Energetic and Thermodynamic Aspects of Hysteresis

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(Received 5 October 1995)

A thermodynamic description of hysteresis phenomena is proposed, where the system evolution is described as a sequence of Barkhausen jumps, and the Preisach model is used to characterize the jump sequence. Expressions for Gibbs energy, entropy, and entropy production are derived. The equilibrium states of the minimum Gibbs energy are defined and the equations for the thermal relaxation of a generic initial state are derived.

PACS numbers: 75.60.Ej, 05.70.Ln, 74.60.Ge

Nonequilibrium thermodynamics has been applied with success to a number of situations, such as chemical reactions or transport phenomena, where suitable relationships between irreversible flows and thermodynamic forces can be assumed [1,2]. It is not so for hysteresis phenomena, where irreversibility arises from the nonlinear and branching character of the system constitutive laws [3,4]. Thermodynamic approaches to hysteresis have been attempted by several authors [5], but no conclusive results have been reached yet. The main difficulty is that a hysteretic transformation, no matter how slow is the variation of the external driving force, is characterized by a certain proportion between the energy reversibly stored or released by the system and the energy irreversibly dissipated as heat because of hysteresis losses, and we have no general principles helping us to separate these two energy contributions.

On general grounds, hysteresis is the consequence of the existence of many metastable free energy minima. These metastable states are the result of the coupling of characteristic structural features (such as magnetic domain walls in ferromagnets, dislocations in mechanical systems, Abrikosov vortices in type-II superconductors, etc.) among themselves or with environmental disorder. The central feature is, in this respect, that of the Barkhausen jump (BJ). This term, commonly used for magnetic systems, indicates an event where the system, locally brought to instability by the driving force, suddenly moves to a new metastable configuration. Two energy terms will be involved in the jump, one representing the difference in free energy between the old and the new metastable configuration, and the other one measuring the amount of energy dissipated as heat during the jump. Our comprehension of the hysteretic behavior of the system is thus intimately related to our knowledge of the BJ statistics. The mathematical tool best suited to this physical picture is the Preisach model (PM) of hysteresis [3]. In PM, the macroscopic hysteresis properties of a given system are expressed as sums over a suitable distribution of elementary switching units. In this Letter, these switching units are used to describe the BJ behavior, and the hysteretic state of the system is defined in terms of a certain line $b(h_c)$, describing the distribution of the switching units over their $+1$ and -1 states.

When attempting a thermodynamic description, it must be realized that a hysteretic system is governed by intrinsically out-of-equilibrium, history-dependent constitutive laws, so that the local equilibrium hypothesis of irreversible thermodynamics has to be abandoned. In its place, we will introduce the PM $b(h_c)$ line as an additional set of internal thermodynamic variables, and we will assume that the usual thermodynamic functions still exist for the hysteretic system, once expressed as functionals of the extended set including $b(h_c)$. In this respect, the present treatment has some analogy with the formalism of rational thermodynamics (see Ref. [2], p. 25). We expect this approach to be useful as a phenomenological description of several hysteresis effects observed in nature. Its main limitation resides in the limits of PM itself as a tool for the description of hysteresis. So far, PM has been successfully applied to ferromagnets [6], superconductors [7], consolidated materials [8], and, at the same time, a number of generalizations of the original PM have been proposed for the treatment of more general and refined input-output relationships [3], dynamic effects [9], and vectorial hysteresis [10]. In [11], it is proven that a system can be described by PM if and only if it obeys two general properties, known as "wiping out" (or "return-point memory" [4]) and "congruency" property [11]. The wiping out property is of quite general character and is obeyed by a broad variety of systems. Conversely, the congruency property imposes more stringent restrictions that, though with some remarkable exceptions such as superconductors [7], are often only approximately satisfied. To overcome, at least partially, such a limitation, in this Letter we introduce an extension of the original PM, where the congruency property takes a much weaker form, expressed by the existence of a field, H_{LS}, dependent on the state of the system, such that congruency is required only with respect to the internal field $H = H_a + H_{\mathrm{LS}}$, where H_a is the externally applied field.

