbf{E} = q_{\mathbf{P}} \mathbf{E}, \quad (112)$$

which shows that it is the analog of the electric part of the Lorentz force density for bound charges. Substituting the expression (112) in the linear relation (106), the latter can be expressed as a Seebeck effect for bound electric charges in an electric insulator, i.e.

$$\mathbf{E} = \varepsilon_{\mathbf{P}} \nabla T, \quad (113)$$

where the Seebeck coefficient for bound electric charges,

$$\varepsilon_{\mathbf{P}} \equiv \frac{\lambda n_A k_B}{q_{\mathbf{P}}}.$$

Moreover, the liquid crystals are subjected to an external electric torque (87) that sets them in rotational motion. Thus, the external torque density is given by,

$$\boldsymbol{\tau}^{\text{ext}} = n_A \mathbf{T}_A^{\text{m}} = \mathbf{P} \times \mathbf{E}. \quad (114)$$

Using equation (113) for the Seebeck effect, the torque density $\boldsymbol{\tau}^{\text{ext}}$ is recast as,

$$\boldsymbol{\tau}^{\text{ext}} = \varepsilon_{\mathbf{P}} (\mathbf{P} \times \nabla T). \quad (115)$$

The rotation occurs in the plane spanned by the electric polarisation \mathbf{P} and the temperature gradient ∇T around the local centre of mass of the liquid crystals. Taking into account the absence of a matter current, i.e. $\boldsymbol{\Theta} = \mathbf{0}$, and substituting the expression (115) for the torque density into Newton’s second law in intrinsic rotation (17), the latter accounts for the Lehmann effect, i.e.

$$\dot{\boldsymbol{\omega}} = \frac{\varepsilon_{\mathbf{P}}}{I} (\mathbf{P} \times \nabla T), \quad (116)$$

where I is the intrinsic angular mass density of the liquid crystals. The intensity of this effect is expressed as

$$\ddot{\theta} = \frac{\varepsilon_{\mathbf{P}} P \nabla T}{I} \sin \theta, \quad (117)$$

where θ is the angle between the electric polarisation \mathbf{P} and the temperature gradient ∇T , i.e. $|\mathbf{P} \times \nabla T| = P \nabla T \sin \theta$.

4.2.2 Electric Lehmann effect

In 1974, de Gennes [32] noticed that liquid crystals were set in rotation by an electric field. This effect is called the electric Lehmann effect [33] since it corresponds to a Lehmann effect where the driving force is an electric force instead of a thermal force. Here, we show how our formalism accounts for the electric Lehmann effect.

We consider the same continuum of liquid crystals as in Section 4.2.1. In contrast to the Lehmann effect, the electric Lehmann effect is an isothermal effect. The temperature gradient ∇T is replaced by an electric potential gradient ∇V that is applied along a direction that is different from the preferred axis of the liquid crystals. By analogy with the Lehmann effect, the electric field must be inhomogeneous in order to generate an electric polarisation force density (112).

In the presence of an applied electric potential gradient ∇V , the electric dipoles \mathbf{p}_A rotate in an asymmetric manner in order to lower the Debye energy $\mathbf{p}_A \cdot \nabla V$, which yields a net electric polarisation \mathbf{P} along the preferred axis $\hat{\mathbf{n}}$ given in equation (103). Moreover, the liquid crystals are subjected to an external electric torque density (114), that sets them in rotational motion. In the absence of a magnetic induction field \mathbf{B} , Faraday's law (108) implies that the torque density (114) yields,

$$\boldsymbol{\tau}^{\text{ext}} = -\mathbf{P} \times \nabla V. \quad (118)$$

The rotation occurs in the plane spanned by the electric polarisation \mathbf{P} and the electric potential gradient ∇V around the local centre of mass of the liquid crystals. Taking into account the absence of a matter current, i.e. $\boldsymbol{\Theta} = \mathbf{0}$, and substituting the expression (118) for the torque density into Newton's second law in intrinsic rotation (17), the latter accounts for the electric Lehmann effect, i.e.

$$\dot{\boldsymbol{\omega}} = -\frac{1}{I} (\mathbf{P} \times \nabla V). \quad (119)$$

The intensity of this effect is expressed as

$$\ddot{\theta} = -\frac{P \nabla V}{I} \sin \theta, \quad (120)$$

where θ is the angle between the electric polarisation \mathbf{P} and the electric potential gradient ∇V , i.e. $|\mathbf{P} \times \nabla V| = P \nabla V \sin \theta$.

It is useful to point out that Quincke [34] observed in 1896 that small dielectric spheres in suspension in a liquid placed inside a parallel-plate capacitor are set in rotational motion by charging the capacitor. The intrinsic rotational dynamics of the Quincke effect is described by the same time evolution equations (119) and (120) as the electric Lehmann effect.

4.2.3 Relaxation of electric dipoles

The first model of the relaxation of electric dipoles \mathbf{p}_A is due to Debye [35]. Here, we show how our formalism accounts for this relaxation.

We consider a homogeneous sample made of a single chemical substance A , consisting of electric dipoles \mathbf{p}_A in the absence of a magnetic induction field, i.e. $\mathbf{B} = \mathbf{0}$. In the frame of sample, the chemical substance A has no translational motion, i.e. $\mathbf{j}_A = \mathbf{0}$.

