

## Free energies in one-dimensional models of magnetic transitions with hysteresis

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**Summary.** — A one-dimensional non-isothermal model for magnetic materials is proposed. It provides a simplified description of transitions from paramagnetic to either ferro- or ferri-magnetic phase which also accounts for hysteresis loops. The temperature enters the model as a parameter leading the transition, so that the compatibility with thermodynamics is ensured by the Clausius-Duhem inequality. Above the critical temperature, the paramagnetic susceptibility is assumed to obey a proper law depending on the material: the Curie-Weiss law for ferromagnets and the Néel-Curie-Weiss law for antiferromagnets and ferrimagnets. At a temperature below the critical point, a bilinear rate-independent o.d.e. rules the evolution of magnetization *versus* magnetic field strength. Because of the special form of its skeleton curve, the model applies to materials whose major hysteresis loop is not rectangular-shaped. In addition, the explicit form of the minimum and maximum free energies is obtained under isothermal conditions for the paramagnetic and hysteretic regimes. This allows us to highlight the amount of work performed on the system which is stored as magnetic energy change.

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### 1. – Introduction

Many important magnetic materials for practical purposes (such as iron, cobalt, nickel and their alloys) exhibit *spontaneous magnetization*. This is the term used to describe the appearance of an ordered spin state (magnetization) at zero applied magnetic field in a ferromagnetic or ferrimagnetic material below a critical point  $\theta_f$ , called *critical temperature*. At temperatures above  $\theta_f$ , the material is paramagnetic and its magnetic behavior is dominated by spin waves or magnons, which are boson collective excitations with energies in the meV range. The magnetization that occurs below  $\theta_f$  is a famous example of the *spontaneous* breaking of a global symmetry and it refers to the choice of

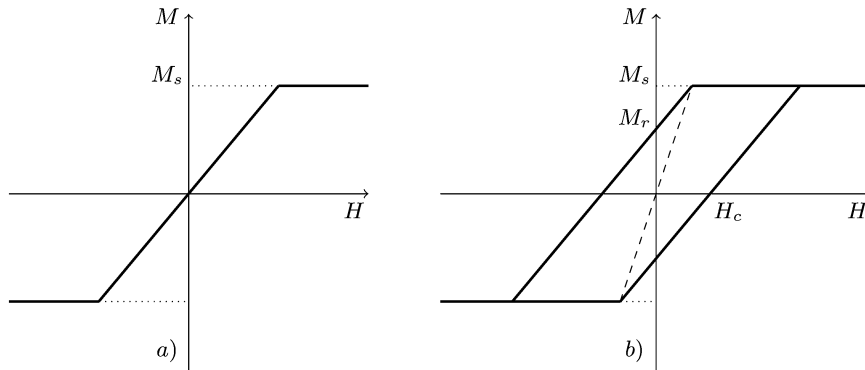


Fig. 1. – a) Paramagnetic regime:  $\theta > \theta_f$ . b) Magnetic hysteresis loop:  $\theta < \theta_f$ .

a magnetization direction by the spins, which have spherical symmetry above  $\theta_f$ , but a preferred axis (the magnetization direction) below  $\theta_f$ . See [1] for more details.

A material is named *ferromagnetic* if all of its magnetic ions lying on each sublattice add a positive contribution to the net magnetization. Ferromagnetic materials, such as iron, cobalt, and nickel have atomic moments that exhibit very strong interactions (due to exchange forces) and result in parallel alignment of atomic moments. This parallel alignment produces a large net magnetization, even in the absence of an applied field, giving rise to a spontaneous magnetization below a critical temperature called *ferromagnetic Curie temperature*.

*Ferrimagnetic* and *antiferromagnetic* materials, in contrast, have antiparallel atomic moments. Their behavior arises from the alignment of spin magnetic moments of electrons in the incompletely filled 3d subshell. Ferrimagnetic materials are commonly iron oxides with a spinel structure. Their sublattices consist of different ions (such as  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ), so that the opposing moments are unequal and a spontaneous magnetization remains. Since ferrimagnetic and antiferromagnetic materials have an uneven number of electrons, they can acquire a permanent magnetization, or remanence, after exposure to a magnetic field. Then, like ferromagnets, they hold a spontaneous magnetization below a critical temperature named *Néel temperature* [2]. In addition, they exhibit hysteresis loops below it and no magnetic order (namely, they are paramagnetic) above it (see, for instance [3-5]).

The hysteretic phenomena, which appear in magnetic materials below the characteristic critical temperature  $\theta_f$ , mean that the relation between the magnetization  $M$  and the external magnetic field  $H$  cannot be expressed in terms of a single-valued function. More precisely, the pair  $(H, M)$  moves along a continuous curve  $\mathcal{C}$  with parametrization

$$t \mapsto (H(t), M(t)), \quad t \in [0, T],$$

which changes according as  $H$  is increasing or decreasing. When the temperature overcomes the threshold value  $\theta_f$ , as a result of thermal agitation, hysteresis disappears and the material becomes paramagnetic, namely the relation between  $M$  and  $H$  becomes a single-valued function. A schematic representation of this behavior is depicted in fig. 1, where  $M_s$  stands for the *saturation magnetization*.

Magnetic materials which are easily magnetized and demagnetized are called *soft*. They exhibit a narrow major hysteresis loop, where both *residual* (or *spontaneous*)

*magnetization*  $M_r$  and *coercive magnetic field*  $H_c$  take small values. The hysteresis loss is measured by the area of the major loop which is proportional to the magnetic product  $H_c M_r$ . Hence, magnetic materials which have small hysteresis loss are soft. By comparison, materials which neither magnetize nor demagnetize easily are called *hard*.

**Aim and plan of the paper.** In this paper we present and scrutinize a one-dimensional thermodynamic model describing temperature-induced transitions in magnetic materials which also accounts for hysteresis loops. For simplicity, the saturation magnetization  $M_s$ , which occurs for large values of the applied magnetic field, is assumed to be temperature independent. Depending on the special choice of the involved material parameters as temperature functions, either ferro-paramagnetic or ferri-paramagnetic transitions are described in sect. 5. Above the critical temperature, the paramagnetic susceptibility obeys standard laws: the Curie-Weiss law for ferromagnets and the Néel-Curie-Weiss law for antiferromagnets and ferrimagnets (see, *e.g.* [1]).

The temperature enters the model as a parameter leading the transition, so that the second law of thermodynamics is verified in the form of the Clausius-Duhem inequality. Starting from this inequality, in sect. 4 we establish the existence of infinitely many magnetic energy functions. Then, we construct their explicit expression and discuss their physical meaning in the ferromagnetic regime. Most papers in the literature consider paramagnetic-ferromagnetic transitions into the framework of Landau's theory. Although this approach turned out to be very fruitful, it is unable to capture the multi-valued structures of the hysteresis loops. Indeed, its basic assumption is the introduction of a fourth-order energy function, whereas in materials with hysteresis infinitely many magnetic energies occur.

Our analysis heavily relies on the complete controllability property of the dynamical system ruling the evolution of the model and describing the hysteresis loops (see sect. 3). This forced us to restrict our attention to the special class of *Duhem models*, which provides a simplified non-isothermal description of the hysteretic behavior. In sect. 2 we briefly discuss the main features of this class of models and their difference with respect to history-differential models. In particular, a bilinear rate-independent o.d.e. is assumed to rule the evolution of magnetization *versus* magnetic field strength at a fixed temperature below the critical point. Since the resulting *skeleton curve* (see fig. 2) is represented either by a single-valued increasing function, or by the inverse of a maximal monotone graph (in the limit case), the model turns out to be well-fitting if applied to materials whose major hysteresis loop is not rectangular-shaped. This is the case when soft ferri- and ferromagnets are taken into account, for instance nichel (Ni) and nichel iron alloys ( $\text{NiFe}_2\text{O}_4$ ).

It is worth noting that all the results proved in this paper can be easily extended to more realistic Duhem models (see, *e.g.* [6, 7]).

## 2. – Isothermal models

For the sake of simplicity, we shall consider just uniaxial (*i.e.* one-dimensional) evolutionary hysteretic phenomena, so that the magnetic field  $H$  and the magnetization  $M$  will be treated as scalar fields. In addition, henceforth the dependence on the space variable is understood and not written. As usual, the magnetic induction  $B$  is given by

$$(1) \quad B = \mu_0 H + M,$$

where the constant  $\mu_0$  denotes the magnetic permeability in the vacuum.

The *hysteresis* phenomenon is a memory effect where the pair  $(H(t), M(t))$  moves along a continuous curve  $\mathcal{C}$  in the  $(H, M)$ -plane and the magnetization  $M(t)$  is determined by the magnetic field through its history up to time  $t$ ,

$$H^t(s) = H(t - s), \quad s \in \mathbb{R}^+.$$

Hysteresis distinguishes from other memory effects because it exhibits *permanent memory*. This means that the initial value of the magnetization is permanently retained and, although it might partially fade in time, it will affect the actual magnetization value even after a constant magnetic history.

In addition, hysteresis obeys the *rate-independent property*, namely  $M(t)$  must depend just on the range of the history  $H^t$  of the magnetic field and on the order in which its values  $H^t(s)$  are taken, but is independent of the speed at which such values follow one another. In other words, a model of hysteresis is rate independent if the scaling

$$\hat{H}(\alpha t) \mapsto H(t), \quad \hat{M}(\alpha t) \mapsto M(t), \quad \alpha > 0,$$

leaves unchanged the graph of the curve  $\mathcal{C}$  and merely transforms its parametrization into  $\tau \mapsto (\hat{H}(\tau), \hat{M}(\tau))$ ,  $0 \leq \tau \leq \alpha T$ . As a consequence, it can be shown that  $M(t)$  depends on  $H^t$  only through

$$\tilde{H}^t = (\tilde{H}_1, \tilde{H}_2, \dots, \tilde{H}_n, \dots),$$

the sequence of the local maxima and minima of  $H^t$  (cf. [8, Chap. 11]).

