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# Thermodynamics, Electrodynamics and Ferrofluid-Dynamics

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**Summary.** Applying thermodynamics consistently and in conjunction with other general principles (especially conservation laws and transformation properties) is shown in this review to lead to useful insights and unambiguous results in macroscopic electromagnetism. First, the static Maxwell equations are shown to be equilibrium conditions, expressing that entropy is maximal with respect to variations of the electric and magnetic fields. Then, the full dynamic Maxwell equations, including dissipative fields, are derived from locality, charge conservation, and the second law of thermodynamics.

The Maxwell stress is obtained in a similar fashion, first by considering the energy change when a polarized or magnetized medium is compressed and sheared, then rederived by taking it as the flux of the conserved total momentum (that includes both material and field contributions). Only the second method yields off-equilibrium, dissipative contributions from the fields. All known electromagnetic forces (including the Lorentz force, the Kelvin force, the rotational torque  $\mathbf{M} \times \mathbf{H}$ ) are shown to be included in the Maxwell stress. The derived expressions remain valid for polydisperse ferrofluids, and are well capable of accounting for magneto-viscous effects.

When the larger magnetic particles cluster, or form chains, the relaxation time  $\tau$  of the associated magnetization  $\mathbf{M}_1$  becomes large, and may easily exceed the inverse frequency or shear rate,  $\tau \gtrsim 1/\omega, 1/\dot{\gamma}$ , in typical experiments. Then  $\mathbf{M}_1$  needs to be included as an independent variable. An equation of motion and the associated modifications of the stress tensor and the energy flux are derived. The enlarged set of equations is shown to account for shear-thinning, the fact that the magnetically enhanced shear viscosity is strongly diminished in the high-shear limit,  $\dot{\gamma}\tau \gg 1$ . There is no doubt that it would account for other high-frequency and high-shear effects as well.

## 1 Introduction

The microscopic Maxwell theory, being the epitome of simplicity and stringency, strikes some as the mathematical equivalent of the divine ordinance: “Let there be light.” The macroscopic version of the Maxwell theory is not held in similar esteem, and far less physicists are willing to accept it as an equally important pillar of modern physics. At the start of his lectures on electrodynamics, a colleague of mine routinely asserts, only half in jest, that the fields  $\mathbf{D}$  and  $\mathbf{H}$  were invented by experimentalists, with the mischievous intent to annoy theorists. He then goes on with his lectures, without ever mentioning these two letters again.

Even if this is an extreme position, his obvious vexation is more widely shared. Both to novices and some seasoned physicists, the macroscopic Maxwell theory seems quietly obscure, only precariously grasped. When it is my turn to teach electromagnetism, this whole muddle occasionally surfaces, keeping me awake at nights, before lectures in which alert and vocal students demand coherent reasoning and consistent rules.

Colleagues more moderate than the previous one maintain, in a similar vein, that of the four macroscopic fields, only  $\mathbf{E}$ ,  $\mathbf{B}$  are fundamental, as these are the spatially averaged

microscopic fields,  $\mathbf{E} = \langle \mathbf{e} \rangle$ ,  $\mathbf{B} = \langle \mathbf{b} \rangle$ . Containing information about the polarization  $\mathbf{P}$  and magnetization  $\mathbf{M}$ , the fields  $\mathbf{D} \equiv \mathbf{E} + \mathbf{P}$ ,  $\mathbf{H} \equiv \mathbf{B} - \mathbf{M}$  are part of the condensed system, and hence rather more complex quantities. As this view stems directly from the textbook method to derive the macroscopic Maxwell equations from spatially averaging (or coarse-graining) the microscopic ones, it is subscribed to by many. Yet, as we shall see in section 2.1, this view implies some rather disturbing ramifications. For now, we only observe that it is hardly obvious why the apparently basic difference between  $\mathbf{D}$  and  $\mathbf{E}$  seems to be of so little consequences macroscopically. For instance, it is (something as humdrum as) the orientation of the system's surface with respect to the external field that decides which of the internal fields,  $\mathbf{D}$  or  $\mathbf{E}$ , is to assume the value of the external one.

The characteristic distinction between micro- and macroscopic theories, as we detail in section 2, is the time-inversion symmetry. The arguments are briefly summarized here: Microscopically, the system may go forward or backward in time, and the equations of motion are invariant under time-inversion. Macroscopic systems break this time-inversion invariance,<sup>3</sup> it approaches equilibrium in the forward-direction of time, and the backward direction is forbidden. This fact is expressed in the equations by the dissipative terms. Consider for instance the macroscopic Maxwell equation,  $\dot{\mathbf{B}} = -c\nabla \times (\mathbf{E} + \mathbf{E}^D)$ , where  $\mathbf{E}^D \sim \dot{\mathbf{D}}$  (similar to the second term in the pendulum equation,  $\ddot{x} + \beta\dot{x} + \omega_0^2 x = 0$ ) breaks the time-inversion symmetry of the equation. Without  $\mathbf{E}^D$ , this Maxwell equation would be time-inversion invariant and hence deficient. We shall call  $\mathbf{E}^D \sim \dot{\mathbf{D}}$  the dissipative electric, and similarly,  $\mathbf{H}^D \sim \dot{\mathbf{B}}$  the dissipative magnetic, field. The basic difficulty with the textbook method of deriving the macroscopic Maxwell equation is,  $\mathbf{E} = \langle \mathbf{e} \rangle$  could not possibly contain a term  $\sim \dot{\mathbf{D}}$ , because starting from the microscopic Maxwell equations that are invariant under time-inversion, it is impossible to produce symmetry-breaking  $\mathbf{H}^D$  and  $\mathbf{E}^D$  by spatial averaging.

After all the course work is done, even a good student must have the impression that electrodynamics and thermodynamics, two areas of classical physics, are completely separate subjects. (The word *entropy* does not appear once in the hundreds of pages of Jackson's classics on electrodynamics [1].) Yet, based on concepts as primary as overwhelming probability, thermodynamic considerations are the bedrock of macroscopic physics, so general there can be little doubt that they must also hold for charges, currents and fields. The success London, Ginzburg and Landau enjoyed in understanding superconductivity is one proof that this is true. As we shall see, thermodynamic considerations are indeed useful for understanding macroscopic electrodynamics, some of which could easily be taught in introductory courses, and would usefully be part of the common knowledge shared by all physicists.

In section 2, the usual derivation of macroscopic electrodynamics employing coarse-graining is first discussed, clarifying its basic ideas and pinpointing its difficulties. Then the thermodynamic approach is introduced, making some simple, useful, and possibly surprising points: • It is the introduction of fields that renders electromagnetism a local description. • The static Maxwell equations are an expression of the entropy being maximal in equilibrium. • The structure of the temporal Maxwell equations,<sup>4</sup>

$$\dot{\mathbf{D}} = c\nabla \times (\mathbf{H} + \mathbf{H}^D), \quad \dot{\mathbf{B}} = -c\nabla \times (\mathbf{E} + \mathbf{E}^D), \quad (1)$$

follows from charge conservation alone. (The electric current will be included in the main text.) The dissipative fields are given as

$$\mathbf{H}^D = \alpha d_t \mathbf{B}, \quad \mathbf{E}^D = \beta d_t \mathbf{D}. \quad (2)$$

<sup>3</sup> Some equations in particle physics are only CPT-invariant – this symmetry is then what the associated macro-equations, should they ever be needed, will break.

<sup>4</sup> Approaching electrodynamics from a purely thermodynamic point of view [2], these dissipative fields were first introduced in 1993, see [3, 4]. Later, they were applied to magnetic fluids [5, 6, 7] and ferro-nematics [8], also understood as the low-frequency limit of dynamics that includes the polarization [9] or magnetization [10] as independent variables, and shown to give rise to effects such as shear-excited sound waves [11, 12]. Part of the introduction and section 2 are similar in content to a popular article in German, which appeared in the Dec/2002 issue of *Physik Journal*.

Confining our considerations in section 2 to the rest frame,  $\mathbf{d}_t$  simply denotes partial temporal derivative,  $\mathbf{d}_t \rightarrow \frac{\partial}{\partial t}$ . In section 5, these expressions are generalized to arbitrary frames, in which the medium's velocity  $\mathbf{v}$  and rotational velocity  $\boldsymbol{\Omega} \equiv \frac{1}{2} \nabla \times \mathbf{v}$  are finite, then

$$\mathbf{d}_t \equiv \left( \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla - \boldsymbol{\Omega} \times \right). \quad (3)$$

Including  $\mathbf{H}^D$  has the additional consequence that the total field  $\mathbf{H} + \mathbf{H}^D$  is no longer necessarily along  $\mathbf{B}$ : In isotropic liquids, the equilibrium field is  $\mathbf{H} = \mathbf{B}/(1 + \chi)$  for linear constitutive relation, and remains along  $\mathbf{B}$  also nonlinearly. The dissipative field takes the form  $\mathbf{H}^D = -\alpha \boldsymbol{\Omega} \times \mathbf{B}$  for a rotating medium exposed to a stationary, uniform field, and is perpendicular to  $\mathbf{B}$ . As we shall see, this is why the magnetic torque is finite, and magneto-viscous effects [13, 14] may be accounted for without having to include the relaxation of the magnetization.

The transport coefficient  $\alpha, \beta$  are functions of thermodynamic variables such as temperature, density and field. In section 7.1, considering a polydisperse ferrofluid with different magnetizations  $M_q$ , each relaxing with  $\tau_q$ , and assuming linear constitutive relations,  $M_q = \chi_q H$ , we estimate  $\alpha$  as  $\sum \tau_q \chi_q / (1 + \sum \chi_q)$ .

Starting from the proposition that the sum of material and field momentum is (in the absence of gravitation) a conserved quantity, and its density obeys the conservation law,  $\dot{g}_i + \nabla_j \Pi_{ij} = 0$ , we identify this flux as the Maxwell stress. (Conservation of total momentum is a consequence of empty space being uniform, see more details in section 5.1.) Because  $-\nabla_j \Pi_{ij}$  is the quantity responsible for the acceleration  $\dot{g}_i$ , we may identify it as the robust and locally valid expression for the force density including electromagnetic contributions. So  $F_i = -\int \nabla_j \Pi_{ij} d^3r = -\oint \Pi_{ij} dA_j$ , integrated either over an arbitrary volume  $V$ , or over the associated surface  $A_i$ , is the force this volume develops.

In section 3, we derive the Maxwell stress  $\Pi_{ij}$  thermodynamically, by considering the energy associated with deforming a polarizable or magnetizable body, by  $\delta r_i$  at the surface. Since  $\Pi_{ij} dA_j$  is the surface force density, the energy is  $\delta U = -\oint \Pi_{ij} dA_j \delta r_i$ . (Note that a constant  $\delta r_i$  translates the body, and does not deform it. We consider a constant  $\delta r_i$  that is finite only for part of the surface enclosing a volume.) If  $\delta r_i$  is along the surface normal,  $\delta r_i \parallel dA_j$ , the volume is compressed, if  $\delta r_i$  is perpendicular to the surface normal,  $\delta r_i \perp dA_j$ , it is sheared, and the shape is changed. Without field and in equilibrium, the Maxwell stress reduces to a uniform pressure,  $\Pi_{ij} \rightarrow P \delta_{ij}$ , implying  $\bullet$  shape changes do not cost any energy, and  $\bullet$  we may take  $P \delta_{ij}$  out of the integral in  $\delta U = -\oint \Pi_{ij} dA_j \delta r_i$ , reducing it to the usual thermodynamic relation,  $\delta U = -P \oint dA_j \delta r_i = -P dV$ . In the presence of fields (that may remain nonuniform even in equilibrium), the field-dependent Maxwell stress  $\Pi_{ij}$  is the thermodynamic quantity to deal with.

In section 3.2, the derived Maxwell stress is shown to reduce to

$$\begin{aligned} \nabla_k \Pi_{ik} &= s \nabla_i T + \rho \nabla_i \mu \\ &- \frac{1}{c} \frac{\partial}{\partial t} (\mathbf{D} \times \mathbf{B}) - (\rho_e \mathbf{E} + \frac{1}{c} \mathbf{j}_e \times \mathbf{B})_i. \end{aligned} \quad (4)$$

Containing (part of) the Abraham force and the macroscopic Lorentz force (where  $\rho_e, \mathbf{j}_e$  denote the electric charge and current density), the second line vanishes for neutral systems and stationary fields. The first line is, remarkably, also a quickly vanishing quantity. Generally speaking, the temperature  $T$  and chemical potential  $\mu$  are functions of the entropy  $s$ , density  $\rho$  and field. Without field, both are constant in equilibrium, and  $f^{\text{bulk}} = s \nabla_i T + \rho \nabla_i \mu = 0$ . Applying a nonuniform field leads to non-uniform  $T$  and  $\mu$ , but (in liquids) a very slight change of the density suffices to eliminate  $f^{\text{bulk}}$  again. As this occurs with the speed of sound, the task of detecting any bulk electromagnetic forces  $f^{\text{bulk}}$  directly is rather difficult – though one may of course measure it indirectly, via the density profile as a response to the applied field, or in the case of ferrofluid, via the concentration profile, see section 4.1.

In section 3.4,  $f^{\text{bulk}}$  is (adhering to conventions, see [15]) written as

$$s \nabla_i T + \rho \nabla_i \mu = -\nabla P(\rho, T) + \mathbf{f}^P \quad (5)$$

$$\mathbf{f}^P \equiv M_i \nabla H_i + \nabla \int (\rho \frac{\partial}{\partial \rho} - 1) M_i dH_i. \quad (6)$$

The idea is to divide the vanishing bulk force into a field-independent “zero-field pressure” and a field-dependent “ponderomotive force.” As discussed there, this step has some problems. (The electric field is assumed absent. It leads to completely analogous expressions.)

First, the zero-field pressure, defined as the pressure that remains when the applied field is switched off, depends on how it is switched off. For instance, doing this adiabatically or at constant temperature lead to zero-field pressures that differ by the term  $\int s \frac{\partial}{\partial s} M_i dB_i$ , comparable to the terms in  $\mathbf{f}^P$ . [Eq (5) is appropriate for constant temperature and density.] Second, assuming  $M \sim \rho$ , or equivalently  $(\rho \frac{\partial}{\partial \rho} - 1)M_i = 0$ , seemingly yields the Kelvin force,  $\mathbf{f}^P = M_i \nabla H_i$ , but does not generally, as an equivalent calculation in section 3.4 leads to  $\mathbf{f}^P = M_i \nabla B_i + \nabla \int (\rho \frac{\partial}{\partial \rho} - 1) M_i dB_i$  and the analogous conclusion,  $\mathbf{f}^P = M_i \nabla B_i$ . Closer scrutiny shows assuming  $M \sim \rho$  is only consistent if the susceptibility is small,  $\chi \ll 1$ , and terms  $\sim \chi^2$  may be neglected. In this case, of course, both forces are equivalent, as they differ by  $\frac{1}{2} \nabla M^2$ . Possibly, the reason this kind of faulty deductions were never refuted is because the sum  $\mathbf{f}^{\text{bulk}} = -\nabla P(\rho, T) + \mathbf{f}^P$ , as mentioned, vanishes quickly. And the explicit form of  $\mathbf{f}^P$  is only reflected in a hard-to-measure weakly-varying density, without any further consequences. In ferrofluids, however, if one waits long enough, an inhomogeneous field leads to a much more strongly varying concentration profile, see section 4.1. This should be a worthwhile experiment.

Density, entropy and fields are discontinuous quantities at a system's boundary. Being a function of them, the Maxwell stress  $\Pi_{ij}$  is also discontinuous there. On the other hand, the force  $-\nabla_j \Pi_{ij}$  vanishes quickly only where  $\Pi_{ij}$  varies smoothly. Consider a magnetized system (denoted as  $^{in}$ ), surrounded by a differently magnetized fluid (denoted as  $^{ex}$ ). One example is a ferrofluid vessel surrounded by air, another is an aluminum object submerged in ferrofluid. Applying the same consideration as above, only taking the integration volume to be the narrow region on both sides of the boundary, we find the total force to be a difference of two surface integrals,  $\mathcal{F}_i = \oint \Pi_{ij}^{in} dA_j - \oint \Pi_{ij}^{ex} dA_j \equiv \Delta \oint \Pi_{ij} dA_j$ . This force does not vanish in equilibrium, and its magnetic part is shown in section 3.3 to assume the form

$$\mathcal{F}^{\text{mag}} = \Delta \oint [ \int M_k dH_k + \frac{1}{2} M_n^2 ] dA. \quad (7)$$

It is equivalent to three known formulas, first to

$$\oint (H_i B_j - \delta_{ij} \int B_k dH_k) dA_j, \quad (8)$$

where the integration is along a surface enclosing the internal object but located in the external fluid [15]. If the external magnetization vanishes, Eq (7) is also equivalent to

$$\int M_i \nabla \mathcal{H}_i d^3 r \quad (9)$$

(where  $\mathcal{H}_i = B_i$  is the external field in the absence of the body [15]), and to

$$\oint \frac{1}{2} M_n^2 dA + \int M_k \nabla H_k d^3 r, \quad (10)$$

where the integration is over the volume of the internal object and its surface. The last expression is typically derived [16] as the sum of the Kelvin force  $M_k \nabla H_k$  and a surface couple  $\frac{1}{2} M_n^2$ . However, given the bona fide force density of  $-\nabla_j \Pi_{ij}$ , all forces of Eqs (7, 8, 9, 10) are located at the surface. And neither  $M_k \nabla H_k$  nor  $M_i \nabla \mathcal{H}_i$  are true densities, as their respective force expressions hold only after integration.

In section 5, the consideration includes dissipation and deviations from equilibrium. Starting from general principles including thermodynamics and conservation laws, the full *hydrodynamic Maxwell theory* with fields and the conserved densities (of energy, mass, momentum) as variables, is derived. The result is a dynamic theory for the low-frequency behavior of dense, strongly polarizable and magnetizable fluids. We have especially obtained the explicit form for the momentum and energy flux, the Maxwell stress and Poynting vector, which include dissipative contributions and corrections from the medium's motion. The off-equilibrium Maxwell stress contains especially the following additional terms,

$$\begin{aligned} \nabla_j \Pi_{ij} = \dots - \frac{1}{2} [ \nabla \times (\mathbf{B} \times \mathbf{H}^D + \mathbf{D} \times \mathbf{E}^D) ]_i \\ + H_k^D \nabla_i B_k + E_k^D \nabla_i D_k. \end{aligned} \quad (11)$$

The first line denotes the stress contribution when the dissipative fields are not along the equilibrium ones. Since  $\mathbf{B} \times \mathbf{H}^D = \mathbf{B} \times (\mathbf{H} + \mathbf{H}^D) = \mathbf{M} \times (\mathbf{H} + \mathbf{H}^D)$ , this is the Shliomis

torque [13]. The second line denotes contributions that arise when the magnetization is along  $\mathbf{B}$  but does not have the equilibrium magnitude.

In section 6, a number of key ferrofluid experiments including “rotational field deflection,” “field-enhanced viscosity” and “magnetic pump” are considered employing the hydrodynamic Maxwell theory, showing it is indeed possible to account for these experiments without introducing the magnetization as an independent variable. In section 7, a macroscopic theory for describing polydisperse, chain-forming ferrofluids is introduced. Section 8 summarizes the results on ferronematics and ferrogels, also on the difference when polarization (instead of magnetization) is considered as an independent variable.

Given the long and tortuous history to come to terms with macroscopic electromagnetism, time and again forcing us to back up from blind alleys, any attempt by us on a comprehensive citation would bear historic rather than scientific interests. Since the thermodynamic treatment sketched above and applied by Landau/Lifshitz [15], Rosensweig [16], de Groot/Mazur [17] and others (cf the review by Byrne [18]), is our method of choice, we take them as our starting point, and consequently, only subject them to scrutiny and criticisms here.

This paper is denoted in the SI units throughout, though with a little twist to render the display and manipulation of the formulas simple. We define and employ the fields, sources and conductivity  $\sigma$ ,

$$\begin{aligned} \mathbf{H} &\equiv \hat{\mathbf{H}}\sqrt{\mu_0}, & \mathbf{B} &\equiv \hat{\mathbf{B}}/\sqrt{\mu_0}, & \rho_e &\equiv \hat{\rho}_e/\sqrt{\varepsilon_0}, \\ \mathbf{E} &\equiv \hat{\mathbf{E}}\sqrt{\varepsilon_0}, & \mathbf{D} &\equiv \hat{\mathbf{D}}/\sqrt{\varepsilon_0}, & \sigma &\equiv \hat{\sigma}/\varepsilon_0, \\ \mathbf{P} &\equiv \mathbf{D} - \mathbf{E} \equiv \hat{\mathbf{P}}/\sqrt{\varepsilon_0}, & \mathbf{M} &\equiv \mathbf{B} - \mathbf{H} \equiv \hat{\mathbf{M}}\sqrt{\mu_0}, \end{aligned} \quad (12)$$

where the quantities with hats are the usual ones, in MKSA. All new fields have the dimension  $\sqrt{\text{J}/\text{m}^3}$ , and sensibly,  $\mathbf{H} = \mathbf{B}$  and  $\mathbf{D} = \mathbf{E}$  in vacuum, while  $\rho_e$  and  $\sigma$  are counted in units of  $\sqrt{\text{J}/\text{m}^5}$  and  $s^{-1}$ , respectively. Written in these new quantities, all formulas are rid of the ubiquitous  $\varepsilon_0, \mu_0$ .

## 2 Thermodynamic Derivation of the Maxwell Equations

### 2.1 Coarse-Graining Revisited

It was Lorentz who first differentiated between two versions of the Maxwell equations: The microscopic ones with two fields and the macroscopic ones with four. He also showed how to obtain the latter from the former, a derivation that is conceptually helpful to be divided into two steps: The first consists only of algebraic manipulations, the second, crossing the Rubicon to macroscopics and irreversibility, is the conceptually subtle one. Starting from the microscopic equations,

$$\nabla \cdot \mathbf{e} = \rho_e, \quad \nabla \cdot \mathbf{b} = 0, \quad (13)$$

$$\dot{\mathbf{e}} = c\nabla \times \mathbf{b} - \mathbf{j}_e, \quad \dot{\mathbf{b}} = -c\nabla \times \mathbf{e}, \quad (14)$$

we divide the charge and current into two parts,  $\rho_e = \rho_1 + \rho_2$ ,  $\mathbf{j}_e = \mathbf{j}_1 + \mathbf{j}_2$  (typically taking 1 as free and 2 as bound, though this is irrelevant at the moment). Next, to eliminate  $\rho_2, \mathbf{j}_2$ , the fields  $\mathbf{p}, \mathbf{m}$  are introduced:  $\rho_2 = -\nabla \cdot \mathbf{p}$ ,  $\mathbf{j}_2 = -(\dot{\mathbf{p}} + c\nabla \times \mathbf{m})$ . Although not unique, this step is always possible if  $\rho_2$  is conserved – the fields’ definitions imply  $\dot{\rho}_2 + \nabla \cdot \mathbf{j}_2 = 0$ . Finally, defining  $\mathbf{h} \equiv \mathbf{b} - \mathbf{m}$ ,  $\mathbf{d} \equiv \mathbf{e} + \mathbf{p}$  eliminates  $\mathbf{p}, \mathbf{m}$  and effectuates the “macroscopic” appearance:

$$\nabla \cdot \mathbf{d} = \rho_1, \quad \nabla \cdot \mathbf{b} = 0, \quad (15)$$

$$\dot{\mathbf{d}} = c\nabla \times \mathbf{h} - \mathbf{j}_1, \quad \dot{\mathbf{b}} = -c\nabla \times \mathbf{e}. \quad (16)$$

Although seemingly more complicated, Eqs (15, 16) are equivalent to (13, 14) and not at all macroscopic. This ends the first of the two steps.

Next, we *coarse-grain* these linear equations, spatially averaging them over a small volume – call it grain – repeating the process grain for grain till the grains fill the volume.

Denoting the coarse-grained fields as  $\mathbf{E}^M \equiv \langle \mathbf{e} \rangle$ ,  $\mathbf{D} \equiv \langle \mathbf{d} \rangle$ ,  $\mathbf{B} \equiv \langle \mathbf{b} \rangle$ ,  $\mathbf{H}^M \equiv \langle \mathbf{h} \rangle$ ,  $\mathbf{P} \equiv \langle \mathbf{p} \rangle$ ,  $\mathbf{M} \equiv \langle \mathbf{m} \rangle$ ,  $\rho_\epsilon \equiv \langle \rho_1 \rangle$ ,  $\mathbf{j}_\epsilon \equiv \langle \mathbf{j}_1 \rangle$ , the seemingly obvious result is the macroscopic Maxwell equations,

$$\nabla \cdot \mathbf{D} = \rho_\epsilon, \quad \nabla \cdot \mathbf{B} = 0, \quad (17)$$

$$\dot{\mathbf{D}} = c\nabla \times \mathbf{H}^M - \mathbf{j}_\epsilon, \quad \dot{\mathbf{B}} = -c\nabla \times \mathbf{E}^M. \quad (18)$$

[The superscript  $M$  may appear whimsical here, but will be seen as sensible soon. It denotes the two fields appearing here, in the temporal Maxwell equations (18).] The sketched derivation leads directly to the conclusion that  $\mathbf{E}^M, \mathbf{B}$  are the averaged microscopic fields, while  $\mathbf{D}, \mathbf{H}^M$  are complicated by  $\mathbf{P}, \mathbf{M}$ . Identifying the latter two (in leading orders) with the electric and magnetic dipole densities, respectively, and employing linear response theory imply a host of consequences, of which the presently relevant one is:  $\mathbf{D}, \mathbf{H}^M$  are functions of  $\mathbf{E}^M, \mathbf{B}$  – pairwise proportional for weak fields, with a “temporally nonlocal dependence.” In other words,  $\mathbf{D}$  depends also on the values of  $\mathbf{E}^M$  a while back, and  $\mathbf{H}^M$  on  $\mathbf{B}$ . This is easily expressed in Fourier space,  $\tilde{\mathbf{D}} = \varepsilon(\omega)\tilde{\mathbf{E}}^M$ ,  $\tilde{\mathbf{H}}^M = \tilde{\mathbf{B}}/\mu^m(\omega)$ , where  $\varepsilon, \mu^m$  are<sup>5</sup> complex functions of the frequency  $\omega$ . [A field with tilde denotes the respective Fourier component, eg.  $D(t) = \int d\omega \tilde{D}(\omega)e^{-i\omega t}/2\pi$ .] One cannot overestimate the importance of these two *constitutive relations*: They determine  $\mathbf{D}, \mathbf{H}^M$  in terms of  $\mathbf{E}^M = \langle \mathbf{e} \rangle$ ,  $\mathbf{B} = \langle \mathbf{b} \rangle$ , dispense with the above mentioned non-uniqueness, close the set of equations for given sources, and introduce dissipation. (Still, remember the constitutive relations are an additional input and not the result of coarse-graining.)

General considerations show the real part of  $\varepsilon$  is an even function of  $\omega$ , and the imaginary part an odd one. Focusing on slow processes in dielectrics, we expand  $\varepsilon$  in the frequency  $\omega$  to linear order, writing  $\tilde{\mathbf{D}} = \bar{\varepsilon}(1 + i\omega\beta\bar{\varepsilon})\tilde{\mathbf{E}}^M$ , where  $\bar{\varepsilon}, \beta$  are real, frequency-independent material parameters. Transformed back into temporal space, the constitutive relation reads

$$\mathbf{D} = \bar{\varepsilon}(\mathbf{E}^M - \beta\bar{\varepsilon}\dot{\mathbf{E}}^M). \quad (19)$$

This is a succinct formula: The temporal non-locality is reduced to the dependence on  $\dot{\mathbf{E}}^M$ ; and we intuitively understand that this term (imaginary in Fourier space) is dissipative, as it resembles the damping term  $\sim \dot{\mathbf{x}}$  in the pendulum equation, in which the restoring force is  $\sim \mathbf{x}$ . (Stability requires  $\bar{\varepsilon} > 0$ , and  $\beta$  is positive if electromagnetic waves are to be damped.)

The microscopic Maxwell equations (13, 14) are invariant under time reversal: If  $\mathbf{e}(t), \mathbf{b}(t)$  are a solution, so are  $\mathbf{e}(-t), -\mathbf{b}(-t)$ . (All microscopic variables possess a “time reversal parity.” If even, the variable stays unchanged under time reversal, if odd, it reverses its sign. A particle’s coordinate is even, its velocity odd. Similarly, as electric fields  $\mathbf{e}$  account for charge distributions, and magnetic ones  $\mathbf{b}$  for currents,  $\mathbf{e}$  is even, and  $\mathbf{b}$  odd. Stipulating  $\mathbf{e}$  as even and  $\mathbf{b}$  as odd, the invariance of the Maxwell equations is obvious, as each of Eqs (13, 14) contains only terms with the same parity, eg. the first of Eqs (14) contains only the odd terms:  $\dot{\mathbf{e}}, \nabla \times \mathbf{b}$ , and  $\mathbf{j}_e$ .) Macroscopic theories are not invariant under time reversal, and a solution running forward in time does not remain one when the time is reversed. This is achieved by mixing odd and even terms. In the case of the macroscopic Maxwell equations, we may take the variable  $\mathbf{E}^M = \langle \mathbf{e} \rangle$  as even, and  $\mathbf{B} = \langle \mathbf{b} \rangle$  as odd, because averaging only reduces a strongly varying field to its envelope, with the parity remaining intact.  $\mathbf{D}$ , on the other hand, given by Eq (19), is a mixture of terms with different parities. When inserted into the Maxwell equations, it destroys the reversibility.

This seems to settle the form of the macroscopic Maxwell equations, but does not: Eq (19) cannot be right, because it contains the unphysical, exploding solution:  $D(t) = D_0$  and

$$E^M(t) = E_0^M \exp(t/\beta\bar{\varepsilon}) \equiv E_0^M \exp(t/\tau), \quad (20)$$

for the initial conditions  $D = D_0, E^M = E_0^M$  at  $t = 0$ . This may be avoided by inverting the constitutive relation,  $\tilde{\mathbf{E}}^M = \tilde{\mathbf{D}}/\varepsilon(\omega)$ , which upon expansion becomes  $\tilde{\mathbf{E}}^M = (1/\bar{\varepsilon} - i\omega\beta)\tilde{\mathbf{D}}$ , or back in temporal space,

$$\mathbf{E}^M = \mathbf{D}/\bar{\varepsilon} + \beta\dot{\mathbf{D}}. \quad (21)$$

<sup>5</sup> The permeability  $\mu^m$  is given a superscript, because we need the bare  $\mu$  to denote the chemical potential, a quantity we shall often consider.

Now  $E^M$  depends on  $D, \dot{D}$ , and although there is still a solution  $D \sim \exp(-t/\tau)$ , it relaxes toward zero and is benign. The above frequency expansion confines the validity of Eqs (19, 21) to coarse temporal resolutions, for which a relaxing mode vanishes, but not an exploding one. Only Eq (21) can be correct. Because of an analogous instability,  $H^M \sim e^{t/\bar{\mu}^m \alpha}$ , the proper magnetic constitutive relation is

$$\mathbf{H}^M = \mathbf{B}/\bar{\mu}^m + \alpha \dot{\mathbf{B}}. \quad (22)$$

Given Eq (21,22), the fields  $D, B$  appear the simple, and  $E^M, H^M$  the composite, quantities – and presumably  $D$  is even,  $B$  odd, while  $E^M, H^M$  lack a unique parity. In fact, the reason for taking  $D$  as even is just as persuasive as taking  $E^M$ , because neither the algebraic manipulations (defining  $d$  from  $e, \rho_2$ ), nor the ensuing spatial averaging could possibly have altered  $D$ 's parity: Eqs (15, 16) are as reversible as Eqs (13, 14).

