# *Contributions to modeling and simulation of physical processes with hysteresis*

HABILITATION THESIS

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Iași - 2014

# Contents

# Summary

The present habilitation thesis includes some of the most important scientific results published by the author since obtaining the doctor in science title. The common ground that defines these results is the study of systems with hysteresis. While the main part of this work is constructed around micromagnetic studies of ferromagnetic hysteresis, there is also a section devoted to translating knowledge from ferromagnetic to ferroelectric hysteresis as well as a separate chapter that describes an original model for a different type of hysteresis – the temperature and pressure hysteresis in spin crossover systems.

The thesis is divided in three sections, from which the first one is presenting, in four chapters, scientific results of the author.

After an introduction that outlines some of the main concepts used in the text – like physical or phenomenological model – the first chapter consists of contributions to understanding some of the most intimate mechanisms involved in magnetization processes, those taking place at nanometric to micrometric level. After a short overview of the most important micromagnetic models we present how these models can help understanding magnetization structures formation in amorphous wires as function of size and magnetic properties of the alloy, how the magnetic domain structures can influence resonant absorption in Permalloy wires arrays or what impact it has on magnetic switching of triangular and elliptical nanoislands. All of these studies became possible only recently, when regular computers became strong enough to solve large finite element problems, and even now we still had to use several workstations in a parallel computing environment to approach them. Nevertheless, in some cases, simpler models that require smaller computing power were able to cope with careful chosen problems and this chapter ends with two examples of results obtained using a macrospin micromagnetic model in explaining the role of atomic clusters in rare earth – transition metal magnetic alloys and some characteristics of the noise in magnetic recording.

The second chapter makes use of the same, rather simple micromagnetic model to explain the limits of the approximations considered in a class of phenomenological models – the Preisach-type models – that were designed for significantly larger scale problems. First, we show that the physical mechanisms that lead to the formulation of a feedback Preisach model – i.e., moving model which considers that the interaction field distribution of a macroscopic particulate sample changes linearly with the total moment of the sample – are related to the geometrical characteristics of the samples. Moreover, the results indicate that the linear dependence is only a first-order approximation that is not valid for strongly interacting systems and we propose a more general new model – the PM2 model – which is numerically more efficient and includes previous models as particular cases. The discussion is extended to vector models and the relation between the vector character of samples' magnetic moment and the reversible magnetization component as defined in scalar hysteresis. Here we introduce a versatile vector model designed to efficiently run on parallel computing environments and we generalize the PM2 model adding a reversible component with physical significance.

The third chapter applies methods and results presented in the first two chapters to a recently introduced experimental method – the first-order reversal curves (FORC) diagram. The method was firstly introduced by Mayergoyz as a parameter identification tool for the classical Preisach model but, recently, Pike presented a numerical algorithm that allowed its generalization to any system with hysteresis. First we present how some of our previous results on the relation between geometry and interaction field distribution correlate with FORC diagrams features,

especially with the negative region that cannot be explained with a classic approach. Then, we introduce a similar diagram based on second order reversal curves and we show how it can help in discerning between irreversible and reversible components of magnetization using this opportunity to check how the PM2 model compares to other models in describing experimentally observed features of FORC diagrams. As we have shown in the previous chapter, there is an intimate relation between scalar reversible magnetization component and its vector behavior and, thus, we analyze how FORC diagram can integrate into phenomenological vector modeling. Here we propose two new diagrams: a 3D FORC diagram and a rotational FORC diagram and we show how they can help studying different aspects of vector modeling. The chapter ends with an example of how knowledge in ferromagnetic hysteresis can be applied to ferroelectric hysteresis, presenting a FORC study of polarization processes in ceramics.

The last chapter includes results on modelling a different kind of hysteresis – the thermal and pressure hysteresis in spin crossover compounds – using a new mechano-elastic model. We show that the model we have proposed is able to describe many observed behavior as static and dynamic thermal effects and external pressure influence.

The second section discusses research perspectives with emphasis on what high performance (parallel) computing can bring to hysteresis modeling in both ferromagnetic and spin crossover materials.

The last section includes the references cited throughout this work.