The absence of a magnetic induction field, i.e. $\mathbf{B} = \mathbf{0}$, implies that the linear phenomenological relation (101) reduces to,

$$\boldsymbol{\Omega}_A^{\text{m}} = \mathbf{L}_{AA}^{\text{mm}} \cdot (\mathbf{p}_A \times \mathbf{E}). \quad (121)$$

The process is irreversible and dissipative, which means that the Debye energy $-\mathbf{p}_A \cdot \mathbf{E}$ has to diminish. This implies that the intrinsic rotation of the local element, to which the electric dipole \mathbf{p}_A is attached, occurs in the plane spanned by \mathbf{p}_A and \mathbf{E} . Thus, the rotation rate density $\boldsymbol{\Omega}_A^{\text{m}}$ is collinear to the torque $\mathbf{p}_A \times \mathbf{E}$ and $\mathbf{L}_{AA}^{\text{mm}} = L_{AA}^{\text{mm}} \mathbf{1}$. Substituting the relation (121) into the time evolution equation (33) for \mathbf{p}_A yields an equation accounting for the Debye relaxation of the electric dipoles \mathbf{p}_A in the presence of an electric field \mathbf{E} , i.e.

$$\dot{\mathbf{p}}_A = -\alpha_A \mathbf{p}_A \times (\mathbf{p}_A \times \mathbf{E}), \quad (122)$$

where

$$\alpha_A = n_A^{-1} L_{AA}^{\text{mm}},$$

is a phenomenological friction coefficient.

The electric field \mathbf{E} is an effective field that is defined with respect to the local infinitesimal system. It consists of two contributions, i.e.

$$\mathbf{E} = \mathbf{E}^{\text{ext}} + \mathbf{E}^{\text{int}}. \quad (123)$$

The first contribution \mathbf{E}^{ext} is due to an external field applied on the whole system. The second contribution \mathbf{E}^{int} is due to the dipolar interaction with the infinitesimal systems that are in the neighbourhood of the local system. Note that this contribution is internal to the whole system, but external to the local infinitesimal system. In the neighbourhood of the local system, the Debye energy density due to the dipolar interaction between the local systems is proportional to the magnitude of the spatial variation of the electric polarisation. Thus, the Debye energy density can be expressed as:

$$-\mathbf{P} \cdot \mathbf{E} = -\mathbf{P} \cdot \mathbf{E}^{\text{ext}} - D \mathbf{P} \nabla^2 \mathbf{P}, \quad (124)$$

where D is a phenomenological parameter. Note that the spatial variation is expressed in terms of a Laplacian ∇^2 and not a gradient ∇ , since otherwise the contributions of local neighbouring systems located on opposite sides of the local system would cancel out. The expressions (123) and (124) imply that the electric field \mathbf{E}^{int} is given by:

$$\mathbf{E}^{\text{int}} = D \nabla^2 \mathbf{P}. \quad (125)$$

For a single homogeneous sample made of a single chemical substance A , consisting of electric dipoles \mathbf{p}_A , the electric polarisation $\mathbf{P} = n_A \mathbf{p}_A$. Thus, the time evolution equation (122) becomes,

$$\begin{aligned} \dot{\mathbf{p}}_A = & -\alpha_A \mathbf{p}_A \times (\mathbf{p}_A \times \mathbf{E}^{\text{ext}}) \\ & - D \alpha_A \mathbf{p}_A \times (\mathbf{p}_A \times \nabla^2 (n_A \mathbf{p}_A)), \end{aligned} \quad (126)$$

where the terms on the RHS describe the relaxation due to an external electric field \mathbf{E}^{ext} and to the interaction with the electric dipoles in the neighbourhood of the local system.

4.2.4 Relaxation of magnetic dipoles and magnetisation waves

The first model of relaxation of magnetic dipoles \mathbf{m}_A in the presence of a magnetic induction field \mathbf{B} is due to Landau and Lifshitz [36]. Later, Gilbert [37] developed a Lagrangian formulation of this damping. Here, we show how our formalism accounts for this relaxation and for the magnetisation waves, which are the classical counterpart of the “spin waves” [38].

We consider a homogeneous sample made of a single chemical substance A , consisting of magnetic dipoles \mathbf{m}_A in the absence of an electric field, i.e. $\mathbf{E} = \mathbf{0}$. In the frame of sample, the chemical substance A has no translational motion, i.e. $\mathbf{j}_A = \mathbf{0}$.

The absence of an electric field, i.e. $\mathbf{E} = \mathbf{0}$, implies that the linear phenomenological relation (101) reduces to,

$$\boldsymbol{\Omega}_A^{\text{M}} = \mathbf{L}_{AA}^{\text{MM}} \cdot (\mathbf{m}_A \times \mathbf{B}). \quad (127)$$

The process is irreversible and dissipative, which means that the Larmor energy $-\mathbf{m}_A \cdot \mathbf{B}$ has to diminish. This implies that the intrinsic rotation occurs in the plane spanned by \mathbf{m}_A and \mathbf{B} . Thus, the rotation rate density $\boldsymbol{\Omega}_A^{\text{M}}$ is collinear to the torque $\mathbf{m}_A \times \mathbf{B}$ and $\mathbf{L}_{AA}^{\text{MM}} = L_{AA}^{\text{MM}} \mathbf{1}$. Substituting the relation (127) into the time evolution equation (40) for \mathbf{m}_A yields the Landau-Lifshitz equation [37] accounting for the relaxation of the magnetic dipoles \mathbf{m}_A in the presence of a magnetic induction field \mathbf{B} , i.e.

$$\dot{\mathbf{m}}_A = \gamma_A \mathbf{m}_A \times \mathbf{B} - \beta_A \mathbf{m}_A \times (\mathbf{m}_A \times \mathbf{B}), \quad (128)$$

where

$$\beta_A = n_A^{-1} L_{AA}^{\text{MM}},$$

is a phenomenological friction coefficient. Note that the relaxation of the magnetic dipoles is due to a mix of bodily rotation [39] composed with Néel-type magnetic relaxation [40].