There is nothing wrong with rewriting the microscopic Maxwell equations as Eqs (15, 16) and averaging them to obtain Eqs (17, 18). But being reversible, these are not yet the macroscopic Maxwell equations. In fact, the actual reason dissipative terms appear is because the constitutive relations,  $\tilde{D} = \varepsilon(\omega)\tilde{E}^M$  and  $\tilde{H}^M = \tilde{B}/\mu^m(\omega)$ , close the Maxwell equations, rendering the dynamics of  $P, M$  implicit. Eliminating fast dynamic variables to consider the low-frequency regime is a consequential step, which breaks the connections  $\mathbf{E}^M = \langle \mathbf{e} \rangle$ ,  $\mathbf{D} = \langle \mathbf{e} + \mathbf{p} \rangle$ ,  $\mathbf{B} = \langle \mathbf{b} \rangle \dots$ , established by coarse-graining. Being a consequence of locality and charge conservation (see below), the macroscopic Maxwell equations are always valid, hence necessarily devoid of specifics. One may conceivably arrive at them with varying constitutive relations, implying differently defined fields. However, the proper fields are the ones that also enter the Poynting vector, the Maxwell stress tensor, and the macroscopic Lorentz force,

$$\mathbf{f}_{\text{ML}} = \rho_e \mathbf{E}^M + \mathbf{j}_e \times \mathbf{B}. \quad (23)$$

On a more basic level, one needs to be aware that the whole idea of averaging microscopic equations of motion to obtain irreversible, macroscopic ones is flawed [19]. The two concepts, • entropy as given by the number of available microstates and • paths in phase space connecting these microstates in a temporal order determined by equations of motion are quite orthogonal. Asking how many microstates there are for given energy, irrespective of how these states are arrived at, obviously implies the irrelevance of paths, hence of equations of motion. One reason for this is the fact that tiny perturbations suffice for the system to switch paths which – in any realistic, chaotic system – deviate exponentially from each other. Frequently, the fact that macro- and microdynamics are disconnected is obvious. For instance, irrespective of how energy is being transferred microscopically, and by which particles – classical or quantum mechanical, charged or neutral – temperature always satisfies a diffusion equation (assuming no spontaneously broken gauge symmetry such as in superfluid helium is present). Although the micro- and macro-electrodynamics appear connected, their shared structure is the result of locality and charge conservation, not an indication that one is the average of the other.

Turning now to macroscopic electromagnetic forces, it is tempting to write it as  $\langle \mathbf{f}_L \rangle = \langle \rho_e \mathbf{e} + \mathbf{j}_e \times \mathbf{b} \rangle$ . Yet this formula is of little practical value, as we do not usually have the detailed information that the microscopically accurate fields  $\rho_e, \mathbf{j}_e, \mathbf{e}, \mathbf{b}$  represent. The macroscopic Lorentz force of Eq (23) is obviously different from  $\langle \mathbf{f}_L \rangle$ , even if one assumes that one may indeed identify  $\langle \mathbf{e} \rangle, \langle \mathbf{b} \rangle$  with  $\mathbf{E}^M, \mathbf{B}$ , as  $\langle \rho_e \mathbf{e} \rangle \neq \langle \rho_e \rangle \langle \mathbf{e} \rangle$  (similarly for  $\langle \mathbf{j}_e \times \mathbf{b} \rangle$ ). This difference is frequently taken to be  $P_i \nabla E_i^M$  in the electric, and  $M_i \nabla H_i^M$  in the magnetic case. (Summation over repeated indices is always implied.) Both are referred to as the Kelvin force, with a derivation that presumes the dilute limit of small polarization: First, one calculates the force exerted by an electric field  $\mathbf{e}$  on a single dipole. Next, one assumes that the dipoles in the medium are too far apart to interact and feed back to the field, so the total force density is simply the sum of the forces exerted on all the dipoles in a unit volume, or  $p_i \nabla e_i$ . Without any feedback, the microscopic field is both the applied and the average field,  $e_i = E_i^M$ . And the Kelvin force is:  $\langle p_i \nabla E_i^M \rangle = \langle p_i \rangle \nabla E_i^M \equiv P_i \nabla E_i^M$  – though one should keep in mind that  $P_i \nabla E_i^M \approx P_i \nabla D_i = P_i \nabla (E_i^M + P_i)$ , as  $P_i$  is small in a dilute system.

Facing all these difficulties with averaging microscopic quantities, it is a relief to remember that thermodynamics works exclusively on the macroscopic level, deriving expressions

and equations from general principles, without reference to the microscopic ones. This is what we shall consider from now on.

## 2.2 The Key Role of Locality

Locality, a key concept of physics, is similarly relevant to subjects far beyond: Market economy and evolution theory use local rules among individuals – contracts or the fight for survival – to create socio-economic and biological patterns. Conversely, planned economy and creationism rely on distant actions.

At the heart of the Maxwell equations lies locality. To understand this, one needs to realize that the Maxwell equations may be seen as part of the hydrodynamic theory of condensed systems. If the system is a neutral fluid, three locally conserved densities serve as variables: energy, mass and momentum. If charges are present, it may appear obvious that this conserved quantity is to be included as an additional variable, yet exactly this would violate locality – hence the need to introduce fields. Consider first the microscopic case.

Taking the charge density  $\rho_e(\mathbf{r}, t)$  as a variable, the change in field energy density is  $\phi d\rho_e$ , with the Coulomb potential  $\phi$  depending not only on the local  $\rho_e$ , but on  $\rho_e$  everywhere. Hence, the associated energy  $\phi d\rho_e$  is not localizable, and  $\rho_e$  not a variable of a local theory. Taking instead the electric field  $\mathbf{e}(\mathbf{r}, t)$  as variable, the energy density is  $\frac{1}{2}e^2$ , an unambiguously local expression. (A preference for one of the two energy densities does not preclude the equality of their spatial integrals.) The Coulomb force, with  $\rho_e$  as its variable, acts from the distance; the Lorentz force, expressed in  $\mathbf{e}$  (and  $\mathbf{b}$ ), is local and retarded. Conspicuously,  $\mathbf{e}$  remains partially indetermined for  $\rho_e = \nabla \cdot \mathbf{e}$  given. Yet this is what enables  $\mathbf{e}(\mathbf{r}, t)$  to travel in a wave packet – even while the charge  $\rho_e(\mathbf{r}, t)$  (the acceleration of which in the past created the wave packet) is stationary again. A local description clearly exacts the price of more variables.

Introducing the magnetic  $\mathbf{b}$ -field, via  $\nabla \cdot \mathbf{b} = 0$ , ensures local conservation of energy and momentum in vacuum: The field energy,  $\frac{1}{2}(e^2 + b^2)$ , satisfies a continuity equation. The associated current is the Poynting vector  $c \mathbf{e} \times \mathbf{b}$  which, being the density of field momentum, is itself conserved. If an electron is present, field energy and momentum are no longer conserved, but the total energy and momentum of field and electron are, with the Lorentz force expressing the momentum's rate of exchange between them. With  $\rho_e$  as variable, it is not possible to uphold local conservation of energy and momentum: Giving electromagnetism its local description is arguably the actual achievement of Maxwell's creative genius.

This understanding not only remains valid for the macroscopic case, it is indispensable. Starting with  $\rho_e$ , the conserved, slowly varying charge density, we define a native macroscopic field  $\mathbf{D}$  via  $\nabla \cdot \mathbf{D} \equiv \rho_e$ , which is (same as  $\rho_e$ ) even under time reversal. The relation between  $\mathbf{D}$  and  $\rho_e$  is the same as that between  $\mathbf{e}$  and  $\rho_e$  – only with  $\mathbf{D}$  as variable is it possible to construct a local theory. A further field variable, now odd, is introduced via  $\nabla \cdot \mathbf{B} = 0$ . This exhausts locality as input, and the next task is to derive the equations of motion for  $\mathbf{D}$  and  $\mathbf{B}$ , or the temporal Maxwell equations (18).

## 2.3 Electro- and Magnetostatics

We denote the locally conserved total energy density as  $u$ , taking as its variables the entropy density  $s$ , mass density  $\rho$  and the fields  $\mathbf{D}, \mathbf{B}$ ,

$$du = Tds + \mu d\rho + \mathbf{E} \cdot d\mathbf{D} + \mathbf{H} \cdot d\mathbf{B}. \quad (24)$$

The conjugate fields  $\mathbf{E} \equiv \partial u / \partial \mathbf{D}$ ,  $\mathbf{H} \equiv \partial u / \partial \mathbf{B}$  are defined in exact analogy to the temperature  $T \equiv \partial u / \partial s$ , or the chemical potential  $\mu \equiv \partial u / \partial \rho$ . As  $T$  and  $\mu$ , the fields  $\mathbf{E}, \mathbf{H}$  are real functions of  $s, \rho, \mathbf{D}, \mathbf{B}$ . We do not assume that  $\mathbf{E}, \mathbf{H}$  are necessarily equal to the Maxwell fields,  $\mathbf{E}^M, \mathbf{H}^M$  of Eq (18). In equilibrium, the entropy  $\int s d^3r$  is maximal. And its variation with respect to  $D, B, u, \rho$  vanishes,

$$\int d^3r \{ \delta s - [\alpha \delta u - \beta \delta \rho - A \delta \nabla \cdot \mathbf{B} + \phi \delta (\nabla \cdot \mathbf{D} - \rho_e)] \} = 0. \quad (25)$$



The two constants  $\alpha, \beta$  and the two functions  $A(\mathbf{r}), \phi(\mathbf{r})$  are Lagrange multipliers. The former ensure conservation of energy and mass,  $\delta \int u d^3r, \delta \int \rho d^3r = 0$ ; the latter the validity of Eqs (17). Inserting Eq (24) for  $\delta s$ , this expression breaks down into a sum of four terms, each vanishing independently. The first two are

$$\int d^3r (T^{-1} - \alpha) \delta u = 0, \quad \int d^3r (\mu/T - \beta) \delta \rho = 0.$$

As  $\delta u, \delta \rho$  are arbitrary, the temperature  $T = 1/\alpha$  and the chemical potential  $\mu = \beta/\alpha$  are constants. After a partial integration, with all fields vanishing at infinity, the third term in the sum reads

$$\int d^3r [-\mathbf{H} + \nabla \cdot A(\mathbf{r})] \cdot \delta \mathbf{B} = 0,$$

or  $\nabla \times \mathbf{H} = 0$ . With  $\delta \rho_\epsilon = 0$ , the fourth term is

$$\int d^3r [\mathbf{E} + \nabla \phi(\mathbf{r})] \cdot \delta \mathbf{D} = 0,$$

implying  $\nabla \times \mathbf{E} = 0$ . Summarizing, the conditions for equilibrium are

$$\nabla T = 0, \quad \nabla \mu = 0, \quad \nabla \times \mathbf{E} = 0, \quad \nabla \times \mathbf{H} = 0. \quad (26)$$

Comparing the last two equations to (18), we see that in equilibrium,<sup>6</sup>

$$\mathbf{H}^M = \mathbf{H}, \quad \mathbf{E}^M = \mathbf{E}. \quad (27)$$

This demonstrates that the static Maxwell equations have the same physical origin as the constancy of temperature or chemical potential – they result from the entropy being maximal in equilibrium. Note that once Eqs (26) are given, the associated boundary conditions ensure that the four thermodynamically introduced fields  $\mathbf{D}, \mathbf{B}, \mathbf{E}, \mathbf{H}$  may be measured in an adjacent vacuum.

If the system under consideration is a conductor, the local density is not constant,  $\delta \rho_\epsilon \neq 0$ , though the total charge is,  $\delta \int \rho_\epsilon d^3r = 0$ . This implies  $\phi(\mathbf{r})$  is constant, hence  $\mathbf{E} = -\nabla \phi = 0$ . In other words, the entropy may be further increased by redistributing the charge and becomes maximal for  $\mathbf{E} = 0$ .

A linear medium is given by expanding  $u$  to second order in the fields,

$$u = u_0 + \frac{1}{2}(D^2/\bar{\epsilon} + B^2/\bar{\mu}^m), \quad (28)$$

implying the constitutive relations,  $\mathbf{E} \equiv \partial u / \partial \mathbf{D} = \mathbf{D}/\bar{\epsilon}$ ,  $\mathbf{H} \equiv \partial u / \partial \mathbf{B} = \mathbf{B}/\bar{\mu}^m$ . (Terms linear in  $D, B$  vanish in Eq (28), because  $u - u_0$  is positive definit).

This eye-popping, purely macroscopic approach to electrodynamics, convincingly proving that electro- and magnetostatics are part of thermodynamics, is fundamentally different from the usual coarse-graining procedure. It can be found in § 18 of [15], though the sections's title is so ill-chosen, that it actually serves to hide the subject. The authors express some reservations there, cautioning that the calculation may be questioned, as non-physical fields ( $\nabla \times \mathbf{E}, \nabla \times \mathbf{H} \neq 0$ ) were used to vary the entropy. We believe that this objection is quite unfounded: Off equilibrium, for  $\dot{\mathbf{B}}, \dot{\mathbf{D}} \neq 0$ , the quantities  $\nabla \times \mathbf{E}, \nabla \times \mathbf{H}$  are indeed finite, und healthily physical.

<sup>6</sup> We have  $\nabla \times \mathbf{H} = 0$  in Eq (26), instead of  $\nabla \times \mathbf{H} = \mathbf{j}_\epsilon/c$ , because electric currents are dissipative and vanish in equilibrium. Only for superconductors, capable of sustaining currents in equilibrium, is the latter the proper condition. To derive it, more variables are needed than have been included in Eq (24). A frame-independent version of this “superfluid thermodynamics” was derived recently and employed to consider the London moment [20, 21]: If a superconductor rotates with  $\Omega$ , it maintains the field  $B = 2mc\Omega/e$  in the bulk, instead of expelling it. Notably,  $m$  is the bare mass of the electron and  $e$  its charge. This is a rare instance in physics that the ratio of two macroscopic quantities,  $B/\Omega$ , is given by fundamental constants.

## 2.4 Electrodynamics and Dissipation

Off equilibrium, the fields  $\mathbf{D}, \mathbf{B}$  vary with time. Remarkably, the structure of the temporal Maxwell equations is completely determined by charge conservation: With  $\frac{\partial}{\partial t} \nabla \cdot \mathbf{B} = \nabla \cdot \dot{\mathbf{B}} = 0$ , the field  $\dot{\mathbf{B}}$  must be given as the curl of another field. Call it  $-c\mathbf{E}^M$  and we have  $\dot{\mathbf{B}} = -c\nabla \times \mathbf{E}^M$ . Analogously, with  $\nabla \cdot \dot{\mathbf{D}} = \dot{\rho}_\epsilon = -\nabla \cdot \mathbf{j}_\epsilon$ , the field  $\dot{\mathbf{D}} + \mathbf{j}_\epsilon$  may also be written as the curl of something, or  $\dot{\mathbf{D}} + \mathbf{j}_\epsilon = c\nabla \times \mathbf{H}^M$ . With Eq (27) in mind, we write

$$\mathbf{E}^M = \mathbf{E} + \mathbf{E}^D, \quad \mathbf{H}^M = \mathbf{H} + \mathbf{H}^D, \quad (29)$$

where  $\mathbf{E}^D, \mathbf{H}^D = 0$  in equilibrium. Deriving the explicit expressions for  $\mathbf{E}^D, \mathbf{H}^D$  needs to invoke the Onsager force-flux relation, and will be given in section 5. Here, we present a simple, intuitive argument, excluding conductors: Since equilibrium is defined by the vanishing of  $\nabla \times \mathbf{E}, \nabla \times \mathbf{H}$ , cf. Eqs (26), the dissipative fields  $\mathbf{E}^D$  and  $\mathbf{H}^D$  will depend on these two vectors such that all four vanish simultaneously. Assuming an isotropic medium, the two pairs of axial and polar vectors will to lowest order be proportional to each other,

$$\mathbf{E}^D = \beta c \nabla \times \mathbf{H}, \quad \mathbf{H}^D = -\alpha c \nabla \times \mathbf{E}. \quad (30)$$

Together with (24, 29), Eqs (30) are the nonlinearly valid, irreversible constitutive relations. Assuming weak fields and neglecting magnetic dissipation, ie. Eq (28) and  $\alpha = 0$ , they reduce to  $\mathbf{E} = \mathbf{D}/\bar{\epsilon}$  and  $\mathbf{E}^D = \beta \dot{\mathbf{D}}$ . Conversely, we have  $\mathbf{H} = \mathbf{B}/\bar{\mu}^m$  and  $\mathbf{H}^D = \alpha \dot{\mathbf{B}}$  for  $\beta = 0$ , both the same as Eqs (21, 22).

We consider the relaxation of the magnetization  $\dot{M} = -(M - M^{eq})/\tau$  to estimate the size of the coefficient  $\alpha$ . In Fourier space, we have  $(1 - i\omega\tau)M = M^{eq}$ , or for small frequencies,  $M = (1 + i\omega\tau)M^{eq}$ . This implies  $M = M^{eq} - \tau\dot{M}^{eq} = M^{eq} - \tau(\partial M^{eq}/\partial B)\dot{B}$ . Inserting this into  $H^M = B - M = B - M^{eq} + \tau(dM^{eq}/dB)\dot{B}$ , and identifying  $B - M^{eq}$  as  $H$ , we find

$$\alpha = \tau(\partial M^{eq}/\partial B) \rightarrow \tau\chi/(1 + \chi), \quad (31)$$

and analogously,  $\beta = \tau(\partial P^{eq}/\partial D)$ . The sign  $\rightarrow$  holds for linear constitutive relation, where  $\chi = M/H$  is the magnetic susceptibility.

## 3 The Maxwell Stress and Electromagnetic Forces

While the material momentum  $\rho\mathbf{v}$  is no longer conserved in the presence of electromagnetic fields, the sum of material and field momentum is. This has been mentioned in the introduction and will be dwelt on in great details in chapter 5. Denoting this conserved, total momentum density as  $g_i$ , we take its continuity equation  $\dot{g}_i + \nabla_j \Pi_{ij} = 0$  to define the associated stress tensor  $\Pi_{ij}$ , and write the force density within a continuous medium as

$$f_i^{\text{bulk}} \equiv \dot{g}_i = -\nabla_j \Pi_{ij}. \quad (32)$$

We refer to  $\Pi_{ij}$  as the *Maxwell stress* – although this name is frequently used for its electromagnetic part only. Our reason is, although  $g_i$  and  $\Pi_{ij}$  are well-defined quantities, dividing them into material and electromagnetic contributions, as we shall see below, is a highly ambiguous operation. We believe only the unique  $\Pi_{ij}$  is worthy of Maxwell as a label. Since all macroscopic electromagnetic forces, including the Lorentz and Kelvin force, are contained in  $\Pi_{ij}$ , we shall consider it carefully, starting from the notion that the total force on an arbitrary, simply connected volume is  $\mathcal{F}_i = \int f_i^{\text{bulk}} dV = -\int \nabla_j \Pi_{ij} dV = -\oint \Pi_{ij} dA_j$ , where the Gauss law is employed to convert the volume integral to one over the surface, with the surface element  $dA_j$  pointing outwards.

After the system reverts to stationarity and equilibrium, the force  $g_i = -\nabla_j \Pi_{ij}$  vanishes. The stress  $\Pi_{ij}$  itself, however, same as the pressure  $P$ , remains finite. (The Maxwell stress reduces to the pressure in the field-free limit of a stationary system,  $\Pi_{ij} \rightarrow P\delta_{ij}$ .) Being a function of densities and fields, the stress is discontinuous at the system's boundary if these are. Such a discontinuity represents a surface force that is operative even in equilibrium,

$$\mathcal{F}_i = \oint (\Pi_{ij}^{\text{in}} - \Pi_{ij}^{\text{ex}}) dA_j \equiv \oint \Delta \Pi_{ij} dA_j, \quad (33)$$

where  $^{in}$  and  $^{ex}$  denote *interior* and *external*, respectively. To derive this expression, we broaden the surface of discontinuity to a thin region enclosed by two parallel surfaces, and flatten the discontinuity  $\Delta\Pi_{ij}$  into a large but finite  $\nabla_j\Pi_{ij}$  between these surfaces. Then we integrate  $-\nabla_j\Pi_{ij}$  over an arbitrary portion of this region, and again employ the Gauss law to convert the volume integral into one over the total surface. Now contracting the region's width, only the two large, adjacent surfaces remain. (With  $dA_j$  pointing outwards, there is an extra minus sign in front of  $\Pi_{ij}^{in}$ .) Defining  $n_j$  as the surface normal along  $dA_j$ , the surface force density is

$$f_i^{\text{surf}} = n_j(\Pi_{ij}^{in} - \Pi_{ij}^{ex}) \equiv n_j\Delta\Pi_{ij}. \quad (34)$$

We shall next derive the explicit expression for the stress  $\Pi_{ij}$ , and use it to consider circumstances involving the force densities,  $\mathbf{f}^{\text{bulk}}$  and  $\mathbf{f}^{\text{surf}}$ .

### 3.1 Derivation of the Stress

Generally speaking, the stress  $\Pi_{ij}$  contains contributions from dissipation and flow of the medium, expressed by terms containing quantities such as the dissipative fields  $\mathbf{H}^D$ ,  $\mathbf{E}^D$  of Eq (30), and the velocity  $\mathbf{v}$ . These will be disregarded for the moment and considered in chapter 5.

Before deriving the explicit form of  $\Pi_{ij}$ , let us first understand how the expression for the pressure is thermodynamically derived. Changing the volume  $V$  of a uniform, closed system, the change in energy is  $dU = -P dV$ . As we keep the total entropy and mass constant,  $d(sV) = d(\rho V) = 0$ , the energy density  $du = Tds + \mu d\rho$  may be written as  $du = -(Ts + \mu\rho)dV/V$ . Inserting this in  $dU = d(uV) = Vdu + udV = (u - Ts - \mu\rho)dV$ , we obtain  $P = -u + Ts + \mu\rho$ . Clearly, the pressure is known if  $u(s, \rho)$  is, and it may be calculated as

$$P \equiv - \left. \frac{\partial(uV)}{\partial V} \right|_{sV, \rho V} = -u + \rho \frac{\partial u}{\partial \rho} + s \frac{\partial u}{\partial s}. \quad (35)$$

This method is easily generalized to include fields – all we need is to find a similar geometry in which all variables, including the fields, are constant, and in which the external Maxwell stress  $\Pi_{ij}^{ex}$  vanishes identically. Then the change in energy, employing Eq (34) with  $\Pi_{ij} \equiv \Pi_{ij}^{in}$ , and  $A_j = An_j$  for a flat surface, is

$$dU = -f_i^{\text{surf}} A dr_i = -\Pi_{ij} A_j dr_i. \quad (36)$$

[For  $\Pi_{ij}^{in} = P\delta_{ij}$ , the formula  $dU = -P\delta_{ij} A_j dr_i = -PdV$  is reproduced. If  $\Pi_{ij}$  or  $dr_i$  were non-uniform, and  $\Pi_{ij}^{ex}$  finite, the energy change is  $\oint \delta r_i (\Pi_{ik}^{ex} - \Pi_{ik}) dA_k$ .]

We proceed as outlined above, though heeding the fact that  $dU$  also depends on the change in form, not only in volume. So we take  $A_k$  and  $\delta r_i$  each to successively point in all three directions, evaluating  $\Pi_{ik} A_k \delta r_i$  for nine different configurations, obtaining enough information for all nine components of  $\Pi_{ik}$ . Generalizing the energy density of Eq (24) to include more than one conserved densities  $\rho_\alpha$ ,  $\alpha = 1, 2, \dots$ ,

$$du = Tds + \mu_\alpha d\rho_\alpha + \mathbf{E} \cdot d\mathbf{D} + \mathbf{H} \cdot d\mathbf{B}, \quad (37)$$

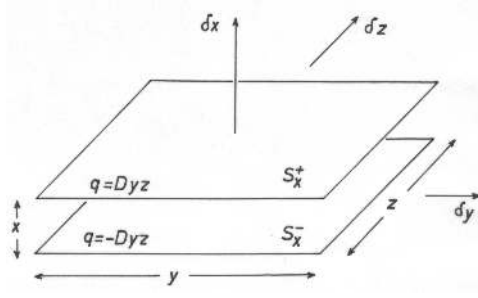
implying a summation over  $\alpha$ , the Maxwell stress will be shown to be

$$\begin{aligned} \Pi_{ik} = \Pi_{ki} = & -E_i D_k - H_i B_k \\ & + (Ts + \mu_\alpha \rho_\alpha + \mathbf{E} \cdot \mathbf{D} + \mathbf{H} \cdot \mathbf{B} - u) \delta_{ik}, \end{aligned} \quad (38)$$

again an expression given in terms of  $u$ , its variables, and the derivatives with respect to these variables.

### Electric Contributions

Consider a parallel-plate capacitor filled with a dielectric medium. Denoting its three linear dimensions as  $x, y, z$ , with  $x \ll y, z$ , the six surfaces  $S_x^\pm, S_y^\pm, S_z^\pm$  (with the outward pointing normal  $\pm\hat{e}_x, \pm\hat{e}_y, \pm\hat{e}_z$ ) have the areas  $A_x = yz, A_y = xz, A_z = xy$ , and the



**Fig. 1.** Two metal plates at  $S_x^+$  and  $S_x^-$  with a dielectric medium between them. Displacing  $S_x^+$  by  $\delta x$  and  $\delta z$  respectively compresses and shears the system.

volume  $V = xyz$ . Taking the two metal plates as  $S_x^\pm$ , the electric fields  $E, D$  are along  $\hat{e}_x$ , see Fig 1. The capacitor is placed in vacuum, so there is neither field nor material outside, with  $\Pi_{ij}^{ex} \equiv 0$ . (The small stray fields at the edges are neglected, because we may place an identical capacitor there, executing the same compression and shear motion.) We now successively displace each of the three surfaces  $S_x^+, S_y^+, S_z^+$ , in each of the three directions,  $\delta r_i = \delta x, \delta y, \delta z$ , while holding constant the quantities: entropy  $sV$ , masses  $\rho_\alpha V$  of the dielectrics, and the electric charges  $\pm q = \pm DA_x$  on the two plates. (The last equality holds because  $q = \int \rho_\epsilon dV = \oint \mathbf{D} \cdot d\mathbf{A}$ .) Displacing the surface  $S_x^+$  by  $\delta x$ , we obtain

$$\delta V = A_x \delta x, \quad \delta s/s = \delta \rho_\alpha / \rho_\alpha = -\delta x/x, \quad \delta D = 0; \quad (39)$$

and we have  $\delta V, \delta s, \delta \rho_\alpha, \delta D = 0$  if the displacement is  $\delta y$  or  $\delta z$  – implying a shear motion of  $S_x^+$ . [There is no summation over  $\alpha$  in Eq (39).] Inserting all three into Eqs (36, 37), we have

$$\Pi_{xx} \delta x = (Ts + \mu_\alpha \rho_\alpha - u) \delta x, \quad \Pi_{yx} = \Pi_{zx} = 0. \quad (40)$$

If the surface is  $S_z^+$  and the displacement  $\delta z$ , we have  $\delta V = A_z \delta z$  and  $\delta s/s, \delta \rho_\alpha / \rho_\alpha, \delta D/D = -\delta z/z$ . If the displacement is  $\delta x$  or  $\delta y$ , we have  $\delta V, \delta s, \delta \rho_\alpha, \delta D = 0$ . Hence

$$\Pi_{zz} \delta z = (Ts + \mu_\alpha \rho_\alpha + E_x D_x - u) \delta z, \quad (41)$$

and  $\Pi_{xz} = \Pi_{yz} = 0$ . Since the directions  $\hat{e}_y$  and  $\hat{e}_z$  are equivalent, we know without repeating the calculation that a displacement of  $S_y^+$  yields  $\Pi_{zz} = \Pi_{yy}$  and  $\Pi_{xy} = \Pi_{zy} = 0$ . (The term  $E_x D_x$  is a result of the metal plates being squeezed, compressing the surface charges,  $\delta q/q = \delta D/D = -\delta z/z$ . The compressibility of the metal – though not that of the dielectric fluid – is taken to be infinite. Otherwise, it would contribute an elastic term in the stress tensor.)

These considerations have yielded all nine components of  $\Pi_{ik}$  for a special coordinate system. Because the stress of Eq (38), for  $\mathbf{D}, \mathbf{E} \parallel \hat{e}_x$  and  $\mathbf{B} = 0$ , produces exactly these components, it is the correct, coordinate-independent expression. This conclusion may appear glib, but is in fact quite solid: If two tensors are equal in one coordinate system, they remain equal in any other. And we have seen that the two are equal in the frame, in which  $\mathbf{E}, \mathbf{D}$  are along  $\hat{x}$ . In other words, the only way to construct a tensor with two parallel vectors, such that  $\Pi_{zz}, \Pi_{yy} = E_x D_x$ , and  $\Pi_{ik} = 0$  otherwise, is to write  $\Pi_{ik} = \delta_{ik} E_j D_j - E_i D_k$ . [There seems to be an ambiguity in the off-diagonal part, as both  $E_i D_k$  and  $E_k D_i$  yields the same nine components derived here; but there is none, because  $\mathbf{E} \parallel \mathbf{D}$  for  $\mathbf{B}, \mathbf{v} = 0$ , therefore  $E_i D_k = E_k D_i$ , cf discussion leading to Eq (164).]

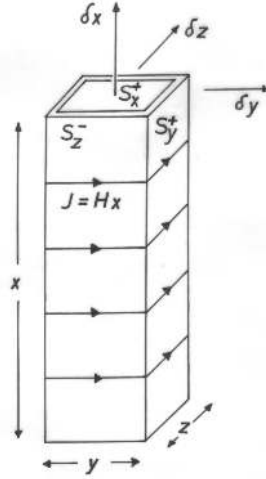
Although this concludes the thermodynamic derivation of the electric part of the Maxwell stress, it is instructive to understand that we could have done it differently, say taking the same capacitor held at a constant voltage  $\phi$ . Considering this modified system must lead to the same stress tensor, because the stress is a local expression which must not depend on whether there is a faraway battery maintaining the voltage. The calculation is similar: One replaces  $u$  in Eq (36) with the potential  $\tilde{u} \equiv u - \mathbf{E} \cdot \mathbf{D}$ , as the system is

no longer electrically isolated<sup>7</sup>.  $E$  is now the field variable, with the constraint  $Ex = \phi$  (which replaces  $DA_x = q$ ). Connecting the capacitor to a heat bath changes the potential to  $\tilde{F} = u - Ts - \mathbf{E} \cdot \mathbf{D}$ , and the constraint changes from constant  $sV$  to  $\delta T = 0$ . ( $\tilde{F}$  is the potential used in [15].) For the explicit calculation see the magnetic case below, implementing  $\mathbf{B} \rightarrow \mathbf{D}$ ,  $\mathbf{H} \rightarrow \mathbf{E}$  in Eqs (42, 43, 44, 45, 46).

If the dielectric medium were simply a vacuum,  $\Pi_{xx} = -\frac{1}{2}E^2$  contracts along  $\hat{e}_x$ , and  $\Pi_{zz} = \frac{1}{2}E^2$  expands along  $\hat{e}_z$ . This reflects the tendency for the differently charged plates to come closer, and the charge in each plate to expand.

## Magnetic Contributions

To obtain the magnetic part of the stress tensor, consider a rod along  $\hat{e}_x$ , of square cross section, made of a magnetizable material and placed in a vacuum. The surfaces  $S_y^\pm, S_z^\pm$  are covered with a sheet of wire-winding that carries a current  $J \perp \hat{e}_x$ . With  $A_x \ll A_y, A_z$ , the magnetic field will be essentially along  $\hat{e}_x$  and confined to the interior of the rod, see Fig 2. So again, there is neither field nor material outside, with  $\Pi_{ij}^{ex} \equiv 0$ . If the system is



**Fig. 2.** A magnetic system with constant current  $J \perp \hat{x}$ , fed by a battery, not shown. Again, it is deformed by displacing  $S_x^+, S_z^+$  along  $\hat{x}$  or  $\hat{z}$ .

isolated, the metal needs to be superconducting to sustain the current, and the constraint on the variable  $B$  during a deformation is constant flux,  $BA_x = \Phi$ . (Compare this to the isolated electric case with  $DA_x = q$ .) If the current  $J$  is held constant by a battery, the attendant potential is (see appendix A)  $\tilde{u} \equiv u - \mathbf{H} \cdot \mathbf{B}$ , and the constraint is  $Hx = J/c$ , from  $J = \int \mathbf{j} \cdot d\mathbf{A} = c \oint \mathbf{H} \cdot d\mathbf{s}$ . (Compare  $Hx = J/c$  to  $Ex = \phi$ .)