0031-9007/96/76(10)/1739(4)$10.00

In PM, a given system is described by a collection of elementary nonsymmetric square loops with up and down switching fields α and β, $\alpha \geq \beta$. Under the action of the field H (e.g., magnetic field in ferromagnets, load in mechanical systems, etc.), a given elementary loop will be in the $+1$ state whenever $H > \alpha$ and in the -1 state whenever $H < \beta$. When $\beta < H < \alpha$, the loop state will depend on the past H history. The output X (magnetic moment, mechanical elongation, etc.) is expressed as an integral over the elementary loop collection

$$X = X_s \int\int_{R_+} d\alpha\, d\beta\, p(\alpha, \beta) - X_s \int\int_{R_-} d\alpha\, d\beta\, p(\alpha, \beta), \qquad (1)$$

where X_s is some characteristic saturation value for the output and R_+, R_- are the regions associated with elementary loops in the $+1$ and -1 states. The boundary between R_+ and R_- is in general a staircase line made up of alternating horizontal and vertical segments [3]. The Preisach distribution $p(\alpha, \beta)$ gives the statistical weight of the various elementary contributions. We assume that $p(\alpha, \beta)$ is positive, normalized to unity, and characterized by the symmetry $p(\alpha, \beta) = p(-\beta, -\alpha)$, which ensures that both $\{H(t), X(t)\}$ and $\{-H(t), -X(t)\}$ are admissible input-output histories for the system. Moreover, we assume that both $p(\alpha, \beta)$ and X_s are independent of H, although they may depend on temperature.

There is no conceptual difficulty in reinterpreting the Preisach distribution as a BJ distribution. To this end, it is convenient to introduce the new field coordinates $h_c = (\alpha - \beta)/2$ and $h_u = (\alpha + \beta)/2$. By taking into account the symmetry $p(\alpha, \beta) = p(-\beta, -\alpha)$, i.e., $p(h_c, h_u) = p(h_c, -h_u)$, we can rewrite Eq. (1) in the form

$$X = X_s \int_0^\infty dh_c \int_0^{b(h_c)} dh_u\, p(h_c, h_u), \qquad (2)$$

where $p(h_c, h_u)$ has been suitably renormalized to include all unimportant numerical factors. $b(h_c)$ represents the mentioned staircase boundary between the R_+ and R_- regions, with slope $|db/dh_c| \leq 1$ and $b(0) = H$. The way a given field history $\{H(t)\}$ determines the $b(h_c)$ line is summarized by the following simple rule (see Fig. 1): Consider the evolution in time of the two lines $h_u = H(t) \pm h_c$; whenever the constraint $H - h_c \leq b(h_c) \leq H + h_c$ is violated for some h_c, modify $b(h_c)$ by a proper amount $\delta b(h_c)$, so that finally $b(h_c) = H - h_c$ [if $\delta b(h_c) > 0$] or $b(h_c) = H + h_c$ [if $\delta b(h_c) < 0$].

In the absence of hysteresis, the thermodynamic state of the system, supposed to be closed and homogeneous, would be fully described by the set of variables $[H, T; X]$. Yet, this set becomes incomplete when the equation of state is hysteretic, and it must be supplemented with a suitable set of internal variables. This set is represented by the boundary line $b(h_c)$ introduced in Eq. (2). Our aim is to express the system's thermodynamic functions as

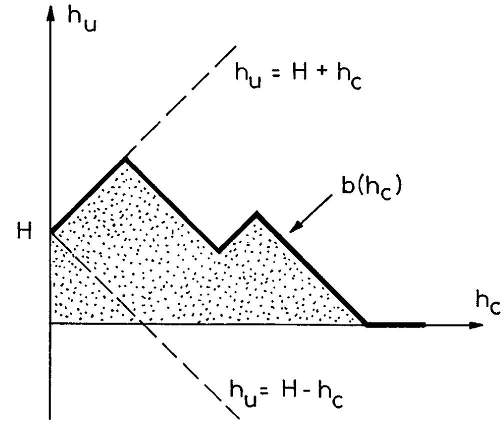

FIG. 1. Preisach representation in the (h_c, h_u) plane, showing the region of integration (point filled) for Eq. (2) and lines controlling $b(h_c)$ behavior according to $H - h_c \leq b(h_c) \leq H + h_c$.