Magnetic materials exhibiting hysteretic phenomena may be described through various kinds of mathematical models [9, 10, 8, 11, 12]. Henceforth, we consider two special classes of them where the path  $(H(t), M(t))$  is a solution of some differential system.

**2.1. History-differential models.** – This class of models, usually referred to as *generalized Duhem models* (see, e.g., [8, 13, 12]), follows a phenomenological approach and is ruled by a rate-type differential equation

$$(2) \quad \dot{M}(t) = \tilde{F}(H(t), M(t), \tilde{H}^t, \dot{H}(t)),$$

where the triplet  $m = (H, M, \tilde{H})$  is named *magnetic state* and the superposed dot stands for the time derivative.

Some examples of history-differential models are devised in [8]. In particular, the simplest case has been proposed in [13] assuming that all the “memory” of the magnet is resumed into the first value  $\tilde{H}_1$  of the sequence  $\tilde{H}^t$  and the magnetic state  $m$  is given by the triplet  $(H, M, \tilde{H}_1)$ . In this case, each inversion point *wipes out* all the previous one, so that only the first term of the sequence may influence the behavior of the material.

Since  $\tilde{H}_1$  is the value of the magnetic field corresponding to the last inversion point where the sign of the magnetic rate  $\dot{H}$  changed, it follows

$$\operatorname{sgn}[H(t) - \tilde{H}_1] = \operatorname{sgn} \dot{H}(t), \quad \operatorname{sgn} P = \begin{cases} 1 & \text{if } P > 0, \\ 0 & \text{if } P = 0, \\ -1 & \text{if } P < 0. \end{cases}$$

Therefore, the rate-independence property yields (see [13])

$$(3) \quad \tilde{F}(H, M, \dot{H}) = \tilde{G}(H, M, \tilde{H}_1, \text{sgn}[H - \tilde{H}_1])\dot{H}.$$

Depending on the value of  $\tilde{H}_1$ , there are infinitely many paths passing through each point of the  $(H, M)$ -plane. This property is typical of the history-differential models.

In spite of the simple form of its evolution equation, the corresponding differential system is not easy to handle. As a consequence, only a few characteristic properties have been hardly proved in its connection [8, 13]. For this reason, we are unable to provide for this model results concerning the explicit expression of the magnetic energy. In particular, the complete controllability of the system is out of reach, at present.

**2.2. Duhem models.** – Classical Duhem models (see, *e.g.*, [10, 8, 12]) are ruled by a rate-type differential equation

$$(4) \quad \dot{M} = F(H, M, \dot{H}),$$

where  $F$  is a function regular enough. They are useful in applied electromagnetics because all functions and parameters involved can be properly chosen to match experimental results. In particular, these models fit well for magnetic materials exhibiting narrow hysteresis loops, such as soft materials (see, *e.g.*, [9, 6, 7]). The property of rate-independence in connection with eq. (4) reads

$$(5) \quad F(H, M, \alpha\dot{H}) = \alpha F(H, M, \dot{H}), \quad \alpha > 0.$$

This allows us to infer that there exists a function  $G$ , called *differential susceptibility* and depending on  $(M, H, \text{sgn } \dot{H})$ , such that

$$F(H, M, \dot{H}) = G(H, M, \text{sgn } \dot{H})\dot{H}.$$

This result easily follows from (5) by letting  $G(H, M, \text{sgn } \dot{H}) = F(H, M, \text{sgn } \dot{H}) \text{sgn } \dot{H}$ . As a consequence, (4) can be written in the form

$$(6) \quad \frac{dM}{dH} = G(H, M, \text{sgn } \dot{H}),$$

which represents the slope of the curve  $\mathcal{C}$  at  $(H, M)$ . Such a slope is well defined at each point where  $\dot{H}$  does not change its sign. Unlike history-differential models, here each point of the  $(H, M)$ -plane lies just on two solution-curves: one related to  $\dot{H} > 0$  (*loading curve*), and the other to  $\dot{H} < 0$  (*unloading curve*).

**2.3. A bilinear model.** – In this paper, we restrict our attention to a bilinear Duhem model for soft magnetic materials devised in [8]. The related hysteresis major loop looks like the picture in fig. 2, where the residual magnetization  $M_r$  and the coercive magnetic field  $H_c$  are, respectively, defined as

$$M_r = \sup\{M : (0, M) \in \Sigma\}, \quad H_c = \sup\{H : (H, 0) \in \Sigma\},$$

where  $\Sigma$  is the closed region bounded from the major hysteresis loop.

The slope of the oblique branches of  $\Sigma$  is denoted by  $\chi$  and represents the *magnetic susceptibility*, whereas  $\kappa$  is the slope of the oblique branch of the dashed curve. They are

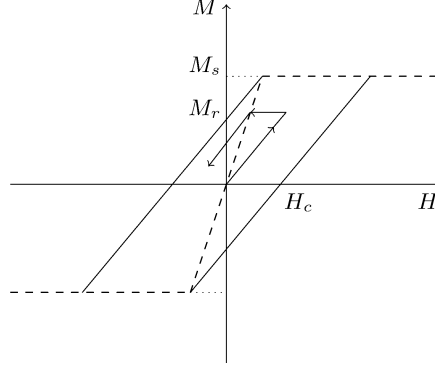


Fig. 2. – Major loop (continuous), skeleton curve (dashed) and hysteresis path (arrowhead).

both positive and  $\kappa > \chi > 0$ . Therefore, we have

$$(7) \quad \nu = \frac{\kappa}{\kappa - \chi} \chi > 0.$$

In order to write the constitutive function  $G$ , we introduce the following sets:

$$\begin{aligned} \Sigma_0 &= \{(H, M) \in \mathbb{R}^2 : M = \kappa H, |M| < M_s\}, \\ \Sigma_1^+ &= \left\{ (H, M) \in \mathbb{R}^2 : M = M_s, H > \frac{M_s}{\kappa} \right\}, \\ \Sigma_1^- &= \left\{ (H, M) \in \mathbb{R}^2 : M = -M_s, H < -\frac{M_s}{\kappa} \right\}, \\ \Sigma_2^+ &= \left\{ \left( \frac{M_s}{\kappa}, M_s \right) \right\}, \quad \Sigma_2^- = \left\{ \left( -\frac{M_s}{\kappa}, -M_s \right) \right\}, \\ \Sigma_3^+ &= \left\{ (H, M) \in \mathbb{R}^2 : M < M_s, \frac{M}{\kappa} < H \leq \frac{1}{\chi} M + \frac{1}{\nu} M_s \right\}, \\ \Sigma_3^- &= \left\{ (H, M) \in \mathbb{R}^2 : M > -M_s, \frac{1}{\chi} M - \frac{1}{\nu} M_s \leq H < \frac{M}{\kappa} \right\}. \end{aligned}$$

Henceforth, we take advantage of the decomposition  $\Sigma = \Sigma^* \cup \Sigma_3^+ \cup \Sigma_3^-$ , where the piecewise linear curve  $\Sigma^* = \Sigma_0 \cup \Sigma_1^+ \cup \Sigma_1^- \cup \Sigma_2^+ \cup \Sigma_2^-$  is called *skeleton curve* and depends only on  $M_s$  and  $\kappa$ . As depicted in fig. 2, the skeleton curve is a single-valued monotone curve. Hence, this model description fits well only a special class of magnetic materials, called *soft*, whose major hysteresis loop is not rectangular-shaped.

By virtue of rate independence, the hysteresis path is ruled by eq. (6), where

$$(8) \quad G(H, M, \text{sgn } \dot{H}) = \begin{cases} \chi, & \text{if } (H, M) \in \Sigma_0 \text{ or} \\ & (H, M) \in \Sigma_2^- \cup \Sigma_3^+ \text{ and } \text{sgn } \dot{H} = 1 \text{ or} \\ & (H, M) \in \Sigma_2^+ \cup \Sigma_3^- \text{ and } \text{sgn } \dot{H} = -1, \\ 0, & \text{otherwise.} \end{cases}$$

This hysteretic system is usually referred to as *bilinear system* [14].

In the sequel, we restrict our attention to this simplified model in order to construct explicit expressions of the magnetic energy and model temperature-induced paramagnetic-ferromagnetic transitions. Nevertheless, all results presented here can be extended to any Duhem model by conveniently adapting our arguments. The key point to do this is represented by the complete controllability of the system (see [15]), which is quite easy to prove for this class of models.

### 3. – Complete controllability

This section is devoted to establish the complete controllability of the bilinear system. To this end, we first need to introduce the concepts of (isothermal) *magnetic process* and *state* of the linked dynamical system.

A *magnetic process*  $\pi$  is a map  $\pi : [0, d_\pi) \rightarrow \mathbb{R}$ , which is piecewise continuous on the time interval  $[0, d_\pi)$ ,  $d_\pi > 0$ , and changes its sign at most a finite number of times. The number  $d_\pi$  denotes the finite duration of  $\pi$ . Henceforth, we set

$$\pi(t) = \dot{H}(t), \quad t \in [0, d_\pi).$$

Since our model is rate independent, we are allowed to consider the set  $\Pi$  of all magnetic processes as composed only by piecewise constant functions. As customary in all Duhem models, the local state of the magnetic material is characterized by the pair

$$\sigma = (H, M) \in \Sigma.$$

Then, given any magnetic process  $\pi \in \Pi$  and any initial state  $\sigma_0 = (H_0, M_0)$ , one can determine the state evolution at time  $t$ ,  $\sigma(t) = (H(t), M(t))$ , by solving the Cauchy problem

$$(9) \quad \begin{cases} \dot{H}(t) = \pi(t), \\ \dot{M}(t) = G(H(t), M(t), \operatorname{sgn} \pi(t))\pi(t), \\ H(0) = H_0, \\ M(0) = M_0. \end{cases}$$

In particular, we have

$$(10) \quad \begin{aligned} H(t) &= H_0 + \int_0^t \pi(s) ds, \\ M(t) &= M_0 + \int_0^t G(H(s), M(s), \operatorname{sgn} \pi(s))\pi(s) ds. \end{aligned}$$

The *state evolution function*,  $\zeta : \Sigma \times \Pi \rightarrow \Sigma$ , is then defined as

$$\zeta(\sigma_0, \pi) = \sigma_\pi,$$

where  $\sigma_\pi = (H(d_\pi), M(d_\pi))$  is computed from (10) by letting  $t = d_\pi$ .