The calculation of the isolated magnetic case repeats the isolated electric one, with all above equations remaining valid taking the replacements  $\mathbf{D} \rightarrow \mathbf{B}$ ,  $\mathbf{E} \rightarrow \mathbf{H}$ . We now consider deformation of the rod at constant current and temperature, so the energy needed to deform the system is

$$\delta(\tilde{F}V) = \tilde{F}\delta V + V\delta\tilde{F} = -A_k \Pi_{ik} \delta r_i, \quad (42)$$

$$\delta\tilde{F} = -s\delta T + \mu_\alpha \delta \rho_\alpha - \mathbf{B} \cdot \delta \mathbf{H}. \quad (43)$$

We again successively displace the three surfaces  $S_x^+, S_y^+, S_z^+$ , in all three directions,  $\delta r_i = \delta x, \delta y, \delta z$ , while holding constant the quantities: temperature, mass and the current, ie

<sup>7</sup> See appendix A on Legendre transformations of field variables.

under the conditions,  $\delta T = 0$ ,  $\delta(\rho_\alpha V) = 0$  and  $\delta(Hx) = 0$ . The first surface to be displaced is  $S_x^+$ . When the displacement is  $\delta x$ , we have

$$\delta V = A_x \delta x, \quad \delta H/H = \delta \rho_\alpha / \rho_\alpha = -\delta x/x; \quad (44)$$

and we have  $\delta V$ ,  $\delta \rho_\alpha$ ,  $\delta H = 0$  if the displacement is  $\delta y$  or  $\delta z$  (implying a shear motion of  $S_x^+$ ). Inserting these into Eqs (42, 43), we obtain

$$\Pi_{xx} \delta x = (Ts + \mu_\alpha \rho_\alpha - u) \delta x, \quad \Pi_{yx} = \Pi_{zx} = 0. \quad (45)$$

If the surface is  $S_z^+$  and the displacement  $\delta z$ , we have  $\delta V = A_z \delta z$ ,  $\delta H = 0$ , and  $\delta \rho_\alpha / \rho_\alpha = -\delta x/x$ . If the displacement is  $\delta x$  or  $\delta y$ , we have  $\delta V$ ,  $\delta \rho_\alpha$ ,  $\delta H = 0$ . Hence

$$\Pi_{zz} \delta z = (Ts + \mu_\alpha \rho_\alpha + H_x B_x - u) \delta z, \quad (46)$$

and  $\Pi_{xz} = \Pi_{yz} = 0$ . Since the directions  $\hat{e}_y$  and  $\hat{e}_z$  are equivalent, we have  $\Pi_{zz} = \Pi_{yy}$  and  $\Pi_{xy} = \Pi_{zy} = 0$ . This consideration again yields all nine components of  $\Pi_{ik}$ . Because the stress of Eq (38) produces the same components for  $\mathbf{B}$ ,  $\mathbf{H} \parallel \hat{e}_x$  and  $\mathbf{D} = 0$ , it is the correct, coordinate-independent expression.

## Conclusions and Comparisons

The above considerations yield the macroscopic Maxwell stress  $\Pi_{ik}$  in equilibrium, for stationary systems  $v \equiv 0$ , with either the electric or the magnetic field present. As we shall see in chapter 5, this expression remains intact with both fields present at the same time – though there are additional off-equilibrium, dissipative terms, and corrections  $\sim v$  to account for a moving medium.

Similar derivations exist in the literature, see Landau and Lifshitz [15] for the electric case, and Rosensweig [16] for the magnetic one. We felt the need for a re-derivation, because the present proof is easier to follow and avoids some inconsistencies: Of the six surfaces enclosing the considered volume, only the capacitor plate is displaced in [15], implying that the surface normal  $\mathbf{n} \parallel \mathbf{A}$  is kept parallel to the  $\mathbf{E}$ -field throughout, and (although a footnote asserts otherwise) only the components  $\Pi_{xx}$ ,  $\Pi_{yx}$  and  $\Pi_{zx}$  have been evaluated. Second, the magnetic terms are obtained in § 35 of [15] by the replacement  $\mathbf{E} \rightarrow \mathbf{H}$ ,  $\mathbf{D} \rightarrow \mathbf{B}$ . The result is correct, as we know, but we also see that the geometry is quite different and the analogy hardly obvious. In § 4 of his book on ferrofluids [16], Rosensweig aims to fill this gap. Unfortunately, he starts from the invalid assumption that a certain winding of the wires he specifies gives rise to a field that is uniform and oblique<sup>8</sup>, though it is not difficult to convince oneself that the field is in fact non-uniform and predominantly parallel to  $\hat{e}_x$ .

<sup>8</sup> Rosensweig's geometry is a slab with  $A_y, A_x \ll A_z$  and current-carrying wires along the surfaces  $S_y^\pm, S_z^\pm$ , see his Fig 4.1. The winding of the wires is oblique, the currents flow along  $\pm \hat{e}_y$  in the two larger plates  $S_z^\pm$ , but has a component along  $\pm \hat{e}_x$  in the two narrow side walls  $S_y^\pm$  – take them to be along  $\pm \hat{\mathbf{m}}$ , a vector in the  $xz$ -plane. Rosensweig maintains that the resultant field is uniform and perpendicular to the surface given by the winding, ie by  $\hat{e}_y$  and  $\hat{\mathbf{m}}$ . We do not agree. First the qualitative idea: If the two much larger plates  $S_z^\pm$  were infinite, the field would be strictly parallel to  $\hat{e}_x$ . This basic configuration should not change much if the plates are made finite, and supplemented with the two narrow side walls  $S_y^\pm$  – irrespective of the currents' direction there. This argument is born out by a calculation to superpose the fields from various portions of the currents. First, divide all currents along  $\pm \hat{\mathbf{m}}$  into two components, along  $\pm \hat{e}_z$  and  $\pm \hat{e}_x$ . Next, combine the first with the currents along  $\pm \hat{e}_y$ , such that the four sections of the four surfaces form a closed loop at the same  $x$ -coordinate. The resultant field of all loops is clearly the main one, and strictly along  $\hat{e}_x$ . The leftover currents are those at  $S_y^\pm$  along  $\pm \hat{e}_x$  and their effect is a small dipole field.

### 3.2 Bulk Force Density and Equilibria

We now employ the expression for the stress, Eq (38), to calculate the bulk force density,  $-\nabla_k \Pi_{ik}$ , cf Eq (32). Introducing the Gibbs potential,

$$G(T, \mu_\alpha, H_i, E_i) = u - Ts - \mu_\alpha \rho_\alpha - H_i B_i - E_i D_i, \quad (47)$$

$$\nabla_k G = -s \nabla_k T - \rho_\alpha \nabla_k \mu_\alpha - B_i \nabla_k H_i - D_i \nabla_k E_i \quad (48)$$

(which in the notation of 3.1 should have been  $\tilde{G}$ ), we write the stress as

$$\Pi_{ik} = -(G \delta_{ik} + H_i B_k + E_i D_k), \quad (49)$$

and its gradient as

$$\begin{aligned} \nabla_k \Pi_{ik} &= s \nabla_i T + \rho_\alpha \nabla_i \mu_\alpha - \rho_e E_i \\ &\quad + B_k (\nabla_i H_k - \nabla_k H_i) + D_k (\nabla_i E_k - \nabla_k E_i) \\ &= s \nabla_i T + \rho_\alpha \nabla_i \mu_\alpha - \frac{1}{c} \frac{\partial}{\partial t} (\mathbf{D} \times \mathbf{B}) \\ &\quad - (\rho_e \mathbf{E} + \frac{1}{c} \mathbf{j}_e \times \mathbf{B})_i. \end{aligned} \quad (50)$$

(Remember that terms  $\sim H^D, E^D$  are generally neglected in this chapter.) This result may first of all be seen as a rigorous derivation of the macroscopic Lorentz force,  $\rho_e \mathbf{E} + \frac{1}{c} \mathbf{j}_e \times \mathbf{B}$ . But there are clearly also additional terms: For neutral systems with stationary fields, the bulk force  $-\nabla_k \Pi_{ik}$  reduces to

$$\mathbf{f}^{\text{bulk}} \equiv -(s \nabla T + \rho_\alpha \nabla \mu_\alpha). \quad (51)$$

Note  $\mathbf{f}^{\text{bulk}}$  is field-dependent, because  $T \equiv \partial u(s, \rho_\alpha, \mathbf{D}, \mathbf{B}) / \partial s$ , and similarly  $\mu_\alpha$ , are functions of the fields. As we shall see in section 3.4, the Kelvin force is contained in  $\mathbf{f}^{\text{bulk}}$ . [The discussion of the force,  $\frac{1}{c} \frac{\partial}{\partial t} (\mathbf{D} \times \mathbf{B})$ , tiny in the context of condensed matter physics, is postponed to Eq (172) and the footnote there.]

The remarkable point about  $\mathbf{f}^{\text{bulk}}$ , however, is that it vanishes quickly – on the order of the inverse acoustic frequencies, as long as mechanical equilibrium reigns. Inhomogeneities in temperature, concentration and field are easily and quickly compensated by an appropriate and small inhomogeneity in the density.

To better understand this, we shall examine various equilibrium conditions below. But we must first specify the chemical potentials. For a two-component system such as a solution, with  $\rho_1, \rho_2$  denoting the solute and solvent density, one may take  $\rho_1$  and  $\rho_2$ , or equivalently,  $\rho_1$  and  $\rho \equiv \rho_1 + \rho_2$  as the independent variables. The same holds for ferrofluids, suspensions of magnetic particles, where  $\rho_1$  denotes the average density of magnetic particles and  $\rho_2$  that of the fluid matrix. (Variation of  $\rho_1$  arises primarily from increasing the number of the particles in a unit volume, not from compressing each particle individually.) In what follows, we shall always take the total density  $\rho$  as one of the variables, hence

$$\mu_\alpha d\rho_\alpha \equiv \mu d\rho + \mu_1 d\rho_1. \quad (52)$$

Besides, we shall no longer display the electric terms explicitly from now on, because these follow from identical considerations in all the ensuing formulas, and are simply given by the replacements

$$\mathbf{B} \rightarrow \mathbf{D}, \mathbf{H} \rightarrow \mathbf{E}, \mathbf{M} \rightarrow \mathbf{P}. \quad (53)$$

For linear constitutive relations (LCR),  $\mathbf{M} = \chi^m \mathbf{H}$ ,  $\mathbf{P} = \chi^e \mathbf{E}$ , the replacements imply  $\bar{\mu}^m = 1 + \chi^m \rightarrow \bar{\varepsilon} = 1 + \chi^e$ . Note that the magnetic force is generally speaking up to five orders of magnitude stronger than the electric one. This is connected to the fact that their respective, easily attainable field values are similar in SI units:  $\hat{E} \approx 10^7 \text{V/m}$ ,  $\hat{H} \approx 10^7 \text{A/m}$ , ie  $E \approx 30 \sqrt{\text{J/m}^3}$ ,  $H \approx 10^4 \sqrt{\text{J/m}^3}$ . Also, both susceptibilities are similar in ferrofluids, and do not usually exceed  $10^4$ , hence we have  $\mu_0^m \hat{H}^2 = H^2 \approx \varepsilon_0 \hat{E}^2 \times 10^5 = E^2 \times 10^5$ . In addition to the greater ease and safety of handling, this frequently makes magnetic fields the preferred ones when applying electromagnetic forces.

## True Equilibrium

Temperature and chemical potentials are constant in equilibrium,

$$\nabla T = 0, \quad \nabla \mu = 0, \quad \nabla \mu_1 = 0, \quad (54)$$

so  $\mathbf{f}^{\text{bulk}} \equiv 0$ , cf Eq (51). Constant chemical potentials  $\mu, \mu_1$  (as functions of  $T, \rho, \rho_1, \mathbf{B}$ ) imply that a non-uniform  $\mathbf{B}$  field needs to be compensated by varying densities  $\rho, \rho_1$ , if  $T$  is kept constant. In a one-component liquid, the field inhomogeneity works against the liquid's compressibility,  $\kappa_T \equiv -\partial\rho^{-1}/\partial\mu$ , hence the density change is a small effect. This is different in two-component systems such as ferrofluids, because concentration may vary without compression, and the field only works against the osmotic compressibility  $\kappa_{\text{os}}$ .

In this context, it is important to realize that in suspensions such as ferrofluids incompressibility does not mean  $\delta\rho, \nabla \cdot \mathbf{v} = 0$ . Here, incompressibility implies the constancy of the two microscopic densities,  $\rho_M$  of the magnetic particles and  $\rho_F$  of the fluid matrix (where  $\rho_1 = \langle\rho_M\rangle$ ,  $\rho_2 = \langle\rho_F\rangle$ ), with the averaging taken over a volume element containing many particles). Because the particles are typically denser than the fluid:  $\rho_M \approx 5\rho_F$ , a variation of the particle concentration changes the total density,  $\rho \equiv \rho_1 + \rho_2$ , without any compression taking place. Since  $\rho_1/\rho_M$  is the fraction of volume occupied by the particles, and  $\rho_2/\rho_F$  that occupied by the fluid, we have  $\rho_1/\rho_M + \rho_2/\rho_F = 1$ . Taking  $\rho_M$  and  $\rho_F$  as constant, this implies

$$d\rho = \gamma d\rho_1, \quad \gamma = 1 - \rho_F/\rho_M. \quad (55)$$

If the concentration changes, so does  $\nabla \cdot \mathbf{v} \sim \dot{\rho} \sim \dot{\rho}_1 \neq 0$ . Inserting Eq (55) into  $\mu_\alpha d\rho_\alpha = \mu^{\text{te}} d\rho = \mu_1^{\text{te}} d\rho_1$ , there is only one independent chemical potential, either

$$\mu^{\text{te}} = \mu + \mu_1/\gamma, \quad (56)$$

or  $\mu_1^{\text{te}} = \gamma\mu + \mu_1$ . ( $^{\text{te}}$  stands for true equilibrium.) And the equilibrium conditions are  $\nabla T = 0, \nabla \mu^{\text{te}} = 0$ . In other words, an incompressible two-component system is formally identical to a compressible one-component system, with the usual compressibility substituted by the osmotic one (larger by 6 orders of magnitude, see 4.1),

$$\kappa_{\text{os}} \equiv -\partial\rho_1^{-1}/\partial\mu_1^{\text{te}} = -\partial\rho^{-1}/\partial\mu^{\text{te}}. \quad (57)$$

Note that since our starting equation remains Eq (37), in which  $\mu_\alpha d\rho_\alpha = \mu d\rho + \mu_1 d\rho_1$  is replaced by  $\mu^{\text{te}} d\rho$ , all ensuing results remain valid, especially the stress  $\Pi_{ij}$  and the bulk force  $\mathbf{f}^{\text{bulk}}$ .

## Quasi-Equilibrium

Establishing equilibrium with respect to  $\rho_1$ , the density of suspended particles, is frequently a slow process. Depending on the field gradient, the geometry of the experiment and the particle size, it may take days to weeks [22, 23]. For a rough estimate, equate the Stokes with the Kelvin force to calculate the velocity  $v$  with which a magnetic particle moves:  $6\pi\eta Rv = (4\pi R^3/3) \cdot \chi\mu_0^m \nabla \hat{H}^2/2$ . Taking the particle radius as  $R = 10\text{nm}$ , the viscosity as  $\eta = 10^{-3}\text{kg/ms}$ , the susceptibility as  $\chi \approx 1$ , the field as  $\hat{B} = 0.1\text{T}$ , and the field gradient  $\nabla \hat{B}$  as  $1\text{T/mm}$ , the velocity is around  $10^{-3}\text{mm/s}$ , and the time the particles need to achieve equilibrium at a distance of  $100\text{mm}$  is  $\tau \approx 100\text{mm}/v \approx 10^5\text{s}$  – though particles  $10^2$  times larger ( $R = 1\mu$ ) are  $10^4$  times faster, with  $\tau \approx 10^1\text{s}$ .

After a ferrofluid with both  $\rho$  and  $\rho_1$  uniform is brought into contact with an inhomogeneous magnetic field, the establishment of heat and mechanical equilibria are comparatively fast processes: The condition  $\mathbf{f}^{\text{bulk}} = -(s\nabla T + \rho\nabla\mu^{\text{qe}}) = 0$  is as mentioned satisfied within the inverse acoustic frequency. Constant temperature takes somewhat longer, it is established after heat had enough time to diffuse through the system. For time scales much smaller than the above  $\tau$ , only the concentration  $c \equiv \rho_1/\rho$  remains unchanged. With  $d\rho_1 = cd\rho$  dependent, the thermodynamics is accounted for by only one chemical potential,  $\mu_\alpha d\rho_\alpha = \mu^{\text{qe}} d\rho$ , and the *quasi-equilibrium conditions* are given as

$$\nabla T = 0, \quad \nabla \mu^{\text{qe}} \equiv \nabla(\mu + c\mu_1) = 0. \quad (58)$$



Again, all formulas including the stress and the bulk force remain valid if we replace  $\mu$  with  $\mu^{\text{qe}}$ . Being essentially incompressible, the two-component ferrofluid in quasi-equilibrium usually maintains homogeneous densities,  $\nabla \rho_1 = c \nabla \rho \approx 0$ . (Only a slight spatial variation of  $\rho$  is necessary to compensate for the presence of an inhomogeneous field.) For the rest of this chapter, we shall confine our formulas to that of a one-component fluid,  $\mu_\alpha d\rho_\alpha \rightarrow \mu d\rho$ , but employ

$$\mu \rightarrow \mu^{\text{te}}, \quad \mu \rightarrow \mu^{\text{qe}} \quad (59)$$

to produce results appropriate for true or quasi-equilibrium, respectively.

### Gravitation

It is not always possible to neglect gravitation when considering electromagnetic forces. Including it in the energy,  $u^{\text{tot}} = u + \rho\phi$ , with  $\phi = gz$ , yields  $du^{\text{tot}} = Tds + (\mu + \phi)d\rho + \dots$ , so maximal entropy now implies constant  $\mu + \phi$ , or

$$\nabla \mu = -\nabla \phi = -g\hat{e}_z, \quad (60)$$

or  $\nabla \mu^{\text{qe}} = -\nabla \phi$ , or  $\nabla \mu^{\text{te}} = -\nabla \phi$ . Inserting Eq (60) in Eq (51), the bulk force density  $\mathbf{f}^{\text{bulk}}$  is seen to reduce to the gravitational force.

### 3.3 The Surface Force Density

Since the non-gravitative part of the bulk force density  $\mathbf{f}^{\text{bulk}}$  vanishes quickly, only the surface force,  $\mathbf{f}^{\text{surf}}$  of Eq (34), remains operative for typical experimental situations. This force is what we shall consider now.

### Stress in Equilibrium

The expression for the surface force may be considerably simplified. With the equilibrium conditions given as in Eqs (54), (58) or (60), the Gibbs potential  $G$  is nonuniform only due to the inhomogeneities in the electromagnetic fields and the gravitational potential  $\phi$ , cf Eq (48).  $G$  may be separated into its *zero-field* and *electromagnetic* contributions, which respectively account for its value in the absence of fields and the modification when a field is turned on,

$$G(T, \mu, \mathbf{H}) = G(0) + G_{\text{em}}, \quad (61)$$

$$G_{\text{em}}(T, \mu, \mathbf{H}) = -\int \mathbf{B} \cdot d\mathbf{H}|_{T, \mu}. \quad (62)$$

As indicated, the integral is to be taken for given  $T, \mu$ . Without gravitation,  $G(0) = -K$  is a spatial constant:  $G(0)$  is a function of  $T, \mu$ , both uniform. With gravitation but no field,  $-\nabla_k \Pi_{ik} = \nabla_i G(0) = \rho \nabla_i \phi$ , cf Eqs (48, 50, 60). Hence

$$G(0) = g \int \rho(0) dz - K, \quad (63)$$

where  $\rho(0)$  is the density without field, the one that prevails when the field is turned off at given  $T, \mu$ . (Note  $\int \rho(0) dz \approx \rho(0)z$  for quasi-equilibrium, but not the true equilibrium.) Inserting Eq (61) into Eq (49) yields

$$\Pi_{ij} = [-G(0) + \int B_i dH_i|_{T, \mu}] \delta_{ij} - H_i B_j, \quad (64)$$

stating that we may calculate  $\Pi_{ij}$  for an arbitrary point of the medium, if we know it at one point (fixing the value of  $K$ ) and the field everywhere.

In true and quasi-equilibrium, the field integral  $\int B_i dH_i|_\mu$  is respectively taken at given  $\mu^{\text{te}}$  and  $\mu^{\text{qe}}$ , cf Eqs (56, 58). It is instructive to compare both integrals, as they lead to rather different  $G_{\text{em}}$ . The magnetization is usually measured varying a uniform external field in an enclosed system with a thermal contact, such that  $\rho, \rho_1, T$  stay constant. So the measured quantity is  $M_i(T, \rho, H_i)$ . To evaluate  $G_{\text{em}} = -(\frac{1}{2}H^2 + \int M_i dH_i|_\mu)$ , we need  $M_i(T, \mu, H_i)$  instead, a quantity to be measured in an open system, one connected to a particle reservoir, which itself is not subject to a field, so its chemical potential  $\mu$  remains

constant. Increasing the field in the subsystem, magnetic particles from the reservoir will enter it, resulting in a larger susceptibility than in a closed one. Of course, instead of measuring  $\partial M/\partial H|_\mu$  directly, we may also measure  $\partial M/\partial H|_\rho$ , and calculate the difference between the two susceptibilities,

$$\left.\frac{\partial M}{\partial H}\right|_\mu = \left.\frac{\partial M}{\partial H}\right|_\rho + \left.\frac{\partial M}{\partial \rho}\right|_H \left.\frac{\partial \rho}{\partial \mu}\right|_H. \quad (65)$$

(Hold  $T$  constant throughout, and assume  $\mathbf{M} \parallel \mathbf{H}$ , with  $M, H$  denoting the magnitudes.) Eq (65) is derived by combining an identity with a Maxwell relation,

$$\left.\frac{\partial M}{\partial H}\right|_\mu = \left.\frac{\partial M}{\partial H}\right|_\rho + \left.\frac{\partial M}{\partial \rho}\right|_H \left.\frac{\partial \rho}{\partial H}\right|_\mu, \quad (66)$$

$$\left.\frac{\partial \rho}{\partial H}\right|_\mu = \left.\frac{\partial B}{\partial \mu}\right|_H = \left.\frac{\partial M}{\partial \rho}\right|_H \left.\frac{\partial \rho}{\partial \mu}\right|_H. \quad (67)$$

For quasi-equilibrium,  $\partial \rho/\partial \mu^{\text{qe}} = \rho^2 \kappa_T$ , with  $\kappa_T$  denoting the smallish isothermal compressibility. So the difference between the two  $\partial M/\partial H$  is negligible,

$$\int \mathbf{B} \cdot d\mathbf{H}|_{\mu^{\text{qe}}} \approx \int \mathbf{B} \cdot d\mathbf{H}|_\rho. \quad (68)$$

This is easy to understand, because the open system does not have time to bring the magnetic particle into the field region, so there can be no great difference to a closed system. For the compressible true equilibrium, circumstances are different,  $\partial \rho/\partial \mu^{\text{te}} = \rho^2 \kappa_{\text{os}}$ , with the inverse osmotic compressibility  $\kappa_{\text{os}}$  larger by around  $10^6$ , see 4.1). So the difference between these two susceptibilities is significant. For LCR, Eq (65) reduces to

$$\chi^{\text{m}}(\mu^{\text{te}}) = \chi^{\text{m}}(\rho) + (\rho \partial \chi^{\text{m}}/\partial \rho)^2 \kappa_{\text{os}} H^2, \quad (69)$$

showing that the difference between the two susceptibilities is of second order in the field, and may be neglected if one strictly adheres to LCR. On the other hand, this calculation also shows when the second term may no longer be neglected: Estimating  $\rho(\partial \chi^{\text{m}}/\partial \rho) \approx \chi^{\text{m}} \approx 1$ , and  $\kappa_{\text{os}}^{-1} \approx 10^3$  Pa (see 4.1 below), we find  $\kappa_{\text{os}} H^2 \approx 1$  for  $\hat{H} \approx 3 \times 10^4$  A/m.

## Boundary Conditions

Assuming absence of surface currents, the field boundary conditions are  $\Delta B_n = 0$ ,  $\Delta H_t = 0$ , where

$$\Delta A \equiv A^{in} - A^{ex} \quad (70)$$

is defined as in Eq (34), and the subscripts  $n$  and  $t$  denote the components normal or tangential to the interface:  $H_t \equiv \mathbf{H} \cdot \mathbf{t}$ ,  $B_n \equiv \mathbf{B} \cdot \mathbf{n}$ . ( $\mathbf{n}, \mathbf{t}$  are the normal and tangential unit vectors.) Inserting them into the stress, Eq (38), we find that the off-diagonal part of the surface force density  $\Delta \Pi_{ij}$  vanishes identically,  $\Delta \Pi_{tn} \equiv \Delta(\Pi_{ikt_i n_k}) = -\Delta(H_t B_n) = -B_n \Delta H_t - H_t \Delta B_n = 0$ . The diagonal part,  $\Delta \Pi_{nn} \equiv \Delta(\Pi_{ik n_i n_k})$ , does not vanish and contributes to the surface force. Starting from Eqs (64),  $\Pi_{nn} = -G(0) + \frac{1}{2} H^2 + \int M_i dH_i - H_n B_n$ , we employ  $\frac{1}{2} \Delta(H_t^2 + H_n^2 - 2H_n B_n) = \frac{1}{2} \Delta(M_n^2 + H_t^2 - B_n^2) = \frac{1}{2} \Delta M_n^2$  to obtain

$$\Delta \Pi_{nn} = \Delta[-G(0) + \int M_i dH_i|_{T,\mu} + \frac{1}{2} M_n^2]. \quad (71)$$

At a free surface, because  $\Delta \Pi_{nn}$  and the surface tension are the only operative forces, the force equilibrium,

$$\Delta \Pi_{nn} = \alpha(R_1^{-1} + R_2^{-1}) \quad (72)$$

with  $\alpha > 0$  denoting the surface tension and  $R_1, R_2$  the principle radii of curvature, is the proper boundary condition. If one side of the interface is air, we have  $M \equiv 0$  and  $-G(0) = P_{\text{atm}} - g \int \rho_{\text{air}}(0) dz \approx P_{\text{atm}}$  being the atmospheric pressure. Then the boundary condition [employing Eq (63)] is

$$\begin{aligned} K - g \int \rho(0) dz + \int M_i dH_i|_{T,\mu} + \frac{1}{2} M_n^2 \\ = P_{\text{atm}} + \alpha(R_1^{-1} + R_2^{-1}). \end{aligned} \quad (73)$$

### The Total Surface Force

We evaluate the force on a magnetizable body, submerged in a fluid that is differently magnetized. Because  $dA_j = n_j dA$ ,  $\Delta \Pi_{tn} \equiv 0$ , and  $\Delta \Pi_{ij} = \delta_{ik} \Delta \Pi_{kj} = (n_i n_k + t_i^1 t_k^1 + t_i^2 t_k^2) \Delta \Pi_{kj}$ , Eq (33) takes the form

$$\mathcal{F} = \oint \Delta \Pi_{nn} d\mathbf{A}. \quad (74)$$

We insert Eqs (63, 71), note that the surface integral over any constant vanishes,  $\oint \Delta K dA_i = 0$ , and separate  $\mathcal{F} = \mathcal{F}^{\text{mag}} + \mathcal{F}^{\text{grav}}$  into a magnetic and a gravitational part, to obtain

$$\mathcal{F}^{\text{mag}} = \Delta \oint [\int M_k dH_k|_{T,\mu} + \frac{1}{2} M_n^2] d\mathbf{A} \quad (75)$$

$$= \Delta \oint [\int M_t dH_t|_{T,\mu} + \int M_n dB_n|_{T,\mu}] d\mathbf{A}, \quad (76)$$

$$\mathcal{F}^{\text{grav}} = -g \hat{e}_z V \Delta \rho(0). \quad (77)$$

Eq (77), with  $V$  the volume of the body and  $\hat{e}_z$  the unit vector pointing upward, may be somewhat of a surprise, as  $\rho(0)$  is the zero-field density, and not the actual one, as effected by magnetic field gradients. Clearly, the difference is hidden in the magnetic part of the force, see Eq (79) below.

Note taking  $\nabla T = 0$ , we have  $\nabla \int B_i dH_i \equiv -\nabla G_{\text{em}}(\mu, T, H_i) = (-\partial G_{\text{em}}/\partial \mu) \nabla \mu + B_i \nabla H_i$ . Furthermore, with  $-\partial G_{\text{em}}/\partial \mu = \rho - \rho(0)$ ,  $\nabla \mu = -g \hat{e}_z$ , and  $\nabla \int M_i dH_i = \nabla \int B_i dH_i - H_i \nabla H_i$ , we have

$$\nabla \int M_i dH_i = M_i \nabla H_i - [\rho - \rho(0)] g \hat{e}_z. \quad (78)$$

Using it to consider a magnetizable body in vacuum, we may first eliminate the  $\Delta$  in Eq (75), and then employ the Gauss law to formerly write

$$\mathcal{F}^{\text{mag}} = \oint \frac{1}{2} M_n^2 d\mathbf{A} + \int M_k \nabla H_k d^3r - [\rho - \rho(0)] g \hat{e}_z V. \quad (79)$$

This is sometimes construed as proof that  $M_k \nabla H_k$  is the bulk Kelvin, and  $\frac{1}{2} M_n^2$  a surface, force density [16]. This is incorrect: All forces are located at the surface.

Although it is sometimes difficult to discern non-locality in a static context, this is an easy task in a dynamic one: The appropriate question is always, which volume element is going to be accelerated if force balance suddenly fails. As long as we adhere to the definition of Eq (32), we are sure that the force is where the accelerated volume element resides. This connection gets lost only after the force density is integrated over, especially when the Gauss law has been employed.

If the magnetizable body is a plate, with the field gradient normal to its surface, refer to Eq (76) to realize that if the field is either predominantly tangential or normal to its surface, the respective magnetic force is  $\int M_k \nabla H_k d^3r$  or  $\int M_k \nabla B_k d^3r$ , see [24]. Conversely, the force on a non-magnetic body submerged in ferrofluid of magnetization  $M$  is

$$\mathcal{F}^{\text{mag}} = - \oint [\int M_t dH_t|_{T,\mu} + \int M_n dB_n|_{T,\mu}] d\mathbf{A}. \quad (80)$$

Two different force expressions are found in the book by Landau and Lifshitz [15]. (The calculations are in electric quantities and assume linear constitutive relation, see §16. They are converted to magnetic quantities and generalized to nonlinear constitutive relation here.) The first is

$$\mathcal{F}_i^{\text{mag}} = \oint dA_j (H_i B_j - \delta_{ij} \int B_k dH_k|_{T,\mu}). \quad (81)$$

It is easily derived if gravitation is neglected: First eliminate  $\Pi_{ij}^{\text{in}}$  in Eq (34), because  $\oint \Pi_{ij}^{\text{in}} dA_j = \int \nabla_j \Pi_{ij}^{\text{in}} d^3r$  vanishes in mechanical equilibrium. Then insert Eq (64) into  $\Pi_{ij}^{\text{ex}}$  to obtain the above expression. Note the integral may be taken rather far away from the enclosed body, as long as the external medium is in mechanical equilibrium, because

the integral over any closed surface within the external medium  $\oint dA_j \Pi_{ij}^{ex}$  vanishes. The second force expression is only valid in vacuum, or any other non-magnetizable medium,

$$\mathcal{F}^{\text{mag}} = \int M_i \nabla \mathcal{H}_i d^3 r, \quad (82)$$

where  $\mathcal{H}_i = \mathcal{B}_i$  is the external field in the absence of the body. The proof in [15], of the equivalence between Eq (81) and (82), is quite protracted, spread over many sections. The essence is: One starts from the energy Eq (37) to deduce the form of the Maxwell stress, from which Eq (81) is deduced. And one may employ this force to calculate the energy change associated with the displacement of the magnetized body,  $\delta U = \mathcal{F}_i^{\text{mag}} \delta r_i$ . Equivalently,  $\delta U$  may be calculated employing Eq (37) directly, integrating it over the whole space for the two positions, before and after the displacement, of the magnetized body. And the difference in energy is  $\delta U$ , same as above. Moreover, as shown in [15], instead of employing Eq (37), one may calculate  $\delta U$  by using  $\delta U = \int \mathbf{M} \cdot \delta \mathcal{H} d^3 r$ , which conveniently prescribes an integration over the magnetizable body only. Since  $\delta \mathcal{H} = (\delta r_i \nabla_i) \mathcal{H}$  with  $\delta r_i$  constant for a solid-body displacement, we have  $\delta U = \delta r_i \int M_k \nabla_i \mathcal{H}_k d^3 r = \mathcal{F}_i^{\text{mag}} \delta r_i$ , or Eq (82). ( $\mathcal{F}^{\text{mag}} = \int M_i \nabla_i \mathcal{H} d^3 r$  is also valid since  $\nabla \times \mathcal{H} = 0$ .)