# Rezumat

Prezenta teză de abilitare include unele dintre cele mai importante rezultate științifice publicate de autor de la obținerea titlului de doctor în științe. Toate aceste rezultate au ca aspect comun studiul sistemelor cu histerezis. Cea mai mare parte a lucrării de față este construită în jurul studiului micromagnetic a histerezisului feromagnetic dar există și o porțiune consacrată prezentării felului în care cunoștințele obținute din studiul histerezisului feromagnetic se pot reflecta în studiul histerezisului feroelectric și, de asemenea, un capitol care descrie un model original pentru un alt fel de histerezis – cel în funcție de temperatură și de presiune prezent în sisteme cu tranziție de spin.

Teza este împărțită în trei secțiuni, dintre care prima prezintă, de-a lungul a patru capitole, rezultate științifice ale autorului.

După o introducere care creionează unele dintre principalele concepte utilizate mai departe în text – precum model fizic sau model fenomenologic – primul capitol constă în contribuții direcționate spre înțelegerea unora dintre cele mai intime mecanisme implicate în procesele de magnetizare, cele care au loc la nivel nanometric. După o scurtă vedere de ansamblu a celor mai importante modele micromagnetice prezentăm cum aceste modele pot ajuta la înțelegerea structurilor de domenii magnetice din fire metalice amorfe în funcție de mărimea lor și de proprietățile magnetice ale aliajului, cum structura de domenii poate influența absorbția rezonantă în rețele de fire din permalloy sau ce impact are aceasta asupra comutării în nanoinsule triunghiulare sau eliptice. Toate aceste studii au devenit posibile relativ recent, când calculatoarele obișnuite au devenit suficient de puternice pentru a rezolva probleme de element finit de mari dimensiuni, și chiar și acum a trebuit să folosim mai multe stații de lucru în paralel pentru a aborda aceste probleme. Cu toate acestea, în unele cazuri, modele mai simple ce solicitau putere de calcul redusă au fost capabile să descrie unele probleme iar acest capitol se încheie cu două exemple de rezultate obținute cu un model micromagnetic cu macrospini în explicarea rolului clusterilor atomici asupra proprietăților aliajelor magnetice de pământ rar cu metal de tranziție și unele caracteristici ale zgomotului în înregistrările magnetice.

În al doilea capitol se folosește același model micromagnetic mai simplu pentru a explica limitele unora dintre aproximațiile luate în considerare într-o clasă de modele fenomenologice – modelele de tip Preisach – care au fost proiectate pentru sisteme cu dimensiuni la o scară semnificativ mai mare. Pentru început arătăm că mecanismul ce a dus la formularea unui model Preisach cu feedback – modelul moving în care se consideră că distribuția interacțiunilor într-o probă macroscopică se modifică liniar cu momentul total al probei – este în relație cu geometria probei. Mai mult, rezultatele indică faptul că dependența liniară este numai o aproximație de prim ordin care nu mai este adevărată în cazul sistemelor cu interacțiuni puternice și aici propunem un nou model – modelul PM2 – mai eficient din punctul de vedere al calculelor necesare și care include modele existente drept cazuri particulare. Discuția este apoi extinsă la modele vectoriale și la relația dintre caracterul vectorial al momentului magnetic și componenta reversibilă a magnetizării așa cum este definită în histerezisul scalar. Aici introducem un nou model vectorial proiectat să ruleze eficient în medii de calcul paralel și generalizăm modelul PM2 adăugând o componentă reversibilă descrisă pornind de la un suport fizic.

Al treilea capitol aplică metode și rezultate prezentate în primele două capitole unei metode experimentale introdusă recent – metoda diagramelor curbelor de întoarcere de prim ordin (FORC). Această metodă a fost introdusă de Mayergoyz pentru identificarea parametrilor modelului Preisach clasic iar, recent, Pike a prezentat un algoritm ce permite generalizarea