*Proposition 3.1.* *System (9) is completely  $\zeta$ -controllable, that is to say, for any given  $\sigma_1, \sigma_2 \in \Sigma$  there exists a process  $\pi \in \Pi$  such that  $\zeta(\sigma_1, \pi) = \sigma_2$ .*

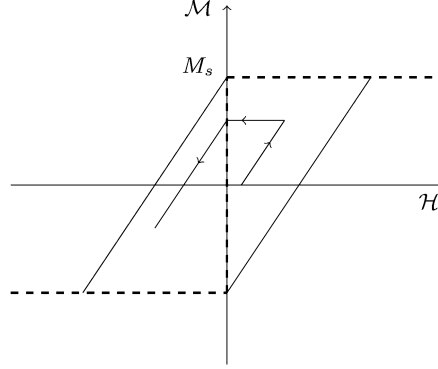


Fig. 3. – Paths of hysteresis and the skeleton curve (dashed).

The proof of this result is very technical, because a lot of parameters enter the model description. Therefore, we transform the original Cauchy problem (9) into a dynamical system involving just two parameters,  $M_s$  and  $\nu$  as defined in (7). To this end, we introduce the following change of variables:

$$(11) \quad \begin{cases} \mathcal{H} = H - \frac{1}{\kappa}M, \\ \mathcal{M} = M. \end{cases}$$

As represented in fig. 3, the new skeleton curve is not a monotone function, but the inverse of a maximal monotone graph. There,  $\nu$  is the slope of the oblique branches.

As before, we introduce the splitting  $\mathcal{S} = \mathcal{S}^* \cup \mathcal{S}_3^+ \cup \mathcal{S}_3^-$ , where  $\mathcal{S}^* = \mathcal{S}_0 \cup \mathcal{S}_1^+ \cup \mathcal{S}_1^- \cup \mathcal{S}_2^+ \cup \mathcal{S}_2^-$  is the graph of the new skeleton curve, and

$$\begin{aligned} \mathcal{S}_0 &= \{(\mathcal{H}, \mathcal{M}) \in \mathbb{R}^2 : \mathcal{H} = 0, |\mathcal{M}| < M_s\}, \\ \mathcal{S}_1^+ &= \{(\mathcal{H}, \mathcal{M}) \in \mathbb{R}^2 : \mathcal{M} = M_s, \mathcal{H} > 0\}, \\ \mathcal{S}_1^- &= \{(\mathcal{H}, \mathcal{M}) \in \mathbb{R}^2 : \mathcal{M} = -M_s, \mathcal{H} < 0\}, \\ \mathcal{S}_2^+ &= \{(0, M_s)\}, \quad \mathcal{S}_2^- = \{(0, -M_s)\}, \\ \mathcal{S}_3^+ &= \{(\mathcal{H}, \mathcal{M}) \in \mathbb{R}^2 : |\mathcal{M}| < M_s, 0 < \mathcal{H} \leq \frac{1}{\nu}(\mathcal{M} + M_s)\}, \\ \mathcal{S}_3^- &= \{(\mathcal{H}, \mathcal{M}) \in \mathbb{R}^2 : |\mathcal{M}| < M_s, \frac{1}{\nu}(\mathcal{M} - M_s) \leq \mathcal{H} < 0\}. \end{aligned}$$

Then, eq. (6) transforms into  $\dot{\mathcal{M}} = \mathcal{G}(\mathcal{H}, \mathcal{M}, \text{sgn } \dot{\mathcal{H}}) \dot{\mathcal{H}}$ , where

$$(12) \quad \mathcal{G}(\mathcal{H}, \mathcal{M}, \text{sgn } \dot{\mathcal{H}}) = \begin{cases} \nu, & \text{if } (\mathcal{H}, \mathcal{M}) \in \mathcal{S}_0 \text{ or} \\ & (\mathcal{H}, \mathcal{M}) \in \mathcal{S}_2^- \cup \mathcal{S}_3^+ \text{ and } \text{sgn } \dot{\mathcal{H}} = 1 \text{ or} \\ & (\mathcal{H}, \mathcal{M}) \in \mathcal{S}_2^+ \cup \mathcal{S}_3^- \text{ and } \text{sgn } \dot{\mathcal{H}} = -1, \\ 0, & \text{otherwise.} \end{cases}$$



Accordingly, from (11) it follows that  $\dot{\mathcal{H}} = \gamma(H, M, \text{sgn } \dot{H})\dot{H}$ , with

$$\gamma(H, M, \text{sgn } \dot{H}) = \begin{cases} \frac{\kappa}{\kappa + \nu}, & \text{if } (H, M) \in \Sigma_0 \text{ or} \\ & (H, M) \in \Sigma_2^- \cup \Sigma_3^+ \text{ and } \text{sgn } \dot{H} = 1 \text{ or} \\ & (H, M) \in \Sigma_2^+ \cup \Sigma_3^-, \text{ and } \text{sgn } \dot{H} = -1, \\ 1, & \text{otherwise.} \end{cases}$$

As a consequence,  $\dot{\mathcal{H}}$  turns out to be piecewise continuous and changes sign at most a finite number of times, as well as  $\dot{H}$ . Finally, since the map  $(H, M) \mapsto (\mathcal{H}, \mathcal{M})$ , as defined by (11), is a bijection, we are allowed to consider either  $(H, M)$  or  $(\mathcal{H}, \mathcal{M})$  as the magnetic state of the material, and either  $\dot{H}$  or  $\dot{\mathcal{H}}$  as the magnetic process. In particular, we can identify  $\mathcal{P} = \Pi$ .

Given any initial state  $m_0 = (\mathcal{H}_0, \mathcal{M}_0) \in \mathcal{S}$  and any process  $p \in \mathcal{P}$ , we can express the solution of the Cauchy problem

$$(13) \quad \begin{cases} \dot{\mathcal{H}} = p, \\ \dot{\mathcal{M}} = \mathcal{G}(\mathcal{H}, \mathcal{M}, \text{sgn } p)p, \\ \mathcal{H}(0) = \mathcal{H}_0, \\ \mathcal{M}(0) = \mathcal{M}_0 \end{cases}$$

as the pair  $m(t) = (\mathcal{H}(t), \mathcal{M}(t))$  given by

$$(14) \quad \begin{aligned} \mathcal{H}(t) &= \mathcal{H}_0 + \int_0^t p(s) ds, \\ \mathcal{M}(t) &= \mathcal{M}_0 + \int_0^t \mathcal{G}(\mathcal{H}(s), \mathcal{M}(s), \text{sgn } p(s)) p(s) ds. \end{aligned}$$

Henceforth, the corresponding state evolution function  $\rho : \mathcal{S} \times \mathcal{P} \rightarrow \mathcal{S}$  is defined as

$$\rho((\mathcal{H}_0, \mathcal{M}_0), p) = (\mathcal{H}(d_p), \mathcal{M}(d_p)).$$

It is easy to check that the *complete  $\zeta$ -controllability* of (9) in  $\Pi$  is perfectly equivalent to the *complete  $\rho$ -controllability* of (13) in  $\mathcal{P}$ . Accordingly, Proposition 3.1 holds true if we prove the following

*Proposition 3.2. The dynamical system (13) is completely controllable in  $\mathcal{P}$ .*

The first step to show the complete controllability of system (13) consists in exhibiting a process  $p$  connecting every pair of states  $(\mathcal{H}_1, \mathcal{M}_1)$ ,  $(\mathcal{H}_2, \mathcal{M}_2)$  on the skeleton curve  $\Sigma^*$ . This is achieved in Appendix A (see Lemma A.1) and the proof of Proposition 3.2 is completed in Appendix B.

#### 4. – Minimum and maximum ferromagnetic energies

In this section we restrict our attention to uniform and isothermal one-dimensional processes, namely  $\nabla\theta = \mathbf{0}, \dot{\theta} = 0$ . If this is the case, absolute temperature enters the

constitutive relations just as a parameter and the local Clausius-Duhem inequality (see, for instance, [16]) reduces to the so-called *electromagnetic dissipation inequality*

$$\dot{\psi}(t) \leq E(t) \dot{D}(t) + H(t) \dot{B}(t) + J(t) E(t),$$

where  $\psi$  is the free-energy density per unit volume,  $D$  the electric displacement and  $J$  the electric current. Disregarding all effects due to the electric field (*i.e.* assuming  $D = J = 0$ ) and taking advantage of (1), we obtain

$$(15) \quad \dot{\psi}(t) \leq \mu_0 H(t) \dot{H}(t) + H(t) \dot{M}(t).$$

In the paramagnetic regime (see fig. 1a),  $M$  is piecewise linear,

$$M = \begin{cases} \chi H, & \text{if } -\frac{M_s}{\chi} \leq H \leq \frac{M_s}{\chi}, \\ M_s, & \text{otherwise,} \end{cases}$$

and all magnetic paths are reversible. As a consequence, the *magnetic energy potential*  $\psi$  is uniquely determined to within a constant. Assuming the normalizing condition  $\psi(0, 0) = 0$ , it reads

$$(16) \quad \psi(H, M) = \frac{\mu_0}{2} H^2 + \frac{1}{2\chi} M^2.$$

On the contrary, because of the multi-valued relation between  $M$  and  $H$ , as described in sect. 2, in the ferromagnetic regime there exists an uncountable set of magnetic energy densities  $\psi$ , all of which are normalized at  $(0, 0)$  and satisfy the dissipation inequality (15). They are referred to as *magnetic energy subpotentials* for the given dynamical system (see, for instance, [15]). For simplicity, in the sequel we consider the bilinear dynamical system (6)-(8), although what follows holds true with minor changes for all Duhem models.