functionals of the extended set $[b(\cdot), T; X]$, with $b(0) = H$. The entropy balance equation is $dS = \delta S_e + \delta S_i$, where dS is the variation of the system entropy, δS_e is the entropy flow into the system, and $\delta S_i \geq 0$ is the internal entropy production. Analogously, the energy balance reads $dU = \delta W + T\delta S_e = \delta W + TdS - T\delta S_i$, where dU is the variation of the system's internal energy and δW is the external work performed on the system $\delta W = H_a dX$, where H_a is the applied field. In terms of the Helmoltz free energy $F = U - TS$, we have $dF = \delta W - SdT - T\delta S_i = H_a dX - SdT - T\delta S_i$. Let us evaluate $\delta W = H_a dX$ from Eq. (2), for an infinitesimal variation $\delta b(h_c)$ at constant temperature. By taking into account that changes of $b(h_c)$ can occur only in correspondence of $H_a = b(h_c) + h_c$ [$\delta b(h_c) > 0$] or $H_a = b(h_c) - h_c$ [$\delta b(h_c) < 0$], we obtain

$$\delta W = X_s \int_0^\infty dh_c\, b(h_c) p[h_c, b(h_c)] \delta b(h_c) + X_s \int_0^\infty dh_c\, h_c p[h_c, b(h_c)] |\delta b(h_c)|. \qquad (3)$$

Given the positiveness of $p(h_c, h_u)$, the last term is always positive. In addition, its integral over a cyclic field variation is precisely equal to the area of the hysteresis loop. It thus represents the entropy production

$$T\delta S_i = X_s \int_0^\infty dh_c\, h_c p[h_c, b(h_c)] |\delta b(h_c)|. \qquad (4)$$

This conclusion is in agreement with the results obtained in [12]. The first term, on the other hand, can be expressed as the variation of the functional

$$F = X_s \int_0^\infty dh_c \int_0^{b(h_c)} dh_u\, h_u p(h_c, h_u) + F_{\mathrm{LS}}. \qquad (5)$$

We conclude that Eq. (5) represents the system free energy. These results make clear the physical reason for the introduction of the fields h_c and h_u. In fact, Eqs. (4)

and (5) show that the variations in dissipated and stored energy brought about by a BJ associated with (h_c, h_u) are $[T\Delta S_i]_{\rm BJ} = h_c|\Delta X_{\rm BJ}|$ and $[\Delta F]_{\rm BJ} = h_u \Delta X_{\rm BJ}$, where $\Delta X_{\rm BJ} = X_s p(h_c, h_u)\Delta h_c \Delta h_u$ is the X variation caused by the jump.

The presence of the term $F_{\rm LS}$ in Eq. (5) is worthy of some attention. Considering the way it was introduced, $F_{\rm LS}$ should be a function of temperature only, but we are naturally led to extend this result. In fact, $F_{\rm LS}$ describes the large-scale free energy behavior of the system, obtained after smoothing out the fluctuating landscape responsible for hysteresis. There is no reason why this large-scale behavior should not exhibit some residual nonhysteretic dependence on X, $F_{\rm LS}(X, T)$. The need for such an extension could have been already foreseen when we tacitly assumed, in the derivation of Eq. (3), that the applied field H_a and the Preisach field H controlling the BJ evolution were coincident. This is not necessarily the case. The presence of $F_{\rm LS}$ gives in fact an additional contribution to the free energy variation dF, equal to $-H_{\rm LS} dX$, with $H_{\rm LS}(X,T) = -[\partial F_{\rm LS}/\partial X]_T$. The effective field acting in the Preisach plane is $H = H_a + H_{\rm LS}$ and, in the derivation of Eq. (3), the decomposition $H_a = b(h_c) \pm h_c$ must be modified into $H \equiv H_a + H_{\rm LS} = b(h_c) \pm h_c$.