The magnetic induction field \mathbf{B} is an effective field that is defined with respect to the local infinitesimal system. It consists of two contributions, i.e.

$$\mathbf{B} = \mathbf{B}^{\text{ext}} + \mathbf{B}^{\text{int}}. \quad (129)$$

The first contribution \mathbf{B}^{ext} is due to an external field applied on the whole system. The second contribution \mathbf{B}^{int} is due to the ferromagnetic interaction with the infinitesimal systems that are in the neighbourhood of the local system. In the neighbourhood of the local system, the Larmor energy density due to the ferromagnetic interaction between the local systems is proportional to the magnitude of the spatial variation of the magnetisation [41]. Thus, the Larmor energy density can be expressed as,

$$-\mathbf{M} \cdot \mathbf{B} = -\mathbf{M} \cdot \mathbf{B}^{\text{ext}} - A \mathbf{M} \nabla^2 \mathbf{M}, \quad (130)$$

where A is a phenomenological parameter called the stiffness constant [38]. The expressions (129) and (130) imply that the magnetic induction field \mathbf{B}^{int} is given by,

$$\mathbf{B}^{\text{int}} = A \nabla^2 \mathbf{M}. \quad (131)$$

For a single homogeneous sample made of a single chemical substance A , consisting of magnetic dipoles \mathbf{m}_A , the magnetisation $\mathbf{M} = n_A \mathbf{m}_A$. Thus, the Landau-Lifshitz equation (128) becomes,

$$\begin{aligned} \dot{\mathbf{m}}_A = & \gamma_A \mathbf{m}_A \times \mathbf{B}^{\text{ext}} - \beta_A \mathbf{m}_A \times (\mathbf{m}_A \times \mathbf{B}^{\text{ext}}) \\ & + A \gamma_A \mathbf{m}_A \times \nabla^2 (n_A \mathbf{m}_A) \\ & - A \beta_A \mathbf{m}_A \times (\mathbf{m}_A \times \nabla^2 (n_A \mathbf{m}_A)). \end{aligned} \quad (132)$$

The first and second terms on the RHS of the Landau-Lifshitz equation (132) describe, respectively, the precession and relaxation due to an external magnetic induction field \mathbf{B}^{ext} . The third term describes magnetisation waves [42] and the fourth term describes the relaxation of magnetisation waves.

4.2.5 Thermally driven magnetisation current

The relaxation of magnetic dipoles in a ferroelectric metal, established in Section 4.2.4, rests on the assumption that the chemical units carrying the magnetic dipoles are at rest. Here, we consider a ferromagnetic conductor made of a fixed lattice and conduction electrons. The magnetic dipoles are carried by the core electrons of the lattice and the conduction electrons (e.g. ‘*d-f*’ and ‘*s-p*’ electrons, respectively). On the scale of interest, the core electrons of the lattice are described as a continuum of chemical type A at rest and the conduction electrons as a fluid of chemical type B . The core electrons carry a magnetic dipole \mathbf{m}_A and the conduction electrons a magnetic dipole \mathbf{m}_B . A quasi-uniform and constant magnetic induction field \mathbf{B} and temperature gradient ∇T are applied on the ferromagnetic conductor. No electric field is applied, i.e. $\mathbf{E} = -\nabla V = \mathbf{0}$.

Since the core electrons are at rest on the scale of interest, the current density of the core electron continuum vanishes, $\mathbf{j}_A = \mathbf{0}$. In our formalism, we neglect the “chemical reactions” a between the core and conduction electrons, i.e. $\omega_a = 0$. Thus, the time evolution equation (40) for the magnetic dipoles \mathbf{m}_A of the core electron continuum A reduces to,

$$\begin{aligned} n_A \dot{\mathbf{m}}_A = & \gamma_A n_A (\mathbf{m}_A \times \mathbf{B}) - \mathbf{m}_A \times \boldsymbol{\Omega}_A^{\text{M}} \\ & + \gamma_{AB} n_A (\mathbf{m}_A \times n_B \mathbf{m}_B). \end{aligned} \quad (133)$$

The absence of electric field, i.e. $\mathbf{E} = \mathbf{0}$, implies that the linear phenomenological relation (98) for the core electron continuum reduces to,

$$\boldsymbol{\Omega}_A^{\text{M}} = \mathbf{L}_{AA}^{\text{MM}} \cdot (\mathbf{m}_A \times \mathbf{B}) + \mathbf{L}_{AB}^{\text{MM}} \cdot (\mathbf{m}_B \times \mathbf{B}). \quad (134)$$

The effect on the rotation of the core electrons of the magnetic torque $\mathbf{m}_B \times \mathbf{B}$ of the conduction electrons is negligible compared to the effect of the magnetic torque $\mathbf{m}_A \times \mathbf{B}$

of the core electrons, i.e. $L_{AB} \ll L_{AA}$, which yields the phenomenological equation (127). The magnetic induction field \mathbf{B} consists of two contributions as established in equation (129) where the magnetisation $\mathbf{M} = n_A \mathbf{m}_A + n_B \mathbf{m}_B$, i.e.