The rigorous definition of a magnetic subpotential for the bilinear system (9) requires the notion of *magnetic work* expended to reach the final state  $\sigma_\pi$  starting from the initial state  $\sigma_0$  by virtue of a process  $\pi$ , namely

$$w(\sigma_0, \pi) = \int_0^{d_\pi} H(t) \dot{B}(t) dt = \int_0^{d_\pi} H(t) [\mu_0 \dot{H}(t) + \dot{M}(t)] dt,$$

where the pair  $(H(t), M(t))$  is given by (10). Thus, we have

$$(17) \quad w(\sigma_0, \pi) = \left[ \frac{\mu_0}{2} H^2(t) \right]_0^{d_\pi} + \int_0^{d_\pi} H(t) \dot{M}(t) dt.$$

*Definition 4.1.* A function  $\psi : \Sigma \rightarrow \mathbb{R}$  is said a *magnetic subpotential* of the dynamical system (9) if it satisfies the following conditions:

- i)  $\psi(0, 0) = 0$ ;
- ii) for any given  $\sigma_1, \sigma_2 \in \Sigma$  and  $\pi \in \Pi$  such that  $\zeta(\sigma_1, \pi) = \sigma_2$ , then

$$\psi(\sigma_2) - \psi(\sigma_1) \leq w(\sigma_1, \pi).$$

In the paramagnetic regime this inequality holds as an equality and the magnetic energy potential (16) is strictly related to the expended magnetic work, in that

$$w((0, 0), \pi) = \frac{\mu_0}{2} H^2 + \frac{1}{2\chi} M^2.$$

For further convenience, we stress that a subpotential  $\psi$  needs not to be continuous. Furthermore, we denote by  $\Psi$  the set of all magnetic subpotentials  $\psi$  satisfying Definition 4.1 and we let

$$\begin{aligned} \overleftarrow{\Pi}_{(H, M)} &= \{\pi \in \Pi : \zeta((H, M), \pi) = (0, 0)\}, \\ \overrightarrow{\Pi}_{(H, M)} &= \{\pi \in \Pi : \zeta((0, 0), \pi) = (H, M)\}, \end{aligned}$$

which represent, respectively, the set of all processes driving the system from  $(H, M)$  to  $(0, 0)$  and vice versa. As will be shown in Appendix C, both  $\overleftarrow{\Pi}_{(H, M)}$  and  $\overrightarrow{\Pi}_{(H, M)}$  are non-empty sets. According to [15], this is enough to conclude that there exist the minimum and maximum magnetic energy subpotentials, defined as follows:

$$(18) \quad \psi_{\min}(H, M) = \sup_{p \in \overleftarrow{\Pi}_{(H, M)}} [-w((H, M), \pi)] = - \inf_{p \in \overleftarrow{\Pi}_{(H, M)}} w((H, M), \pi),$$

$$(19) \quad \psi_{\max}(H, M) = \inf_{p \in \overrightarrow{\Pi}_{(H, M)}} w((0, 0), \pi).$$

The function  $\psi_{\min}$  represents the *maximum amount of work* which may be extracted from the system when moving it from  $(H, M)$  to  $(0, 0)$ , whereas  $\psi_{\max}$  represents the *minimum amount of storage energy* which is required to supply the system when attaining  $(H, M)$  from  $(0, 0)$ . In general (see [15] and Corollary 4.3),  $\Psi$  is a convex set and

$$\psi_{\min} \leq \psi \leq \psi_{\max},$$

for any subpotential  $\psi \in \Psi$ . In the paramagnetic regime,  $\overleftarrow{\Pi}_{(H, M)} = -\overrightarrow{\Pi}_{(H, M)}$  because of reversibility, and  $\psi_{\min} = \psi_{\max}$  so yielding the uniqueness of  $\psi$ .

**4.1. Explicit expressions of  $\psi_{\min}$  and  $\psi_{\max}$ .** – We take advantage here from the change of variables (11) which simplifies the evolution of the system. Indeed, we first transform the expression of the expended work  $w(\sigma_0, \pi)$  by means of (11). Then we achieve the expression of the minimum and maximum magnetic energy in terms of  $(\mathcal{H}, \mathcal{M})$  and finally, by reverting (11), we find out the expression of  $\psi_{\min}$  and  $\psi_{\max}$  in terms of  $(H, M)$ .

The work  $w(\sigma_0, \pi)$  in (17) is contributed by two terms. It is convenient to express both of them in terms of  $\mathcal{H}$  and  $\mathcal{M}$ , namely

$$w(m_0, p) = \left[ \frac{\mu_0}{2} \left( \mathcal{H}(t) + \frac{1}{\kappa} \mathcal{M}(t) \right)^2 + \frac{1}{2\kappa} \mathcal{M}^2(t) \right]_0^{d_p} + \int_0^{d_p} \mathcal{H}(t) \dot{\mathcal{M}}(t) dt,$$

where  $\mathcal{H}(t), \mathcal{M}(t)$  are given by (14). This work expression can be split into the sum of a conservative and a dissipative term, namely

$$(20) \quad w(m_0, p) = w_0(m_0, m(d_p)) + w_D(m_0, p),$$

where

$$(21) \quad w_D(m_0, p) = \int_0^{d_p} \mathcal{H}(t) \dot{\mathcal{M}}(t) dt,$$

$$(22) \quad w_0(m_0, m(d_p)) = \left[ \frac{\mu_0}{2} \left( \mathcal{H}(t) + \frac{1}{\kappa} \mathcal{M}(t) \right)^2 + \frac{1}{2\kappa} \mathcal{M}^2(t) \right]_0^{d_p}.$$

It is worth noting that  $w$  is dissipative. Indeed, for every close cycle  $p \in \mathcal{P}$  such that  $\rho(m_0, p) = m_0$ , in view of (12) and (20), we have

$$w(m_0, p) = w_D(m_0, p) = \oint_0^{d_p} \mathcal{G}(\mathcal{H}(t), \mathcal{M}(t), \text{sgn } \dot{\mathcal{H}}(t)) \mathcal{H}(t) \dot{\mathcal{H}}(t) dt \geq 0.$$

*Theorem 4.2.* The explicit expressions of  $\psi_{\min}$  and  $\psi_{\max}$  are given by

$$\begin{aligned} \psi_{\min}(\mathcal{H}, \mathcal{M}) &= \frac{\mu_0}{2} \left[ \mathcal{H} + \frac{1}{\kappa} \mathcal{M} \right]^2 + \frac{1}{2\kappa} \mathcal{M}^2, \\ \psi_{\max}(\mathcal{H}, \mathcal{M}) &= \begin{cases} \psi_{\min}(\mathcal{H}, \mathcal{M}) & \text{if } (\mathcal{H}, \mathcal{M}) \in \mathcal{S}^*, \\ \psi_{\min}(\mathcal{H}, \mathcal{M}) + \frac{\nu}{2} \mathcal{H}^2 & \text{if } (\mathcal{H}, \mathcal{M}) \in \mathcal{S}_3^+ \cup \mathcal{S}_3^-. \end{cases} \end{aligned}$$

*Corollary 4.3.* All  $\psi \in \Psi$  satisfy  $\psi_{\min} \leq \psi \leq \psi_{\max}$  and can be written in the form  $\psi = \psi_{\min} + \varphi$ , where  $\varphi$  satisfies the following conditions:

- a)  $\varphi(m) = 0$  if  $m = (\mathcal{H}, \mathcal{M}) \in \mathcal{S}^*$ ;
- b)  $\varphi(m_2) - \varphi(m_1) \leq w_D(m_1, p)$ , for all  $m_1, m_2 \in \mathcal{S}$  such that  $\rho(m_1, p) = m_2$ ;
- c) for any  $\mathcal{M} \in [-M_s, M_s]$ , the function  $\varphi(\cdot, \mathcal{M})$  is increasing when  $\mathcal{H} \geq 0$  and decreasing when  $\mathcal{H} \leq 0$ .

The proof of Theorem 4.2 and Corollary 4.3 are given in Appendix C.

We clarify here the physical meaning and implications of these results. Because of the splitting (20) of the work performed on the system, the minimum magnetic energy  $\psi_{\min}$  at  $m = (\mathcal{H}, \mathcal{M})$  represents the amount of work which is stored during any process starting from the virgin state  $(0, 0)$ . Indeed, from (22)

$$\psi_{\min}(\mathcal{H}, \mathcal{M}) = w_0(m_0, m(d_p)).$$

On the other hand, the residual term  $w_D(m_0, p)$  gives the amount of work which is irreversibly dissipated and properly depends on the process  $p$ . In particular, for any closed cycle the stored magnetic energy vanishes and  $w_D$  gives the area of the loop, as expected. By virtue of the decomposition  $\psi = \psi_{\min} + \varphi$ , item (b) in Corollary 4.3 states that the change in  $\varphi$  gives a lower bound for the dissipated work  $w_D$ .