Let us now discuss the properties of the equilibrium states under constant temperature and applied field, corresponding to minimum Gibbs energy $G = F - H_a X$. By applying standard variational methods to Eqs. (5) and (2), we find that G is at an extremum when $b(h_c) = H \equiv H_a + H_{\rm LS}$ [13]. In analogy with the terminology used in magnetism, we will call this solution the anhysteretic state. When $H_{\rm LS} \approx 0$, the anhysteretic state can be approximately attained by applying an oscillating field of amplitude slowly decreasing from infinity to zero, superimposed to the constant field H_a. The fact that field histories of this sort should produce low energy states is often assumed and exploited in the literature, although not always with a clear physical justification.

So far, no considerations have been made on the role of temperature. Some conclusion on this point can be drawn if we reconsider the description of hysteresis as due to the coupling of certain structural features (domain walls, dislocations, and vortices) with quenched-in disorder. We expect that temperature will affect the parameters controlling the structure-disorder coupling much more than the statistical distribution of the quenched-in disorder itself. In terms of the PM description, this means that a temperature variation should mainly produce an overall rescaling of the Preisach distribution, without affecting too much its functional dependence on h_c and h_u. In other words, there will exist two characteristic fields $H_c(T)$ and $H_u(T)$ such that any dependence on the PM fields h_c and h_u will be expressible in terms of the combinations h_c/H_c and h_u/H_u only. Let us consider the dimensionless quantities $p(h_c, h_u)\, dh_c\, dh_u \to \hat{p}(x_c, x_u)\, dx_c\, dx_u$, $b(h_c)/H_u \to \hat{b}(x_c)$, where $x_c = h_c/H_c$, $x_u = h_u/H_u$. The dimensionless Preisach density $\hat{p}(x_c, x_u)$ is independent of temperature and characterizes the system's internal structure. The dimensionless boundary $\hat{b}(x_c)$ is also temperature independent, and describes, together with the temperature T, the state of the system. In particular, any transformation under fixed $\hat{b}(x_c)$ is a transformation with zero entropy production. By expressing X and F in terms of $\hat{p}(x_c, x_u)$ and $\hat{b}(x_c)$, and by considering a nondissipative infinitesimal transformation where $\delta S_i = 0$, i.e., $\delta\hat{b}(x_c) = 0$, we obtain the following expression for the system entropy [14]:

$$S = HX\,\frac{d\ln(X_s)}{dT} - (F - F_{\rm LS})\,\frac{d\ln(X_s H_u)}{dT} - \left[\frac{\partial F_{\rm LS}}{\partial T}\right]_X . \quad (6)$$

The state described by $b(h_c)$ is in general a metastable state, with the natural tendency to evolve towards the anhysteretic state of minimum Gibbs energy. A BJ driven by the applied field H_a is characterized by $[\Delta G]_{\rm BJ} + [T\Delta S_i]_{\rm BJ} = 0$. During thermal relaxation, H_a is not sufficient to induce directly the jump and the missing energy must be supplied by the thermal bath. Let us term $\Delta E = [\Delta G]_{\rm BJ} + [T\Delta S_i]_{\rm BJ} > 0$ this energy barrier. The BJ will cause a localized change of $b(h_c)$ around some h_c value, and a corresponding output variation $\Delta X_{\rm BJ}$. By taking into account that $[T\Delta S_i]_{\rm BJ} = h_c|\Delta X_{\rm BJ}|$ and $[\Delta F]_{\rm BJ} = h_u \Delta X_{\rm BJ}$, we obtain $\Delta E \equiv \Delta E_+ = [b(h_c) + h_c - H]|\Delta X_{\rm BJ}|$ if $\Delta X_{\rm BJ} > 0$, $\Delta E \equiv \Delta E_- = [-b(h_c) + h_c + H]|\Delta X_{\rm BJ}|$ if $\Delta X_{\rm BJ} < 0$. The relative probability with which the two types of events will occur is controlled by the Boltzmann factor $\exp(-\Delta E/k_B T)$. The average $b(h_c)$ variation per unit time will thus be proportional to $|\Delta b(h_c)|[\exp(-\Delta E_+/k_B T) - \exp(-\Delta E/k_B T)]$, where $|\Delta b(h_c)|$ is defined by the relation $|\Delta X_{\rm BJ}| = X_s p[h_c, b(h_c)]|\Delta b(h_c)|^2$. This leads to the relaxation equation