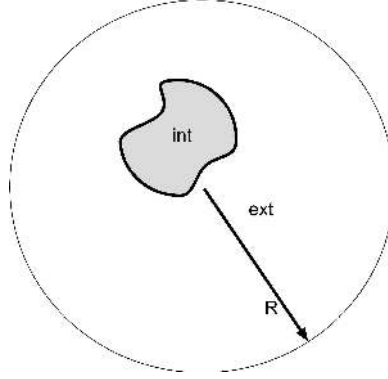


Fig. 3. force on a magnetizable body

Given this long-winding reasoning, there has been some misunderstanding in the literature [25, 26], and it is useful to provide a direct proof [27] of the equivalence between Eq (81) and (82): Writing  $\mathbf{H} \equiv \mathcal{H} + \mathbf{h}$  and  $\mathbf{B} \equiv \mathcal{B} + \mathbf{b}$  defines the additional fields  $\mathbf{h}, \mathbf{b}$  which arise due to the presence of the magnetizable body, represented as the shaded area (marked “int” for internal) in Fig 3. With  $\nabla \cdot \mathbf{B} = 0$ ,  $\nabla \times \mathbf{H} = 0$  and  $\nabla \cdot \mathcal{B} = 0$ ,  $\nabla \times \mathcal{H} = 0$ , we also have  $\nabla \cdot \mathbf{b} = 0$ ,  $\nabla \times \mathbf{h} = 0$ . Besides,  $\mathcal{H} \equiv \mathcal{B}$  hold generally, and  $\mathbf{H} \equiv \mathbf{B}$ ,  $\mathbf{h} = \mathbf{b}$  hold outside the magnetized body (in the region marked “ext” for external). This is why we may write the magnetic force, Eq (81), as  $\oint_{ext} dA_j (H_i B_j - \frac{1}{2} H^2 \delta_{ij})$ . (The subscript  $ext$  notes that we are to take the values of the discontinuous field on the external side of the body.) Dividing the fields as defined, the force is

$$\mathcal{F}_i^{\text{mag}} = \oint_{ext} (\mathcal{H}_i \mathcal{B}_j - \frac{1}{2} \mathcal{H}^2 \delta_{ij} + h_i b_j - \frac{1}{2} h^2 \delta_{ij} + \mathcal{H}_k h_k \delta_{ij} - \mathcal{H}_i b_j - h_i \mathcal{B}_j) dA_j. \quad (83)$$

With  $\mathcal{B}_i, \mathcal{H}_i$  continuous, we may employ the Gauss law to show that the first two terms cancel:  $\int d^3 r \nabla_j (\mathcal{H}_i \mathcal{B}_j - \frac{1}{2} \mathcal{H}^2 \delta_{ij}) = \int d^3 r (\mathcal{B}_j - \mathcal{H}_j) \nabla_j \mathcal{H}_i = 0$ . The next two terms also vanish, as the surface of integration may be displaced into infinity:  $\oint_{ext} dA_j (h_i b_j - \frac{1}{2} h^2 \delta_{ij}) = \oint_{R \rightarrow \infty} dA_j (h_i b_j - \frac{1}{2} h^2 \delta_{ij}) + \int_{ext} d^3 r (h_j - b_j) \nabla_j h_i$ . The first term falls off as  $R^{-3} R^{-3} R^2 = R^{-4}$ , because its longest reaching contribution is dipolar; the second term is zero because  $h_j = b_j$ . The last three terms are continuous and may be written as

$\oint_{int} dA_j (\mathcal{H}_k h_k \delta_{ij} - \mathcal{H}_i b_j - h_i \mathcal{B}_j) = \int_{int} d^3r [(\mathcal{B}_j - \mathcal{H}_j) \nabla_j h_i + (b_j - h_j) \nabla_j \mathcal{H}_i]$  which, with  $\mathcal{B}_j = \mathcal{H}_j$  and  $M_i = B_i - H_i = b_i - h_i$ , is the proof of  $\mathcal{F}_i^{\text{mag}} = \int d^3r M_i \nabla_j \mathcal{H}_i$ . (These three terms are continuous because, with  $dA_j \parallel n_j$ ,  $n_j$  being the normal vector, they may be written as  $(\mathcal{H}_t h_t + \mathcal{H}_n h_n) n_i - \mathcal{H}_i b_n - h_i \mathcal{H}_n$ , where the subscripts  $t$  and  $n$  refer to the normal and tangential components, respectively. This vector is given by two components:  $\mathcal{H}_t h_t + \mathcal{H}_n h_n - \mathcal{H}_n b_n - h_n \mathcal{H}_n = \mathcal{H}_t h_t - \mathcal{H}_n b_n$  along  $\mathbf{n}$ , and  $-\mathcal{H}_t b_n - h_t \mathcal{H}_n$  perpendicular to it. It is continuous because  $\mathcal{H}_i = \mathcal{B}_i$ ,  $h_t$ , and  $b_n$  are all continuous.)

Summarizing, we note that all the above formulas account for the same force, strictly located at the surface.  $M_k \nabla H_k$  and  $M_i \nabla H_i$  are valid only after a volume integration, they should not be interpreted as body force densities.

### 3.4 Zero-Field Pressure and the Kelvin Force

In this section, we shall consider two ambiguous quantities which are nevertheless frequently employed: the zero-field pressure and the Kelvin force. Their introduction is based on a seemingly self-evident premise, that one can divide the Maxwell stress,  $\Pi_{ij}$ , into a field-independent part, called zero-field pressure,  $P\delta_{ij}$ , and a field-dependent one, given less obviously by the Kelvin force,  $M_j \nabla_i H_j$ . Accepting this, the momentum balance,  $\dot{g}_i + \nabla_j \Pi_{ij} = 0$ , is frequently written as [28]

$$\dot{g}_i + \nabla_i P - M_j \nabla_i H_j = 0. \quad (84)$$

(There is also a third term,  $\frac{1}{2} \nabla \times (\mathbf{M} \times \mathbf{B})$ , deemed to enter the equation only when the fluid contains the degree of freedom of ‘‘internal rotation.’’ We shall discuss it with other off-equilibrium terms, in next chapter.) Eq (84) and the premise leading to it are fallacious on three accounts: • First, however one defines the zero-field pressure, it is never a universally field-independent quantity, and some field effects are always contained in  $P$ . • Even neglecting the first point, writing  $\nabla_j \Pi_{ij}$  as  $\nabla_i P - M_j \nabla_i H_j$  requires preconditions far more severe than is usually acknowledged. As a consequence, they are frequently violated when the Kelvin force is applied. • Finally, as we discussed in section 3.2, the bulk force density  $\mathbf{f}^{\text{bulk}} = -\nabla_j \Pi_{ij}$  vanishes quickly, within the inverse acoustic frequency. So whatever the field-dependent part of the stress is,  $M_j \nabla_i H_j$  or not, it does not account for any electromagnetic action at longer time spans. (The following consideration neglects gravitation and again treats true and quasi-equilibrium simultaneously, with  $\mu \rightarrow \mu^{\text{qe}}, \mu^{\text{te}}$  where appropriate.)

### Different Zero-Field Pressures

As with the Gibbs energy, Eq (61), one may also divide the free energy densities  $F \equiv u - Ts$  and  $\tilde{F} \equiv F - \mathbf{H} \cdot \mathbf{B}$  into a field-free and field-induced part,

$$F = F(0) + F_{\text{em}}, \quad F_{\text{em}} = \int \mathbf{H} \cdot d\mathbf{B}|_{T,\rho}, \quad (85)$$

$$\tilde{F} = \tilde{F}(0) + \tilde{F}_{\text{em}}, \quad \tilde{F}_{\text{em}} = -\int \mathbf{B} \cdot d\mathbf{H}|_{T,\rho}, \quad (86)$$

with  $F(0) = \tilde{F}(0)$  a function of  $T, \rho$  and the integrals taken at given  $T, \rho$ . With  $-G(0)$  given as  $-u(0) + Ts(0) + \mu\rho(0)$ , cf Eq (61), it appears quite natural to refer to it as the zero-field pressure  $P(0)$ . In a similar vein, one may take  $(\rho \frac{\partial}{\partial \rho} - 1)F(0) \equiv \rho\mu(0) - F(0)$  as  $P(0)$ . Unfortunately, these two pressures are different.  $P(\mu) \equiv -G(0)$  is the pressure that remains turning off the field at given  $\mu, T$ , while  $P(\rho) \equiv (\rho \frac{\partial}{\partial \rho} - 1)F(0)$  is the pressure at given  $\rho, T$ . Because the chemical potential  $\mu$  changes turning off the field at given density, and vice versa, the density changes at given  $\mu$ , these two pressures differ by a field-dependent quantity. [If the field is turned off adiabatically, rather than isothermally, the result is yet another zero-field pressure:  $P(s, \rho) \equiv (\rho \frac{\partial}{\partial \rho} + s \frac{\partial}{\partial s} - 1)u(0)$ .] Generally speaking, when choosing an appropriate set of variables, it is useful to keep in mind that  $\rho$  is the variable that remains unchanged when a field is turned on in a closed system. Therefore,  $F(0)$  is frequently a temporal constant, in contrast to  $G(0)$ , a spatial constant.

Writing  $G$ , either in terms of  $F$ ,  $\tilde{F}$ , or directly, we have

$$-G = (\rho \frac{\partial}{\partial \rho} - 1)F - \mathbf{H} \cdot \mathbf{B} = P(\rho) + \frac{1}{2}H^2 - \frac{1}{2}M^2 + \int d\mathbf{B} \cdot (1 - \rho \frac{\partial}{\partial \rho})\mathbf{M}|_\rho, \quad (87)$$

$$-G = (\rho \frac{\partial}{\partial \rho} - 1)\tilde{F} = P(\rho) + \frac{1}{2}H^2 + \int d\mathbf{H} \cdot (1 - \rho \frac{\partial}{\partial \rho})\mathbf{M}|_\rho, \quad (88)$$

$$-G = P(\mu) + \frac{1}{2}H^2 + \int d\mathbf{H} \cdot \mathbf{M}|_\mu, \quad (89)$$

where the density derivatives are taken at constant  $B$  in Eq (87), and at constant  $H$  in (88). Equating Eq(88) with (89), we find

$$P(\mu) = P(\rho) - \int (\rho \frac{\partial}{\partial \rho} \mathbf{M}) \cdot d\mathbf{H}|_\rho + \Delta, \quad (90)$$

$$\Delta \equiv \int d\mathbf{H} \cdot \mathbf{M}|_\rho - \int d\mathbf{H} \cdot \mathbf{M}|_\mu \quad (91)$$

where  $\Delta$  is negligible only for quasi-equilibrium,  $\mu = \mu^{\text{qe}}$ , see Eq (68).

There are in fact two basic problems with the notion of a zero-field pressure. First, there are simply no universally field-independent quantities: Choosing one set of independent thermodynamic variables, the dependent ones are field-dependent. Second, while temperature and chemical potential are well-defined in the presence of field, cf Eq (37), the pressure is not. The usual notion of pressure,  $P \equiv -\partial \int u d^3r / \partial V$ , is ill-defined, because ponderable systems in the presence of field are anisotropic, and the energy depends not only on volume changes,  $\delta V$ , as implied by this definition, but also on variations of the shape, cf section 3.1. And the appropriate quantity to deal with is the Maxwell stress.

In isotropic liquids, the pressure is directly measurable, and it encompasses many concepts that we find convenient, even intuitive: as the surface force density, as the momentum current, and as a quantity that is continuous across interfaces. Hence there is widespread reluctance to abandon the pressure at finite fields. Unfortunately, though there are numerous ways to generalize the pressure that will preserve some of these properties, none covers all. So one may either define many different pressures – an approach [16] we eschew as it requires great care and tend to confuse – or face up to the Maxwell stress, as we do here.

There is a second pitfall worthy of attention. In dilute systems, the magnetization is usually proportional to the density, or  $M = \rho(\partial M / \partial \rho)$ . Inserting this into Eqs (87) and (88) respectively, we find

$$-G = P(\rho) + \frac{1}{2}(H^2 - M^2), \quad -G = P(\rho) + \frac{1}{2}H^2, \quad (92)$$

a clear contradiction. It arises because  $M = \rho(\partial M / \partial \rho)$  at given  $B$  or  $H$  are mathematically inequivalent. Physically, when we assume  $M \sim \rho$ , this is meant as an approximation for dilute systems, implying that higher order terms  $\sim \rho^2, \rho^3 \dots$  may be neglected. But consistency dictates that we must neglect all higher order terms, including  $M^2 \sim \rho^2$ . This implies that  $M$  is also to be treated as a small quantity,  $M \ll H$  and  $\chi^{\text{m}} \ll 1$ . Then the difference of  $\frac{1}{2}M^2$  is quadratically small, and the contradiction evaporates. Inserting the second of Eq (92) in  $\Pi_{ik} = -[G\delta_{ik} + H_i B_k]$ , we find a popular form for the Maxwell stress tensor,  $\Pi_{ik} = (P + \frac{1}{2}H^2)\delta_{ik} - H_i B_k$ . Clearly, it is to be taken with a large grain of salt, as it is valid only for dilute systems, and vanishing magnetizations,  $M \ll H$ .

## Different Kelvin Forces

Although we know that the bulk force density,  $\mathbf{f}^{\text{bulk}} = -\nabla_j \Pi_{ij} = -s\nabla T - \rho\nabla\mu$ , is a quantity that vanishes quickly, we may nevertheless (taking the temperature as constant), divide the term  $\rho\nabla\mu$  into the gradient of the zero-field pressure and a field-induced ‘‘ponderomotive force’’  $\mathbf{f}^{\text{P}}$ . The fact that their sum vanishes is then accepted as force equilibrium. Employing  $\rho, T$  as variables, we write

$$\mathbf{f}^{\text{bulk}} = -\rho\nabla[\mu(0) + \mu_{\text{em}}] = -\nabla P(\rho) + \mathbf{f}^{\text{P}}, \quad (93)$$

$$-\mathbf{f}^{\text{P}}/\rho \equiv \nabla[\partial F_{\text{em}}/\partial \rho]_B = \nabla[\partial \tilde{F}_{\text{em}}/\partial \rho]_H. \quad (94)$$

Because  $\nabla F_{\text{em}} = (\partial F_{\text{em}}/\partial \rho)\nabla\rho + H_i \nabla B_i$ , we may also write  $\mathbf{f}^{\text{P}}$  as  $\nabla[F_{\text{em}} - \rho\partial F_{\text{em}}/\partial \rho] - H_i \nabla B_i$ , or similarly, as  $\nabla[\tilde{F}_{\text{em}} - \rho\partial \tilde{F}_{\text{em}}/\partial \rho] + B_i \nabla H_i$ , implying

$$\mathbf{f}^P = M_i \nabla H_i + \nabla \int (\rho \frac{\partial}{\partial \rho} - 1) M_i dH_i, \quad (95)$$

$$= M_i \nabla B_i + \nabla \int (\rho \frac{\partial}{\partial \rho} - 1) M_i dB_i, \quad (96)$$

which are equivalent expressions for the Helmholtz force. For LCR,  $M = \chi^m H = \chi^m B / (1 + \chi^m)$ , both reduce to  $\mathbf{f}^P = \nabla (\frac{1}{2} H^2 \rho_\alpha \partial \chi^m / \partial \rho_\alpha) - \frac{1}{2} H^2 \nabla \chi^m$ . Assuming  $\rho \partial M / \partial \rho = M$ , for either given  $H$  or  $B$ , the respective Kelvin force is

$$\mathbf{f}^P = M_i \nabla H_i, \quad \mathbf{f}^P = M_i \nabla B_i. \quad (97)$$

Again, as discussed below Eq (92),  $\rho \partial M / \partial \rho \approx M$  is a physically sensible approximation only for  $M \ll H, B$ . But then of course  $M_i \nabla H_i \approx M_i \nabla B_i$ .

This point is not widely appreciated. Many authors take  $M_i \nabla H_i$  to be valid also for  $M \approx H$ , while the expression  $M_i \nabla B_i$  is usually spurned for no good reasons [29, 30]. Similarly,  $P_i \nabla E_i$  is preferred over  $P_i \nabla D_i$ . (One source of confusion may be Eq (79), which seemingly states that the bulk force density is  $M_i \nabla H_i$ , irrespective how large  $M_i$  is in comparison to  $H_i$ . But as discussed there, this is a seriously flawed interpretation.)

## The Magnetic Bernoulli Equation

The magnetic Bernoulli equation (MBE) by Rosensweig [16],

$$\rho g z + P(\rho) - \rho \frac{\partial}{\partial \rho} \int M_i dH_i = \text{constant}, \quad (98)$$

is a useful, extensively employed relation. It is contained in Eq (63), or  $g \int \rho(0) dz - G(0) = K$ . The connection is given by Eqs (89, 90), showing

$$g \int \rho(0) dz + P(\rho) - \int (\rho \frac{\partial}{\partial \rho} M_i) dH_i|_\rho + \Delta = K. \quad (99)$$

For quasi-equilibrium,  $\rho(0) \approx \rho$  is a constant, and  $\Delta \approx 0$ , see Eq (91). So the equation's left side reduces to the MBE. For true equilibrium, MBE needs to be taken as given by Eq (99). [The velocity dependent terms in the original MBE are not included here, because considerations of mass currents in ferrofluids need to include viscosities. Besides, some of the velocity dependent terms in the stress tensor are missing in [16], cf Eq (158).]

This section ends here. No summary is given, though the reader is advised to revisit the introduction, which should now provide a rather clear overview of all the derived results.

## 4 Static Experiments

Four experiments are collected here which either have been, or are well suited for being, carried out in ferrofluids. They serve the purpose of illustrating the expressions derived above, for both the bulk, and the surface, force density.

### 4.1 Field Induced Variations in Densities

Density variation of a magnetizable liquid in the presence of field inhomogeneity is referred to as electro- or magnetostriction. It is frequently calculated with the Kelvin force,  $M_i \nabla H_i$ , assuming it balances the gradient of the zero-field pressure, arising from a density variation. As discussed above, this force expression is not valid for large magnetization. Besides, it is quite easy to use the generally valid equilibrium conditions (60) instead. For a one-component liquid, taking the chemical potential  $\mu$  as a function of  $\rho, T, H$ , we have  $\nabla \mu = [\partial \mu / \partial \rho] \nabla \rho + [\partial \mu / \partial H] \nabla H = -\nabla \phi$ . With  $\partial \mu / \partial \rho = 1 / (\rho^2 \kappa_T)$  and  $\partial \mu / \partial H = -\partial B / \partial \rho$ , this implies the density gradient

$$\nabla \rho = \rho^2 \kappa_T \left( \frac{\partial B}{\partial \rho} \Big|_H \nabla H - \nabla \phi \right). \quad (100)$$

This differential relation may be integrated to  $\Delta \rho = \rho^2 \kappa_T \Delta (\frac{1}{2} H^2 \partial \chi^m / \partial \rho - g z)$  for LCR, with  $\Delta$  denoting the difference of any quantity behind it at two points in the liquid, if  $\kappa_T$  and  $\rho$  may be taken as essentially constant (ie,  $\Delta \rho \ll \rho$ ).

In a two-component fluid, not much changes for quasi-equilibrium, but circumstances are quite different in true equilibrium. Although the calculation remains formally unchanged, the much larger  $\kappa_{\text{os}}$  replaces  $\kappa_T$  in Eq (100).

To our knowledge, field-induced density variation has only been measured in one-component liquids [31], not in any two-component ones. This is unfortunate, because the variation of solute or particle density is so much more pronounced. To estimate its magnitude in ferrofluids, we write the linearized version of Eq (100) as  $\Delta\rho_1/\rho_1 = \kappa_{\text{os}}(\rho_1\partial\chi^{\text{m}}/\partial\rho_1)\Delta(\mu_0^{\text{m}}\frac{1}{2}\hat{H}^2)$ . Approximating  $(\rho_1\partial\chi^{\text{m}}/\partial\rho_1) \approx 1$ ,  $\kappa_{\text{os}} \approx 10^{-3}/\text{Pa}$ , we find  $\Delta\rho_1/\rho_1 \approx 0.1$  for  $\hat{B} = 10^{-2}\text{T}$ . And with Eq (55), a similar size effect in the total density,  $\Delta\rho/\rho \approx 0.1$ . Contrast this with the tiny change in the total density,  $\Delta\rho/\rho = 5 \cdot 10^{-8}$  at the same field – a result of the small compressibility,  $\kappa_T = 5 \cdot 10^{-10}/\text{Pa}$ . [The value for  $\kappa_{\text{os}}$  is obtained by considering a ferrofluid with 10% of its volume occupied by magnetic particles of the radius  $r = 10\text{nm}$ , so the particle density is  $n_1 = 0.1/(4\pi r^3/3)$ . Assuming ideal gas behavior, the inverse osmotic compressibility  $\kappa_{\text{os}}^{-1}$  is equal to the osmotic pressure,  $P_{\text{os}} = n_1 k_B T$ , so  $\kappa_{\text{os}} = 10^{-3}/\text{Pa}$  if  $T = 300\text{K}$ .]

## 4.2 Current Carrying Vertical Wire

We consider a vertical wire that goes through a dish filled with ferrofluid. Feeding the wire with an electric current  $J$  will drag the ferrofluid toward the wire (at  $r = 0$  and along  $e_z$ , in cylindrical coordinates). The ferrofluid surface is given by  $z(r)$ , with  $z(r \rightarrow \infty) \equiv 0$ . We find  $K = P_{\text{atm}}$  evaluating the boundary condition Eq (73) for  $z = 0$ , because the magnetization vanishes with the field, and the curvature radii diverge. The boundary condition at  $z(r)$ , with  $M_n \equiv 0$ , yields

$$\int M_i dH_i|_{\mu} = \alpha(R_1^{-1} + R_2^{-1}) + g \int \rho(0) dz, \quad (101)$$

a display of force balance between gravitation, magnetic surface force and surface tension. Neglecting  $\alpha$  and assuming quasi-equilibrium,  $\rho - \rho(0) \ll \rho(0)$ ,  $\int M_i dH_i|_{\mu} \approx \int M_i dH_i|_{\rho}$ , see Eq (68), this equation is the same as the associated result in [16]. Especially for LCR,  $M_i = \chi^{\text{m}} H_i$ , the left side reduces to  $\frac{1}{2} \chi^{\text{m}} H^2$ , with  $H = J/(2\pi r)$ . And we have a hyperbolic profile of the interface,  $8\pi^2 \rho g z = J^2 \chi^{\text{m}} / r^2$ .

The effect of  $\alpha$  is more important for weak currents,  $J$  small. It may be neglected in any case for  $z \rightarrow 0$ , where both curvature radii are large enough to be ignored. For  $z$  large and  $r$  small, one curvature radius is  $r$ , and the other  $\infty$ . So this part of the ferrofluid column is accounted for by  $g \int \rho(0) dz + \alpha/r = J^2 \chi^{\text{m}} / (8\pi^2 r^2)$ , with the term  $\sim r^{-2}$  being asymptotically ( $r \rightarrow 0$ ) the dominant one. In between, where the actual bend from the horizontal to the vertical takes place, both curvature radii (of different signs) are finite and should be included.

## 4.3 Hydrostatics in the Presence of Fields

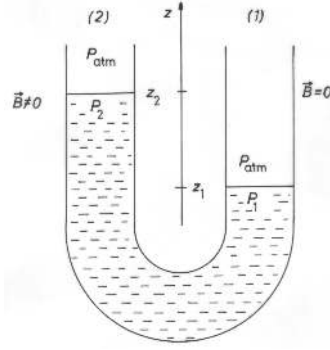
In a system of two connected tubes, with only the second subject to a magnetic field, we expect the ferrofluid column to be higher in this tube, as ferrofluid is attracted to the region of stronger fields, see Fig 4. To calculate the level difference, we employ the boundary condition Eq (73) for the (flat) liquid-air interface in both tubes. Since the field vanishes in the first, the boundary condition states  $P_{\text{atm}} = K - g \int_0^{z_1} \rho(0) dz$ . Inserting this into the boundary condition for tube 2, we obtain

$$g \int_{z_1}^{z_2} \rho(0) dz = \int M_i dH_i|_{\mu} + \frac{1}{2} M_n^2, \quad (102)$$

again a display of force balance. If the field is predominantly tangential ( $H_n, B_n \approx 0$ ) or normal ( $H_t, B_t \approx 0$ ) to the liquid surface, the right side is respectively given as  $\int M_i dH_i$  and  $\int M_i dB_i$ , or  $\frac{1}{2} H^2 \chi^{\text{m}}$  and  $\frac{1}{2} H^2 \chi^{\text{m}} (1 + \chi^{\text{m}})$  for LCR.

Next we consider the quantity that a pressure gauge measures in a ferrofluid. Characterized by a stress tensor, field-exposed ferrofluids do not possess a unique pressure, yet a pressure gauge will still give some reading, and the question is what this is. Think of the gauge as an enclosed volume of air, at the pressure  $P_{\text{atm}}$ , see Fig 5. One side of



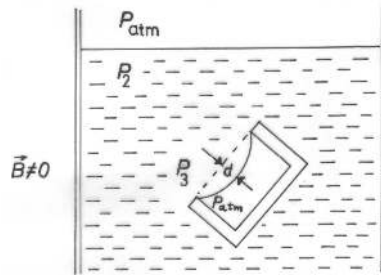


**Fig. 4.** A U-tube filled with ferrofluid, with only the left arm exposed to a field.

this volume is an elastic membrane, which is displaced if the external stress tensor deviates from the internal one. A finite displacement  $d$  stores up the elastic energy  $kd^2/2$  per unit area of the membrane. (Take the membrane to be stiff, ie  $k$  large and  $d$  small, then we need not worry about the pressure change inside.) The elastic energy implies a force density  $kd$ , rendering the boundary condition across the membrane as  $\Delta\Pi_{nn} = kd$ , or  $K - P_{\text{atm}} + \int M_i dH_i|_{\mu} + \frac{1}{2}M_n^2 - g \int \rho(0)dz = kd$ , see Eq (73). ( $d$  is taken to be positive when the membrane protrudes into the gauge.) We have  $d = 0$  in the atmosphere down to the liquid surface of tube 2, and also just below the surface – take this as point 2. Moving further down the liquid column, to an arbitrary point 3,  $d$  becomes finite to maintain force equilibrium. Employing the above boundary condition for both point 2 and 3, and subtracting one from the other, we find

$$kd = \Delta(\int M_i dH_i|_{\mu} + \frac{1}{2}M_n^2 - g \int \rho(0)dz), \quad (103)$$

with  $\Delta A \equiv A(3) - A(2)$ . Note  $\Delta M_n^2 \equiv M_n^2(3) - M_n^2(2)$ , where  $M_n(3)$  is the magnetization at point 3 normal to the pressure gauge membrane, and  $M_n(2)$  the magnetization at point 2 normal to the liquid surface – both components are not necessarily parallel. If the field is uniform, if the membrane of the gauge is parallel to the liquid surface, and if quasi-equilibrium holds, Eq (103) reduce to the zero-field hydrostatic relation,  $g\rho(z_2 - z_3) = kd$ ; otherwise, field contributions abound – even if the pressure gauge is simply rotated at point 3 in the presence of a uniform field. (All fields are the actual ones, distorted by the gauge's presence.)

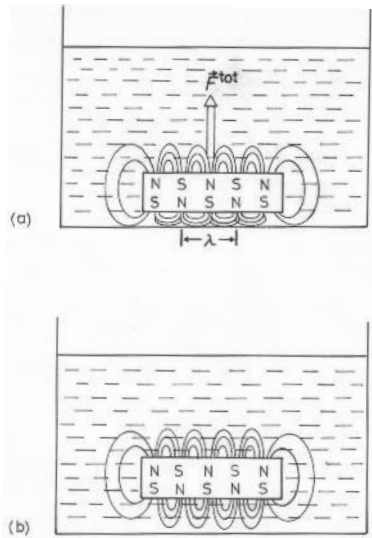


**Fig. 5.** A vessel filled with ferrofluid is subject to a field. The pressure inside the ferrofluid, as measured by the membrane displacement of a pressure gauge, changes with height, orientation and the field strength.

#### 4.4 Magnetic O-Rings and Scrap Separation

In this section, we address the physics of some technical applications: magnetic O-rings, self-levitation and scrap separation. Consider scrap separation first. An inhomogeneous magnetic field which becomes weaker with increasing height may lift non-magnetizable bodies submerged in ferrofluids off the ground, and hold them at specific heights which depend on the shape and density of the bodies. Balancing the gravitational and magnetic force,  $\mathcal{F}^{\text{mag}} + \mathcal{F}^{\text{grav}} = 0$ , we have  $\mathcal{F}^{\text{grav}}$  given by Eq (77), and  $\mathcal{F}^{\text{mag}}$  by (80).

Because scrap separation is an equilibrium phenomenon, we may also consider energy instead of force. Take first the fact that ferrofluid is attracted to regions of higher magnetic field. If the field  $B$  is given, the field energy is  $B^2/2$  in air, and  $B^2/(2\bar{\mu}^m)$  in ferrofluids. With  $\bar{\mu}^m > 1$ , the second expression is always smaller. So given the choice, a volume element of ferrofluid will occupy the region with the highest possible field. Conversely, a non-magnetizable object submerged in ferrofluid will tend to occupy the region of lowest field strength. If a difference in height is involved, all these happen only as long as the gain in field energy is larger than the loss in gravitational energy. (If instead  $H$  is given,  $\bar{F}_{\text{em}} \equiv F_{\text{em}} - HB$  is larger in vacuum than in the ferrofluid:  $-H^2/2 > -\bar{\mu}^m H^2/2$ .) Similarly,



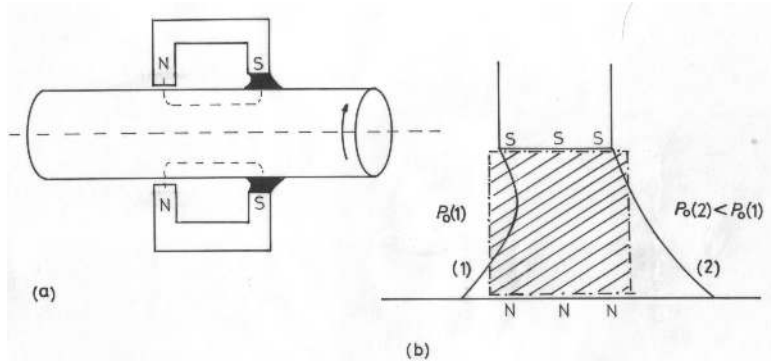
**Fig. 6.** Self-levitation in ferrofluid of a magnetizing body.

a permanently magnetized body submerged in ferrofluid tends to collect as much liquid in the region of its field as possible – even at the price of levitating itself off the bottom, a phenomenon that is sometime referred to as “self-levitation” [16]. if the magnetic body consists of a periodic array of north and south poles, with periodicity  $\lambda$ , the field extends one or two  $\lambda$  into the ferrofluid. Levitated approximately that far from the bottom, the body will usually have reached its equilibrium position, as no further gain in field energy may be achieved levitating itself yet higher, see Fig 6.

Magnetic O-rings may be found in most computer hard disk drives and are perhaps the most widely deployed ferrofluid device. In these, some ferrofluid is positioned as an O-ring between a highly permeable rotating shaft and the pole of a permanent magnet, see Fig 7. Serving as a pressure seal, it enables the rotary component to work in vacuum. Note that we are dealing with a metastable state here, as it is always energetically more favorable to remove the ferrofluid seal first, have the pressure equalized, and then return the seal to its original place at the poles. So the quantity of interest is the lowest energy barrier that must be overcome, which in any realistic problems is notoriously difficult to find. The correct force balance, on the other hand, is

$$A\Delta P_{\text{atm}} = \Delta \int \left( \frac{1}{2} M_n^2 + \int M_k dH_k |_{\mu} \right) dA, \quad (104)$$

between the difference in the magnetic surface force, Eq (75), and the difference in external pressure. [Clearly, the appropriate replacement is given by  $\mu \rightarrow \mu^{te}$ , cf Eq (59).] The field is strongest in the middle of the O-ring and decays towards both ends. If  $\Delta P_{\text{atm}}$  were zero, the force  $\mathcal{F}^{\text{mag}}$  would be the same on both surfaces, and the ferrofluid stays in the middle of the O-ring. Increasing the pressure on the left (surface 1) pushes the ferrofluid towards right, such that surface 1 is in the region of higher, and surface 2 is in the region of weaker, fields. Equilibrium is achieved when the difference in  $\mathcal{F}^{\text{mag}}$  balances  $\Delta P_{\text{atm}}$ . The strongest pressure difference maintainable is when one surface is at the field maximum and the other is field-free. Assuming for simplicity that the magnetic field is predominantly tangential, and that LCR holds, we have  $\Delta P = \frac{1}{2} \chi^m H^2$ . With  $\chi^m \approx 1$ ,  $H^2 = \mu_0^m \hat{H}^2$ , and taking  $\hat{H}$  as of order  $10^5$  A/m, this pressure difference is about  $10^5$  N/m<sup>2</sup>, approximately the atmospheric pressure.