Finally, in terms of  $H$  and  $M$  the minimum and maximum free energies can be expressed as follows:

$$\psi_{\min}(H, M) = \frac{\mu_0}{2}H^2 + \frac{1}{2\kappa}M^2,$$

$$\psi_{\max}(H, M) = \begin{cases} \frac{\mu_0}{2}H^2 + \frac{1}{2\kappa}M^2 & \text{if } (H, M) \in \Sigma^*, \\ \frac{\mu_0 + \nu}{2}H^2 - \frac{\nu}{\kappa}HM + \frac{\nu + \kappa}{2\kappa^2}M^2 & \text{if } (H, M) \in \Sigma_3^+ \cup \Sigma_3^-. \end{cases}$$

Alternately, we have

$$\psi_{\max}(H, M) = \begin{cases} \psi_{\min}(H, M) & \text{if } (H, M) \in \Sigma^*, \\ \psi_{\min}(H, M) + \frac{\nu}{2} \left( H - \frac{1}{2\kappa}M \right)^2 & \text{if } (H, M) \in \Sigma_3^+ \cup \Sigma_3^-. \end{cases}$$

In the paramagnetic regime, when  $\kappa = \chi$  and  $\Sigma$  reduces to  $\Sigma^*$ , both  $\psi_{\min}$  and  $\psi_{\max}$  transform into (16).

## 5. – Hysteresis and temperature-induced transitions

In the literature the mathematical model of transitions from a paramagnetic to a ferromagnetic phase is usually described according to the so-called *Landau's theory of  $\lambda$ -point transitions* which traces back to [17] (see, for instance, [10]). Although this approach has been recently improved and generalized in [16], it is unable to capture the multi-valued structure of the hysteresis loops. This is mainly due to the uniqueness (up to an additive constant) of the free-energy function which is assumed there (see [18]). On the contrary, infinitely many free energies occur when hysteretic phenomena are involved, as shown in the previous section.

In order to model the magnetic transition as a phenomenon induced by temperature variations, here we let the material parameters  $\chi$  and  $\kappa$  depend on  $\theta$ . In the sequel, we discuss separately the transitions from paramagnetic to ferromagnetic, ferrimagnetic and antiferromagnetic phases, since different expressions of functions  $\chi(\theta)$  and  $\kappa(\theta)$  are respectively involved (see, for instance, [1]).

All magnetic materials such as ferromagnets, ferrimagnets and antiferromagnets exhibit paramagnetic behavior above their *critical temperature*,  $\theta_f$ . According to the bi-linear model presented in the previous sections, this behavior can be represented by assuming (see fig. 1a)

$$\kappa(\theta) = \chi(\theta) > 0, \quad \theta \geq \theta_f.$$

On the contrary, when temperature is lowered below the critical point, the hysteretic phenomena occur and, according to the isothermal model (see fig. 1b)

$$\kappa(\theta) > \chi(\theta) > 0, \quad 0 < \theta < \theta_f.$$

Vice versa, when  $\theta$  increases and overcomes the value  $\theta_f$ , then hysteresis loops disappear and the major loop reduces to the skeleton curve.

In order to describe the passage between the paramagnetic regime and the hysteretic one as temperature decreases, we write  $\chi$  in the form

$$\chi(\theta) = \kappa(\theta)[1 - \tau(\theta)],$$

where  $\tau$  is a suitable continuous function satisfying the following conditions:

- $0 < \tau(\theta) < 1$  for any  $0 < \theta < \theta_f$ ;
- $\tau(\theta) = 0$  for any  $\theta \geq \theta_f$ .

From (7) we have  $1/\nu(\theta) = 0$  and then  $\Sigma_3^-(\theta) = \Sigma_3^+(\theta) = \emptyset$  for any  $\theta \geq \theta_f$ . Finally, the differential constitutive equation ruling the evolution of the magnetization at any temperature  $\theta$  can be expressed by

$$\dot{M} = G(\theta, H, M, \operatorname{sgn} \dot{H})\dot{H},$$

where

$$G(\theta, H, M, \operatorname{sgn} \dot{H}) = \begin{cases} \kappa(\theta)[1 - \tau(\theta)], & \text{if } (H, M) \in \Sigma_0(\theta) \text{ or} \\ & (H, M) \in \Sigma_2^-(\theta) \cup \Sigma_3^+(\theta) \text{ and } \operatorname{sgn} \dot{H} = 1 \text{ or} \\ & (H, M) \in \Sigma_2^+(\theta) \cup \Sigma_3^-(\theta) \text{ and } \operatorname{sgn} \dot{H} = -1, \\ 0, & \text{otherwise.} \end{cases}$$

In particular, the residual magnetization  $M_r$  can be evaluated as the intersection of the major hysteresis loop with the vertical magnetization axis, namely

$$(23) \quad M_r(\theta) = M_s \left[ 1 - \frac{\chi(\theta)}{\kappa(\theta)} \right] = M_s \tau(\theta).$$

Analogously, the coercive magnetic field  $H_c$  can be evaluated as the intersection of the major hysteresis loop with the horizontal axis, namely

$$H_c(\theta) = M_s \left[ \frac{1}{\chi(\theta)} - \frac{1}{\kappa(\theta)} \right] = \frac{M_s \tau(\theta)}{\kappa(\theta)[1 - \tau(\theta)]}.$$

When  $\theta \geq \theta_f$ , both  $M_r$  and  $H_c$  vanish.

*Remark.* The explicit expression for  $\tau$  depends strongly on the magnetic material properties. For a large class of ferro- and ferri-magnetic materials, the spontaneous magnetization  $M_r$  obeys approximately the generalized Bloch law (see, for instance, [1, p. 246])

$$(24) \quad M_r(\theta) = M_{r0} \left[ 1 - \left( \frac{\theta}{\theta_f} \right)^\alpha \right], \quad 0 < \theta < \theta_f,$$

where  $M_{r0} < M_s$  and  $\alpha > 1$ . From quantum mechanics,  $\alpha = 3/2$  (see [19, 1]). By comparing (24) with (23), we easily obtain

$$\tau(\theta) = \frac{M_{r0}}{M_s} \left[ 1 - \left( \frac{\theta}{\theta_f} \right)^\alpha \right],$$

so that the ratio  $\chi/\kappa$  takes the form

$$(25) \quad \frac{\chi(\theta)}{\kappa(\theta)} = 1 - \tau(\theta) = \begin{cases} 1 - \frac{M_{r0}}{M_s} \left[ 1 - \left( \frac{\theta}{\theta_f} \right)^\alpha \right], & 0 < \theta < \theta_f, \\ 1, & \theta \geq \theta_f. \end{cases}$$

For various ferromagnetic and ferrimagnetic garnets the dependence of the spontaneous magnetization on temperature differs from Bloch's law (see, for instance, [1, p. 517]). Nevertheless, even in these cases the expression for  $\tau$  can be exactly evaluated by comparing the experimental graphs with (23). This means that once the expression of  $\kappa(\theta)$  is given, we can recover the expression of  $\chi(\theta)$  and vice versa.

The behavior of the susceptibility at temperatures above  $\theta_f$  is very different when ferromagnetic, ferrimagnetic or antiferromagnetic materials are considered. Henceforth, we distinguish two cases:

- a)  $\lim_{\theta \rightarrow \theta_f} \chi(\theta) = \lim_{\theta \rightarrow \theta_f} \kappa(\theta) < \infty$ ;
- b)  $\lim_{\theta \rightarrow \theta_f} \chi(\theta) = \lim_{\theta \rightarrow \theta_f} \kappa(\theta) = \infty$ .

The first case is typical of antiferromagnetic materials (see [1, p. 449]), where the critical temperature is represented by the Néel temperature  $\theta_{fN}$  and

$$(26) \quad \chi(\theta) = \frac{\lambda}{\theta + \theta_*}, \quad \theta > \theta_{fN}.$$

Here  $\theta_* > \theta_{fN}$  is the so-called *paramagnetic Curie temperature*. Since hysteretic phenomena have been recently investigated in connection with antiferromagnetism (see, for instance [4, 5]), we are allowed to include this case into our analysis.

*Example a.1. Antiferromagnetic transition.* Letting  $\kappa_N = \kappa(\theta_{fN}) = \chi(\theta_{fN})$ , we assume

$$\kappa(\theta) = \begin{cases} \kappa_N, & 0 < \theta < \theta_{fN}, \\ \frac{\lambda}{\theta + \theta_*}, & \theta \geq \theta_{fN}, \end{cases}$$

where  $\lambda = \kappa_N(\theta_{fN} - \theta_*)$ . As a consequence, from (25) we have

$$\chi(\theta) = \begin{cases} \kappa_N \left\{ 1 - \frac{M_{r0}}{M_s} \left[ 1 - \left( \frac{\theta}{\theta_f} \right)^\alpha \right] \right\}, & 0 < \theta < \theta_{fN}, \\ \kappa(\theta), & \theta \geq \theta_{fN}. \end{cases}$$

This curve is depicted in fig. 4 and well approaches the usual form of the susceptibility  $\chi$  for antiferromagnetic materials (see [1, p. 460]).

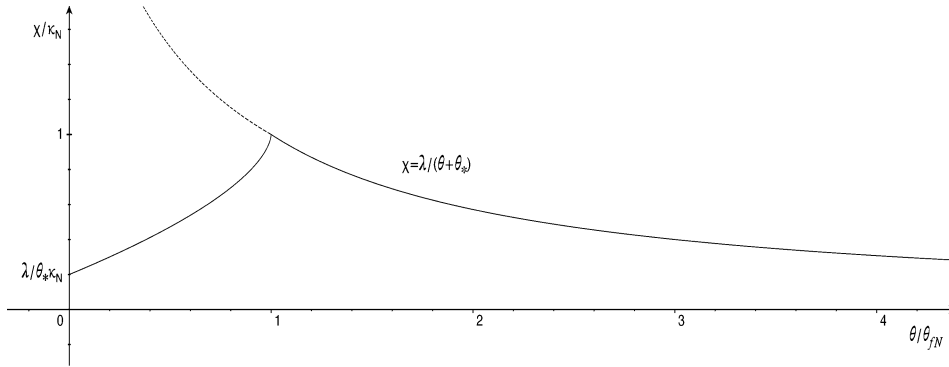


Fig. 4. – A typical susceptibility curve in antiferromagnetics.