$$\mathbf{B} = \mathbf{B}^{\text{ext}} + A \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B). \quad (135)$$

Following the same lines of thought as in Section 4.2.4, the local time evolution equation (133) for the magnetic dipoles of the core electron continuum is found to be,

$$\begin{aligned} \dot{\mathbf{m}}_A = & \gamma_A (\mathbf{m}_A \times \mathbf{B}^{\text{ext}}) - \beta_A \mathbf{m}_A \times (\mathbf{m}_A \times \mathbf{B}^{\text{ext}}) \\ & + A \gamma_A \mathbf{m}_A \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B) \\ & - A \beta_A \mathbf{m}_A \times (\mathbf{m}_A \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B)) \\ & + \gamma_{AB} (\mathbf{m}_A \times n_B \mathbf{m}_B). \end{aligned} \quad (136)$$

The last term on the RHS of equation (136) is expected to lead to the magnetisation transfer torque, which is the classical counterpart of the ‘‘spin transfer torque’’, generated by the magnetic dipoles \mathbf{m}_B of the conduction electron fluid on the magnetic dipoles \mathbf{m}_A of the core electron continuum [43]. Note that the time evolution of the magnetic dipoles \mathbf{m}_A of the core electron continuum has no explicit dependence on the temperature gradient because these electrons do not undergo transport. However, as shown below, the dynamics of the magnetic dipoles \mathbf{m}_B depends on the temperature gradient.

Since we neglect the ‘‘chemical reactions’’ a between the core and conduction electrons, i.e. $\omega_a = 0$, the time evolution equation (40) for the magnetic dipoles \mathbf{m}_B of the core electron continuum B is given by:

$$\begin{aligned} n_B \dot{\mathbf{m}}_B = & \gamma_B n_B (\mathbf{m}_B \times \mathbf{B}) - \mathbf{m}_B \times \Omega_B^M - (\mathbf{j}_B \cdot \nabla) \mathbf{m}_B \\ & + \gamma_{BA} n_B (\mathbf{m}_B \times n_A \mathbf{m}_A). \end{aligned} \quad (137)$$

Following the same procedure as for the core electrons, we recast the local time evolution equation (133) for the magnetic dipoles of the conduction electron fluid as:

$$\begin{aligned} \dot{\mathbf{m}}_B = & \gamma_B (\mathbf{m}_B \times \mathbf{B}^{\text{ext}}) - \beta_B \mathbf{m}_B \times (\mathbf{m}_B \times \mathbf{B}^{\text{ext}}) \\ & + A \gamma_B \mathbf{m}_B \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B) \\ & - A \beta_B \mathbf{m}_B \times (\mathbf{m}_B \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B)) \\ & - n_B^{-1} (\mathbf{j}_B \cdot \nabla) \mathbf{m}_B + \gamma_{BA} (\mathbf{m}_B \times n_A \mathbf{m}_A). \end{aligned} \quad (138)$$

In order to find an explicit expression for the third term on the RHS of the time evolution equation (138), we describe the transport of the conduction electron fluid in the presence of a uniform magnetic induction field \mathbf{B} and a temperature gradient ∇T . In the absence of an electric field, i.e. $\nabla V = \mathbf{0}$, the linear phenomenological relation (100) reduces to,

$$\mathbf{j}_B = -\frac{1}{q_B} \boldsymbol{\sigma}_B \cdot \boldsymbol{\varepsilon}_B \cdot \nabla T, \quad (139)$$

where the isothermal electric conductivity tensor $\boldsymbol{\sigma}_B$ and the thermopower tensor $\boldsymbol{\varepsilon}_B$ are defined, respectively

as [19],

$$\begin{aligned} \boldsymbol{\sigma}_B & \equiv q_B^2 \mathbf{L}_{BB}, \\ \boldsymbol{\varepsilon}_B & \equiv \frac{1}{q_B} \mathbf{L}_{BB}^{-1} \cdot \mathbf{L}_{Bs}, \end{aligned} \quad (140)$$

and satisfy the following symmetries [44],

$$\begin{aligned} \boldsymbol{\sigma}_B(\mathbf{B}) & = \boldsymbol{\sigma}_B^T(-\mathbf{B}), \\ \boldsymbol{\varepsilon}_B(\mathbf{B}) & = \boldsymbol{\varepsilon}_B^T(-\mathbf{B}), \end{aligned}$$

where the exponent T stands for transpose. Thus, these phenomenological tensors can be split into symmetric and antisymmetric parts according to,

$$\begin{aligned} \boldsymbol{\sigma}_B \cdot \mathbf{a} & = \sigma_{B\parallel} \mathbf{a} + \sigma_{B\perp} (\hat{\mathbf{B}} \times \mathbf{a}), \\ \boldsymbol{\varepsilon}_B \cdot \mathbf{a} & = \varepsilon_{B\parallel} \mathbf{a} + \varepsilon_{B\perp} (\hat{\mathbf{B}} \times \mathbf{a}), \end{aligned} \quad (141)$$

where the magnetic induction field $\mathbf{B} = B \hat{\mathbf{B}}$ and \mathbf{a} is an arbitrary vector. By symmetry the collinear factors $\sigma_{B\parallel}$ and $\varepsilon_{B\parallel}$ are independent of the intensity B of the magnetic induction field. The Hall and Nernst effects [19,44] require the orthogonal factors $\sigma_{B\perp}$ and $\varepsilon_{B\perp}$ respectively to be inversely proportional to B , i.e. $\sigma_{B\perp} \propto B^{-1}$ and $\varepsilon_{B\perp} \propto B^{-1}$. Using the splittings (141), the linear phenomenological relation (139) becomes,