**Fig. 7.** Magnetic pressure seal: (a) principle and (b) enlarged view of the ferrofluid plug.

## 5 Electrostatics Including Dissipation

Turning now to dynamics and off-equilibrium phenomena, we first note that there are two types of theories: the high-resolution theory for a low density system, with at most one particle per infinitesimal volume element (or per grain, as in photographs); and the low resolution theory for a high density system, with many particles per grain. The microscopic Maxwell equations and the Newtonian equation of motion (including the Lorentz force) belong to the first type. There are no hidden charges, polarization or magnetization here, and we know the whereabouts of every single particle. The second type includes the macroscopic Maxwell equations, or any thermodynamic and hydrodynamic theories. The problems about dissipation and the coarse-grained electromagnetic force arise here.

We shall consider three systems with increasing densities, to be accounted for by theories of decreasing resolution and growing complexities: The first system (section 5.1) is a dilute gas of charged particles, well accounted for by the Newtonian equation of motion and the microscopic Maxwell equations. The next system (section 5.2) is a slightly dissociated liquid of particles possessing negligible electric and magnetic dipole moments. So there are many neutral particles, but at most one charge carrier, per grain. The appropriate theory here is of a mixed type, a combination of the hydrodynamic theory and the microscopic Maxwell equations. The first accounts for all particles, the latter for the spatially slowly varying field, produced by the charge carriers that are few and far apart. Finally, in section 5.3, we consider a dense system possessing dipole moments and hidden sources, which needs to be accounted for by the genuinely low-resolution, hydrodynamic-type Maxwell theory that is our goal to derive and consider in details. The advantage of

this successive approach is, finding the more complicated expressions in the dense medium is rendered very much simpler and transparent by the considerations of the previous two dilute systems – but we do not, even loosely, derive one from the other.

### 5.1 Rarefied Plasmas

The microscopic Maxwell equations (13, 14) account for the time evolution of the finely resolved fields  $\mathbf{e}$  and  $\mathbf{b}$  for given sources, the electric charge density and its current; while the feedback of the fields on the motion of the sources is given by the Newtonian equation of motion,

$$m^\alpha \dot{\mathbf{v}}^\alpha = q^\alpha (\mathbf{e} + \mathbf{v}^\alpha \times \mathbf{b}/c), \quad (105)$$

one for each particle  $\alpha$ . Eqs (13, 14, 105) represent a closed theory, frequently rather too detailed but conceptually simple. However, it does contain a notational inconsistency: The Maxwell equations are an Euler type theory, accounting for the time evolution of fields at a given point in space, while the Newtonian equation is of the Lagrange type, which concentrates on a given particle. (So the term  $\mathbf{j}_e = \rho_e \mathbf{v}$  in Eq (14) denotes the electric current at a space point, while  $\mathbf{v}^\alpha$  in Eq (105) is the velocity of particle  $\alpha$ .) As only the Euler formulation lends itself to a reduction of the resolution, we shall first find the Euler version of Eq (105), with a resolution high enough that each grain contains at most one particle. (This theory is meant as a starting point, to clarify a few concepts important for the more complex theories of the following chapters. So we shall simply discard the possibility that even in a rarefied gas, two particles will occasionally come close to each other.)

#### The Eulerian Newtonian Equation

Taking the volume of the grain as  $V_G$ , we may identify its velocity, mass and charge with that of the particle occupying it at a given instance, and take all three to be zero if there is no particle:  $\mathbf{v}^\alpha \rightarrow \mathbf{v}(\mathbf{r}, t)$ ,  $m^\alpha/V_G \rightarrow \rho(\mathbf{r}, t)$ ,  $q^\alpha/V_G \rightarrow \rho_e(\mathbf{r}, t)$ , defining three highly discontinuous fields. After some very moderate coarse-graining rendering these fields differentiable, the many Newtonian equations of motion reduce to one field equation,

$$\rho \frac{d}{dt} \mathbf{v} \equiv \rho [\dot{\mathbf{v}} + (\mathbf{v} \cdot \nabla) \mathbf{v}] = \rho_e (\mathbf{e} + \mathbf{v} \times \mathbf{b}/c). \quad (106)$$

It is now essential to include the continuity equation,

$$\dot{\rho} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad (107)$$

which in the Lagrange version is implicit, nearly incidentally contained in the fact that one does not lose any of the many equations (105). The energy and momentum density are respectively,

$$u^M = \rho(c^2 + v^2/2), \quad \mathbf{g}^M = \rho \mathbf{v}, \quad (108)$$

where the energy comprises of the rest energy and the (non-relativistic) kinetic energy. Employing Eqs (106, 107), we find

$$\dot{u}^M + \nabla \cdot \mathbf{Q}^M = \mathbf{j}_e \cdot \mathbf{e}, \quad (109)$$

$$\dot{g}_i^M + \nabla_k \Pi_{ik}^M = (\rho_e \mathbf{e} + \mathbf{j}_e \times \mathbf{b}/c)_i, \quad (110)$$

$$\mathbf{Q}^M = u^M \mathbf{v}, \quad \Pi_{ik}^M = g_i^M v_k. \quad (111)$$

#### The Field Contributions

The field contribution to the energy and momentum density are

$$u^F = \frac{1}{2}(e^2 + b^2), \quad \mathbf{g}^F = \mathbf{e} \times \mathbf{b}/c. \quad (112)$$

From the Maxwell equations (13, 14) we deduce

$$\dot{u}^F + \nabla \cdot \mathbf{Q}^F = -\mathbf{j}_e \cdot \mathbf{e}, \quad \mathbf{Q}^F = c \mathbf{e} \times \mathbf{b}, \quad (113)$$

$$\dot{g}_i^F + \nabla_k \Pi_{ik}^F = -(\rho_e \mathbf{e} + \mathbf{j}_e \times \mathbf{b}/c)_i, \quad (114)$$

$$\Pi_{ik}^F = (e^2 + b^2 - u^F) \delta_{ik} - e_i e_k - b_i b_k. \quad (115)$$

Note the relationship  $\mathbf{g}^F = \mathbf{Q}^F/c^2$ . This is far from accidental and derives from the symmetry of the relativistic energy-momentum 4-tensor,  $\Pi_{\alpha\beta}^F = \Pi_{\beta\alpha}^F$ , because  $c g_k^F = \Pi_{k4}^F$ ,  $Q_k^F/c = \Pi_{4k}^F$ . Less formally,  $\mathbf{g}^F = \mathbf{Q}^F/c^2$  may also be seen as field angular momentum conservation for  $\rho_e \rightarrow 0$ . The angular momentum density  $\ell^F \equiv \mathbf{r} \times \mathbf{g}^F$  is a locally conserved quantity in neutral systems. Rewriting Eq (114) (with  $\rho_e \rightarrow 0$ ) as  $\partial \ell_i^F / \partial t + \nabla_m (\varepsilon_{ijk} r_j \Pi_{km}^F) = \varepsilon_{ijk} \Pi_{kj}^F$ , we observe that the angular momentum  $\ell_i^F$  satisfies a continuity equation only if the stress tensor is symmetric. Although this argument seems to require merely the symmetry of the momentum 3-tensor,  $\Pi_{ik}^F = \Pi_{ki}^F$ , a nonvanishing  $\Pi_{k4}^F - \Pi_{4k}^F$  in one inertial system will foul up the symmetry of the 3-tensor in other systems, as the antisymmetric parts of any 4-tensors mix in a Lorentz transformation. Yet angular momentum is conserved in every inertial system.

Since this reasoning is so general, it also holds for the material part,  $\mathbf{g}^M = \mathbf{Q}^M/c^2$ . Hence the expression for the momentum density is in fact  $\mathbf{g}^M = \rho \mathbf{v} [1 + v^2/(2c^2)]$ , cf Eqs (108, 111) – though we are quite justified to neglect the second term in the nonrelativistic limit. Later, when we have no prior knowledge of the form of the momentum density, we shall deduce it from that of the energy flux, as angular momentum is also conserved in dense systems.

We register the fact that while the expression for the energy density  $u^F = \frac{1}{2}(e^2 + b^2)$  is a genuine input – independent of and in addition to, the Maxwell equations, the formula  $\mathbf{g}^F = \mathbf{e} \times \mathbf{b}/c$  is not, since  $\mathbf{Q}^F$  is given once  $u^F$  is. Moreover, since Eq (114) then follows from the Maxwell equations, and because total momentum is conserved, the Lorentz force is a result as well.

## Energy and Momentum Conservation

The preceding two sections allow the simple and noteworthy conclusion that our starting equations imply local conservation of total energy, momentum and angular momentum in the presence of charge,  $\rho_e \neq 0$ . Taking  $u \equiv u^F + u^M$  and  $\mathbf{g} \equiv \mathbf{g}^F + \mathbf{g}^M$ , we find

$$\dot{u} + \nabla \cdot \mathbf{Q} = 0, \quad \dot{g}_i + \nabla_k \Pi_{ik} = 0, \quad (116)$$

$$\Pi_{ik} = \Pi_{ki} = \Pi_{ik}^F + \Pi_{ik}^M, \quad \mathbf{Q} = \mathbf{Q}^F + \mathbf{Q}^M, \quad (117)$$

$$\mathbf{Q}/c^2 = \mathbf{g} \approx \rho \mathbf{v} + \mathbf{e} \times \mathbf{b}/c. \quad (118)$$

These results have been collected here because local conservation of these quantities is always true, independent of the above derivation tailored to a dilute and finely resolved system. So we may use them as input next.

## 5.2 Weakly Dissociated Liquids

Now we consider a dense macroscopic system in its hydrodynamic regime: To the above gas of dilute charge carriers we add a dense system of neutral particles with vanishing electric and magnetic dipole moments. This is still a comparatively simple system, as the highly resolving, vacuum Maxwell equations (13, 14) remain valid – and with them all the equations under the title “The Field Contributions,” including the expressions on field energy and field momentum. The other equations concerning the material contributions, under the title “The Eulerian Newtonian Equation”, must be modified. These are now accounted for by three smooth and slowly varying hydrodynamic variables: the coarse-grained densities of mass, energy and momentum  $\langle \rho \rangle$ ,  $\langle u^M \rangle$ ,  $\langle \mathbf{g}^M \rangle$ , while the entropy density  $s$ , being a function of these variables, is not independent. (The coarse-graining brackets are dropped below to keep the notation simple.)

## The Material Contributions

We first consider the hydrodynamic theory of a neutral, isotropic liquid. It consists of continuity equations for the densities of mass, energy, momentum:  $\rho, u^M, \mathbf{g}^M$ , and a balance equation for the entropy  $s$ , with a source term  $R^D/T$ ,

$$\dot{\rho} + \nabla \cdot \mathbf{j} = 0, \quad \dot{s} + \nabla \cdot \mathbf{f} = R^D/T, \quad (119)$$

$$\dot{u}^M + \nabla \cdot \mathbf{Q}^M = 0, \quad \dot{g}_i^M + \nabla_k \Pi_{ik}^M = 0. \quad (120)$$

The fluxes  $\mathbf{j}, \mathbf{f}, \mathbf{Q}^M, \Pi_{ik}^M$  and the entropy production  $R^D$  are not yet specified and need to be determined. As we retrace how they are derived [32], we learn to use the same method to set up the equations for our third system, to derive the hydrodynamic Maxwell theory.

The present theory is in two points fundamentally different from the previous one. First, dissipative terms appear in the equations of motion, breaking their time-reversal symmetry. These will be marked by a superscript  $D$ . Second is our ignorance of the explicit form of  $u^M$ , the material energy. Nevertheless,  $\mathbf{g}^M = \rho \mathbf{v}$  remains an excellent approximation, because the energy flux  $\mathbf{Q}^M$  is still dominated by the term  $c^2 \rho \mathbf{v}$ , accounting for the transport of rest energy, cf Eqs (108, 111). In nonrelativistic physics, it is not customary to include the rest energy in  $u^M$ . This represents a shift to a different set of conserved quantities: from  $u^M, \rho$  to  $\bar{u}^M = u^M - \rho c^2, \rho$ , implying  $\bar{\mathbf{Q}}^M = \mathbf{Q}^M - c^2 \rho \mathbf{v}$ . This changes the link between the momentum density and the energy flux to  $\mathbf{g}^M = \bar{\mathbf{Q}}^M/c^2 + \rho \mathbf{v} \approx \rho \mathbf{v}$ . We follow this convention, but drop the bar, from here on.

To compensate for our ignorance of  $u^M$ , we resort to thermodynamics. Assuming that the energy is a function of  $s$  and  $\rho$  in the rest frame of the liquid (denoted by the subscript  $0$ ), we write

$$du_0^M = \frac{\partial u_0^M}{\partial s} ds + \frac{\partial u_0^M}{\partial \rho} d\rho \equiv T ds + \mu_0 d\rho. \quad (121)$$

In a liquid with a finite velocity  $\mathbf{v}$ , we have  $u^M = u_0^M + (\mathbf{g}^M)^2/2\rho$ . Because  $d[(\mathbf{g}^M)^2/2\rho] = \mathbf{v} \cdot d\mathbf{g}^M - \frac{1}{2}v^2 d\rho$ , Eq (121) is generalized to

$$du^M = T ds + \mu d\rho + \mathbf{v} \cdot d\mathbf{g}^M, \quad (122)$$

where  $\mu = \mu_0 - \frac{1}{2}v^2$  is the lab-frame chemical potential.

Taking the time derivative of Eq (122),  $\dot{u}^M = T\dot{s} + \mu\dot{\rho} + \mathbf{v} \cdot \dot{\mathbf{g}}^M$ , we have a relation which Eqs (119, 120) must identically satisfy – irrespective how  $u^M$  (and  $T, \mu, \mathbf{v}$ ) depend on  $s, \rho, \mathbf{g}^M$ . This is therefore a strong constraint, enough to unambiguously determine all the fluxes. (This approach is sometimes referred to as the “standard procedure,” see [17, 32]. It was first applied to electromagnetism in [2, 3] to derive the dissipative terms.) A remarkable result is that the fluxes are given in terms only of the variables and the conjugate variables appearing in Eq (122). Without an explicit expression for  $u^M$ , the equations clearly must be written in these general quantities, and we may take this observation as an indication that hydrodynamic theories contain only conservation laws and thermodynamics as input – the reason for its general validity.

Defined as the energy change if the volume changes at constant entropy and mass, the pressure  $P$  is related to the variables and conjugate variables of  $u^M$  via the Duhem-Gibbs (or Euler) relation, cf Eq (35),

$$\begin{aligned} P &\equiv -\partial(\int u_0^M d^3r)/\partial V = -u_0^M + Ts + \mu_0\rho \\ &= -u^M + Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g}^M. \end{aligned} \quad (123)$$

Specifying that one term in the momentum flux of Eq (120) is the pressure,  $\Pi_{ij}^M = P\delta_{ij} + \pi_{ij}$  (there is no loss of generality, as  $\pi_{ij}$  must now be determined), we combine Eqs (122, 123) to obtain  $\nabla P = s\nabla T + \rho\nabla\mu + g_k^M\nabla v_k$ , and write  $\dot{u}^M = T\dot{s} + \mu\dot{\rho} + \mathbf{v} \cdot \dot{\mathbf{g}}^M$  as

$$\begin{aligned} \nabla \cdot \mathbf{Q}^M &= \nabla_i(\mu j_i + T f_i + v_k \pi_{ik}) - (j_i - \rho v_i)\nabla_i \mu \\ &\quad - R^D - (f_i - s v_i)\nabla_i T - (\pi_{ik} - v_k g_i)v_{ik}, \end{aligned} \quad (124)$$

where  $v_{ik} \equiv \frac{1}{2}(\nabla_i v_k + \nabla_k v_i)$ . Postponing the question of uniqueness for a moment, we identify  $Q_i^M = \mu j_i + T f_i + v_k \pi_{ik}$ , set  $f_i^D \equiv s v_i - f_i$ ,  $\Pi_{ik}^D \equiv v_k g_i - \pi_{ik}$ , and  $j_i - \rho v_i = 0$ ,

to obtain  $R^D = f_i^D \nabla_i T + \Pi_{ik}^D v_{ik}$ . (The dissipative mass current,  $j_i^D \equiv j_i - \rho v_i$ , is a relativistically small quantity that we shall neglect, see [33] and the footnote in section 5.2.) Identifying  $f_i^D$ ,  $\Pi_{ik}^D$  as the dissipative fluxes,  $\nabla_i T$ ,  $v_{ik}$  as the thermodynamic forces, we follow the Onsager prescription to take them as pairwise proportional [34], yielding heat conduction and the viscous stress,

$$\mathbf{f}_i^D = \kappa \nabla T, \quad \Pi_{ik}^D = 2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell} \delta_{ik}, \quad (125)$$

where  $v_{ij}^0 \equiv v_{ij} - \frac{1}{3} \delta_{ij} v_{\ell\ell}$ . Clearly, all fluxes have now been determined, especially  $f_i = s v_i - f_i^D$  and  $\Pi_{ik}^M = P \delta_{ik} + v_k g_i - \Pi_{ik}^D$ . Without the dissipative terms, the energy and momentum flux are  $Q_i^M = (u^M + P) v_i$ ,  $\Pi_{ik}^M = g_i^M v_k + P \delta_{ik}$ , and the only difference to Eqs (111) is the pressure  $P$ . This and the dissipative terms are the indicators for a dense and interacting system.

The question remains whether the conclusion we draw from Eq (124) is unique. The answer is yes, because all terms must be written such that they can be assigned to one of two groups. Either it is the divergence of something, then it belongs to the energy flux  $\mathbf{Q}^M$ ; or it vanishes in equilibrium, like  $\nabla_i T$ ,  $v_{ij}$ , then it is part of  $R^D$ . For instance, the term  $T \dot{s} = -T \nabla_i f_i + \dots$  must be written as  $f_i \nabla_i T - \nabla_i (T f_i)$ , with the first term contributing to  $R^D$ , and the second to  $\mathbf{Q}^M$ . All terms in Eq (124) clearly belong to one of the two groups, and there is no possibility to change them such that this feature is maintained.

## The Field-Dependent Contributions

Next, we consider finite electric charges and currents. Collecting all terms derived above, and adding the ones that become finite with  $\rho_e, \mathbf{j}_e$ , we have

$$\dot{\rho} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad \dot{s} + \nabla \cdot (s \mathbf{v} - \mathbf{f}^D) = R^D / T, \quad (126)$$

$$\dot{u}^M + \nabla_i [(u^M + P) v_i - T f_i^D - v_k \Pi_{ik}^D] = \mathbf{j}_e \cdot \mathbf{e}, \quad (127)$$

$$\dot{\mathbf{g}}_i^M + \nabla_k (P \delta_{ik} + g_i^M v_k - \Pi_{ik}^D) = (\rho_e \mathbf{e} + \mathbf{j}_e \times \mathbf{b}/c)_i, \quad (128)$$

$$R^D = f_i^D \nabla_i T + \Pi_{ik}^D v_{ik} + (\mathbf{j}_e - \rho_e \mathbf{v}) \cdot \mathbf{e}_0. \quad (129)$$

The right sides of Eqs (127, 128) have the given form because the source terms of Eqs (113, 114) remain unchanged, and summing up the respective right sides must yield nil, as in Eqs (116), such that total energy and momentum remain conserved. The third term in  $R^D$  of Eq (129) goes beyond the sum of microscopic Maxwell equations and the hydrodynamics of neutral fluids. It needs to be derived. The reason for this term is,  $\mathbf{j}_e$  is no longer given by  $\rho_e \mathbf{v}$  as in section 5.1, because  $\mathbf{v}$  is now the velocity of a volume element containing many particles. Given a finite electric field,  $\mathbf{e}_0 = \mathbf{e} + \mathbf{v} \times \mathbf{b}/c$ , in the rest frame of the volume element, electric charge carriers will move with respect to this volume. This is indeed what we obtain starting from Eq (129), identifying  $\mathbf{j}_e - \rho_e \mathbf{v}$  as the rest-frame current,  $\mathbf{e}_0$  as the thermodynamic force, and taking them as proportional,

$$(\mathbf{j}_e - \rho_e \mathbf{v})_i \equiv (\mathbf{j}_e^D)_i = \sigma_{ij} (\mathbf{e} + \mathbf{v} \times \mathbf{b}/c)_j, \quad (130)$$

where  $\sigma_{ij} = \sigma_{\parallel} b_i b_j + \sigma_{\perp} (\delta_{ij} - b_i b_j) + \sigma_H \varepsilon_{ijk} b_k$ , with  $\sigma_{\parallel}, \sigma_{\perp}$  being the conductivity along and perpendicular to  $\mathbf{b}$ , and  $\sigma_H$  accounting for the Hall effect.

Now, we derive the third term in  $R^D$ , Eq (129), by taking the temporal derivative of Eq (122),  $\dot{u}^M = T \dot{s} + \mu \dot{\rho} + \mathbf{v} \cdot \dot{\mathbf{g}}^M$ , finding  $\nabla_i Q_i^M = -R^D + \mathbf{e} \cdot \mathbf{j}_e - \mathbf{v} \cdot (\rho_e \mathbf{e} + \mathbf{j}_e \times \mathbf{b}/c) \dots$ , where the second term is from  $\dot{u}^M$ , the third from  $\dot{\mathbf{g}}^M$ , and the dots denote terms not containing  $\mathbf{e}$  or  $\mathbf{b}$ . Combing them as  $\mathbf{j}_e \cdot (\mathbf{e} - \mathbf{b} \times \mathbf{v}/c) - \mathbf{e} \cdot \rho_e \mathbf{v} = (\mathbf{j}_e - \rho_e \mathbf{v}) \cdot \mathbf{e}_0$ , because  $\mathbf{e} \cdot \mathbf{v} = \mathbf{e}_0 \cdot \mathbf{v}$ , we obtain  $\nabla_i Q_i^M = -R^D \dots + (\mathbf{j}_e - \rho_e \mathbf{v}) \cdot \mathbf{e}_0$ . Because  $\mathbf{e}_0 = 0$  in equilibrium, the last term is part of  $R^D$ , concluding the proof.

Inserting  $\nabla P = s \nabla T + \rho \nabla \mu + g_j^M \nabla v_j$  and the first of Eq (126) into (128), we obtain an equation that will prove useful for later comparison,

$$\begin{aligned} \rho \frac{d}{dt} v_i + s \nabla_i T + \rho \nabla_i \mu + g_k^M \nabla_i v_k \\ = \nabla_k \Pi_{ik}^D + (\rho_e \mathbf{e} + \mathbf{j}_e \times \mathbf{b}/c)_i. \end{aligned} \quad (131)$$

Adding the Maxwell equations (13, 14), the system of equation is closed and the description complete. With  $\rho_e$  given by  $\nabla \cdot \mathbf{e}$ , and  $u^M$  by Eq (122), the independent variables

are  $\rho, s, \mathbf{g}^M, \mathbf{e}, \mathbf{b}$ . Referred to as magneto-hydrodynamics, these are realistic equations, used for describing plasma at low frequencies [15]. Usually,  $\sigma_{\parallel}, \sigma_{\perp}$  are large, hence  $\rho_e, \mathbf{e}_0 = \mathbf{e} + \mathbf{v} \times \mathbf{b}/c$  negligibly small.

### The 2-field Theory

In the low-resolution theory of the next chapter, we shall be dealing with dense systems containing hidden charges and dipole moments. The obvious consequence is the appearance of four fields ( $\mathbf{E}, \mathbf{D}, \mathbf{B}, \mathbf{H}$ ) – replacing  $(\mathbf{e}, \mathbf{b})$  here. An equally important difference is the fact that (the possibility of an expansion notwithstanding) we no longer have a general expression for the field energy  $u^F$  – which was important for arriving at the right-side terms of Eqs (127, 128), especially the Lorentz force. In fact, it will not even be possible to separate the total, conserved energy density  $u$  into  $u^F$  and  $u^M$ , such that the former depends on the field variables alone, and the latter only on the material ones. In the simple example  $u = \tilde{u}(\rho, s) + \frac{1}{2}B^2/\bar{\mu}^m(\rho, s)$ , the temperature  $T = \partial u/\partial s + \frac{1}{2}B^2\partial(1/\bar{\mu}^m)/\partial s$  is a function of  $B$ , and  $H = B/\bar{\mu}^m$  depends on  $s$ . As a result, none of the variables or conjugate variables is unambiguously assigned to either field or material. (This is the reason why attempts to divide the total, conserved momentum density  $\mathbf{g}$  into  $\mathbf{g}^M$  and  $\mathbf{g}^F$  is such a futile exercise, see the discussion in [2].)

Fortunately, neither the Lorentz force nor a division of  $u$  and  $\mathbf{g}$  into material and field contributions are necessary for deriving the hydrodynamic Maxwell theory. As a first step, we shall convince ourselves that the present theory can be equally well written in the variables  $u \equiv u^F + u^M, \mathbf{g} \equiv \mathbf{g}^F + \mathbf{g}^M$ , such that the Lorentz force never appears. We do this by adding to the material equations the field contributions from 5.1, same as we did there, obtaining

$$\dot{u} + \nabla_i(Q_i^R - Q_i^D) = 0, \quad (132)$$

$$\dot{\mathbf{g}}_i + \nabla_k(\Pi_{ik}^R - \Pi_{ik}^D) = 0, \quad (133)$$

$$\mathbf{Q}^R \equiv (Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g}^M)\mathbf{v} + c\mathbf{e} \times \mathbf{b}, \quad (134)$$

$$\begin{aligned} \Pi_{ik}^R &\equiv g_i^M v_k - e_i e_k - b_i b_k + \delta_{ik} \times \\ &\quad (Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g}^M - u + \mathbf{e}^2 + \mathbf{b}^2). \end{aligned} \quad (135)$$

$$\mathbf{g} = \mathbf{g}^M + \mathbf{e} \times \mathbf{b}/c = \rho\mathbf{v} + \mathbf{Q}^R/c^2. \quad (136)$$

The dissipative terms,  $\Pi_{ik}^D$  and  $Q_i^D \equiv T f_i^D + v_k \Pi_{ik}^D$ , remain as given above. Note  $\mathbf{Q}^D$  does not contribute to the total momentum density,<sup>9</sup> see Eq (136). The expression for  $du$  is obtained by adding the first of Eqs (112) to Eq (122),

$$du = Tds + \mu d\rho + \mathbf{v} \cdot d\mathbf{g}^M + \mathbf{e} \cdot d\mathbf{e} + \mathbf{b} \cdot d\mathbf{b} \quad (137)$$

$$= Tds + \mu d\rho + \mathbf{v} \cdot d\mathbf{g} + \mathbf{e}_0 \cdot d\mathbf{e} + \mathbf{b}_0 \cdot d\mathbf{b}. \quad (138)$$

Eq (138) is algebraically identical to Eq (137), because  $\mathbf{v} \cdot d(\mathbf{g}^M - \mathbf{g})c = -\mathbf{v} \cdot d(\mathbf{e} \times \mathbf{b}) = -(\mathbf{v} \times \mathbf{e}) \cdot d\mathbf{b} + (\mathbf{v} \times \mathbf{b}) \cdot d\mathbf{e}$ , where  $\mathbf{e}_0 \equiv \mathbf{e} + (\mathbf{v}/c) \times \mathbf{b}$ ,  $\mathbf{b}_0 \equiv \mathbf{b} - (\mathbf{v}/c) \times \mathbf{e}$  are the respective rest frame fields.

Containing  $\rho, s, u, \mathbf{g}, \mathbf{e}, \mathbf{b}$  as variables, the new theory is given by the Maxwell equations (13, 14), the equations for mass and entropy (126), and all the displayed equations of this section. It is closed, complete and quite equivalent to the old one. We may refer to it as the “2-field” theory, and require that the genuinely low-resolution “4-field” theory reduces to it by setting  $\mathbf{E}^D, \mathbf{H}^D = 0$ , and

<sup>9</sup> This is a subtle point. The quick argument is, as a variable,  $\mathbf{g}$  must not contain terms of different time-inversion parity. The more complete argument is, the existence of a finite  $\mathbf{Q}^D$  is in fact the result of subtracting the rest energy from the total – without which the dissipative energy flux vanishes identically. As shown in [33], there is a connection between this and the vanishing of the dissipative mass current,  $\mathbf{j}^D$ : Taking the mass, energy and total energy conservation as  $\dot{\rho} + \nabla_i(\rho v_i + j^D) = 0, \dot{u} + \nabla_i(Q_i - Q_i^D) = 0, \dot{u}^{tot} + \nabla_i \tilde{Q}_i = 0$ , we have  $\dot{u}^{tot} = u + \rho c^2$  and  $\tilde{Q}_i = Q_i + \rho v_i c^2 = g_i c^2$ , with  $\tilde{Q}_i^D = 0$ , or  $j_i^D c^2 - Q_i^D = 0$ . Because  $j_i^D = Q_i^D/c^2$  is a relativistically small quantity, it is usually neglected.



$$\mathbf{E} = \mathbf{D} \rightarrow \mathbf{e}, \quad \mathbf{H} = \mathbf{B} \rightarrow \mathbf{b}, \quad (139)$$

implying also  $\mathbf{E}_0 = \mathbf{D}_0 \rightarrow \mathbf{e}_0, \mathbf{H}_0 = \mathbf{B}_0 \rightarrow \mathbf{b}_0$ .

### Radiation Damping

Let us briefly address the old problem of radiation damping, which has created considerable confusion in some textbooks, with the result that a good number of physicists take it to be an indication for the inconsistency of the Maxwell theory. The basic physics of radiation damping is in fact given by the equations derived in 5.2, stating conservation of total energy and momentum. So it is an easy conclusion to draw that if a charged body's acceleration generates radiation, it will lack exactly the energy and momentum that is being carried away by the radiation. This is all we need to know about radiation damping, and there is no reason whatever why it is not consistently accounted for by the above equations.

The problems such as discussed in [1] arise from two sources: First and foremost, it is the usual yet unreasonable insistence to reduce the above derived set of differential equations to a term or two,  $\sim v, \ddot{v}$ , in the Newtonian equation of motion for the charged body. Failure of this oversimplified description is not connected to any inconsistencies of the Maxwell theory. Second, and less centrally, it also stems from the fruitless attempt to describe point charges employing the Maxwell theory – constructed to account for finite charge densities. Although electrons may well be infinitely small, one can at most criticize the Maxwell theory for not being realistic (in this extreme quantum limit), but not for being inconsistent.

### 5.3 The Hydrodynamic Maxwell Theory

The low-resolution, hydrodynamic Maxwell theory we are going to derive and consider is a fairly general one, valid for arbitrarily strong fields and nonlinear constitutive relations, with the medium moving and the electromagnetic field time-dependent. However, it is a local-equilibrium theory with a confined frequencies range. To go beyond it, additional, nonhydrodynamic variables such as magnetization and polarization need to be included, as in sections 7 and 8.

#### Galilean versus Lorentz Transformation

Another constraint of the hydrodynamics Maxwell theory is that it is only non-relativistically valid, confined to small medium velocities,  $v \ll c$ . Unfortunately, the first-order Lorentz transformation is quite different from the Galilean transformation. For instance, with  $\nabla_0$  denoting the rest-frame spatial derivative, the former takes it to change as  $\nabla = \nabla_0 - \frac{1}{c^2} \mathbf{v} \partial_t$ , while the latter sees no difference,  $\nabla = \nabla_0$ . The Lorentz transformation is of course the fundamentally correct one. And we could, as a matter of principle, construct a fully covariant theory, and employ the theory expanded to linear order in  $v/c$ . This has in fact been done [35, 36, 37], but the result is unnecessarily complicated, and highly unpractical as an account for laboratory physics – there is simply no need to include relativistic terms distinguishing  $\nabla_0$  from  $\nabla$  say in Eqs (119, 120). And the question remains whether the theory may be simplified in a consistent fashion.