*Example a.2.* (see fig. 5) Some special forms of the susceptibility for ferromagnetic materials fall into case (a). For instance, they can be described by assuming

$$\chi(\theta) = \begin{cases} \chi_0 \left( 1 - a \sqrt[3]{\frac{\theta}{\theta_f} - 1} \right) & 0 < \theta \leq \theta_f, \\ \chi_0 \left( 1 - \frac{3}{4} \sqrt[3]{\frac{\theta}{\theta_f} - 1} \right) & \theta_f < \theta < 2\theta_f, \\ \frac{\chi_0}{4 \left( \frac{\theta}{\theta_f} - 1 \right)} & \theta \geq 2\theta_f, \end{cases}$$

where  $a, \chi_0 > 0$ . The corresponding expression of  $\kappa$  can be easily recovered from (25).

Usually, however, both in ferromagnetic and ferrimagnetic materials the susceptibility exhibits a singularity at the critical point so that they fall into case (b). In particular,

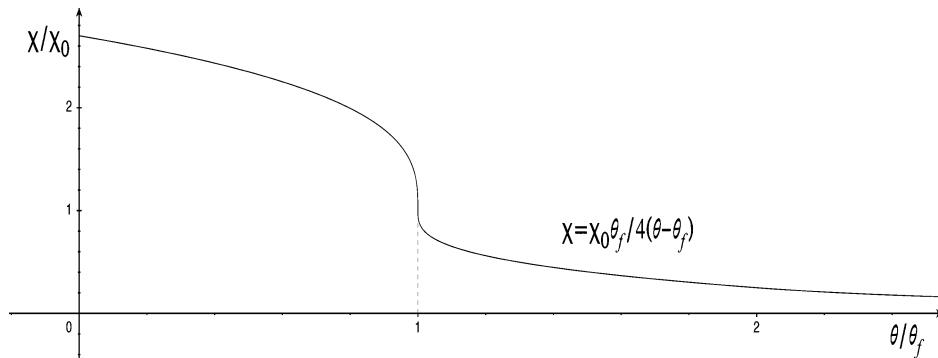


Fig. 5. – The susceptibility curve of *Example a.2.*



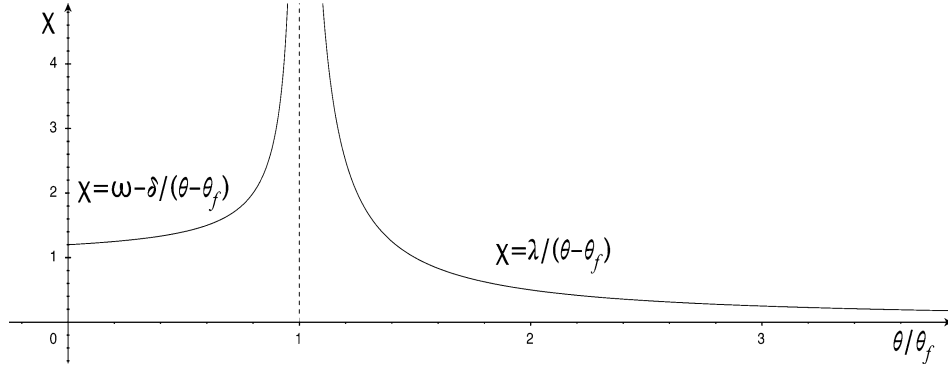


Fig. 6. – A susceptibility curve in ferromagnetic-paramagnetic transitions.

in ferromagnetic materials there follows the Curie-Weiss law, written as

$$(27) \quad \chi(\theta) = \frac{\lambda}{\theta - \theta_f}, \quad \theta > \theta_f,$$

where  $\lambda > 0$  is a suitable positive constant and  $\theta_f$  is usually called *ferromagnetic Curie temperature*. Actually, eq. (27) fits well the paramagnetic behavior well above  $\theta_f$ , but it breaks down in the region very close to  $\theta_f$  (see, for instance, [1, p. 269]). The behavior of  $\chi$  in ferrimagnetic materials is more complicated. For temperature values far above the *critical Néel temperature*  $\theta_{fN}$ , it looks like in antiferromagnetics. According to [1], in this range  $\chi$  is given by

$$(28) \quad \chi(\theta) = \frac{\lambda(\theta - \theta_d)}{(\theta - \theta_d)(\theta + \theta_*) + \xi} = \frac{\lambda(\theta - \theta_d)}{(\theta - \theta_{fN})(\theta + \theta_a)}, \quad \theta > \theta_{fN},$$

where  $\theta_d$  is the Domb-Fisher temperature,  $\xi, \theta_a > 0$  and  $\theta_*$  is called *paramagnetic Curie temperature* because of the relation

$$\chi(\theta) \approx \frac{\lambda}{\theta + \theta_*}, \quad \theta \gg \theta_{fN}.$$

*Example a.3.* (see fig. 6) *Ferro- and ferri-magnetic transitions.* In order to model a paramagnetic-ferromagnetic transition, we let

$$\kappa(\theta) = \begin{cases} \infty, & 0 < \theta \leq \theta_f, \\ \frac{\lambda}{\theta - \theta_f}, & \theta > \theta_f, \end{cases} \quad \chi(\theta) = \begin{cases} \omega - \frac{\delta}{\theta - \theta_f}, & 0 < \theta \leq \theta_f, \\ \frac{\lambda}{\theta - \theta_f}, & \theta > \theta_f, \end{cases}$$

with  $\omega, \delta > 0$ . On the other hand, replacing (27) with (28), ferrimagnetic transitions can be modeled, too. In both cases, the hysteresis curves at  $\theta = \theta_f$  look like those in fig. 3.

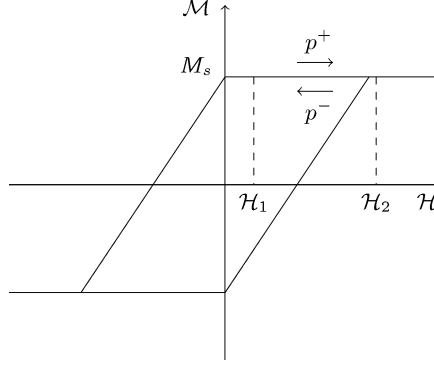


Fig. 7. – The processes  $p^+$  and  $p^-$ .

## APPENDIX A.

### A preliminary Lemma

In order to show the complete controllability of dynamical system (12), we first prove a preliminary result in which processes connecting states in different regions of  $\Sigma$  are explicitly constructed. Although the amount of expended or absorbed work during the process is meaningless in connection with controllability, we establish here some estimates of these amounts in view of the construction of the minimum and maximum free energies, as devised in Appendix C.

For later convenience, given two processes  $p_1, p_2 \in \mathcal{P}$  of duration  $d_1, d_2$  respectively, we denote by  $p_1 * p_2$  the process of duration  $d_1 + d_2$  defined as

$$p_1 * p_2 = \begin{cases} p_1(t) & t \in [0, d_1), \\ p_2(t - d_1) & t \in [d_1, d_1 + d_2). \end{cases}$$

*Lemma 1.1.* *The following conditions hold.*

- i) *Every couple of states belonging to  $\mathcal{S}_1^+ \cup \mathcal{S}_2^+$  [or  $\in \mathcal{S}_1^- \cup \mathcal{S}_2^-$ ] is connected by reversible processes, that is if  $(\mathcal{H}_1, \mathcal{M}_1), (\mathcal{H}_2, \mathcal{M}_1) \in \mathcal{S}_1^+ \cup \mathcal{S}_2^+$  [or  $\in \mathcal{S}_1^- \cup \mathcal{S}_2^-$ ], then there exists at least a process,  $p \in \mathcal{P}$  such that*

$$\rho(\mathcal{H}_1, \mathcal{M}_1), p) = (\mathcal{H}_2, \mathcal{M}_1) \quad \text{and} \quad w_D((\mathcal{H}_1, \mathcal{M}_1), p) = 0.$$

- ii) *Every couple of states belonging to  $\mathcal{S}_0 \cup \mathcal{S}_2^+ \cup \mathcal{S}_2^-$  is connected by quasi-reversible processes, that is if  $(0, \mathcal{M}_1), (0, \mathcal{M}_2) \in \mathcal{S}_0$ , then, for any  $\varepsilon > 0$ , there exists at least a process,  $p^\varepsilon \in \mathcal{P}$  such that*

$$\rho(0, \mathcal{M}_1), p^\varepsilon) = (0, \mathcal{M}_2) \quad \text{and} \quad 0 \leq w_D((\mathcal{H}_1, \mathcal{M}_1), p^\varepsilon) < \varepsilon.$$

*Proof.* We split the proof into two parts.

- i) Let  $(\mathcal{H}_1, \mathcal{M}_*), (\mathcal{H}_2, \mathcal{M}_*) \in \mathcal{S}_1^+ \cup \mathcal{S}_1^- \cup \mathcal{S}_2^+ \cup \mathcal{S}_2^-$ ,  $\mathcal{H}_1 < \mathcal{H}_2$ . To fix ideas, we suppose  $\mathcal{M}_* = M_s$  (see fig. 7).