$$\mathbf{j}_B = -\frac{\sigma_{B\parallel} \varepsilon_{B\parallel}}{q_B} \nabla T - \frac{\sigma_{B\parallel} \varepsilon_{B\perp} + \sigma_{B\perp} \varepsilon_{B\parallel}}{q_B} (\hat{\mathbf{B}} \times \nabla T) \quad (142)$$

where the first term on the RHS describes the Soret effect [19] for the component of the magnetic induction field \mathbf{B} that is collinear to the temperature gradient ∇T and does not affect the transport, and the second term describes the Ettingshausen effect [19] per unit of electric charge q_B for the components of the magnetic induction field \mathbf{B} that are orthogonal to the temperature gradient ∇T and affect the transport.

Since the core electrons A do not participate to the transport, the magnetisation current density tensor \mathbf{j}_M is entirely due to the transport of the conduction electrons B . Using the relations (37) and (142), \mathbf{j}_M is found to be,

$$\begin{aligned} \mathbf{j}_M = & -\frac{\sigma_{B\parallel} \varepsilon_{B\parallel}}{q_B} \mathbf{m}_B \odot \nabla T \\ & - \frac{\sigma_{B\parallel} \varepsilon_{B\perp} + \sigma_{B\perp} \varepsilon_{B\parallel}}{q_B} \mathbf{m}_B \odot (\hat{\mathbf{B}} \times \nabla T). \end{aligned} \quad (143)$$

Substituting the linear phenomenological equation (142) into the local time evolution equation (138) for the magnetic dipoles \mathbf{m}_B of the conduction electron fluid B , the

latter becomes,

$$\begin{aligned}
\dot{\mathbf{m}}_B = & \gamma_B (\mathbf{m}_B \times \mathbf{B}^{\text{ext}}) - \beta_B \mathbf{m}_B \times (\mathbf{m}_B \times \mathbf{B}^{\text{ext}}) \\
& + A \gamma_B \mathbf{m}_B \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B) \\
& - A \beta_B \mathbf{m}_B \times (\mathbf{m}_B \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B)) \\
& + \gamma_{BA} (\mathbf{m}_B \times n_A \mathbf{m}_A) + \frac{\sigma_{B\parallel} \varepsilon_{B\parallel}}{q_B n_B} \nabla T \cdot \nabla \mathbf{m}_B \\
& + \frac{\sigma_{B\parallel} \varepsilon_{B\perp} + \sigma_{B\perp} \varepsilon_{B\parallel}}{q_B n_B} (\hat{\mathbf{B}} \times \nabla T) \cdot \nabla \mathbf{m}_B,
\end{aligned} \quad (144)$$

where the first and second terms on the RHS account respectively for the precession and the relaxation of the magnetic dipoles \mathbf{m}_B . The third and fourth terms account respectively for the magnetisation waves and the relaxation of the magnetisation waves. The fifth term accounts accounts for the interaction between the magnetic dipoles \mathbf{m}_A and \mathbf{m}_B of the core and conduction electrons respectively. The sixth and seventh terms account for the transport of \mathbf{m}_B and describe the magnetisation accumulation, which is the classical counterpart of the “spin accumulation”, generated by the temperature gradient ∇T in the presence of a magnetic induction field \mathbf{B} . The sixth term describes the magnetisation accumulation collinear to the temperature gradient and the seventh term describes the magnetisation accumulation orthogonal to the temperature gradient and the magnetic induction field.

The times scales associated to the precession and the relaxation of the conduction electrons are much smaller than the time scales associated to the magnetisation accumulation and the magnetisation transfer torque. Thus, on the latter time scales, the first, second, third and fourth terms on the RHS of the time evolution equation (144) can be neglected, i.e.

$$\begin{aligned}
\dot{\mathbf{m}}_B = & \gamma_{BA} (\mathbf{m}_B \times n_A \mathbf{m}_A) + \frac{\sigma_{B\parallel} \varepsilon_{B\parallel}}{q_B n_B} \nabla T \cdot \nabla \mathbf{m}_B \\
& + \frac{\sigma_{B\parallel} \varepsilon_{B\perp} + \sigma_{B\perp} \varepsilon_{B\parallel}}{q_B n_B} (\hat{\mathbf{B}} \times \nabla T) \cdot \nabla \mathbf{m}_B.
\end{aligned} \quad (145)$$

4.2.6 Electrically driven magnetisation current

The time evolution of the magnetic dipoles of a ferromagnetic conductor, made of a fixed lattice and conduction electrons, in the presence of a quasi-uniform magnetic field \mathbf{B} and a temperature gradient ∇T was established in Section 4.2.5. Here we consider an isothermal conductor, i.e. $\nabla T = \mathbf{0}$, in the presence of a quasi-uniform electric field, i.e. $\mathbf{E} = -\nabla V$. On the scale of interest, the core electrons of the lattice are described as a continuum of chemical type A at rest and the conduction electrons as a fluid of chemical type B . The core electrons carry a magnetic dipole \mathbf{m}_A and the conduction electrons a magnetic dipole \mathbf{m}_B .