The semi-macroscopic theory in section 5.2, on weakly dissociated liquids, has an easy way out. Because the material and field variables are cleanly separated there, we may simply employ the Galilean transformation for the material variables, and the Lorentz transformation for the field variables, applying the same dichotomy to  $\nabla$ , depending on what kind of variables it is applied to. This does not work for a dense, strongly interacting system, as it becomes quite impossible to distinguish field from material variables. For instance, since temperature or chemical potential are functions of  $s, \rho, \mathbf{B}$ , we would not know which transformation to prescribe for  $\nabla \mu = [\partial \mu / \partial \rho] \nabla \rho + [\partial \mu / \partial H] \nabla H + \dots$ .

Facing this dilemma, a practical solution is to assume true Galilean behavior, taking the variables of the two temporal Maxwell equations (18) as invariant,

$$\mathbf{D} = \mathbf{D}_0, \quad \mathbf{B} = \mathbf{B}_0, \quad (140)$$

and its fluxes to transform as

$$\mathbf{E} \equiv \mathbf{E}_0 - (\mathbf{v}/c) \times \mathbf{B}, \quad \mathbf{H} \equiv \mathbf{H}_0 + (\mathbf{v}/c) \times \mathbf{D}. \quad (141)$$

(Compare this with the continuity equation (107), in which the density  $\rho$  is invariant, while the flux transforms as  $\mathbf{j} = \rho\mathbf{v} + \mathbf{j}_0$ .) As we shall presently see, this transformation maintains the form-invariance of the Maxwell equations. (See [38] for a thoughtful, more formal derivation.) As a result, the theory to be derived and considered is fully Galilean invariant. And one may convince oneself that all relevant terms from the covariant equations in [35, 36, 37] are included, with the excluded ones quite obviously numerically insignificant.

Unfortunately, there is one inconsistency. Starting from the rest-frame energy, Eq (24), to find the associated expression for an arbitrary frame, and requiring it to reduce to Eq (138) under the prescription of Eq (139), the only possibility is

$$du = Tds + \mu d\rho + \mathbf{v} \cdot d\mathbf{g} + \mathbf{E}_0 \cdot d\mathbf{D} + \mathbf{H}_0 \cdot d\mathbf{B}, \quad (142)$$

where  $\mathbf{g}$  is the conserved total momentum density. The inconsistency is, this expression is a result of a first-order Lorentz transformation, see appendix B. However, since there is no clear prescription for transforming the energy in Galilean-invariant systems, we may simply take Eq (142) as the correct starting expression, from which to derive the hydrodynamic Maxwell theory. [Eq (142) is not needed after the equations are set up, since all constitutive relations,  $T, \mu, \mathbf{E}_0, \mathbf{H}_0$  as functions of  $s, \rho, \mathbf{D}, \mathbf{B}$  are obtained from the rest frame energy.]

### Frame-Independent Thermodynamics

Starting from Eq (142), the associated equilibrium conditions are

$$\nabla \times \mathbf{H}_0 = 0, \quad \nabla \times \mathbf{E}_0 = 0 \quad (\text{or } \mathbf{E}_0 = 0), \quad (143)$$

$$\nabla T = 0, \quad v_{ij} = 0, \quad \dot{\mathbf{v}} + \nabla\mu = 0. \quad (144)$$

The three equations of the first line are deduced starting from Eq (142), going through the same steps as those leading to Eqs (26). Clearly, in spite of the more general starting point, these conditions have not changed, and are still given by setting, in the local rest frame, either the curl or the field itself to zero, nicely illustrating that the fact of being in equilibrium does not depend on the frame of reference.  $\nabla T = 0$  is also unchanged, and the last two equations are obtained by keeping momentum, angular momentum and the booster constant: Vanishing  $v_{ij}$  allows only motions that are combinations of translation and solid-body rotation. Not being Galilean invariant, the chemical potential  $\mu$  has additional terms when the system rotates and translates. The gradient of these terms is canceled by  $\dot{\mathbf{v}}$ . (The details are in [33], and not reproduced here, because it is off the present focus on the Maxwell theory.)

First, given the fact that the charge density  $\rho_\epsilon = \nabla \cdot \mathbf{D}$  is (to linear order in  $v/c$ ) a scalar,  $\mathbf{D}$  must also be one. Since  $\nabla \cdot \mathbf{B} = 0$  is also a scalar, so is  $\mathbf{B}$ . In equilibrium and for a medium at rest, because both  $\dot{\mathbf{B}} = -c\nabla \times \mathbf{E}^M$  and  $\nabla \times \mathbf{E}_0$  vanish, we deduced in section 2.3 that  $\mathbf{E}^M = \mathbf{E}^0$  and  $\dot{\mathbf{B}} = -c\nabla \times \mathbf{E}_0$ ; similarly, also  $\dot{\mathbf{D}} = c\nabla \times \mathbf{H}_0$ . For a translating and rotating medium in equilibrium,  $\mathbf{D}$  and  $\mathbf{B}$  move with the medium, such that each material point keeps their values, and the temporal derivatives in the local rest frame,  $d_t\mathbf{D}$  and  $d_t\mathbf{B}$  vanish. They relate to the laboratory derivatives as given in Eq (3),

$$d_t\mathbf{D} \equiv \dot{\mathbf{D}} + (\mathbf{v} \cdot \nabla)\mathbf{D} - \boldsymbol{\Omega} \times \mathbf{D} = 0, \quad (145)$$

$$d_t\mathbf{B} \equiv \dot{\mathbf{B}} + (\mathbf{v} \cdot \nabla)\mathbf{B} - \boldsymbol{\Omega} \times \mathbf{B} = 0, \quad (146)$$

where  $\boldsymbol{\Omega} \equiv \frac{1}{2}(\nabla \times \mathbf{v})$ . So the properly generalized equations are

$$d_t\mathbf{D} = c\nabla \times \mathbf{H}_0, \quad d_t\mathbf{B} = -c\nabla \times \mathbf{E}_0. \quad (147)$$

On the other hand, because of locality and charge conservation, we know that the Maxwell equations always have the structure

$$\dot{\mathbf{B}} = -c\nabla \times \mathbf{E}^M, \quad \dot{\mathbf{D}} + \mathbf{j}_\epsilon = c\nabla \times \mathbf{H}^M. \quad (148)$$

Defining  $\mathbf{E} = \mathbf{E}^M$ ,  $\mathbf{H} = \mathbf{H}^M$  to be the Maxwell fields of an arbitrary inertial frame in equilibrium, we find that Eqs (148) and (147) are equivalent with the identifications  $\mathbf{j}_\epsilon = \rho_\epsilon \mathbf{v}$  and Eq (141). (Terms  $\sim v_{ij}$  are neglected, since it vanishes in equilibrium.)

In a satellite containing a superconducting coil and a magnetizable medium, the field is in general non-uniform. Yet if coil and medium co-rotate and co-translate,  $\mathbf{d}_t \mathbf{D}, \mathbf{d}_t \mathbf{B} = 0$ , the system may stay in equilibrium,  $\nabla \times \mathbf{E}_0, \nabla \times \mathbf{H}_0 = 0$ , maintaining its motion for ever, and keeping its momentum and angular momentum constant. In an earth-bound laboratory, the magnetic coil is frequently fixed, and the field stationary in the lab-frame  $\dot{\mathbf{B}}, \dot{\mathbf{D}} = 0$ . If the medium rotates, with  $\boldsymbol{\Omega} \times \mathbf{B} \neq 0$ , all four quantities,  $\mathbf{d}_t \mathbf{D}, \mathbf{d}_t \mathbf{B}, \nabla \times \mathbf{E}_0, \nabla \times \mathbf{H}_0$  are finite, and the system is not in equilibrium, implying the motion is dissipative and will eventually stop. This combination of field dissipation and transformation is the basic physics for magneto-viscous effects such as field-enhanced viscosity and rotational field deflection, see section 6.2 and 6.3. As this consideration is so general, there is no doubt that any magnetic suspensions (Brownian or not), or in fact any uniform magnetizable fluids, are capable of showing these effects, though the associated magnitude will depend on the relaxation time of the magnetization and be very different. Usually, of course, magneto-viscous effects are derived by considering the term  $\boldsymbol{\Omega} \times \mathbf{M}$  in the relaxation equation for the magnetization, set up for the specific case of suspended particles relaxing in a Brownian fashion (ie, with the magnetization fixed to the particles and the particle rotating against the fluid matrix) [13, 16]. As a result, the false impression was established in the ferrofluid community that Brownian relaxation is a necessary precondition for magneto-viscous effects.

With Eqs (140, 141), we may rewrite the energy, Eq (142), as

$$du = Tds + \mu d\rho + \mathbf{v} \cdot d\mathbf{g}^M + \mathbf{E} \cdot d\mathbf{D} + \mathbf{H} \cdot d\mathbf{B}, \quad (149)$$

$$\mathbf{g}^M \equiv \mathbf{g} - \mathbf{D} \times \mathbf{B}/c, \quad (150)$$

generalizing Eq (137). Note Eq (150) is only a shorthand, the result of going from Eq (142) to (149). As we shall see, in all the fluxes of the hydrodynamic equations, this  $\mathbf{g}^M$  will replace the material momentum density  $\mathbf{g}^M = \rho \mathbf{v}$  of section 5.2. But there is no further implication, especially not that this  $\mathbf{g}^M$  were the (unambiguous) material part of the momentum density.

In their classic book, De Groot and Mazur devoted a chapter to considering macroscopic electromagnetism, aiming to set up a hydrodynamic theory capable of accounting for polarizable and magnetizable condensed systems. Yet because they oversaw the crucial difference between  $\mathbf{g}$  and  $\mathbf{g}^M$ , and started from the superficially plausible thermodynamic relation,  $du = \dots + \mathbf{v} \cdot d\mathbf{g} + \mathbf{E} \cdot d\mathbf{D} + \mathbf{H} \cdot d\mathbf{B}$ , and because they did not include the dissipative fields,  $\mathbf{H}^D$  and  $\mathbf{E}^D$ , their results differ significantly from those given below.

## The Standard Procedure

The complete set of equations of motion, valid also off equilibrium, is

$$\dot{\mathbf{D}} = c \nabla \times \mathbf{H}^M - \mathbf{j}_\epsilon, \quad \dot{\mathbf{B}} = -c \nabla \times \mathbf{E}^M, \quad (151)$$

$$\dot{\rho} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad \dot{s} + \nabla \cdot \mathbf{f} = R^D/T, \quad (152)$$

$$\dot{u} + \nabla \cdot \mathbf{Q} = 0, \quad \dot{g}_i + \nabla_k \Pi_{ik} = 0, \quad (153)$$

stating that conserved quantities satisfy continuity equations, while the field variables satisfy the Maxwell equations – expressing locality and charge conservation. To obtain the fluxes,  $\mathbf{H}^M, \mathbf{E}^M, \mathbf{f}, \mathbf{Q}, \Pi_{ik}$ , and the entropy production  $R^D$ , we first introduce some fluxes with a superscript  $D$ , as a pure definition, though they will turn out to be the dissipative contributions,

$$\mathbf{f} = s\mathbf{v} - \mathbf{f}^D, \quad \Pi_{ik} = \Pi_{ik}^R - \Pi_{ik}^D, \quad \mathbf{j}_\epsilon = \rho_\epsilon \mathbf{v} + \mathbf{j}_\epsilon^D, \quad (154)$$

$$\mathbf{E}^M = \mathbf{E} + \mathbf{E}^D, \quad \mathbf{H}^M = \mathbf{H} + \mathbf{H}^D, \quad (155)$$

then we proceed as in 5.2, inserting the equations of motion into the temporal derivative of Eq (142),  $\dot{u} = T\dot{s} + \mu\dot{\rho} + \mathbf{v} \cdot \dot{\mathbf{g}} + \mathbf{E}_0 \cdot \dot{\mathbf{D}} + \mathbf{H}_0 \cdot \dot{\mathbf{B}}$ , and sorting the terms to be either part of a divergence, or proportional to a thermodynamic force that vanishes in equilibrium. The

result is  $\nabla \cdot \mathbf{Q} = \nabla \cdot \mathbf{Q}^1 + R^D - R^1 = \nabla \cdot \mathbf{Q}^2 + R^D - R^2$ , and contains a non-uniqueness, with  $\mathbf{Q}^2 = \mathbf{Q}^1 + \mathbf{E}^D \times \mathbf{H}^D$ ,  $R^2 = R^1 + \nabla \cdot (\mathbf{E}^D \times \mathbf{H}^D)$ , where

$$R^1 \equiv (\mathbf{j}_\epsilon - \rho_\epsilon \mathbf{v}) \cdot \mathbf{E}_0 - (f_i - sv_i) \nabla_i T - (\Pi_{ik} - \Pi_{ik}^R) v_{ik} + (\mathbf{E}^M - \mathbf{E}) \cdot c \nabla \times \mathbf{H}_0 - (\mathbf{H}^M - \mathbf{H}) \cdot c \nabla \times \mathbf{E}_0, \quad (156)$$

$$Q_i^1 \equiv T f_i + (\mu \rho + \mathbf{v} \cdot \mathbf{g}^M) v_i + (\Pi_{ik} - \Pi_{ik}^R) v_k + c[\mathbf{E} \times \mathbf{H} + (\mathbf{E}^M - \mathbf{E}) \times \mathbf{H}_0 + \mathbf{E}_0 \times (\mathbf{H}^M - \mathbf{H})]_i. \quad (157)$$

$$\Pi_{ik}^R \equiv g_i^M v_k - E_i D_k - H_i B_k + \delta_{ik} \times (Ts + \mu \rho + \mathbf{v} \cdot \mathbf{g}^M - u + \mathbf{E} \cdot \mathbf{D} + \mathbf{H} \cdot \mathbf{B}), \quad (158)$$

$$R^2 \equiv \mathbf{f}^D \cdot \nabla T + \tilde{\Pi}_{ik}^D v_{ik} + \mathbf{H}^D \cdot \mathbf{d}_t \mathbf{B} + \mathbf{E}^D \cdot \mathbf{d}_t \mathbf{D} + \mathbf{j}^D \cdot \mathbf{E}_0^M, \quad (159)$$

$$Q_i^2 \equiv T f_i + (\mu \rho + \mathbf{v} \cdot \mathbf{g}^M) v_i - \tilde{\Pi}_{ik}^D v_k + c[\mathbf{E}^M \times \mathbf{H}^M]_i + \frac{1}{2}(B_i H_k^D - B_k H_i^D + D_i E_k^D - D_k E_i^D) v_k \quad (160)$$

$$\Pi_{ik} \equiv \Pi_{ik}^R - \tilde{\Pi}_{ik}^D + (B_j H_j^D + D_j E_j^D) \delta_{ik} - \frac{1}{2}(B_k H_i^D + B_i H_k^D + D_k E_i^D + D_i E_k^D). \quad (161)$$

We may either conclude  $\mathbf{Q} = \mathbf{Q}^1$ ,  $R^D = R^1$ , or  $\mathbf{Q} = \mathbf{Q}^2$ ,  $R^D = R^2$ , with both satisfying all general principles, and the difference mainly notational. In the first case, the thermodynamic forces are  $c(\nabla \times \mathbf{H}_0)$ ,  $c(\nabla \times \mathbf{E}_0)$ . In the second, they are the rest-frame derivatives,  $\mathbf{d}_t B_i$  and  $\mathbf{d}_t D_i$ , connected to the first pair as

$$\begin{aligned} \mathbf{d}_t B_i &= -c(\nabla \times [\mathbf{E}_0 + \mathbf{E}^D])_i - B_i v_{kk} + B_k v_{ik}, \\ \mathbf{d}_t D_i &= (c \nabla \times [\mathbf{H}_0 + \mathbf{H}^D] - \mathbf{j}^D)_i - D_i v_{kk} + D_k v_{ik}. \end{aligned} \quad (162)$$

[ $\mathbf{d}_t B_i$  and  $\mathbf{d}_t D_i$  are defined by Eqs (145, 146), and the two above expressions are the off-equilibrium generalization of Eqs (147).] Therefore, a diagonal Onsager matrix in the second will have off-diagonal terms in the first, and vice versa.<sup>10</sup> We choose the second representation, as ferrofluids tend to be diagonal in it.

In equilibrium, all forces vanish,  $\nabla T$ ,  $v_{ik}$ ,  $\nabla \times \mathbf{H}_0$ ,  $\nabla \times \mathbf{E}_0$ ,  $\mathbf{d}_t B_i$ ,  $\mathbf{d}_t D_i \rightarrow 0$ , (possibly also  $\mathbf{E}_0 \rightarrow 0$ ), and we have:  $\mathbf{f} = s\mathbf{v}$ ,  $\mathbf{j}_\epsilon = \rho_\epsilon \mathbf{v}$ ,  $\mathbf{E}^M = \mathbf{E}$ ,  $\mathbf{H}^M = \mathbf{H}$ , especially  $\Pi_{ik} = \Pi_{ik}^R$  and

$$\mathbf{Q} = (Ts + \mu \rho + \mathbf{v} \cdot \mathbf{g}^M) \mathbf{v} + c \mathbf{E} \times \mathbf{H}. \quad (163)$$

These expressions correctly reduce to the 2-field theory, especially Eqs (134, 135). Also, for  $\mathbf{v} = 0$ , the stress  $\Pi_{ik}^R$  reduces to that of Eq (38).

For vanishing fluid velocity and linear constitutive relations,  $\mathbf{v} \equiv 0$ ,  $\bar{\varepsilon} \mathbf{E} = \mathbf{D}$  and  $\bar{\mu}^m \mathbf{H} = \mathbf{B}$ , the symmetry of the stress tensor is obvious. Consider the rotational invariance of the energy  $u$  to see that the stress is generally symmetric,  $\Pi_{ik}^R = \Pi_{ki}^R$ : Rotating the system by an infinitesimal angle  $d\theta$ , the scalars are invariant,  $du, ds, d\rho = 0$ , while the vectors change as  $d\mathbf{g}^M = \mathbf{g}^M \times d\theta$ ,  $d\mathbf{D} = \mathbf{D} \times d\theta$ ,  $d\mathbf{B} = \mathbf{B} \times d\theta$ . Inserting these into Eq (149) yields,

$$\varepsilon_{ijk} \Pi_{kj} = (\mathbf{v} \times \mathbf{g}^M + \mathbf{E} \times \mathbf{D} + \mathbf{H} \times \mathbf{B})_i = 0. \quad (164)$$

With dissipation,  $R^D \neq 0$ , the fluxes are amended by dissipative contributions. Identifying  $\mathbf{Q} = \mathbf{Q}^2$ ,  $R^D = R^2$ , and assuming a diagonal matrix, we have

$$\mathbf{H}^D = \alpha \mathbf{d}_t \mathbf{B}, \quad \mathbf{E}^D = \beta \mathbf{d}_t \mathbf{D}, \quad \mathbf{j}^D = \sigma \mathbf{E}_0^M, \quad (165)$$

$$\mathbf{f}^D = \kappa \nabla T, \quad \tilde{\Pi}_{ik}^D = 2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell} \delta_{ik}, \quad (166)$$

<sup>10</sup> Since the energy flux and the entropy production were slightly altered, by the term  $\nabla \cdot (\mathbf{E}^D \times \mathbf{H}^D)$ , the two sets of expressions are in fact inequivalent – if both  $\mathbf{E}^D$  and  $\mathbf{H}^D$  are finite. [The term  $\nabla \cdot (\mathbf{E}^D \times \mathbf{H}^D)$  is both the divergence of something and vanishes in equilibrium, it can therefore be part of either the energy flux  $\mathbf{Q}$  or the entropy production  $R^D$ , a rare instance of inconclusiveness of the standard procedure.] This mainly concerns the so-called sq-modes, considered in [6, 7] and relevant for systems, in which both the magnetic and electric dissipation are sufficiently large, such that neither  $\alpha$  nor  $\beta$  may be neglected.

where  $\alpha, \beta, \sigma, \kappa, \eta_1, \eta_2$  are transport coefficients [or matrices as in Eq (130)], and functions of thermodynamic variables. Note  $v_{ij}^0 \equiv v_{ij} - \frac{1}{3}\delta_{ij}v_{\ell\ell}$  as before. Insert the expressions into Eq (155), to find  $\mathbf{E}^M, \mathbf{H}^M$ ; into Eq (161) to find the stress  $\Pi_{ik}$ ; into Eq (154) to find the entropy flux  $\mathbf{f}$  and electric current  $\mathbf{j}_\epsilon$ . Though off-diagonal terms are generally needed for a complete account, these fluxes are frequently a good approximation, eg for ferrofluids exposed to weak fields.

## Summary of the Results

First, we write down the explicit formulas for the energy and momentum flux,

$$Q_i = (sT + \mu\rho + \mathbf{v} \cdot \mathbf{g}^M)v_i - \kappa\nabla_i T - (2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell}\delta_{ik})v_k + \left[ c\mathbf{E}^M \times \mathbf{H}^M + \frac{1}{2}\mathbf{v} \times (\alpha\mathbf{B} \times d_t\mathbf{B} + \beta\mathbf{D} \times d_t\mathbf{D}) \right]_i, \quad (167)$$

$$\begin{aligned} \Pi_{ik} = & A\delta_{ik} - (2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell}\delta_{ik}) + g_i^M v_k - E_i^M D_k - H_i^M B_k \\ & + \frac{1}{2}(\alpha B_k d_t B_i - \alpha B_i d_t B_k + \beta D_k d_t D_i - \beta D_i d_t D_k). \end{aligned} \quad (168)$$

$$A \equiv Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g}^M + \mathbf{E}^M \cdot \mathbf{D} + \mathbf{H}^M \cdot \mathbf{B} - u.$$

For stationary media,  $\mathbf{v} \equiv 0$ , the energy flux is  $\mathbf{Q}^2 = c\mathbf{E}^M \times \mathbf{H}^M - T\mathbf{f}^D$ , demonstrating the validity of the Poynting theorem in the presence of dissipation. However, for a moving medium, the additional terms  $\sim vd_t B$  do show up the limit of the Poynting theorem. For a solid-body rotation, these terms are  $\sim \mathbf{v} \times \boldsymbol{\Omega} \cdot (\alpha\mathbf{B}^2 + \beta\mathbf{D}^2)$  and imply an energy current either converging onto, or escaping from, the center, possibly causing a temperature gradient to form. The stress may also be written as

$$\begin{aligned} \Pi_{ik} = & A\delta_{ik} - (2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell}\delta_{ik}) \\ & - \frac{1}{2}[E_i^M D_k + E_k^M D_i - g_i^M v_k + (i \leftrightarrow k)], \end{aligned} \quad (169)$$

which renders it explicitly symmetric, and seemingly a straight-forward extension of Eq (38). In this context it is of some interest to revisit the discussion in §80, 81 of [15], in which the authors lamented our ignorance about the form of the Maxwell stress in the presence of dissipation. It is also instructive to retrace the considerations of section 3.2, to obtain the off-equilibrium expression for the bulk force density, the generalization of Eq (50) and (131). Inserting  $\dot{g}_i^M + \nabla_k(g_i^M v_k) = \rho \frac{d}{dt} g_i^M / \rho$  and  $\frac{\partial}{\partial t}(\mathbf{D} \times \mathbf{B})_i / c = \nabla_k(B_k H_i^M + D_k E_i^M) - B_k \nabla_i H_k^M - D_k \nabla_i E_k^M - \rho_\epsilon E_i^M - (\mathbf{j}_\epsilon \times \mathbf{B})_i / c$  into  $\dot{g}_i + \nabla_k \Pi_{ik} = 0$ , with  $\Pi_{ik}$  given by Eq (168), we find

$$\begin{aligned} & \rho \frac{d}{dt}(g_i^M / \rho) + s\nabla_i T + \rho_\alpha \nabla_i \mu_\alpha + g_k^M \nabla_i v_k - \nabla_k(2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell}\delta_{ik}) \\ & = (\rho_\epsilon \mathbf{E}^M + \mathbf{j}_\epsilon \times \mathbf{B} / c)_i - \alpha d_t B_k \nabla_i B_k - \beta d_t D_k \nabla_i D_k \\ & \quad + \frac{1}{2}[\nabla \times (\alpha\mathbf{B} \times d_t\mathbf{B} + \beta\mathbf{D} \times d_t\mathbf{D})]_i. \end{aligned} \quad (170)$$

Clearly, the bulk force is  $s\nabla_i T + \rho_\alpha \nabla_i \mu_\alpha + g_k^M \nabla_i v_k$ , and the Lorentz force is now given in terms of  $\mathbf{E}^M$ . Two purely dissipative forces are operative, one longitudinal and the other transversal. For the magnetic case, the first is  $\alpha d_t B_k \nabla_i B_k$ , finite if  $d_t \mathbf{B} \parallel \mathbf{B}$ ; the second is  $\frac{1}{2}\nabla \times (\alpha\mathbf{B} \times d_t\mathbf{B})$ , finite when  $d_t \mathbf{B} \perp \mathbf{B}$ . This is the case when the field rotates while the medium is stationary, or more usually, when the medium rotates in the presence of a stationary field,  $\dot{\mathbf{B}} = 0$ ,  $\boldsymbol{\Omega} \neq 0$ . If  $\mathbf{B} \parallel \mathbf{H}$ , one can write this force as  $\frac{1}{2}\nabla \times (\mathbf{M} \times \mathbf{B})$ , because  $\mathbf{B} \times d_t \mathbf{B} = \mathbf{B} \times \mathbf{H}^D = \mathbf{B} \times \mathbf{H}^M \equiv \mathbf{M} \times \mathbf{B}$ . This force is routinely used in ferrofluids to account for dissipative behavior, see [16], and widely believed to be a consequence of ferrofluids being suspensions, as it is derived from the internal angular momentum of the particles [13]. Yet as we now realize, it is quite generically the transversal part of the dissipative force. There is no reason whatever it should not exist in homogeneous systems, fluid or solid.

To generalize the boundary conditions discussed in 3.3, we note that the connecting conditions for fields are now  $\Delta D_n, \Delta B_n, \Delta E_t^M, \Delta H_t^M = 0$ . As a result, especially the condition  $\Delta \Pi_{tn} = 0$  is no longer automatically satisfied and hence contains useful information. At a free surfaces contiguous to atmosphere, inserting Eq (168) with  $v_n = 0$  into  $\Delta \Pi_{tn} = 0$  (ie. neglecting surface tension), we find

$$B_n H_t^D - B_t H_n^D + D_n E_t^D - D_t E_n^D = \eta_1 (\nabla_n v_t + \nabla_t v_n). \quad (171)$$

We shall use this boundary condition for instance to consider the shear force exerted by a rotating field on the surface of a magnetizable liquid, in section 6.2.

### The Total Momentum Density

Since all expressions of this section are derived starting from Eq (142), without the total, conserved momentum density  $\mathbf{g}$  ever being specified, they remain valid irrespective of its explicit form, whether  $\mathbf{g}$  is given by  $\rho\mathbf{v} + \mathbf{E} \times \mathbf{H}/c$ , or say  $\rho\mathbf{v} + \mathbf{D} \times \mathbf{B}/c$ , to pick a historically popular form, see the discussion in [2]. Yet the rigorous identity,  $\mathbf{g} = \mathbf{Q}/c^2$  discussed in 5.1, or its rewritten version, the second of Eq (136), really leaves us with no choice other than

$$\mathbf{g} = \rho\mathbf{v} + \mathbf{E} \times \mathbf{H}/c \quad (172)$$

[again neglecting  $(Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g}^M)/\rho c^2 \ll 1$ ]. Being a term of zeroth order in the velocity,  $\mathbf{E} \times \mathbf{H}/c$  may not be neglected – though the difference to  $\mathbf{E}_0 \times \mathbf{H}_0/c$  may. Our considerations make abundantly clear that the conserved momentum density is the sum of material and field contributions, with the Maxwell tensor being the associated flux. Nevertheless, the numbers in the context of condensed matter are such that  $\rho\mathbf{v} \gg \mathbf{E} \times \mathbf{H}/c$ , and the second term may usually be neglected.<sup>11</sup> Given Eq (150), or  $\mathbf{g}^M = \rho\mathbf{v} + (\mathbf{E} \times \mathbf{H} - \mathbf{D} \times \mathbf{B})/c$ , Eq (170) yields the term  $\frac{\partial}{\partial t}(\mathbf{E} \times \mathbf{H} - \mathbf{D} \times \mathbf{B})/c$ , the Abraham force [39]. And since  $\mathbf{D} \times \mathbf{B}$  and  $\mathbf{E} \times \mathbf{H}$  are of the same order of magnitude, it is again a negligible quantity.

## 6 Off-Equilibrium Experiments

Having been derived from thermodynamics, conservation laws, and the transformation properties, the expressions of section 5 are fairly general, valid for all magnetizable and polarizable liquids, from single-component paramagnetic fluids to suspensions such as ferrofluids, and their respective electric counterparts. In the case of ferrofluids, although one is tempted to think that the properties of the ferromagnetic particles, the magnetic moment and internal angular momentum, would be important, this is true only at higher frequencies or at higher resolutions. On a coarse scale relevant for many experiments, of which four are discussed below, the present theory is quite adequate, even appropriate for being not unnecessarily detailed.

### 6.1 Induction and Incompatible Equilibria

First, we examine the familiar case of the Faraday law of induction and the eddy-current break, from the perhaps unusual view angle of equilibria in different frames [40, 41]. As similar “transformational physics” is also at work in the ensuing sections on various experiments in (non-conducting) ferrofluids, this example builds a useful analogy.

The third of Eqs (143) states that equilibrium requires the electric field in the conductor’s local rest-frame to vanish,

$$\mathbf{E}_0 = \mathbf{E} + \mathbf{v} \times \mathbf{B}/c = 0. \quad (173)$$

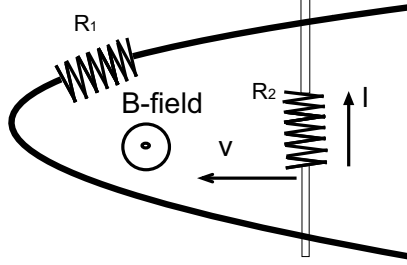
If  $\mathbf{E}_0$  is finite, the entropy is not maximal, and an electric current  $\mathbf{j}_e = \mathbf{j}^D = \sigma\mathbf{E}_0$  is cranked up to redistribute the charge, forcing  $\mathbf{E}_0$  towards zero. Inserting  $\mathbf{j}_e = \sigma\mathbf{E}_0$  into

<sup>11</sup> Taking  $\rho$  as 1 g/cm<sup>3</sup>,  $v$  as 1 cm/s,  $\hat{H} = 10^7$  A/m,  $\hat{E} = 10^7$  V/m (ie  $H$  as  $10^4$ , and  $E$  as  $30 \sqrt{\text{J}/\text{M}^2}$ ), we still have  $\rho v c / E H \approx 3000$ . And if we are to compare  $\partial(\mathbf{E} \times \mathbf{H})/c \partial t$  with the dissipative force  $\nabla \times (\mathbf{B} \times \alpha \hat{\mathbf{B}})$ , we find their quotient to scale with the small quantity,  $t/\tau$ , where  $t = L/c$  is the time light needs to cross the system of extension  $L$ , while  $\tau$  is the relaxation time of magnetization or polarization, because  $\nabla \sim 1/L \sim 1/ct$  and  $\alpha \sim \tau$ .

$\dot{\mathbf{D}} = \nabla \times \mathbf{H}^M - \mathbf{j}_e$  (and taking  $\mathbf{D} = \mathbf{E}$  as appropriate for metals) leads to a relaxation equation for  $\mathbf{E}$ ,

$$\dot{\mathbf{E}} = c \nabla \times \mathbf{H}^M - \mathbf{E}_0 / \tau, \quad (174)$$

with  $\tau \equiv 1/\sigma$  around  $10^{-19}$ s for copper. This fact allows us to cast a fresh look on the Faraday's law of induction.



**Fig. 8.** Equilibrium requires the rest-frame electric field of both the stationary and moving section of the wire to vanish. As this cannot happen simultaneously, a current is generated “out of frustration.”