We choose a process  $p^+$  of duration  $d^+ = \mathcal{H}_2 - \mathcal{H}_1$  such that  $p^+(t) = 1$ ,  $t \in [0, d^+)$ . In view of (12) and (21), we deduce  $\dot{\mathcal{M}} = 0$  and  $w_D((\mathcal{H}_1, M_s), p^+) = 0$ . Moreover, from (14) it follows

$$\begin{aligned}\mathcal{H}(d) &= \mathcal{H}(0) + \int_0^{d^+} p^+(s) ds = \mathcal{H}_1 + d^+ = \mathcal{H}_2, \\ \mathcal{M}(d) &= \mathcal{M}(0) = M_s,\end{aligned}$$

that is

$$\rho((\mathcal{H}_1, M_s), p^+) = (\mathcal{H}_2, M_s).$$

Similarly, by letting

$$p^-(t) = -1, \quad t \in [0, \mathcal{H}_2 - \mathcal{H}_1),$$

we obtain

$$\rho((\mathcal{H}_2, M_s), p^-) = (\mathcal{H}_1, M_s), \quad \text{and} \quad w_D((\mathcal{H}_2, M_s), p^-) = 0.$$

By repeating the same arguments, one can easily prove the statement in the case  $(\mathcal{H}_1, \mathcal{M}_*), (\mathcal{H}_2, \mathcal{M}_*) \in \mathcal{S}_1^- \cup \mathcal{S}_2^-$ .

- ii) Let  $(0, \mathcal{M}_1), (0, \mathcal{M}_2) \in \mathcal{S}$ , with  $\mathcal{M}_1 < \mathcal{M}_2$  and  $p^+, p^-$  be two processes of duration  $d$  defined as  $p^+(t) = -p^-(t) = 1$ ,  $t \in [0, d)$ . The duration  $d$  will be chosen in the sequel. We show that

$$\rho((0, \mathcal{M}_1), p^*) = (0, \mathcal{M}_1 + \nu d), \quad w_D((d, \mathcal{M}_1 + \nu d), p^*) = \frac{\nu}{2} d^2,$$

where the process  $p^*$  of duration  $2d$  is defined as  $p^*(t) = p^+ * p^-(t)$ . First we prove

$$\rho((0, \mathcal{M}_1), p^+) = (d, \mathcal{M}_1 + \nu d).$$

Indeed, (14) yields

$$\begin{aligned}\mathcal{H}(d) &= \mathcal{H}(0) + \int_0^d p^+(s) ds = d, \\ \mathcal{M}(d) &= \mathcal{M}(0) + \nu[\mathcal{H}(d) - \mathcal{H}(0)] = \mathcal{M}_1 + \nu d.\end{aligned}$$

Furthermore, we have

$$w_D((0, \mathcal{M}_1), p^+) = \int_0^d \mathcal{H}(s) \dot{\mathcal{M}}(s) ds = \int_0^d \nu \mathcal{H}(s) \dot{\mathcal{H}}(s) ds = \left[ \frac{\nu}{2} \mathcal{H}^2(s) \right]_0^d = \frac{\nu}{2} d^2.$$

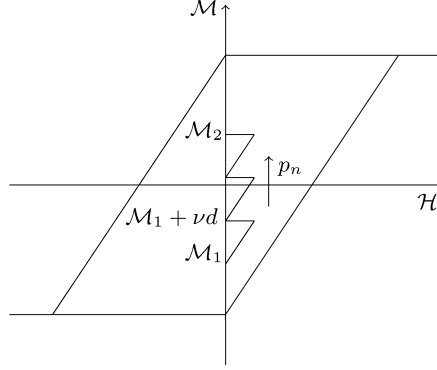


Fig. 8. – The graph of process  $p_n$  when  $n = 3$ .

Now we calculate  $\rho((d, \mathcal{M}_1 + \nu d), p^-)$ . Since  $(d, \mathcal{M}_1 + \nu d) \in \mathcal{S}_3^+$  and  $p^- \equiv -1$ , eq. (12) leads to  $\dot{\mathcal{M}} = 0$ . In view of (14), this implies

$$\begin{aligned} \mathcal{M}(d) &= \mathcal{M}_1 + \nu d, \\ \mathcal{H}(d) &= \mathcal{H}(0) + \int_0^d p^-(s) ds = d - d = 0, \\ 0 &= w_D((d, \mathcal{M}_1 + \nu d), p^-). \end{aligned}$$

Accordingly, the following equalities hold:

$$\begin{aligned} \rho((0, \mathcal{M}_1), p^*) &= (0, \mathcal{M}_1 + \nu d), \\ w_D((0, \mathcal{M}_1), p^*) &= w_D((0, \mathcal{M}_1), p^+) + w_D((d, \mathcal{M}_1 + \nu d), p^-) = \frac{\nu}{2} d^2. \end{aligned}$$

By repeating the same technique  $n$  times, one can prove that

$$\rho((0, \mathcal{M}_1), p_n) = (0, \mathcal{M}_1 + \nu n d), \quad w_D((0, \mathcal{M}_1), p_n) = \frac{\nu}{2} n d^2,$$

where  $p_n$  is the process of duration  $n d$  represented in fig. 8 and defined as

$$p_n = \underbrace{p^* * \cdots * p^*}_{n \text{ times}}.$$

We fix  $\varepsilon > 0$  and we choose  $d = \frac{\mathcal{M}_2 - \mathcal{M}_1}{n\nu}$ . Letting  $n > \frac{(\mathcal{M}_2 - \mathcal{M}_1)^2}{2\nu\varepsilon}$ , we have

$$0 \leq w_D((0, \mathcal{M}_1), p_n) < \varepsilon.$$

Finally, let  $(0, \mathcal{M}_1), (0, \mathcal{M}_2) \in \mathcal{S}$ , with  $\mathcal{M}_1 > \mathcal{M}_2$ . We denote by  $\bar{p}_n^*$  the process

$$\bar{p}_n^* = \underbrace{\bar{p}^* * \cdots * \bar{p}^*}_{n \text{ times}}, \quad \bar{p}^* = p^- * p^+,$$

where  $p^+(t) = -p^-(t) = 1$ ,  $t \in [0, d]$ ,  $d = \frac{\mathcal{M}_1 - \mathcal{M}_2}{n\nu}$ . It is easy to prove

$$\rho(0, \mathcal{M}_1, \bar{p}_n^*) = (0, \mathcal{M}_2) \quad \text{and} \quad 0 \leq w_D((\mathcal{H}_1, \mathcal{M}_1), \bar{p}_n^*) < \varepsilon$$

with  $n > \frac{(\mathcal{M}_1 - \mathcal{M}_2)^2}{2\nu\varepsilon}$ . □

## APPENDIX B.

### Proof of Proposition 3.2

*Proof.* It is sufficient to prove that for every state  $(\mathcal{H}, \mathcal{M}) \in \mathcal{S}$  there exist at least two processes  $\overleftarrow{p}, \overrightarrow{p} \in \mathcal{P}$  satisfying

$$\rho((\mathcal{H}, \mathcal{M}), \overleftarrow{p}) = (0, 0), \quad \rho((0, 0), \overrightarrow{p}) = (\mathcal{H}, \mathcal{M}).$$

If  $\mathcal{H} = 0$ , the thesis is guaranteed by Lemma A.1. Let  $(\mathcal{H}, \mathcal{M}) \in \mathcal{S}$  with  $\mathcal{H} \neq 0$ . First we exhibit a process  $\overleftarrow{p}$  such that  $\rho((\mathcal{H}, \mathcal{M}), \overleftarrow{p}) = (0, 0)$ . To fix ideas, we suppose that  $\mathcal{H} > 0$ . We denote by  $p_{\mathcal{H}}^-$  the process defined as  $p_{\mathcal{H}}^-(t) = -1$ ,  $t \in [0, \mathcal{H}]$ . Thanks to (12) and (14), we obtain

$$\rho((\mathcal{H}, \mathcal{M}), p_{\mathcal{H}}^-) = (0, \mathcal{M}).$$

Furthermore, by applying Lemma A.1, we deduce the existence of a process  $p^\varepsilon$  satisfying

$$\rho((0, \mathcal{M}), p^\varepsilon) = (0, 0).$$

Accordingly, the process  $\overleftarrow{p}$  defined as  $\overleftarrow{p} = p_{\mathcal{H}}^- * p^\varepsilon$  connects the initial state  $(\mathcal{H}, \mathcal{M})$  to the final state  $(0, 0)$ .

If  $\mathcal{H} < 0$ , we substitute the process  $p_{\mathcal{H}}^-$  with  $p_{-\mathcal{H}}^+$ , defined as  $p_{-\mathcal{H}}^+(t) = 1$ ,  $t \in [0, -\mathcal{H}]$ , and we let  $\overleftarrow{p} = p_{-\mathcal{H}}^+ * p^\varepsilon$ .