The core electron continuum is at rest and the local time evolution equation (136) for the magnetic dipoles

\mathbf{m}_A is independent of the transport. The time evolution for the magnetic dipoles \mathbf{m}_B of the conduction electron fluid is given by equation (138).

In order to find an explicit expression for the third term on the RHS of the time evolution equation (138), we describe the transport of the conduction electron fluid in the presence of a magnetic induction field \mathbf{B} and an electric potential gradient ∇V . In the absence of a temperature gradient, i.e. $\nabla T = \mathbf{0}$, the linear phenomenological relation (100) reduces to,

$$\mathbf{j}_B = -\frac{1}{q_B} \sigma_B \cdot \nabla V. \quad (146)$$

Using the first splitting (141), the linear phenomenological relation (146) becomes,

$$\mathbf{j}_B = -\frac{\sigma_{B\parallel}}{q_B} \nabla V - \frac{\sigma_{B\perp}}{q_B} (\hat{\mathbf{B}} \times \nabla V) \quad (147)$$

where the first term on the RHS is Ohm’s law [19] per unit of electric charge q_B for the component of the magnetic induction field \mathbf{B} that is collinear to the electric potential gradient ∇V and does not affect the transport, and the second term describes the Hall effect [19] per unit of electric charge q_B for the components of the magnetic induction field \mathbf{B} that are orthogonal to the electric potential gradient ∇V and affect the transport.

Since the core electrons A do not participate to the transport, the magnetisation current density tensor \mathbf{j}_M is entirely due to the transport of the conduction electrons B . Using the relations (37) and (147), \mathbf{j}_M is found to be,

$$\mathbf{j}_M = -\frac{\sigma_{B\parallel}}{q_B} \mathbf{m}_B \odot \nabla V - \frac{\sigma_{B\perp}}{q_B} \mathbf{m}_B \odot (\hat{\mathbf{B}} \times \nabla V). \quad (148)$$

Substituting the linear phenomenological equation (147) into the local time evolution equation (138) for the magnetic dipoles \mathbf{m}_B of the conduction electron fluid B , the latter becomes,

$$\begin{aligned}
\dot{\mathbf{m}}_B = & \gamma_B (\mathbf{m}_B \times \mathbf{B}^{\text{ext}}) - \beta_B \mathbf{m}_B \times (\mathbf{m}_B \times \mathbf{B}^{\text{ext}}) \\
& + A \gamma_B \mathbf{m}_B \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B) \\
& - A \beta_B \mathbf{m}_B \times (\mathbf{m}_B \times \nabla^2 (n_A \mathbf{m}_A + n_B \mathbf{m}_B)) \\
& + \gamma_{BA} (\mathbf{m}_B \times n_A \mathbf{m}_A) + \frac{\sigma_{B\parallel}}{q_B n_B} \nabla V \cdot \nabla \mathbf{m}_B \\
& + \frac{\sigma_{B\perp}}{q_B n_B} (\hat{\mathbf{B}} \times \nabla V) \cdot \nabla \mathbf{m}_B,
\end{aligned} \quad (149)$$

where the sixth and seventh terms account for the transport of \mathbf{m}_B and describe the magnetisation accumulation generated by the electric potential gradient ∇V in the presence of a magnetic induction field \mathbf{B} . The seventh term describes the magnetisation accumulation collinear to the electric potential gradient and the fifth term describes the magnetisation accumulation orthogonal to the electric potential gradient and the magnetic induction field.

On the time scales associated to the magnetisation accumulation and the magnetisation transfer torque, the time evolution equation (149) reduces to, i.e.

$$\begin{aligned} \dot{\mathbf{m}}_B = & \gamma_{BA} (\mathbf{m}_B \times n_A \mathbf{m}_A) + \frac{\sigma_{B\parallel}}{q_B n_B} \nabla V \cdot \nabla \mathbf{m}_B \\ & + \frac{\sigma_{B\perp}}{q_B n_B} (\hat{\mathbf{B}} \times \nabla V) \cdot \nabla \mathbf{m}_B. \end{aligned} \quad (150)$$

4.2.7 Thermodynamics of magnetic vortices

The magnetic counterpart of electrically polarised liquid crystals are magnetic vortices or skyrmions [45]. Skyrmions were observed recently [46] in the insulating ferromagnet Cu_2OSeO_3 . Our formalism predicts the precession and relaxation of magnetic vortices in the presence of a temperature gradient.

We consider magnetic vortices that can be treated as a continuum. They form a gyromagnetic medium with a vorticity axis of unit vector $\hat{\mathbf{n}}$. We assume that $\hat{\mathbf{n}}$ is uniform. The magnetic vortices are made of ions and core electrons and are considered as an electrically neutral substance of type A , i.e. $q_A = 0$. In the absence of an external interaction, the vortices have no net magnetisation. On a macroscopic scale, the continuum is homogeneous, i.e. $\nabla \mu_A = \mathbf{0}$. We assume that the viscosity can be neglected, i.e. $\nabla \nu_A = \mathbf{0}$ and $\nabla \omega_A = \mathbf{0}$. Moreover, since the magnetic dipoles are carried by the electrons, that have no intrinsic angular mass, we do not consider the rotation of the matter but we take into account only the rotation of the magnetisation. In an electrical insulator, the magnetic vortices have no translational motion, i.e. $\mathbf{j}_A = \mathbf{0}$. Furthermore, a temperature gradient ∇T is applied along a direction that is different from the vorticity axis of the magnetic vortices.