metodei la orice sistem cu histerezis. Pentru început prezentăm felul în care rezultate prezentate în capitolele anterioare se corelează cu aspecte observate în diagramele FORC, în special cu regiunile negative ce nu pot fi explicate în modelul clasic. Apoi introducem o diagramă similară pornind de la curbe de magnetizare de ordinul al II-lea și demonstrăm cum aceasta poate fi folosită în a discerne componentele reversibilă și ireversibilă ale magnetizării, folosind această oportunitate și pentru a verifica felul în care modelul PM2 descrie diagrame FORC experimentale, în comparație cu alte modele. După cum am arătat anterior, există o relație intimă între componenta reversibilă scalară a magnetizării și comportarea vectorului magnetizație; în acest capitol extindem această discuție la felul în care diagrama FORC se integrează în modelele fenomenologice vectoriale. Astfel, propunem două noi diagrame: o diagramă FORC 3D și o diagramă FORC rotațională și arătăm cum acestea pot ajuta studiului diferitelor aspecte ale modelelor vectoriale. Capitolul se încheie cu un exemplu al felului în care cunoștințe dobândite în studiul histerezisului feromagnetic pot fi aplicate în histerezisul feroelectric prezentând un studiu FORC al procesului de polarizare în materiale ceramice.

Ultimul capitol include concluzii în studiul unui alt tip de histerezis – cel determinat de temperatură și de presiune în compuși cu tranziție de spin – cu ajutorul unui nou model mecano-elastic. Demonstrăm cum modelul propus de noi este capabil să descrie multe dintre comportările observate precum efecte termice statice și dinamice sau influența presiunii externe.

Cea de a doua secțiune aduce în discuție perspective de profesionale de cercetare cu accent pe contribuția potențială a calculului de înaltă performanță (paralel) în modelarea histerezisului în materiale feromagnetice și cu tranziții de spin.

Ultima secțiune include referințe citate de-a lungul tezei.

# Section I – Scientific and professional results

## 1 Introduction

The philosophers of science have identified many *categories of models* used in physics, starting from a multitude of criteria, like whether the model uses physical of fictional objects, if the model is a preliminary or a complement of a theory and many more. In the framework of the present work – the hysteretic processes – there are two categories of models of a particular interest, i.e. the *phenomenological* and the *physical* models.

Phenomenological models, in general, are defined [1] as "*models that only describe observable properties of their targets and abstain from postulating any hidden mechanisms that might govern the studied phenomenon*" and several such models have been extensively used in magnetism (as the main area of physics that drives the study of hysteresis), especially due to their facile implementation.

Often seen as a disjunctive class of models, the physical models have a different approach to describing phenomena, starting from known physical theories valid for small parts of a complex system and trying to explain the behavior of the ensemble.

In the case of magnetism, one of the most prominent theory have the roots in the works of William Fuller Brown, Jr. [2, 3] published in 1940, influenced by early contributions of Landau and Lifshitz. The theory leads to a system of nonlinear three-dimensional coupled differential equations which was impossible to solve in the general case at the time of its publication. Since the computers have gained an important role in physics, the field of modeling magnetic properties of materials have flourished and several micromagnetic models appeared. Nevertheless, these physical models – based on numerical solutions of the Brown's equations – require a significant computing power in order to describe rather small, up to micrometric size samples and, thus, even though the numerical solutions became available, the micromagnetic models were difficult to apply for real, macroscopic size samples.

The need for models able to give a fast response when characterizing large samples was fulfilled by a set of phenomenological models of hysteresis, like the Preisach Model [4], the Jiles-Atherton Model [5] or the Energetic (Hauser) Model [6]. None of these models fit well into the previous definition given for the phenomenological models in the sense that all of them start from some physical considerations, none of them is completely independent of a theory [1]. Still, all of them are considered phenomenological models because all sacrifice the complexity of the physical reality, resorting to strong simplifications in order to give fast response that fits the observed behavior.

One can simplify saying that the phenomenological models of hysteresis usually have a *top-down* approach while the physical models of hysteresis are built using a *bottom-up* strategy. This interpretation might not be valid for the two categories of models as a whole, but applies very well for the two representatives classes – one of each category – that are the main focus of this work: the (physical) micromagnetic models and the (phenomenological) Preisach-type models.