$$\frac{\partial b(h_c, t)}{\partial t} = -2\,\frac{H_\nu}{\tau_\nu}\sinh\!\left[\frac{b(h_c, t) - H}{H_\nu}\right]\exp\!\left(-\frac{h_c}{H_\nu}\right), \quad (7)$$

where $H_\nu = k_B T/|\Delta X_{\rm BJ}|$, $\tau_\nu = \tau_0 H_\nu \{X_s p[h_c, b(h_c, t)]/|\Delta X_{\rm BJ}|\}^{1/2}$, and $1/\tau_0$ is some attempt frequency associated with the jump. The structure of Eq. (7) is rather complex, because the unknown $b(h_c, t)$ is present in τ_ν, H_ν, and $H \equiv H_a + H_{\rm LS}$. These dependences should be analyzed case by case. Yet, the basic properties of the equation are illustrated well by the simple case where τ_ν and H_ν are approximated by constants, and the time dependence of H is known. Let us consider in particular the case where H is decreased from $+\infty$ down to some value H_0, which is then kept constant in time. In this case, $b(h_c, t = 0) = H_0 + h_c$. Let us look for a solution of Eq. (7) of the form $b(h_c, t) = H_0 + h_c - \Delta b(h_c, t)$.

At sufficiently high h_c, the hyperbolic sine function can be approximated by an exponential and

$$\Delta b \approx H_\nu \ln\left(1 + \frac{t}{\tau_\nu}\right). \tag{8}$$

This solution is valid down to $h_c \sim h_c^* = H_\nu \ln(1 + t/\tau_\nu)$. For $h_c < h_c^*$, $\Delta b \approx h_c$ and $b(h_c, t) \approx H_0$. In the initial relaxation stages, when Δb is small and Eq. (8) applies almost everywhere, the output variation ΔX can be expressed as

$$\Delta X \approx -\left[\frac{dX}{dH}\right]_{H_0} H_\nu \ln\left(1 + \frac{t}{\tau_\nu}\right), \tag{9}$$

where dX/dH represents the system response under appreciable field rates $dH/dt \sim H_\nu/\tau_\nu$. Deviations from this logarithmic behavior will take place in general as Δb increases. A more detailed analysis will be given elsewhere [14].

One can envisage a number of applications of the present approach. An example is the prediction of the heat flow $T\delta S_e$ out of a ferromagnetic specimen, while it is driven along the hysteresis loop. This is not just the heat associated with hysteresis losses. Accurate experiments, reported in several textbooks [15], give evidence of a much richer structure, for which only qualitative interpretations have been proposed. These experimental data can be interpreted by expressing $T\delta S_e$ as $T\delta S_e = TdS - T\delta S_i$, and by making use of Eqs. (6) and (4). Another case is the description of vortex pinning and hysteresis in hard type-II superconductors. Critical state models, of which the Bean model is the leading example [16], have been widely employed for this purpose. Remarkably, critical state models are particular cases of the Preisach model [7]. This is the starting point for a reinterpretation of critical state models in light of the present approach. Equation (5) can be used to describe the vortex-vortex interaction energy and Eq. (4) to estimate the energy dissipation taking place when a vortex line penetrates into the system and triggers a burst of internal vortex rearrangements. Finally, Eqs. (7)–(9) provide a natural description of flux creep phenomena. Details of these applications will be given in a forthcoming more expanded work [14].

The author wishes to thank Martino Lo Bue for a number of helpful suggestions. This work was partially supported by the European Union, PECO Contract No. CIPA-CT93-0239.

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