In this case, the linear phenomenological relation (100) reduces to:

$$\mathbf{m}_A \nabla \mathbf{B} = (\mathbf{L}_{AA}^{-1} \cdot \mathbf{L}_{As}) \cdot \nabla T, \quad (151)$$

where the temperature gradient ∇T induces a magnetic induction field \mathbf{B} and the magnetic dipoles \mathbf{m}_A rotate in an asymmetric manner in order to lower the Larmor energy $-\mathbf{m}_A \cdot \mathbf{B}$. Thus, a net magnetisation \mathbf{M} is generated along the vorticity axis $\hat{\mathbf{n}}$, i.e.

$$\mathbf{M} = n_A \mathbf{m}_A = n_A (\mathbf{m}_A \cdot \hat{\mathbf{n}}) \hat{\mathbf{n}}, \quad (152)$$

and the linear relation (151) is recast as:

$$\mathbf{M} \nabla \mathbf{B} = (n_A \mathbf{L}_{AA}^{-1} \cdot \mathbf{L}_{As}) \cdot \nabla T. \quad (153)$$

The spatial symmetry requires the magnetisation force $\mathbf{M} \nabla \mathbf{B}$ and the thermal force $-\nabla T$ to be collinear, i.e.

$$\mathbf{M} \nabla \mathbf{B} = n_A \mathbf{L}_{AA}^{-1} \mathbf{L}_{As} \nabla T, \quad (154)$$

where $\mathbf{L}_{AA}^{-1} = L_{AA}^{-1} \mathbb{1}$ and $\mathbf{L}_{As} = L_{As} \mathbb{1}$. Thus, the phenomenological relation (154) can be recast as,

$$\mathbf{M} \nabla \mathbf{B} = \lambda n_A k_B \nabla T, \quad (155)$$

where λ is a dimensionless parameter by the relation (107). The magnetisation force density $\mathbf{M} \nabla \mathbf{B}$ satisfies the vectorial identity,

$$\begin{aligned} \mathbf{M} \nabla \mathbf{B} = & \nabla (\mathbf{M} \cdot \mathbf{B}) - \mathbf{B} \nabla \mathbf{M} - \mathbf{B} \times (\nabla \times \mathbf{M}) \\ = & -\nabla_{\mathbf{M}} \cdot (\mathbf{M} \odot \mathbf{B} - (\mathbf{M} \cdot \mathbf{B}) \mathbb{1}) + (\nabla \times \mathbf{M}) \times \mathbf{B} \end{aligned} \quad (156)$$

where we used Thomson's law, i.e.

$$\nabla \cdot \mathbf{B} = 0, \quad (157)$$

and the index \mathbf{M} denotes that there is a dot product between the covariant differential operator ∇ and the magnetisation \mathbf{M} . The term $\nabla_{\mathbf{M}} \cdot (\mathbf{M} \odot \mathbf{B} - (\mathbf{M} \cdot \mathbf{B}) \mathbb{1})$ in the identity (156) corresponds to a surface contribution after integration over the volume of the continuum of magnetic vortices [31]. Thus, it can be neglected in the bulk of the continuum where the magnetisation force density $\mathbf{M} \nabla \mathbf{B}$ is expressed as:

$$\mathbf{M} \nabla \mathbf{B} = (\nabla \times \mathbf{M}) \times \mathbf{B}. \quad (158)$$

Moreover, using the definition of the bound electric current density $\mathbf{j}_{\mathbf{M}}$, i.e.

$$\mathbf{j}_{\mathbf{M}} \equiv \nabla \times \mathbf{M}, \quad (159)$$

the magnetisation force density (158) is recast as [31],

$$\mathbf{M} \nabla \mathbf{B} = \mathbf{j}_{\mathbf{M}} \times \mathbf{B}, \quad (160)$$

which shows that it is the analog of the magnetic part of the Lorentz force density for bound currents. Substituting the expression (160) in the linear relation (154), the latter becomes,

$$\mathbf{j}_{\mathbf{M}} \times \mathbf{B} = \lambda n_A k_B \nabla T, \quad (161)$$

which implies that

$$\mathbf{j}_{\mathbf{M}} \times (\mathbf{j}_{\mathbf{M}} \times \mathbf{B}) = \lambda n_A k_B (\mathbf{j}_{\mathbf{M}} \times \nabla T), \quad (162)$$

and that the thermally induced magnetic induction field \mathbf{B} is orthogonal to the bound current $\mathbf{j}_{\mathbf{M}}$, i.e.

$$\mathbf{j}_{\mathbf{M}} \cdot \mathbf{B} = 0.$$

Using the vectorial identity

$$\mathbf{j}_{\mathbf{M}} \times (\mathbf{j}_{\mathbf{M}} \times \mathbf{B}) = (\mathbf{j}_{\mathbf{M}} \cdot \mathbf{B}) \mathbf{j}_{\mathbf{M}} - \mathbf{j}_{\mathbf{M}}^2 \mathbf{B} = -\mathbf{j}_{\mathbf{M}}^2 \mathbf{B},$$

the linear relation (162) describing the magnetic field \mathbf{B} induced by a temperature gradient ∇T on magnetic vortices $\mathbf{j}_{\mathbf{M}}$ can be recast as a magnetic Seebeck effect for bound currents in an electric insulator, i.e.

$$\mathbf{B} = \varepsilon_{\mathbf{M}} \times \nabla T, \quad (163)$$

where the magnetic Seebeck vector for bound electric currents,

$$\varepsilon_{\mathbf{M}} \equiv -\lambda n_A k_B \mathbf{j}_{\mathbf{M}}^{-1}.$$

Note that the magnetic Seebeck effect (163) is the magnetic analog of the Seebeck effect (113) in an electric insulator.