# 2 Micromagnetic models

## 2.1 General remarks on micromagnetic modeling

In the last decades the magnetic devices industry witnessed two major changes. In a first instance, the market has more than doubled – if until about ten years ago the only market demanding information storage devices was the computer industry, in the last years these devices have been attached to more and more common electronics like portable MP3 players and TV sets with digital recording facilities. Secondly, the storage devices industry has lost a large market segment when many producers of portable commercial products decided not to use hard-drives for their new generation of devices and they have used instead semiconductor Flash memories. For the same storing capacity, the Flash memory is up to 100 times more expensive and has a smaller number of guaranteed read-write cycles compared to the magnetic memory but has the advantage of being smaller and without any moving parts.

As a consequence, the research in magnetic devices was aimed more towards the mechanical parts miniaturization and the power consumption reduction.

The physical magnetic support of data has changed a lot since the beginning of the magnetic recording, leading to a more than 1000 times increase of the recording density, but the actual information holder remained the same: the information is stored in the orientation of the magnetic moment of a small portion of material. In the same time, the information writing process has remained practically the same: if certain information is to be written on one segment of material that represents a bit, then a magnetic field strong enough to switch the magnetic moment is applied and maintained for a certain period of time.

The race toward smaller magnetic storage devices lead to the *magnetic recording trillema*:

- In order to increase the recording density one must decrease the volume of the magnetic grains that make the medium.
- Smaller magnetic volume particles have smaller energy barrier separating the two magnetic moment equilibrium minima so the probability that the magnetic moment switches under the influence of the thermal movement is increasing. While the value of the thermal energy can be considered a constant (decreasing working temperature is not a practical solution for these devices), one must increase the energy barrier by increasing the magnetic anisotropy of the material $\left(K_{anizo}\right)$.
- Larger $K_{anizo}$ also means larger coercive field so significantly larger magnetic fields are required to write the information but these values are limited by the writing head design.

There are also other different challenges to overcome in the way towards a higher information recording density. For example, even if one succeeds in decreasing the bit size while maintaining its thermal stability and its writability, its readability becomes a problem: due to the small quantity of magnetic material in one bit one must either (or both) increase the quality of the magnetic read head detector and increase the magnetic saturation value of the material.

No matter what technical solution will prevail in future magnetic storage devices, the micromagnetic analysis of the magnetic moments static structure as well as their switching behavior are key points in magnetism research, bringing understanding and contributing to innovation.

For several decades, the phenomenological models were preferred in the study of hysteresis because of their considerably small computing power requirements for samples large enough to be relevant for applications.

In recent applications, like the current hard disk storage media or the existing nanostructured materials made of a mixture of different magnetic phases, the typical grain size is in the range of 10 to 100 nm. At this scale, the quantum effects, usually seen in the form of exchange interactions, have to be taken into account. The macroscopic phenomenological hysteresis models fail here because most of them were built for bulk samples and the micrometric devices lack the statistical significance assumed in bulk. Also, the complete *ab initio* quantum approach is impossible to apply to such samples that, even though they are small, they still contain $10^7$ atoms or more.

Driven by the strong increase in computing power in the last 20 years a different class of physical models, at an intermediate scale, gained popularity. The *micromagnetic models* are based on the notion of macrospin – the magnetic moment of a small ferromagnetic region where all the atomic moments are considered aligned – studying the way they interact and evolve in time. At the moment, computers can be used to solve small size problems but, by carefully chosen approximations, the problem size can be increased up to tens or hundreds of micrometers.

One of the free micromagnetic applications that became a standard in the scientific community is the Object Oriented MicroMagnetic Framework – *OOMMF* – built by NIST, USA. One of its main drawbacks is that it uses the finite difference method to solve the problems and, thus, it describes well only samples with rectangular geometry because the solving method divides the sample in rectangular cells.

We have focused our attention on a newer, more promising open source micromagnetic framework – *magpar* – developed at the Technical University Vienna, Austria. Magpar uses the finite element method, based a tetrahedral mesh, and, also, was designed to run in parallel computing environments, allowing us to use the resources of our computer cluster to solve problems too big for ordinary computers.