If a metallic object starts to move with the velocity  $\mathbf{v}$  in the presence of a magnetic field  $\mathbf{B}$ , the equilibrium condition,  $\mathbf{E}_0 = 0$ , is established very quickly, as described by Eq (174) – implying a finite lab-frame field,  $\mathbf{E} = -\mathbf{v} \times \mathbf{B}/c$ , and an appropriate charge separation. Now consider a loop, with only a section of it moving, as in the classic setup of Fig. (8). Because there are two inequivalent paths, “frustration” sets in: The moving section, as just discussed, strives to establish a finite lab-frame field  $\mathbf{E}$  by separating charges, while the stationary part works just as hard to eliminate it. It is the incompatibility of these two equilibria that maintains a field and gives rise to circulating current. Both exist as long as the velocity of the moving section  $\mathbf{v}$  is finite.

Integrating the Maxwell equation in the form  $\dot{\mathbf{B}} = -c \nabla \times (\mathbf{E}_0 - \mathbf{v} \times \mathbf{B}/c)$  around a conducting loop, we arrive at  $\int d\mathbf{A} \cdot \dot{\mathbf{B}} + \oint \mathbf{B} \cdot (\mathbf{v} \times d\boldsymbol{\ell}) = -c \oint \mathbf{E}_0 \cdot d\boldsymbol{\ell}$ . Identifying the conductor's velocity  $\mathbf{v}$  with that the area  $\mathbf{A}$  changes, the two terms on the left may be combined as  $\frac{d}{dt} \int \mathbf{B} \cdot d\mathbf{A}$ . The term on the right,  $-c \oint \mathbf{j}_e \cdot d\boldsymbol{\ell}/\sigma$ , may be integrated assuming constant current  $I$ . In the example of Fig. 8, this gives  $-c(R_1 + R_2)I$ , with  $R_1, R_2$  denoting the respective resistance. The result is the Faraday's law of induction,

$$\frac{d}{dt} \phi \equiv \frac{d}{dt} \int \mathbf{B} \cdot d\mathbf{A} = -c(R_1 + R_2)I. \quad (175)$$

Frequently, the term *motional electro-motive force* is used for  $\int \mathbf{E}_0 \cdot d\boldsymbol{\ell}$ . It is a label (and not an understanding) for a quantity that looks like a potential yet cannot possibly be one, as  $\oint \mathbf{E}_0 \cdot d\boldsymbol{\ell}$  is non-zero. Yet the point is, every portion of  $\oint \mathbf{E}_0 \cdot d\boldsymbol{\ell}$  that is in the same rest frame is certainly a healthy potential in this frame. Their sum is finite simply because they are potentials in different frames.

Eq (173) may be understood in two different ways. The first takes  $\mathbf{E}_0$  as the field in its local rest frame:  $\mathbf{E}_0$  is different from  $\mathbf{E}$  in the moving wire, but the same in the stationary one. The second takes  $\mathbf{E}_0$  as a Lorentz-transformed field. It is the field of the frame moving with  $\mathbf{v}$  in the lab frame, and always different from  $\mathbf{E}$ . To distinguish this second field from the first, we refer to it as  $\mathbf{E}_2$ . Both  $\mathbf{E}_2$  and the lab-frame field  $\mathbf{E}$  are stationary in our example and possess a potential:  $\mathbf{E}_2 = -\nabla U_2$ ,  $\mathbf{E} = -\nabla U_1$ , but  $\mathbf{E}_0$  does not. So  $\oint \mathbf{E}_0 \cdot d\boldsymbol{\ell}$  is finite, while  $\oint \mathbf{E} \cdot d\boldsymbol{\ell} = \oint \mathbf{E}_2 \cdot d\boldsymbol{\ell} = 0$ . Nevertheless, there is no reason why we may not identify  $\int \mathbf{E}_0 \cdot d\boldsymbol{\ell}$  across  $R_2$  with  $\int \mathbf{E}_2 \cdot d\boldsymbol{\ell} = \Delta U_2 = R_2 I$ ,  $\int \mathbf{E}_0 \cdot d\boldsymbol{\ell}$  across  $R_1$  with  $\int \mathbf{E} \cdot d\boldsymbol{\ell} = \Delta U_1 = R_1 I$ , and rewrite the law of induction as  $\frac{d}{dt} \phi = \Delta U_1 + \Delta U_2$ , with each of the two two potential drops well defined – although their sum does not vanish, because they are given in different frames.

There are two limiting cases worth a brief pause. In the first,  $R_2 \gg R_1$ , the resistance of the sliding bar is much larger than that of the stationary arc. Because the latter wins out fighting to maintain its equilibrium, we have  $\mathbf{E} \approx 0$ , field and potential are negligible in the lab frame. Vice versa, for  $R_2 \ll R_1$ ,  $\mathbf{E}_2 \approx 0$  holds; there is no field or potential in

the moving frame. However, in neither case does  $\mathbf{E}_0$  vanish, and the current that flows remains the same as long as the sum  $R_1 + R_2$  is the same, and given by Eq (175).

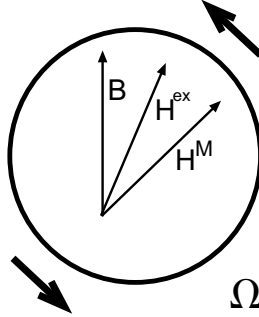
The next example is the eddy-current break, a metal plate moving with  $\mathbf{v}$ , with only part of the metal exposed to a stationary magnetic field. Typically, the plate is rotating, with  $\Omega$ , and the field-exposed region sits off-center, at radius  $R$ . Equilibrium is given by  $\mathbf{E} = 0$  outside the field-exposed region, and by  $\mathbf{E} = -\mathbf{v} \times \mathbf{B}/c$  inside it, same as in the previous example. The only difference is, the inhomogeneity is now in  $\mathbf{B}$  rather than  $\mathbf{v}$ . Again, the field-exposed region wants to maintain a charge separation, while the field-free region works to eliminate it, and the result is a frustration-induced eddy current. For homogeneous conductivity, both regions work equally hard, and the two residual fields are equal in magnitude:  $\mathbf{E} = -\mathbf{E}_0$ . So the current is  $\mathbf{j}_e = \sigma \mathbf{E}_0 = \frac{1}{2} \sigma \mathbf{v} \times \mathbf{B}/c$ , see [42]. To calculate the breaking force of the eddy current, start from Eq (131), assume  $\rho_e = 0$ , constant  $T$  and  $\mu$ , small velocity  $\mathbf{v}$ , and  $\mathbf{b} = \mathbf{B}$ , to arrive at  $\rho \frac{\partial}{\partial t} \mathbf{v} = \mathbf{j}_e \times \mathbf{B}/c$ , or

$$\rho \partial \mathbf{v} / \partial t = \frac{1}{2} \sigma (\mathbf{v} \times \mathbf{B}) \times \mathbf{B} / c^2. \quad (176)$$

This implies a relaxation time  $\tau = 2\rho c^2 / \sigma B^2$  for  $\mathbf{v} \perp \mathbf{B}$ . Assuming  $\sigma = \hat{\sigma} / \varepsilon_0 \approx 10^{19} \text{s}$ ,  $\rho \approx 10^4 \text{kg/m}^3$ ,  $c = 10^8 \text{m/s}$ , and  $\hat{B}$  in T, we have  $\tau \approx 10^{-5} \hat{B}^{-2} \text{s}$ .

If the magnetic field (ie. the field-producing coil) also moves, and is stationary in the frame of the metal plate, the total system is in equilibrium. There is then no current, dissipation or force. If it moves with  $\mathbf{u}$ , the current is  $\mathbf{j}_e = \frac{1}{2} \sigma (\mathbf{v} - \mathbf{u}) \times \mathbf{B}$ , proportional to the metal's velocity in the frame of the field. The dissipation  $R^D$  is always  $\mathbf{j}_e \mathbf{E}_0 = \mathbf{j}_e^2 / \sigma$ , see Eq(129).

## 6.2 Rotational Field deflection



**Fig. 9.** Due to dissipation, a real (rather than imaginary) contribution in the permeability  $\mu^m$ , the fields  $\mathbf{B}$  and  $\mathbf{H}^M$  are misaligned in a rotating ferrofluid.

Similar “transformational physics” also exists in non-conductors. As we shall see, in all three following experiments, it is the need for field and medium to be stationary in the same frame before they can equilibrate. (All experiments are magnetic, though of course the exact same electric ones, obtained by employing Eqs (53) and  $\alpha \rightarrow \beta$ , also exist.)

Consider a long cylinder filled with ferrofluid, rotated along the cylinder axis and subject to a perpendicular external field  $\mathbf{H}^{\text{ex}}$ , see Fig. (9). The internal fields  $\mathbf{H}^M$ ,  $\mathbf{B}$  are related to the external one as  $\mathbf{H}^M + \mathbf{B} = 2\mathbf{H}^{\text{ex}}$ , see §8 of [15]. With Eq (165), the internal fields are related as  $\mathbf{H}^M = \mathbf{H}(\mathbf{B}) - \alpha \boldsymbol{\Omega} \times \mathbf{B}$ . So  $\mathbf{H}^M$ ,  $\mathbf{H}$  may be expressed by  $\mathbf{B}$ , which in turn is given by  $\mathbf{H}^{\text{ex}}$ . For LCR,  $(1 + \chi)\mathbf{H} = \mathbf{B}$ , we have

$$\mathbf{H}^M = \left[ \frac{1}{(1+\chi)} - \alpha \boldsymbol{\Omega} \times \right] \mathbf{B}, \quad (177)$$

$$\mathbf{H}^{\text{ex}} = \frac{1}{2} \left[ 1 + \frac{1}{(1+\chi)} - \alpha \boldsymbol{\Omega} \times \right] \mathbf{B}. \quad (178)$$

The expression in the bracket of the first formula contains the inverse permeability. Note the term  $\alpha \boldsymbol{\Omega} \times \mathbf{B}$  that is both dissipative and real. If the external field  $\mathbf{H}^{\text{ex}}$  also rotates,



resulting in  $\dot{\mathbf{B}} = \boldsymbol{\omega} \times \mathbf{B}$ , we have instead  $\mathbf{H}^M = \mathbf{H}(\mathbf{B}) + \alpha(\boldsymbol{\omega} - \boldsymbol{\Omega}) \times \mathbf{B}$ , see Eqs (146, 165). And dissipation vanishes for  $\boldsymbol{\omega} = \boldsymbol{\Omega}$ .

To calculate the torque  $\mathbf{T}$  exerted by the rotating field on the container, dragging it to co-rotate,  $\boldsymbol{\Omega} \rightarrow \boldsymbol{\omega}$ , we consider the entropy produced in the sample,  $R^D V = (\mathbf{H}^D)^2 V / \alpha = \alpha B^2 (\boldsymbol{\omega} - \boldsymbol{\Omega})^2 V$ , see Eqs (159, 165), and equate it with the change in kinetic energy,  $\dot{U}_{\text{kin}} = (\boldsymbol{\omega} - \boldsymbol{\Omega}) \cdot \mathbf{T}$ , to obtain

$$\theta \dot{\boldsymbol{\Omega}} \equiv \mathbf{T} = \alpha B^2 (\boldsymbol{\omega} - \boldsymbol{\Omega}) V. \quad (179)$$

Clearly, the time the system needs to come to a stop is  $\theta / \alpha B^2 V$ , with  $\theta$  denoting the system's moment of inertia. Although this time has the same field dependence as that of the eddy-current break, see Eq (176), it is, with  $\alpha = \tau (\frac{\chi}{1+\chi})$  [see section 7:  $\chi$  is the susceptibility and  $\tau$  the relaxation time of the magnetization], typically a few orders of magnitude larger.

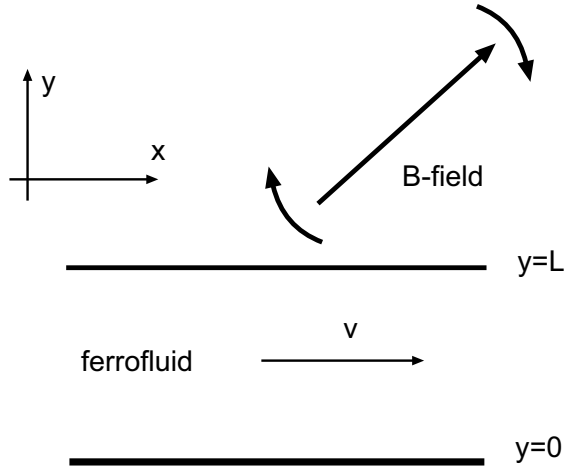
### 6.3 Field-Enhanced Viscosity

Next, we consider how field dissipation (terms  $\sim \alpha$ ) gives rise to an enhanced viscosity [43]. Take (i) neutrality, (ii) small, stationary, incompressional flow, and (iii) time-independent and spatially constant field, temperature, and chemical potential in Eq (170) to obtain [5]

$$\nabla_k \{ 2\eta_1 v_{ik} + \frac{1}{2} \alpha [B_i (\mathbf{B} \times \boldsymbol{\Omega})_k - B_k (\mathbf{B} \times \boldsymbol{\Omega})_i] \} = 0. \quad (180)$$

The viscosity is  $\eta$ , if  $\mathbf{B} \parallel \boldsymbol{\Omega}$ , and it is maximally enhanced if  $\mathbf{B} \perp \boldsymbol{\Omega}$ . For a plane shear flow,  $|v_{ik}| = |\Omega| = \frac{1}{2} \nabla_x v_y$ , the effective viscosity is  $\eta_e = \eta + \frac{1}{2} \alpha B^2$ . This result was first derived by Shliomis [13], as mentioned, from considering the angular momentum and the magnetic moment of the suspended particles in ferrofluids explicitly. Neither is obviously necessary, and a one-component paramagnetic fluids will in principle display the same enhancement. However, due to the time scales typical in these fluids,  $\alpha$  is many orders of magnitude smaller.

### 6.4 A Magnetic Pump



**Fig. 10.** Due to dissipation, a real (rather than imaginary) contribution in the permeability  $\mu^m$ , the fields  $\mathbf{B}$  and  $\mathbf{H}^M$  are misaligned in a rotating ferrofluid.

Finally, we consider an experiment probing the force exerted by a time-dependent, uniform field. Assume planar geometry and consider a slab of ferrofluid extending from a solid bottom at  $y = 0$  to the free surface at  $y = L$ . Given a  $\mathbf{B}$ -field rotating in the  $\hat{x}/\hat{y}$  plane, there will be a velocity  $v$  along  $\hat{x}$ : Because the field is uniform, Eq (170) reduces to

the shear flow equation,  $\rho\dot{v} = \eta_e \nabla^2 v$  (with  $\eta_e = \eta + \frac{1}{2}\alpha B^2$ , cf 6.3). We solve the equation employing the boundary conditions:

$$\eta_e \nabla_n v = \frac{1}{2}\alpha(B_n \dot{B}_t - B_t \dot{B}_n) \quad (181)$$

at  $y = L$ , cf Eq (171), and  $v = 0$  at  $y = 0$ . (The physics is, since the coefficient  $\alpha$  is discontinuous at the free surface, the dissipative field  $\mathbf{H}^D = \alpha \mathbf{d}_t \mathbf{B}$  deliver a finite contribution there, and represents a surface force.) Take the rotation frequency of the rotary field as  $\omega$ , we have  $B_n \dot{B}_t - B_t \dot{B}_n = B^2 \omega$ . For large penetration depths,  $\sqrt{\eta_e/\rho\omega} \gg L$ , the flow field is linear,

$$v = \frac{1}{2}\alpha B^2 \Omega y / \eta_e, \quad (182)$$

with its maximal value  $v = \frac{1}{2}\alpha B^2 \Omega L / \eta_e$  at the surface. This experiment has already been performed in a special geometry, with excellent agreement, see [44, 45].

A static, normal field  $B_n \parallel \hat{\mathbf{y}}$ , and an oscillatory, tangential one,  $B_t = \Delta B_t \times \exp(-i\omega t) \parallel \hat{\mathbf{x}}$ , lead to the time-dependent velocity  $v(x) = (\alpha x / 2\eta_e) B_n \dot{B}_t$ . The results for curved interfaces (causing a spin-up of the ferrofluid) or for higher frequencies may be found in [5].

## 7 Ferrofluid-Dynamics

As discussed and demonstrated in details in the last two sections, the hydrodynamic Maxwell theory is well capable of accounting for magneto- and electro-viscous effects, when the fields,  $\mathbf{H}^M$  and  $\mathbf{B}$ , or  $\mathbf{E}^M$  and  $\mathbf{D}$ , are not in equilibrium with each other, either being misaligned or not having the appropriate magnitude. Yet this is only true for small frequencies,  $\omega\tau \ll 1$ , and small shear rates,  $\dot{\gamma}\tau \ll 1$ , where  $\tau$  is the relaxation time of either the magnetization  $\mathbf{M}$  or the polarization  $\mathbf{P}$ . At higher frequencies,  $\omega\tau \gtrsim 1$ , or higher shear rates,  $\dot{\gamma}\tau \gtrsim 1$ ,  $\mathbf{M}$  or  $\mathbf{P}$  need to be included as independent variables, complete with an own equation of motion. In this section, we discuss the inclusion of the magnetization. (The inclusion of the Polarization is done somewhat differently, and found in section 8.1.)

There are broadly speaking three theories applied to understanding ferrofluids: The *quasi-equilibrium theory* [16] was introduced by Rosensweig, who employed it in the first seven chapters of his book[16] to account for a wide range of static effects. In this theory, the magnetization is taken in local equilibrium with the magnetic field,  $\mathbf{M}(\mathbf{r}, t) = \mathbf{M}^{\text{eq}}[\mathbf{H}(\mathbf{r}, t)]$ , and not an independent variable. Although it is widely believed that this is the reason the *quasi-equilibrium theory* does not account for magneto-viscous effects, we have seen that the reason is rather the neglect of the dissipative field,  $\mathbf{H}^D \sim \mathbf{d}_t \mathbf{B}$ .

To account for magneto-viscous effects, especially the fact that the shear viscosity is enhanced by a static magnetic field [43], Shliomis [13] (see also [46, 47]) started from the intuitive picture of magnetic particles rotating against the viscosity of the carrier liquid as the actual source of dissipation, and included both the magnetization  $\mathbf{M}$  and the angular momentum density  $\mathbf{S}$  as additional variables (though the latter is usually adiabatically eliminated afterwards). This theory contains the relaxation equation for magnetization and an extra term in the momentum flux,  $\Delta \Pi_{ij} = \frac{1}{2}\varepsilon_{ijk}(\mathbf{H} \times \mathbf{M})_k$ . Frequently referred to as the *Shliomis theory*, it does its job well for  $\omega\tau \ll 1$ , but is less competent to quantitatively account for “negative viscosity” [48, 49], a phenomenon at  $\omega\tau \gtrsim 1$ . As a consequence, Shliomis referred to a more elaborate evolution equation for  $\mathbf{M}$ , see [50, 51], derived from a microscopic, statistical investigation of rotating magnetic particles. Since the equation was solved with the assistance of the effective field method, this second variant is commonly denoted as the *effective-field theory*, or EFT. The EFT considerably improved the agreement to the “negative viscosity”-experiment. More recently, it was found that if the ferrofluid is polydisperse, the larger magnetic particles form short chains when subject to field, showing as a consequence polymer-like behavior such as shear-thinning and normal stress differences [52, 55, 56, 57, 58], see the accompanying article on rheology by Ilg. So the conclusion seems that the Shliomis theory is only valid for low frequency,  $\omega\tau \ll 1$ , and that even EFT fails for non-Newtonian ferrofluids.

Some of these common believes have been contradicted in [10], especially: (1) the Shliomis theory and EFT share the same basic structure. And the only difference lies in the coefficients the relaxation equation for the magnetization is adorned with – because they

were derived from differing microscopic assumptions. (2) Frequently, the most efficient way to construct a macroscopic theory is to establish its structure from general principles, employ a few experiments to determine its coefficients, and avoid a microscopic derivation altogether. Take for instance the Navier-Stokes equation, which may be set up in accordance to momentum conservation and the second law of thermodynamics. Then one measures the viscosity in a simple geometry – rather than trying to calculate it. With both pieces of information present, this theory develops great predictive power in a broad range of circumstances, for any geometry and boundary conditions. (3) Including the magnetization as an independent variable, one may employ thermodynamics to yield the general structure for its relaxation equation, which contains especially a coefficient  $\lambda_2$  that accounts for the coupling to elongational flows (measured to be between 0 and 0.88 in [59]). (4) Choosing appropriate values for the relaxation time  $\tau$  and coefficient  $\lambda_2$ , experiments in the range of  $\omega\tau \gg 1$  are well accounted for, see [60, 61, 62]. There is no reason why this should not be the case also with polydisperse, non-Newtonian ferrofluids.

### 7.1 Polydispersity

In this and the next section, we present a theory for polydisperse, chain-forming ferrofluids that consists of (1) a relaxation equation for the magnetization  $\mathbf{M}_1$  of the chains (or any other aggregates), and (2) the hydrodynamic Maxwell theory to account for the particles that remain single. The latter possess a wide range of relaxation times, all smaller than  $\tau_2$ , typically at around  $10^{-4}$ s. And  $\mathbf{M}_1$ 's relaxation time  $\tau_1$  is orders of magnitude larger than  $\tau_2$ , as it is determined not only by how fast the chains may be oriented, but also how quickly particles can be transported and assembled, to form chains or aggregates of the appropriate shape and size. Although all particles above a certain critical radius would contribute to chain formation, and they also possess a range of intrinsic relaxation times before chain formation sets in, we may expect, and shall at any rate assume, that chains are formed with a mixture of particles, and the associated relaxation time is fairly well defined. ( $\tau_1$  is probably typically around a few seconds – though the actual numbers are less important than the fact  $\tau_1 \gg \tau_2$ .) Clearly, the resulting theory is valid for arbitrary values of  $\omega\tau_1$  and  $\dot{\gamma}\tau_1$ , as long as  $\omega\tau_2, \dot{\gamma}\tau_2 \ll 1$  hold.

When the hydrodynamic Maxwell theory was derived in section 5, no assumption whatever was made with respect to the microscopic makeup of the magnetic fluid, certainly not that it must consist of magnetic particles of identical radius. So it is naturally valid for polydisperse ferrofluids. Here, we only generalize the evaluation of  $\alpha$ , from that given around Eq (31) to a ferrofluid characterized by many different magnetizations  $M_q$ , each relaxing with  $\tau_q$ . Starting from  $\dot{M}_q = -(M_q - M_q^{eq})/\tau_q$  with  $q = 1, 2, 3, \dots$ , we have  $(1 - i\omega\tau_q)M_q = M_q^{eq}$ , or for small frequencies,  $M_q = (1 + i\omega\tau_q)M_q^{eq}$ . This implies  $M_q = M_q^{eq} - \tau_q\dot{M}_q^{eq} = M_q^{eq} - \tau_q(dM_q^{eq}/dB)\dot{B}$ . Inserting this into  $H^M = B - \sum M_q = B - \sum[M_q^{eq} + \tau_q(dM_q^{eq}/dB)\dot{B}]$ , and identifying  $B - \sum M_q^{eq}$  as  $H$ , we find

$$\alpha = \sum \tau_q(dM_q^{eq}/dB) \rightarrow \sum \tau_q\chi_q/(1 + \sum \chi_q), \quad (183)$$

where the sign  $\rightarrow$  (here and below) holds for linear constitutive relations. This formula clearly holds however finely graded the particle populations are defined, one can even substitute the sums by integrals. Of relevance is only the insight that the effect of magnetization relaxation in a polydisperse ferrofluid is well accounted for by a single coefficient  $\alpha$ , a quantity that is easily measured.

### 7.2 Magnetization of the Chain-Forming Particles

The hydrodynamic Maxwell theory of section 5 describes, as it is, a polydisperse ferrofluid without chain-formation. Adding the associated magnetization  $\mathbf{M}_1$  as an independent variable, the energy density of Eq (149) has the additional term,

$$du = \dots + \mathbf{h} \cdot d\mathbf{M}_1, \quad (184)$$

where  $\dots$  stand for the terms of the hydrodynamic Maxwell theory as given in section 5. For linear constitutive relations and in the rest frame,  $u \rightarrow u_0(\rho, s) + \frac{1}{2}B^2/\mu^m - \mathbf{B} \cdot$

$\mathbf{M} + \frac{1}{2}\mu^m(1 + \chi_1)\mathbf{M}_1^2/\chi_1$ , we have  $\mathbf{H} \equiv \partial u/\partial \mathbf{B} \rightarrow \mathbf{B}/\mu^m - \mathbf{M}_1$  and  $\mathbf{h} \equiv \partial u/\partial \mathbf{M}_1 \rightarrow \mu^m(1 + \chi_1)\mathbf{M}_1/\chi_1 - \mathbf{B}$ . (The magnetic permeability  $\mu^m$  is from the non-chain-forming particles.) The total field is still given as  $\mathbf{H}^M = \mathbf{H} + \mathbf{H}^D = \mathbf{H} + \alpha d_t \mathbf{B}$ , but now depends on  $\mathbf{M}_1$  via  $\mathbf{H}$ .

The relaxation equation for  $\mathbf{M}_1$  is

$$d_t \mathbf{M}_1 = \mathbf{X}^D, \quad (185)$$

where  $d_t$  is the rest frame derivative of Eq (3), while  $\mathbf{X}^D$  couples to the dissipative stress  $\tilde{\Pi}_{ij}^D$  in a Onsager matrix-relation, and is given as

$$\tilde{\Pi}_{ik}^D = 2\eta_1 v_{ik}^0 + \eta_2 v_{\ell\ell} \delta_{ik} + \frac{1}{2} \lambda_2 (M_i h_k + M_k h_i) \quad (186)$$

$$X_i^D = -\zeta h_i + \lambda_2 M_k v_{ik}^0, \quad (187)$$

where  $v_{ij}^0 \equiv v_{ij} - \frac{1}{3} \delta_{ij} v_{\ell\ell}$ . Characterizing the anisotropy of the system, the  $M_i$  here are the total magnetization. In fact, because of the anisotropy, many more coefficients are allowed by symmetry, see [10]. Aiming to keep the theory as simple as possible, only the respective term  $\sim \lambda_2$  is included, as the only off-diagonal one, because they are demonstrably large and relevant in the presence of chain formation [59].

Given the above expressions, the energy flux, the stress tensor, and the entropy production of Eqs (167, 168, 159) are necessarily modified. The added terms are

$$Q_i = \dots - \frac{1}{2} \lambda_2 (M_i h_k + M_k h_i) v_k + \frac{1}{2} [\mathbf{v} \times (\mathbf{h} \times \mathbf{M})]_i, \quad (188)$$

$$\Pi_{ik} = \dots - \frac{1}{2} \lambda_2 (M_i h_k + M_k h_i) + \frac{1}{2} (h_k M_i - h_i M_k), \quad (189)$$

$$R^D = \dots + \tilde{\Pi}_{ik}^D v_{ik} - \mathbf{X}^D \cdot \mathbf{h}. \quad (190)$$

These are the complete set of equations for a polydisperse, chain-forming ferrofluid sporting a dielectric fluid matrix.

### 7.3 Shear-Thinning

Because ferrofluids display non-Newtonian behavior when the magnetic particles form chains [52, 57, 58], the analogy to polymer solutions appears fairly obvious, and one is lead to expect the need to amend ferrofluid-dynamics with some ingredients from polymer physics. On the other hand, polymer solutions and ferrofluids do differ in fundamental ways: Polymer strands are entangled without shear, but get aligned along the flow by it, while magnetic chains are aligned along the field without shear, and broken into pieces by it [52]. Being a negative statement, the term ‘‘non-Newtonian’’ lacks specificity, and there may well be different versions of it requiring different descriptions.

In this section, we demonstrate<sup>12</sup> that ferrofluid-dynamics already has the proper structure to account for shear-thinning [40, 66]. We consider the equations,

$$\frac{d}{dt} M_i + (\mathbf{M} \times \boldsymbol{\Omega})_i - \lambda_2 M_j v_{ij} = -(M_i - M_i^{\text{eq}})/\tau, \quad (191)$$

$$\begin{aligned} \Pi_{ij} = & \tilde{P} \delta_{ij} - 2\eta_1 v_{ij} - H_i B_j + \\ & + \frac{1}{2} [(M_i h_j - M_j h_i) - \lambda_2 (M_i h_j + M_j h_i)], \end{aligned} \quad (192)$$

and for simplicity neglect the contribution from the single particles that do not form chains, by setting  $\alpha = 0$ . (A finite  $\alpha$  only delivers a constant field-induced viscosity enhancement

<sup>12</sup> Polymers are characterized by *transient elasticity*, and its rheology is well-accounted for by a relaxing strain field [63, 64]. There are magnetic fluids which also need the strain field as an extra variable: If the magnetic particles are large enough, they will, in the presence of a strong field, form long chains bridging the whole system. This is the jamming transition, after which the system is truly elastic [65]. Close to it, the strain should be a critical, relaxing variable (because two chains, neither quite bridging the system, temporarily get in the way of each other). This implies transient elasticity. We shall deal with magnetic fluids here that are either incapable of the jamming transition, or far enough away from it, and refer to them as *ferrofluids*. The accepted term for the polymer-like magnetic fluids is *Magneto-rheological fluids*.

that is independent of shear, and hence off the present focus.) We also confine our considerations to incompressible flow,  $\nabla_i v_i = 0$ , and linear constitutive relations. This makes an analytic solution possible, which alone could incontrovertibly establish the fact that the observed non-Newtonian behavior of ferrofluids may indeed be accounted for by magneto-relaxation. The scalar  $\tilde{P}$  in Eq (192) contains all diagonal terms. They are not further specified, as they are relevant only for compressional flows such as considered in [11, 12].

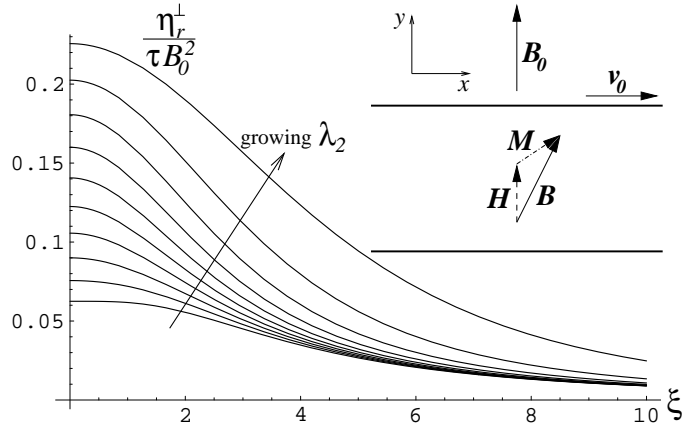
We employ Eqs (191,192) to consider simple shear,  $\mathbf{v} = \dot{\gamma}y\hat{x}$ , with the velocity along  $\hat{x}$  and the gradient along  $\hat{y}$  (see inserts of Fig 11, 12), so the equilibrium magnetization  $\mathbf{M}^{\text{eq}}$  is also in the  $xy$ -plane. Assuming stationarity,  $\frac{d}{dt}\mathbf{M} = 0$ , Eq (191) is a linear, 2x2 matrix equation,  $\underline{\underline{A}}\mathbf{M} = \mathbf{M}^{\text{eq}}$ . Inverted, it reads

$$M_x = \frac{4M_x^{\text{eq}} + 2(1 + \lambda_2)\xi M_y^{\text{eq}}}{4 + (1 - \lambda_2^2)\xi^2}, \quad (193)$$

$$M_y = \frac{4M_y^{\text{eq}} - 2(1 - \lambda_2)\xi M_x^{\text{eq}}}{4 + (1 - \lambda_2^2)\xi^2}, \quad (194)$$

where  $\xi \equiv \dot{\gamma}\tau$ . These two expressions already contain the essence of shear thinning: The magnetization goes to zero in the limit of strong shear,  $\xi \rightarrow \infty$ , implying the vanishing of any magneto-viscous effect, because the second line of Eq (192) also vanishes.

The force density on an infinitely extended plate in the  $xz$ -plane, being dragged along  $\hat{x}$  on top of a ferrofluid layer, is  $\Delta\Pi_{xy} \equiv \Pi_{xy}^{\text{air}} - \Pi_{xy}^{\text{ff}}$ . The stress of air,  $\Pi_{xy}^{\text{air}}$ , is  $-H_x B_y$ , that of the ferrofluid,  $\Pi_{xy}^{\text{ff}}$ , is given by Eq (192). (Because  $H_x B_y$  is continuous,  $\Delta\Pi_{xy}$  is calculated from the second line, in addition to the term  $-2\eta_1 v_{ij}$ .) Taking the total viscosity as  $\eta_l + \eta_r \equiv -\Delta\Pi_{xy}/\dot{\gamma}$ , the magneto-viscous contribution,  $\eta_r$ , is evaluated by inserting Eqs (193,194) into (192) for given boundary conditions, of which we consider two, perpendicular and parallel external field  $B_0$ , (see inserts of Fig 11 and 12).