Now we prove the existence of a process  $\overrightarrow{p} \in \mathcal{P}$  satisfying  $\rho((0, 0), \overrightarrow{p}) = (\mathcal{H}, \mathcal{M})$  for any  $(\mathcal{H}, \mathcal{M}) \in \mathcal{S}$ . We suppose  $\mathcal{H} > 0$ . We choose  $(0, \widetilde{\mathcal{M}}) \in \mathcal{S}_0$  and a process  $\tilde{p} \in \mathcal{P}$  such that  $(\mathcal{H}, \mathcal{M})$  is reachable from  $(0, \widetilde{\mathcal{M}})$  through the process  $\tilde{p}$ . To this aim, we let

$$\tilde{p}(t) = p_{\mathcal{H}}^+(t) = 1, \quad t \in [0, \mathcal{H}], \quad \widetilde{\mathcal{M}} = \mathcal{M} - \nu\mathcal{H}.$$

Therefore, (12) and (14) imply

$$\rho((0, \mathcal{M} - \nu\mathcal{H}), p_{\mathcal{H}}^+) = (\mathcal{H}, \mathcal{M}).$$

If  $\mathcal{H} < 0$ , it is sufficient to replace  $p_{\mathcal{H}}^+$  with  $p_{-\mathcal{H}}^-$ , defined by  $p_{-\mathcal{H}}^-(t) = -1$  for any  $t \in [0, -\mathcal{H}]$ . In view of *ii*) of Lemma A.1, there exists a process  $p^\varepsilon$  which connects  $(0, 0)$  to  $(0, \widetilde{\mathcal{M}})$ . Thus, by defining  $\overrightarrow{p} = p^\varepsilon * \tilde{p}$ , we have

$$\rho((0, 0), \overrightarrow{p}) = (\mathcal{H}, \mathcal{M})$$

and we reach the conclusion. □

## APPENDIX C.

**Proof of Theorem 4.2.**

*Proof.* Henceforth, we respectively denote by  $\overrightarrow{\mathcal{P}}_{(\mathcal{H}, \mathcal{M})}$  and  $\overleftarrow{\mathcal{P}}_{(\mathcal{H}, \mathcal{M})}$  the sets of all processes which attain  $(\mathcal{H}, \mathcal{M})$  from  $(0, 0)$  and vice versa.

In view of (20) the minimum and maximum free energies defined in (18) and (19) assume the form

$$\begin{aligned}\psi_{\min}(\mathcal{H}, \mathcal{M}) &= \psi_0(\mathcal{H}, \mathcal{M}) - \inf_{p \in \overleftarrow{\mathcal{P}}_{(\mathcal{H}, \mathcal{M})}} w_D((\mathcal{H}, \mathcal{M}), p), \\ \psi_{\max}(\mathcal{H}, \mathcal{M}) &= \psi_0(\mathcal{H}, \mathcal{M}) + \inf_{p \in \overrightarrow{\mathcal{P}}_{(\mathcal{H}, \mathcal{M})}} w_D((0, 0), p),\end{aligned}$$

where

$$\psi_0(\mathcal{H}, \mathcal{M}) = \frac{\mu_0}{2} \left( \mathcal{H} + \frac{1}{\kappa} \mathcal{M} \right)^2 + \frac{1}{2\kappa} \mathcal{M}^2.$$

First, we find out the expression of  $\psi_{\min}$ . Our goal consists in showing that

$$(C.1) \quad \inf_{p \in \overleftarrow{\mathcal{P}}_{(\mathcal{H}, \mathcal{M})}} w_D((\mathcal{H}, \mathcal{M}), p) = 0.$$

To fix ideas we suppose  $\mathcal{H} > 0$ . The cases  $\mathcal{H} < 0$  and  $\mathcal{H} = 0$  are similar and they will be omitted. For any  $\varepsilon > 0$ , we consider the process  $\overleftarrow{p} = p_{\mathcal{H}}^- * p^\varepsilon$ , defined as in the proof of Proposition 3.2, that connects  $(\mathcal{H}, \mathcal{M})$  to  $(0, 0)$ . Thus, we have

$$0 \leq \inf_{p \in \overleftarrow{\mathcal{P}}_{(\mathcal{H}, \mathcal{M})}} w_D((\mathcal{H}, \mathcal{M}), p) \leq w_D((\mathcal{H}, \mathcal{M}), p_{\mathcal{H}}^-) + \inf_{p^\varepsilon \in \overleftarrow{\mathcal{P}}_{(0, \mathcal{M})}} w_D((0, \mathcal{M}), p^\varepsilon).$$

Lemma A.1 guarantees that

$$\inf_{p^\varepsilon \in \overleftarrow{\mathcal{P}}_{(0, \mathcal{M})}} w_D((0, \mathcal{M}), p^\varepsilon) = 0.$$

Moreover, the choice of the process  $p_{\mathcal{H}}^-$  yields

$$w_D((\mathcal{H}, \mathcal{M}), p_{\mathcal{H}}^-) = 0.$$

As a consequence, (C.1) is satisfied and

$$\psi_{\min}(\mathcal{H}, \mathcal{M}) = \psi_0(\mathcal{H}, \mathcal{M}).$$

Now we find out the expression of  $\psi_{\max}$ . Let  $(\mathcal{H}, \mathcal{M}) \in \mathcal{S}^*$ . By applying Lemma A.1, we deduce the existence of two processes  $\overleftarrow{p}, p^\varepsilon$ ,  $\varepsilon > 0$  such that

$$\begin{aligned}\rho((0, \mathcal{M}), \overleftarrow{p}) &= (\mathcal{H}, \mathcal{M}), & w_D((\mathcal{H}, \mathcal{M}), \overleftarrow{p}) &= 0, \\ \rho((0, 0), p^\varepsilon) &= (0, \mathcal{M}), & w_D((0, 0), p^\varepsilon) &< \varepsilon.\end{aligned}$$

Accordingly,

$$0 \leq \inf_{p \in \vec{\mathcal{P}}(\mathcal{H}, \mathcal{M})} w_D((0, 0), p) \leq \inf_{p^\varepsilon \in \vec{\mathcal{P}}(\mathcal{H}, \mathcal{M})} w_D((0, 0), p^\varepsilon) + w_D((0, \mathcal{M}), \bar{p}) = 0$$

and  $\psi_{\max} = \psi_0$  in  $\mathcal{S}^*$ . Finally, we suppose  $(\mathcal{H}, \mathcal{M}) \in \mathcal{S}_3^+ \cup \mathcal{S}_3^-$ . Our aim consists in showing that

$$\inf_{p \in \vec{\mathcal{P}}(\mathcal{H}, \mathcal{M})} w_D((0, 0), p) = \frac{\nu}{2} \mathcal{H}^2.$$

Let  $p \in \vec{\mathcal{P}}(\mathcal{H}, \mathcal{M})$  be a process of duration  $d_p$ . By calculating  $w_D((0, 0), p)$ , we obtain

$$w_D((0, 0), p) = \int_0^{d_p} \mathcal{H}(t) \dot{\mathcal{M}}(t) dt = \int_I \nu \mathcal{H}(t) \dot{\mathcal{H}}(t) dt,$$

where  $I = \{t \in [0, d_p] : \dot{\mathcal{M}}(t) = \nu \dot{\mathcal{H}}(t)\}$ . In view of (12),  $\nu \mathcal{H}(t) \dot{\mathcal{H}}(t) \geq 0$  for any  $t \in I$ . Therefore,

$$w_D((0, 0), p) = \int_I \frac{\nu}{2} |d\mathcal{H}^2| = \frac{\nu}{2} V_I(\mathcal{H}^2),$$

where  $V_I(\mathcal{H}^2)$  denotes the total variation of  $\mathcal{H}^2$  on  $I$  (see [12]), *i.e.*

$$V_I(\mathcal{H}^2) = \sup_I \left\{ \sum_{k=1}^N |\mathcal{H}^2(t_k) - \mathcal{H}^2(t_{k-1})| : t_0 < \dots < t_N \in I \right\}.$$

Since the equalities

$$\min_{t \in I} \mathcal{H}^2(t) = 0 = \mathcal{H}^2(0), \quad \mathcal{H}^2(d_p) = \mathcal{H}^2$$

hold, we deduce that  $V_I(\mathcal{H}^2) \geq \mathcal{H}^2$  and hence

$$w_D((0, 0), p) \geq \frac{\nu}{2} \mathcal{H}^2.$$

In order to prove that  $w_D((0, 0), p) \leq \nu \mathcal{H}^2/2$ , we choose the process  $\vec{p} = p^\varepsilon * \bar{p}$  as in the proof of Proposition 3.2. Lemma A.1 yields

$$w_D((0, 0), \vec{p}) = w_D((0, 0), p^\varepsilon) + w_D((0, \mathcal{M} - \nu \mathcal{H}), \bar{p}) < \varepsilon + \frac{\nu}{2} \mathcal{H}^2.$$

Therefore,

$$\inf_{p \in \vec{\mathcal{P}}(\mathcal{H}, \mathcal{M})} w_D((0, 0), p) \leq \frac{\nu}{2} \mathcal{H}^2$$

and  $\psi_{\max}(\mathcal{H}, \mathcal{M}) = \psi_0(\mathcal{H}, \mathcal{M}) + \nu \mathcal{H}^2/2$  in  $\mathcal{S}_3^+ \cup \mathcal{S}_3^-$ .  $\square$

**Proof of Corollary 4.3.**

*Proof.* Property (a) is due to the fact that the minimum and maximum free energies coincide along the skeleton curve  $\mathcal{S}^*$ . Property (b) is a consequence of the definition of subpotentials. Now we prove (c). Letting  $\mathcal{H}_1 < \mathcal{H}_2 < 0$  and  $p(t) = 1$ ,  $t \in [0, \mathcal{H}_2 - \mathcal{H}_1)$ , we have

$$\rho((\mathcal{H}_1, \mathcal{M}), p) = (\mathcal{H}_2, \mathcal{M}) \quad \text{and} \quad w_D((\mathcal{H}_1, \mathcal{M}), p) = 0.$$

Accordingly, in view of (b), we deduce

$$\varphi(\mathcal{H}_2, \mathcal{M}) \leq \varphi(\mathcal{H}_1, \mathcal{M}).$$

Similarly, letting  $\mathcal{H}_1 > \mathcal{H}_2 > 0$  and choosing  $p(t) = -1$ ,  $t \in [0, \mathcal{H}_1 - \mathcal{H}_2)$ , we obtain

$$\varphi(\mathcal{H}_2, \mathcal{M}) \geq \varphi(\mathcal{H}_1, \mathcal{M}). \quad \square$$

\* \* \*

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