*This chapter includes two categories of micromagnetic results:*

- *finite element micromagnetic analysis of different structures, like different kind of ferromagnetic wires*[1,2]*, patterned media*[3] *and nanodots*[4]*, and*
- *macrospin approach to micromagnetic analysis of "exchange spring"-type permanent magnets*[5] *and magnetic noise in recording media*[6]*.*

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# 3 Phenomenological models of hysteresis

## 3.1 General remarks on scalar and vector phenomenological hysteresis models

The Preisach Model [4], a simple model representing the main contribution to science of the Hungarian physicist Ferenc Preisach [43], is in the same time one of the most important models of hysteresis for at least two reasons: it allowed an uncountable number of improvements and it gives an excellent overview of the magnetization processes through its graphical, "Preisach plane" description.

The *classical Preisach model* is a one-dimensional model which describes a system of perfectly aligned single domain ferromagnetic particles. The external magnetic field is considered to act on the alignment direction, which also corresponds to each particle's magnetic easy axis. This is why the hysteresis loop of each particle – named "*hysterons*" by Pescetti [44] – is considered to be rectangular, each such particle switching abruptly between two possible states.

The inter-particles interactions can be expressed as a magnetic field, is also considered to act on the easy axes direction and do not change during any magnetization process. In this hypothesis the interaction field is algebraically added to the external applied field and this can be represented as a shift of the elementary hysteresis loops along the field axis.

While the particles are not taken to be identical, the width of the hysteresis loops – corresponding to the coercive field values – can differ from one particle to another.

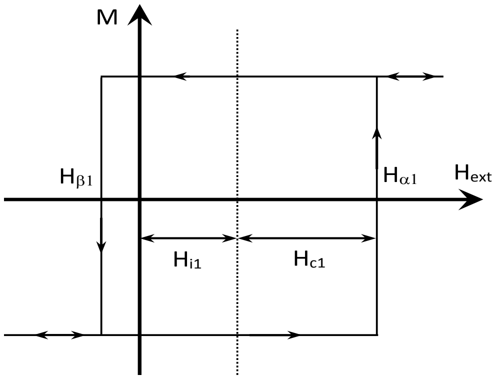


***Fig. 3.1** – The elementary hysteresis loop of a particle with an interaction field $H_{i1}$ and a coercive field $H_{c1}$.*

Each hysteron is completely characterized by two numbers, either the $\left(H_{\alpha 1}, H_{\beta 1}\right)$ pair of switching field values or the $\left(H_{c1}, H_{i1}\right)$ pair of coercive and interaction field values (Fig. 3.1). There are some very simple relations connecting these two pairs:

$$\begin{cases} H_{\alpha 1} = \left(H_{c1} - H_{i1}\right) \\ H_{\beta 1} = -\left(H_{c1} + H_{i1}\right) \end{cases} \text{ or } \begin{cases} H_{c1} = \left(H_{\alpha 1} - H_{\beta 1}\right)/2 \\ H_{i1} = -\left(H_{\alpha 1} + H_{\beta 1}\right)/2 \end{cases} \tag{3.1.1}$$

Using any pair of fields characterizing a hysteron as coordinates, one can associate to each hysteron a point in a plane (called *Preisach plane*). Considering only realistic physical values, the plane reduces to a triangle (Fig. 3.2) limited by two values – i.e. the maximum $H_{\alpha i}$ and the minimum $H_{\beta j}$ values – and by the first bisector of the coordinate system (considering that always $H_{\alpha k} \geq H_{\beta k}$ ). For a certain statistical ensemble of hysterons one can define a distribution of sizes, which translates into a distribution of coercive field values, and a distribution of interaction field values, thus, depending on these distributions, to each point in the Preisach point one can associate more or less hysterons. This distribution of switching elements, often represented as a contour plot in the Preisach plane (Fig. 3.2), or as two dimensional slices along the $H_c$ and $H_i$ axes, it is called the *Preisach distribution*, a function of the two coordinates $P\left(H_\alpha, H_\beta\right)$.