Substituting the expression (163) for the induced magnetic field \mathbf{B} into the local time evolution equation of the magnetic dipoles (128), we obtain an expression for the thermally induced dynamics of the magnetic vortices, i.e.

$$\begin{aligned} \dot{\mathbf{m}}_A = & \gamma_A \mathbf{m}_A \times (\boldsymbol{\varepsilon}_M \times \nabla T) \\ & - \beta_A \mathbf{m}_A \times \left(\mathbf{m}_A \times (\boldsymbol{\varepsilon}_M \times \nabla T) \right), \end{aligned} \quad (164)$$

where the first term and second terms on the RHS describe, respectively, the precession and the relaxation of the magnetic dipoles of the core electrons of the magnetic vortices due to magnetic field \mathbf{B} induced by a temperature gradient ∇T .

4.2.8 Thermally driven magnetisation waves

We consider a ferromagnetic insulator made of a fixed lattice with core electrons that carry a magnetic dipole \mathbf{m}_A . A constant external magnetic induction field \mathbf{B}^{ext} and a temperature gradient ∇T are applied. The time evolution of the magnetic dipoles is given by the Landau-Lifschitz equation (128).

The magnetic induction field \mathbf{B} appearing in the time evolution equation (128) is an effective field that is defined with respect to the local infinitesimal system. It consists of three contributions, i.e.

$$\mathbf{B} = \mathbf{B}^{\text{ext}} + \mathbf{B}^{\text{int}} + \mathbf{B}^{\text{ind}}. \quad (165)$$

The first contribution \mathbf{B}^{ext} is due to an external field applied on the whole system. The second contribution \mathbf{B}^{int} is due to the ferromagnetic interaction with the infinitesimal systems that are in the neighbourhood of the local system. For a core electron continuum, the magnetisation $\mathbf{M} = n_A \mathbf{m}_A$ and the expression for \mathbf{B}^{int} follows from equation (131), i.e.

$$\mathbf{B}^{\text{int}} = A \nabla^2 (n_A \mathbf{m}_A). \quad (166)$$

The third contribution \mathbf{B}^{ind} is induced by the temperature gradient ∇T . For a core electron continuum, the expression for \mathbf{B}^{ind} follows from equation (163), i.e.

$$\mathbf{B}^{\text{ind}} = \boldsymbol{\varepsilon}_M \times \nabla T, \quad (167)$$

where the bound current density yields,

$$\mathbf{j}_M = \nabla \times (n_A \mathbf{m}_A). \quad (168)$$

Note that the magnetic induction field \mathbf{B}^{ind} is induced only in the presence of a bound current density \mathbf{j}_M . This current density arises in the presence of magnetisation waves generated by the applied external magnetic field \mathbf{B}^{ext} .

Substituting the expressions (166) and (167) for the different contributions to the effective magnetic field \mathbf{B} into the Landau-Lifschitz equation (128), the latter

becomes,

$$\begin{aligned} \dot{\mathbf{m}}_A = & \gamma_A (\mathbf{m}_A \times \mathbf{B}^{\text{ext}}) - \beta_A \mathbf{m}_A \times (\mathbf{m}_A \times \mathbf{B}^{\text{ext}}) \\ & + \gamma_A \mathbf{m}_A \times \left(A \nabla^2 (n_A \mathbf{m}_A) + \boldsymbol{\varepsilon}_M \times \nabla T \right) \\ & - \beta_A \mathbf{m}_A \times \left(\mathbf{m}_A \times \left(A \nabla^2 (n_A \mathbf{m}_A) + \boldsymbol{\varepsilon}_M \times \nabla T \right) \right), \end{aligned} \quad (169)$$

where the third and fourth terms describe, respectively, how magnetisation waves and their relaxation are driven by a temperature gradient ∇T . The effect is maximal when the temperature gradient is orthogonal to the bound current \mathbf{j}_M generated by the applied external magnetic field \mathbf{B}^{ext} .

5 Conclusion

The thermodynamics of irreversible processes is considered for an electrically charged continuous medium containing spontaneous electric and magnetic dipoles in the presence of electromagnetic fields. Expressing the extensive matter state fields in terms of their chemical constituents yields explicit expressions for the current densities. Three types of irreversible terms are derived from these expressions. These are scalar, vectorial and pseudo-vectorial terms that describe, respectively, irreversible chemical processes, irreversible transport processes and irreversible relaxation processes. These processes are coupled through the time evolution equations. Note that with such an approach, the mathematical structure of the irreversible thermodynamics is uncovered physically without imposing it a priori using irreducible representations of the Euclidean group.

As an illustration of our formalism, we describe notably the Lehmann and electric Lehmann effects, the relaxation of the electric and magnetic dipoles. In particular, we are able to predict the effect of a temperature gradient on the time evolution of the magnetic dipoles of conduction electrons interacting with core electrons, which leads to precise expressions for the thermal and electronic magnetisation accumulations. We also predict the precession and relaxation of magnetic vortices induced by a temperature gradient in the absence of an applied magnetic induction field, which is very innovative. Finally, in the presence of an applied magnetic induction field, we show explicitly how a temperature gradient drives magnetisation waves.

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