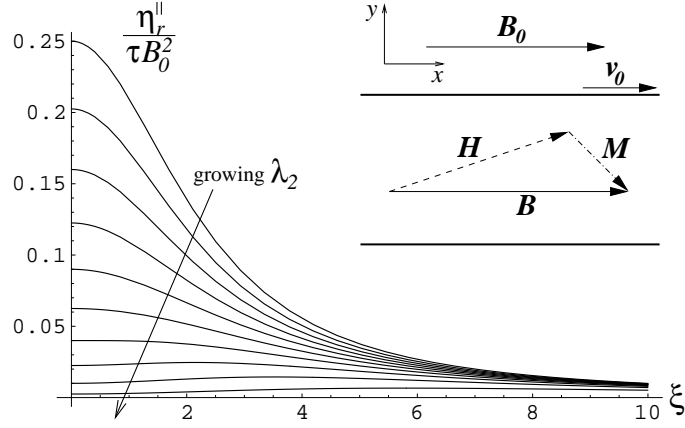


**Fig. 11.** The magneto-viscous contribution to the shear viscosity (in units of  $\tau B_0^2$ ) as a function of  $\xi \equiv \dot{\gamma}\tau$ , from  $\lambda_2 = 0$  to  $\lambda_2 = 0.9$ , in 0.1-steps, for a perpendicular external field  $B_0$ , with  $\chi=1$  (from [66]). Shear-thinning is obvious.

The experimentally most convenient configuration is given by  $B_0$  along  $\hat{y}$ , perpendicular to the plate. Because  $B_y, H_x$  are continuous, the internal fields are  $\mathbf{B} = (M_x, B_0)$ ,  $\mathbf{H} = (0, B_0 - M_y)$ , and  $\mathbf{M}^{\text{eq}} = \chi(0, B_0 - M_y)$ . Using these in Eqs (192,193,194), we find

$$\eta_r^\perp = \frac{(1 + \lambda_2)^2 [4 + (1 - \lambda_2)^2 \xi^2]}{[4(1 + \chi) + (1 - \lambda_2^2)\xi^2]^2} \tau \chi B_0^2. \quad (195)$$

For vanishing shear,  $\xi \rightarrow 0$ , the viscosity  $\eta_r^\perp$  grows with  $\tau, \chi B_0^2$  and  $\lambda_2$ . More generally,  $\eta_r^\perp$  decreases monotonically with shear if  $\chi < (1 + 3\lambda_2)/(1 - \lambda_2)$ , and displays shear-thickening



**Fig. 12.** The magneto-viscous contribution to the shear viscosity as a function of  $\xi \equiv \dot{\gamma}\tau$ , from  $\lambda_2 = 0$  to  $\lambda_2 = 0.9$ , for a parallel external field  $B_0$ , with  $\chi=1$  (from [66]). A growing  $\lambda_2$  diminishes the effect, possibly because the chains are already aligned along the flow, same as polymer strands at high shear.

(ie. a maximum) otherwise. Fig 11 shows the monotonic decay of the magneto-viscous contribution, as a function of shear, for  $\chi = 1$  and ten different values of  $\lambda_2$ , from 0 to 0.9.

If the external field  $B_0$  is parallel to the plate, along  $\hat{x}$ , again because  $B_y, H_x$  are continuous, the internal fields are  $\mathbf{B} = (B_0 + M_x, 0)$ ,  $\mathbf{H} = (B_0, -M_y)$ , and  $\mathbf{M}^{\text{eq}} = \chi(B_0, -M_y)$ . Using these in Eqs (192,193,194), we find

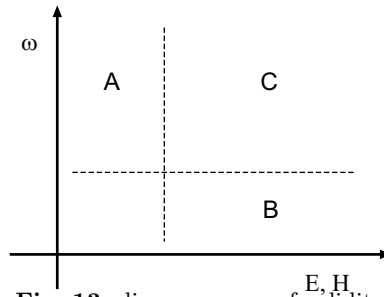
$$\eta_r^{\parallel} = \frac{(1 - \lambda_2)^2 [4(1 + \chi)^2 + (1 + \lambda_2)^2 \xi^2]}{[4(1 + \chi) + (1 - \lambda_2^2) \xi^2]^2} \tau \chi B_0^2. \quad (196)$$

In the limit of low shear,  $\xi \rightarrow 0$ ,  $\eta_r^{\parallel}$  still grows with  $\tau, \chi B_0^2$ , but now decreases with  $\lambda_2$ . (For  $\lambda_2 = 0$  and  $\chi \ll 1$ , both shear viscosities are the same,  $\eta_r^{\perp} = \eta_r^{\parallel}$ , as they should.) For finite shear,  $\eta_r^{\parallel}$  decreases monotonically with  $\xi$ , if  $\chi > \frac{1}{2}(3\lambda_2 - 1)/(1 - \lambda_2)$ , see Fig 12.

## 8 Extensions

### 8.1 The High-Frequency Regime

If we draw a diagram of field strength versus frequency, see Fig 13, we have a vertical



**Fig. 13.** diverse ranges of validity

**A**-stripe along the  $\omega$ -axis — arbitrary frequency but small field strength — that is the range of validity for the linear-response theory,  $\mathbf{E}^M \sim \mathbf{D}$ ,  $\mathbf{H}^M \sim \mathbf{B}$ . The hydrodynamic

theory presented above is valid in the horizontal **B**-stripe, given by arbitrary field strength and small frequencies: With only terms linear in  $\omega$  included, dissipation is accounted for, but not dispersion.

Curiously, only limited results about field energy and momentum exist in the vertical **A**-stripe, for linear-response theory. Assuming • linear constitutive relations, • lack of dissipation (ie real  $\varepsilon, \mu^m$ ), • quasi-monochromacy (small variation around one frequency) and • stationarity (no fluid motion,  $v \equiv 0$ ), Brillouin showed in 1921 that the additional energy due to the presence of fields is

$$\frac{1}{2}\langle E^2 \rangle d(\omega\varepsilon)/d\omega + \frac{1}{2}\langle H^2 \rangle d(\omega\mu^m)/d\omega, \quad (197)$$

where the average is temporal, over a period of oscillation. Forty years later, Pitaevskii showed that under essentially the same assumptions, the stress tensor retains its form from equilibrium, Eq(38), and remarkably, does not contain any frequency derivatives. Both results may be found in §80, 81 of [15]. Note neither expression is valid in the entire **A**-stripe, only in patches disjunct from the field-axis. This is because leaving the field-axis ( $\omega = 0$ ), the first corrections are linear in  $\omega$  – therefore odd and dissipative.

The **C**-space electrodynamics must be a theory that can simultaneously account for dissipation and dispersion, allowing nonlinear constitutive relations and finite velocities for the medium. Although one might expect principal difficulties — due to the apparent lack of a small parameter — the system is in fact, up to the optical frequency  $\sim 10^{15}$ Hz, still in the realm of macroscopic physics, as the associated wavelengths remain large compared to the atomic graininess, in spite of the the clearly ballistic frequency. So a universal, hydrodynamic-type theory should still be possible, and would be useful, for answering questions such as what the average force on a volume element exerted by a laser beam is. (The averaging has a temporal resolution larger than the time needed to establish local equilibrium, and much larger than the light's oscillatory period.) A first step toward such a theory has been quite successful [9]. It includes the polarization as an additional variable, the equation of motion of which is a nonlinear differential equation of second order in the time. The theory reduces to the hydrodynamic Maxwell theory of section 5 for small frequencies, and reproduces the linear-response results of Brillouin and Pitaevskii for small field strength and vanishing dissipation. Further work is needed here to make connections to experiments, clarifying questions such as whether the Poynting theorem holds for dissipative systems, cf [67] and the discussion of Eq (167).

## 8.2 Ferro-Nematics and Ferro-Gels

The works on Ferro-Nematics and Ferro-Gels, by Harald Pleiner, Helmut Brand and co-workers, are reported in this section. Ferrofluids are suspensions of magnetic nanoparticles in a suitable carrier liquid. They are polymer coated or charged in order to prevent coagulation. They show various distinct material properties, like superparamagnetism (large magnetic susceptibility, high saturation magnetization in rather low fields), sensitivity to magnetic Kelvin forces, and a rather complicated influence of magnetic fields on their flow behavior [16, 52]. This has led to a host of important applications as seals (in hard disk drives), active dampers (suspensions), thermal conductors (loudspeakers), as well as in the medical sector (hypothermia, drug targeting in cancer therapy). The favorable ferrofluid properties are generally preserved when dealing with more complex systems, like ferronematics and ferrogels. In addition, new features are coming into play leading to quite unusual and novel type of behavior. Ferronematics are obtained, if the carrier liquid is a lyotropic or thermotropic nematic liquid crystal, while ferrogels are polymer melts swollen by ferrofluids and crosslinked into a gel [68]. If the latter process is done in an external field, a uniaxial ferrogel with a frozen-in magnetization is obtained [69]. Pleiner and Brand are interested in the unconventional macroscopic dynamic behavior of usual ferrofluids and, in particular, in the unusual behavior of unconventional "ferrofluids", like ferronematics and ferrogels.

Viewed as binary mixtures of magnetic particles and carrier fluid, ferrofluids have rather extreme properties. With the grain size being large on molecular length scales, the particle mobility (or concentration diffusivity) is extremely small (very small Lewis

number  $L$ ), allowing to disregard the concentration dynamics in most cases [70, 71]. However, this simplification does not hold for thermal convection, since due to the pronounced Soret effect of these materials in combination with a considerable solutal expansion (large separation ratio  $\psi$ ), the resulting solutal buoyancy forces are dominant. By considering the classical Rayleigh Bénard setup it is shown [72] that both the linear as well as the nonlinear convective behavior is significantly altered by the concentration field as compared to single-component systems. Starting from an initial motionless configuration with a uniform concentration distribution, convective perturbations are found to grow even at Rayleigh numbers well below the threshold  $Ra_c^0$  of pure-fluid convection. It turned out that the actual critical Rayleigh number  $Ra_c$  is drastically smaller, but experimentally inaccessible due to the extremely slow growth of convection patterns for  $Ra \gtrsim Ra_c$ , requiring extremely large observation times. On the other hand, operating the ferrofluid convection experiment at Rayleigh numbers  $Ra_c < Ra \lesssim Ra_c^0$ , reveals considerable positive growth rates, which lead to a saturated nonlinear state almost as fast as pure-fluid convection does at  $Ra > Ra_c^0$ .

In an external magnetic field the apparent imperfection of the bifurcation is even more pronounced. Magnetophoretic effects as well as magnetic stresses have been taken into account in the static and dynamic parts of the equations leading to rather pronounced boundary layer profiles (with respect to the concentration and magnetic potential). This boundary layer couples effectively to the bulk behavior due to the magnetic boundary condition [73].

Another interesting case are ferrofluids with negative separation ratio (negative Soret coefficient). When heating from below molecular binary mixture with negative separation ratio, the thermal and solutal density gradients are opposed such that the linear stationary thermal instability is suppressed for  $\psi < -1$ . Instead, this antagonistic behavior leads to a linear convective instability of the oscillatory type at  $Ra_c^0$ , the critical Rayleigh number for the onset of convection in the single fluid case. This feature is found for ferrofluids, too, but the nonlinear treatment shows that the linearly unstable oscillatory states are transients only and decay after some time, rendering the final convection-free state stable [74]. Above a second threshold, somewhat higher than  $Ra_c^0$ , a finite amplitude stationary instability is found, while small amplitude disturbances do not destroy the convection-free state. When heating from above molecular binary mixtures with negative separation ratio  $\psi < -1$ , a linear stationary instability is found, which is basically driven by the solutal buoyancy and only slightly modified by thermal variations. In ferrofluids, however, the concentration and temperature dynamics show completely different behavior. Thus, this stationary instability is very different from that obtained by heating from below with a positive separation ratio. In the former case small scale structures arise at very high Ra numbers, whose wavelength decreases strongly with increasing Ra. For smaller Ra numbers ( $|Ra| \sim Ra_c^0$ ) the procedure of using the separation of thermal conduction and concentration diffusion times, breaks down.

In the nonlinear domain the question of pattern formation and competition has been discussed numerically and using model amplitude equations for ferrofluids with positive and negative separation ratios [75].

## Ferronematics

If the carrier liquid is a nematogen, several phases are possible. Using a Landau-type free energy function one can describe the phase transitions from an isotropic (superparamagnetic) ferrofluid to a ferromagnetic nematic liquid crystal either directly or via a superparamagnetic nematic liquid crystal [76]. These two nematic phases are usually both called 'ferronematic', although they are distinct phases. Both show nematic ordering, but only the ferromagnetic phase shows spontaneous magnetic ordering, additionally. In the presence of a strong external magnetic field these transitions are smeared out and the different ferronematic phases become rather similar to each other. In nature no ferromagnetic ferronematic phase has been found until today, so the theoretical considerations have been restricted to the superparamagnetic variant.

In equilibrium the orientation of the nematic director and of the magnetization (induced by an external magnetic field) are locked. The macroscopic dynamics of ferronemat-



ics can be described on two different levels, either assuming the magnetization to be relaxed to its static value, or it can be treated as an additional dynamic (relaxing) degree of freedom. In the latter case the equations are structurally the same as for an ordinary nematic liquid crystal, except for the fact that the influence of the magnetic field is much more intense. However, in ordinary nematics magnetic field effects in the dynamics have never been discussed or detected, possibly due to their smallness. In ferronematics there is a good chance to find those effects, which come by various linear field (Hall-like) contributions to the fluxes [77]. They are of opposite thermodynamic signature (reversible/irreversible) compared to the field-free contributions. These new dynamic effects predicted come in four classes. First, the alignment of the nematic director in shear flow is modified by an external field such that the director has a component out of the shear plane, even if the field is in the shear plane. Second, the heat flux due to a temperature gradient, in a magnetic field orthogonal to the latter, induces an additional reversible heat current that is perpendicular to both. Third, a linear field contribution to viscosity leads, for a magnetic field orthogonal to the propagation direction of a sound wave, to a force on a tracer particle in the third direction. Fourth, when the director is reoriented in an external magnetic field, its relaxation is accompanied by an oscillation not observed in usual nematics.

Another possibility to probe the dynamic linear field contributions is the detection of their qualitatively new effects on some well-known instabilities [78]. In the Rayleigh-Bénard instability with the temperature gradient adverse to gravity one gets, in addition to convection flow in the form of one-dimensional rolls, a vorticity flow. As a consequence, in the homeotropic case (the director parallel to the field) the streamlines are oblique to the roll cross-section, while in the planar case (the director perpendicular to the magnetic, but parallel to an electric field) the rolls themselves are tilted with respect to the director depending on the magnetic field strength. In the Saffman-Taylor viscous fingering instability of a growing interface between fluids of different density, the new linear magnetic-field contributions lead to a rotation of the finger structure.

The complete set of macroscopic dynamic equations for ferronematics includes the magnetization as an independent slowly relaxing variable [79]. Orientational changes of the magnetization are coupled to nematic director reorientations not only in the statics, but in the dynamics as well. In addition, there are reversible and dissipative dynamic cross-couplings between (compressional, shear and elongational) flow, (rotations and changes of the absolute value of the) magnetization and director reorientations. Some of these couplings are only possible, when an external magnetic field is present. Some combinations of the parameters that describe these crosscouplings can be measured employing sound waves and the rheology of shear. For a sound wave propagating in a direction oblique to the preferred directions (equilibrium magnetization, nematic director) compressional flow (and changes of the absolute value of the magnetization) are coupled to shear flow (and rotations of the director and the magnetization). There is also a field dependent contribution to sound damping. In addition, the linear response of the system to oscillatory shear flow has been discussed concentrating on frequencies below the transverse magnetization relaxation frequency. This shows directly the influence of the magnetic dynamic degree of freedom on the director dynamics. Even without a magnetic field the modified nematic director diffusion couples to the flow and the apparent viscosity is different from the bare one. In the presence of an external field the director diffusion/relaxation is shifted to a finite frequency, which approximately increases with the third power of the field strength.

## Ferrogels

Due to the crosslinking in ferrogels a network is created that gives rise to elasticity. The truly hydrodynamic variable describing elasticity is the displacement field, or more appropriate for nonlinear theories, the strain tensor. Isotropic ferrogels are superparamagnetic and the magnetization is an additional independent slowly relaxing variable, which allows us to study the system for high frequencies as well [80]. The fact that magnetic grains are attached to the network is expressed by the static coupling of the magnetization and the strain tensor (magnetostriction). This leads to an additional field-dependent contribution to the sound spectrum. The contribution to the transverse sound modes depends on the relative angle between an external field and the wave vector. From the low frequency

limit of the sound spectrum one can obtain information about the effective, magnetic-field-dependent elastic moduli. However, these moduli are different from those measured by static elongations or shear deformations in an external field. The reason is that due to the finite magnetostriction the linear response theory is not applicable. Only in the limit of a vanishing field are they equal and match the true elastic moduli. In the high frequency limit one gets a shift in the sound velocities proportional to the dynamic coupling between the flow and the magnetization. This reflects the fact that the magnetization is an independent variable. Finally, a shear excitation experiment in an oscillating temperature gradient plus a gradient of the magnetic field has been proposed.

Ferromagnetic gels are uniaxial, if the frozen-in magnetization denotes the only preferred direction. Such materials are potentially very interesting for a variety of applications. Uniaxial magnetic gels show on the one hand similarities to other anisotropic gels, like nematic elastomers, and to isotropic ferrofluids and ferrogels, but the combination of preferred direction, magnetic degree of freedom, and elasticity makes them unique and very peculiar. Prominent features are [81] the relative rotations between the magnetization and the elastic network, which couple dynamically flow, shear, and magnetic reorientation. As a result, shear flow in a plane that contains the frozen-in magnetization induces a rotation of the magnetization, not only within the shear plane, but also out of the shear plane. This behavior is qualitatively different from that of other types of materials. The basic results hold, even if the constant shear flow is replaced by an oscillating one, which is very likely done in actual experiments, although the formulas for that case will become much more complicated. Another outstanding aspect of the hydrodynamics of this material is the difference between the mass current density (mass density times velocity) and the momentum density due to a nonvanishing magnetization vorticity. Unheard of in other classical condensed phases, it is known from some uniaxial quantum fluids, where, however, experiments on this aspect are impossible. In uniaxial ferromagnetic gels the static susceptibilities for momentum fluctuations (the long wavelength limit of the static momentum correlation functions) are given by the (bare) density for some geometries only, but show an increased renormalized effective density for other directions. Finally, an oscillating external magnetic field induces not only an oscillation of the magnetization in the direction of the external field, but also oscillating shear strains. The latter are found in planes that contain the frozen-in magnetization and either the external field or the third, perpendicular direction. In addition, the external field also induces a magnetization component perpendicular to both the field and the frozen-in magnetization. The reversible transport coefficient that governs this effect can be calculated by referring to the microscopic quantum mechanical spin-type dynamics for magnetic moments and using the projector formalism to evaluate the frequency matrix. This coefficient vanishes with the magnetization and is, thus, characteristic for this type of ferromagnetic gel.

Surface undulations of the free surface of viscous liquids are known to be able to propagate as gravity or capillary waves. In more complex systems like viscoelastic liquids or gels the transient or permanent elasticity allows for modified transverse elastic waves at free surfaces [82]. They are excited e.g. by thermal fluctuations or by imposed temperature patterns on the surface. In ferrofluids magnetic stresses at the surface come into play. In particular, in an external magnetic field normal to the surface there is a focusing effect on the magnetization at the wave crests of an undulating surface with the tendency to increase the undulations [16]. At a critical field strength no wave propagation is possible and the surface becomes unstable with respect to a stationary pattern of surface spikes (Rosensweig or normal field instability). The same linear instability mechanism is operative when dealing with (isotropic) ferrogels where elasticity comes into play as an additional stabilizing factor. Using linearized dynamic equations and appropriate boundary conditions one gets [83] the general surface wave dispersion relation for ferrogels (in a normal external field), which contains as special cases those for ferrofluids and non-magnetic gels and can be generalized to viscoelastic ferrofluids and magnetorheological fluids. A linear stability analysis reveals the threshold condition, above which stationary surface spikes grow. This critical field depends on gravity, surface tension and on the elastic (shear) modulus of the gel, while the critical wavelength of the emerging spike pattern is independent of the latter. As in the case of ferrofluids neither the threshold nor the critical wavelength depends on the viscosity.

A linear theory neither can determine the actual spike pattern, nor the true nature of the instability (forward, backward etc.). The standard weakly nonlinear (amplitude expansion) theory that provides suitable amplitude equations, by which those questions can be answered, is hampered in the present situation by two problems. First the driving force of the instability is manifest in the boundary conditions, but not in the bulk equations, and second the surface profile (the location where the boundary conditions have to be applied) changes with the order of the amplitude expansion. Thus, for ferrofluids a different path has been chosen [84]. Neglecting the viscosity (and any other dissipative process) from the beginning, the system is Hamiltonian and its stability governed by a free energy, more precisely by the surface free energy, since the magnetic destabilization acts at the surface. This approach is generalized to (uniaxial) ferrogels by taking into account the elastic free energy, additionally. The results have to be taken with the caveat that the neglect of the viscosity is justified at the (linear) instability threshold, but is an unproven assumption for the nonlinear domain and for the pattern forming and selecting process.

### 8.3 Microscopics of Magnetization Relaxation

The results of Berkov and co-workers are reported here, who studied the magnetization relaxation after switching off the external field, and showed that the magnetization decay time (rather the initial slope) rapidly increases with the particle concentration [85]. Such a behaviour results from the formation of clusters due to the magnetodipolar particle interaction. The average equilibrium size of clusters grows with the particle concentration (if the applied field magnitude is kept constant), so that after switching the field off the magnetization relaxation of the system is slower for larger concentrations. The concentration dependence of the ac-susceptibility  $\chi(w, T)$  of a ferrofluid is also studied. According to simulations, ac-susceptibility itself increases with the particle concentration  $c$  due to a collective response involving many particles (“dynamic clusters”). The peak of the  $Im(\chi(w))$ -dependence (for  $T = \text{const}$ ) is slightly shifted towards higher frequencies with increasing concentration. This behaviour is probably due to two competing trends: remagnetization of a cluster should occur slower than for a single particle (larger size), but demagnetizing interaction inside the clusters should lead to a faster magnetization relaxation. Comparing the above results with the corresponding dependencies obtained for rigid dipoles model (where the magnetic moment is fixed with respect to the particle), Berkov and coauthors demonstrated that for the magnetic anisotropy values typical for commonly used ferrofluid materials (like magnetite) inclusion of ‘magnetic’ degrees of freedom is essential to obtain a correct description of ferrofluid dynamics [86]. Using the theory of magnetization relaxation in fine magnetic particle systems consisting of mechanically fixed particles, a new and powerful method for the measurement of the energy barrier density in polydisperse ferrofluids and their characterization is developed [87, 88].

## A Legendre Transformations of Fields

To derive the Maxwell stress, some steps in section 3.1 involve the Legendre transformations  $u - \mathbf{E} \cdot \mathbf{D}$  and  $u - \mathbf{H} \cdot \mathbf{B}$ . They may not seem quite self-evident, because the constancy of the conjugate variable (say Temperature  $T$ ) is usually a crucial input. Yet only very few geometries sport a constant field. And the question is, are Legendre transformations generally valid for the field variables?

First we summarize the considerations of a usual Legendre Transformation, taking the free energy  $F$  as an example. The energy change of a closed system is  $dU = \int d^3r du = \int d^3r T ds = T \int d^3r ds = T dS$ , as  $T$  is a spatial constant in equilibrium. Two systems that come into thermal contact will exchange entropy,  $\Delta S_1 + \Delta S_2 = 0$ , to achieve equal temperature. If one of the two systems is much larger than the other – call them *bath* and *subsystem*, respectively – the bath temperature will not be changed by the contact,  $T_{bath} = T = \text{constant}$ , though its entropy will,  $\Delta S_{bath} + \Delta S = 0$ . (The quantities without a subscript belong to the subsystem.) So its energy may be written as a function of the subsystem’s variables,  $\Delta U_{bath} = T \Delta S_{bath} = -T \Delta S = -\Delta(TS)$ . The change in combined energy  $\Delta(U + U_{bath})$  is therefore

$$\Delta(U + U_{bath}) = \Delta(U - TS) = \int d^3r(u - Ts) = \Delta F(T), \quad (198)$$

the same as the change in the free energy, making it a very useful thermodynamic quantity.

This consideration does not appear transferable to field variables, as  $\mathbf{E}$  (unlike the temperature  $T$ ) is not generally constant in equilibrium. And our inability to write the energy change  $dU = \int d^3r \mathbf{E} \cdot d\mathbf{D}$  as an expression analogous to  $TdS$  stalls an analogous deduction at its very first step. To circumvent this difficulty, we partially integrate the expression,  $dU = \int d^3r \mathbf{E} \cdot d\mathbf{D} = \int d^3r \phi d\rho_\epsilon = \phi \int d^3r d\rho_\epsilon = \phi dQ$ , shifting the energy from the dielectric region to that of the charge carrying metal, where the potential  $\phi$  is a spatial constant. The charge  $Q$  is a conserved quantity, so  $\Delta Q_1 + \Delta Q_2 = 0$  holds when two pieces of metal are brought into contact. Now the analogy  $T \rightarrow \phi$ ,  $S \rightarrow Q$  works, and a Legendre transformed potential  $U - \phi Q$  makes sense, say for a smallish capacitor as the subsystem, and a very large one (or a good battery) as the bath that maintains the potential. Clearly,  $U - \phi Q$  is, as in the case of the heat bath, equal to the change of the combined energy,

$$\Delta(U + U_{bath}) = \Delta(U - \phi Q) = \int d^3r(u - \phi \rho_\epsilon) = \int d^3r(u - \mathbf{E} \cdot \mathbf{D}). \quad (199)$$

The last step, again involving a partial integration, shows that the combined change in energy density may also be written as  $(u - \mathbf{E} \cdot \mathbf{D})$ , which is therefore a perfectly healthy potential. In fact, if we insist on a potential density that is local valid, only  $\tilde{u} \equiv u - \mathbf{E} \cdot \mathbf{D}$  with  $d\tilde{u} = Tds + \mu d\rho - \mathbf{D} \cdot d\mathbf{E}$  will do.

For magnetic variables, there is a similar need to consider systems in which the current is confined to a certain region, say a coiled wire. As we need to consider a closed system in equilibrium maintaining a persistent current, we start with superconducting coils. Defining the magnetic flux as  $\Phi = \int \mathbf{B} \cdot d\mathbf{A} = \int \mathcal{A} \cdot d\mathbf{s}$ , where  $\mathcal{A}$  is the vector potential and  $\mathbf{s}$  the line element, we employ  $\nabla \times \mathbf{H} = \mathbf{j}_\epsilon/c$  to write the magnetic energy as

$$\int d^3r du = \int d^3r \mathbf{H} \cdot d\mathbf{B} = \int d^3r \mathbf{j}_\epsilon \cdot d\mathcal{A}/c = Jd\Phi/c. \quad (200)$$

Again, the second equal sign involves a partial integration, while the third takes the current  $J = \int \mathbf{j}_\epsilon \cdot d\mathbf{A}$ , obtained by integrating  $\mathbf{j}_\epsilon$  over the cross section of the wire, as a constant, because  $\nabla \cdot \mathbf{j}_\epsilon = \nabla \cdot (c\nabla \times \mathbf{H}) = 0$ . The thermodynamic relation  $dU = \dots + \phi dQ + Jd\Phi/c$  shows  $Q, \Phi$  are the quantities remaining constant in an isolated system, that the magnetic counterpart to the conserved charge  $Q$  is the flux  $\Phi$ , both remaining constant when the system is compressed or sheared – as is known to be true for superconductors.

Breaking the currents in both circuits and reconnecting them such that the current go through them consecutively may, in “thermodynamic talk,” may seen as: Two connected coils “exchanging flux  $\Phi$  to equalize their current  $J$ .” And there is also the possibility of a flux-bath that imposes its current onto a coil with far fewer windings.<sup>13</sup> So the replacements  $T \rightarrow J$ ,  $S \rightarrow \Phi$  are thermodynamically sound,

$$\Delta(U + U_{bath}) = \Delta(U - J\Phi/c) = \int d^3r(u - \mathbf{A} \cdot \mathbf{j}_\epsilon/c) = \int d^3r(u - \mathbf{H} \cdot \mathbf{B}), \quad (201)$$

and the potential  $\tilde{u}(\mathbf{H}) \equiv u - \mathbf{H} \cdot \mathbf{B}$  denotes the energy change of the total system.

Of course, we do not usually deal with superconducting coils. But we may substitute the flux-bath by a battery (that maintains a constant voltage, and because of the Ohm law also a constant current), the subsystem-coil by one made of normal metal. Concentrating on the physics outside the wires, where the magnetizable medium only probes the local field, not how it is generated, there is no reason why  $\tilde{u}(\mathbf{H}) \equiv u - \mathbf{H} \cdot \mathbf{B}$  does not yield a perfectly valid thermodynamic description. [The closed system, described by  $u(\mathbf{B})$ , needs to be emulated by an adjusting battery that maintains the flux.]

<sup>13</sup> The more windings, the larger the flux for a given current  $J$ . A coil with many windings, or a large  $\partial\Phi/\partial J$ , therefore corresponds to a system with a big heat capacity, a large  $\partial S/\partial T$ .

## B The General-Frame Thermodynamic Energy

The validity of Eq (142) is shown here directly by transforming the rest frame expression. More specifically, we demonstrate  $\partial(u - \mathbf{v} \cdot \mathbf{g})/\partial \mathbf{D} = \mathbf{E}_0$ , holding  $s$ ,  $\rho_\alpha$ ,  $\mathbf{v}$ , and  $\mathbf{B}$  constant. We start with the pleasingly simple expression

$$u = u_0(\mathbf{D}_0 \rightarrow \mathbf{D}, \mathbf{B}_0 \rightarrow \mathbf{B}) + \frac{1}{2}\rho v^2, \quad (202)$$

that is a result of the accidental cancellation of the terms from the first-order-Lorentz transformation with that of the Taylor expansion,

$$\begin{aligned} u(\mathbf{D}, \mathbf{B}) &= u_0(\mathbf{D}_0, \mathbf{B}_0) + 2\mathbf{v} \cdot \mathbf{g}_0 + \frac{1}{2}\rho v^2 \\ &= u_0(\mathbf{D}_0, \mathbf{B}_0) + 2\mathbf{v} \cdot (\mathbf{E} \times \mathbf{H})/c + \frac{1}{2}\rho v^2 \\ &= u_0(\mathbf{D}, \mathbf{B}) + \frac{1}{2}\rho v^2. \end{aligned}$$

Assuming LCR, or  $u = \frac{1}{2}(D^2/\epsilon + \rho v^2)$  with  $\mathbf{D}_0 = \epsilon \mathbf{E}_0$ , we have

$$\begin{aligned} u - \mathbf{v} \cdot \mathbf{g}^{tot} &= \frac{1}{2}(D^2/\epsilon - \rho v^2) - \mathbf{v} \cdot (\mathbf{E} \times \mathbf{H})/c \\ &= \frac{1}{2}(D^2/\epsilon - \rho v^2) - \mathbf{v} \cdot (\mathbf{D} \times \mathbf{H})/c\epsilon + \mathcal{O}(v^2/c^2), \end{aligned} \quad (203)$$

and deduce

$$\begin{aligned} \partial(u - \mathbf{v} \cdot \mathbf{g}^{tot})/\partial \mathbf{D} \\ = (\mathbf{D} + \mathbf{v} \times \mathbf{H}/c)/\epsilon = \mathbf{D}_0/\epsilon = \mathbf{E}_0. \end{aligned} \quad (204)$$

Higher order terms (such as one  $\sim D^4$  in the energy  $u$ ) do not invalidate this result. The terms in the magnetic field behave analogously.

In Eq (203), the explicit form of  $\mathbf{g}$  in the lab frame was employed to deduce the lab frame energy, Eq (142), from which then the lab frame energy flux, Eq (157), is deduced. This may appear as an inconsistency, but is not, because with the rest frame expression for  $\mathbf{g}_0$  given, we already know that the term  $\sim \mathbf{v}$  is from the rest mass. No detailed information about the energy flux is necessary here.

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