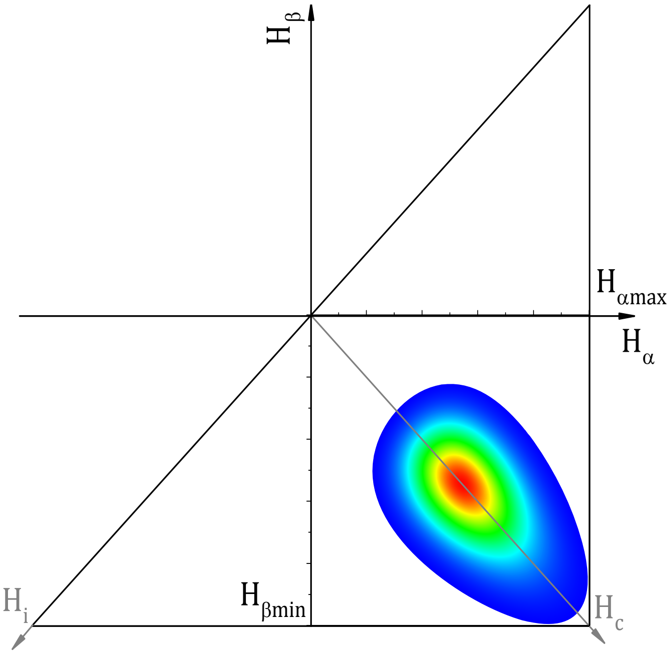


***Fig. 3.2*** – The Preisach plane and a typical Preisach distribution.

Considering that each element (or particle) has a rectangular hysteresis loop, the only possible values of its magnetic moment are $+M_S$ and $-M_S$. All the particles in one point of the Preisach plane will always have the same orientation of their magnetic moments (either positive or negative) so they will contribute to the total moment of the system with $\pm M_S P\left(H_\alpha, H_\beta\right)$.

Even though for real systems the Preisach distribution would be discrete, for facile mathematical characterization this distribution is taken as a continuous functions so the summation of all the hysterons translates to integration of this function over the Preisach plane.

$$M(H) = \iint\limits_{H_{\beta\min} < H_\beta \leq H_\alpha < H_{\alpha\max}} \gamma_{\alpha\beta}(H) P\left(H_\alpha, H_\beta\right) dH_\alpha dH_\beta \tag{3.1.2}$$

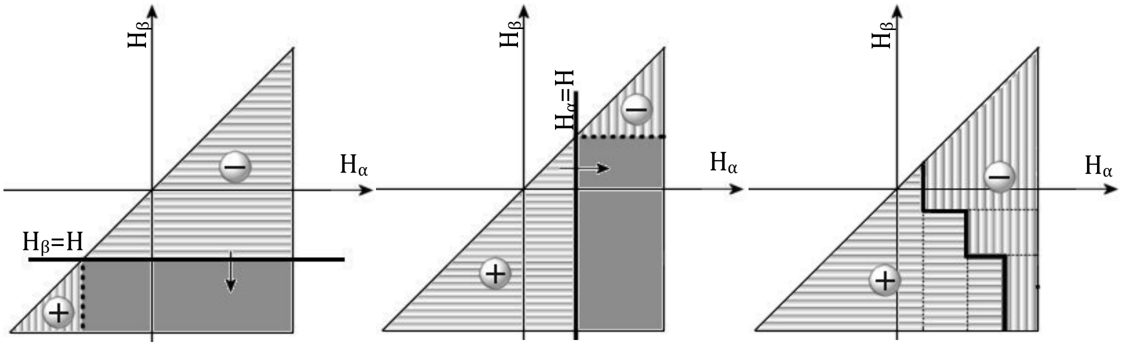
where $\gamma_{\alpha\beta}(H)$ takes values of $+M_S$ or $-M_S$ depending on the state of the hysterons at the coordinates $\left(H_\alpha, H_\beta\right)$ at the $H$ applied external field. One must remark that the values of the

$\gamma_{\alpha\beta}(H)$ function are depending not only on the coordinates $(H_\alpha, H_\beta)$ and the present values of the external field but also on its previous values.

Beside the simplicity, another strong asset of the Preisach model is its graphical interpretation that arises from the rules that can be derived analyzing the switching behavior in the Preisach plane:

- when $H$ decreases, all hysterons characterized by $H_\beta > H$ (situated above the horizontal line $H_\beta = H$ ) must be in the negative state (Fig. 3.3(a));
- when $H$ increases, all the hysterons with $H_\alpha < H$ (to the left of the vertical line $H_\alpha = H$ ) must be in the positive state (Fig. 3.3(b)).

The hysterons situated in the Preisach plane area that is not above the $H_\beta = H$ line nor to the left of the vertical line $H_\alpha = H$ are unperturbed, they maintain the state given by previous external fields – this is the *memory region* of the Preisach plane. Mayergoyz demonstrated [45] that at any time the Preisach plane contains two areas – corresponding to the positive and negative polarized particles – areas that are separated by a staircase line whose vertices have coordinates given by previous local maxima and minima of the applied field (Fig. 3.3(c)).



*(a)* *(b)* *(c)*

***Fig. 3.3*** *– The evolution of the Preisach plane for a decreasing (a), an increasing field (b) and an arbitrary state after the application of a sequence of fields (c).*

In the same time, Mayergoyz [45] has obtained two properties of the systems described by the CPM:

- the *wiping-out property* which states that applied field minimum (or maximum) erases from history previous minima (maxima) greater (or smaller in the case of maxima) than the current field. In other words, minor hysteresis loops must close perfectly, and,
- the *congruency property* which states that all the minor hysteresis loops drawn between the same extreme field values must be congruent.

One of the first remarks of the experimentalists was that in the vast majority of the real systems, the wiping-out property is verified while the congruency property is not verified and that lead to the need of better Preisach-type models, able to describe the experimentally observed behavior.

One must mention here two of the most important improvements brought to the classical Preisach model:

The *generalized Preisach model* (GPM) [46] started from the remark that while any change of the total magnetic moment is due to a certain number of hysterons switching from one state to another, the Preisach model, its classical form, is able to describe only irreversible processes. A simple, widely used, approach to including reversible phenomena in the Preisach model was proposed also by Mayergoyz [45] and consists in considering a separated distribution of completely reversible hysterons, characterized by $H_\alpha = H_\beta$. In the Preisach plane this form a singular distribution along the first bisector of the coordinate system. In this case equation (3.1.2) becomes:

$$M(H) = S \cdot \iint\limits_{\substack{H_{\beta\min} < H_\beta \leq \\ \leq H_\alpha < H_{\alpha\max}}} \gamma_{\alpha\beta}(H) P(H_\alpha, H_\beta) dH_\alpha dH_\beta + (1 - S) \cdot \int\limits_{\substack{H_{\beta\min} < H_\alpha \\ < H_{\alpha\max}}} \gamma_{\alpha\alpha}(H) f(H_\alpha) dH_\alpha \tag{3.1.3}$$

where $S$ is the *squareness* of a given system, usually computed as the ratio between the values of the magnetization at remanence and at saturation.

Another important improvement was introduced by Della Torre [47] which started from the assumption that the actual field acting on each component of a magnetic system is influenced not only by the external magnetic field source but also by the system itself which produces a magnetic field due to its own magnetization. On these premises, Della Torre built the *moving Preisach model* (MPM), a feedback model which simply takes the input $H$ as function of the output $M$:

$$H = H_{ext} + \alpha M \tag{3.1.4}$$

where $H_{ext}$ is the applied (external) field and $\alpha$ is a constant named *moving parameter*.

The two presented extensions are addressing different aspects of the Preisach model and, thus, are not excluding one another. In fact, the *Generalized Moving Preisach Model* (GMPM) is one of the most used variants of the Preisach-type models to date.

*Vector Preisach-type models*

The fact that the most successful phenomenological hysteresis models are the scalar ones – even though the magnetization processes have an intrinsically vector nature – can be justified by the need to simulate the most widely used experimental measurements, those obtained using scalar devices. The scalar approach is often sufficient when keeping the same direction of the applied field throughout the experiment. The first problems are encountered when changing the field direction: two sets of scalar measurements on the same sample for two different directions of the external field can be explained by a phenomenological scalar model only by using two different sets of parameters and thus, finding physical meanings in the parameters becomes more difficult.

In contrast to this approach, the vector phenomenological hysteresis modeling is having as ideal the discovery of a model that, after identification, is able to describe using the same set of parameters both scalar-type processes for any direction of the external field as well as fully vector processes, like the magnetization in rotating external fields.

A number of vector extensions of the Preisach model have been developed [48], [49], [50], [51] but none of them proved to be in the same time robust and numerically efficient.

*This chapter sums contributions of the author on micromagnetic validation of the Preisach-type model parameters with consequences in interaction analysis*[7] *– which also lead to proposing a new variant of Preisach-type model*[8] *– in vector modeling*[9] *as well as in the study of reversible magnetization phenomena*[10]*.*

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# 4 Understanding and using the first-order reversal curves (FORC) diagram method

## 4.1 General remarks on FORC experimental method

For a phenomenological model, the parameters identification procedures are, in fact, a core component of the model itself as these procedures are the main link of the model with the physical reality.

In the case of the Preisach-type models the identification of the parameters means, essentially, finding the Preisach distribution $P(H_\alpha, H_\beta)$. Most attention was paid to the Preisach distributions parameters when the function was written in terms of coercive field and interaction $P(H_c, H_i)$, with the hope that identifying these parameters for a certain sample would give insight on some of its physical parameters.

The strategies for Preisach-type models parameters identification went in two general directions.

In the early stages, until the publication of the Pike's paper on the first-order reversal curves that we shall focus on later, the foremost strategy was to try fitting the main hysteresis curves starting from a given mathematical form for the Preisach distribution (e.g. a product of a Gauss distribution for the interactions and a log-normal distribution for the coercive fields). The quasi-general failure to find the parameters of such functions able to describe the various observed behavior lead to a series of improved Preisach-type models, from the most simple and successful ones as those already described in the previous section, to the most complicated ones that remained as theoretical projects due to difficult implementations.

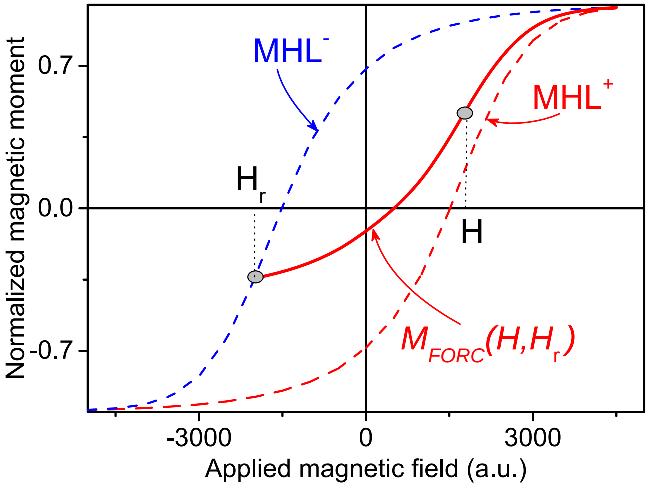

**Fig. 4.1** – *The definition of a first-order reversal curve.*

When establishing the mathematical basis of the Preisach model, Mayergoyz [45] also presented an identification strategy based on a set of measured curves called *first-order reversal curves (FORC)*. The best defined magnetic state of a system it is its magnetic saturation – in the terms of the Preisach model in this point the entire history of previous applied fields is erased. Starting from this point, the major hysteresis loop curve can be defined as a zero-order reversal curve. If one stops the variation of the external field in any point along the major hysteresis curve and then one changes the sign of field variation back towards saturation, one obtains a first-order reversal curve. The value of the magnetization on any point of such a curve is function of both the actual value of the applied field and the value of the reversal field $M_{FORC}\left(H_r,H\right)$ (Fig. 4.1).

Then, the Preisach distribution, in the hypotheses of the Classical Preisach Model, can be defined as the second order mixed derivative:

$$P\left(H,H_r\right)=-\frac{1}{2}\frac{\partial^2 M_{FORC}\left(H,H_r\right)}{\partial H\,\partial H_r} \tag{4.1.1}$$

In time, the interest in the FORC method diminished because of the inherent numerical errors introduced when computing the second-order derivative of experimental data.

In 1999, Pike [69], [70] proposed a simple method to calculate the distribution starting from a set of experimental data which eliminates the previously mentioned errors by using an interpolation procedure. The measurements are considered on a grid of points in the $\left(H,H_r\right)$ system of coordinates. For each point one chooses a certain number of neighbors around the reference point obtaining a rectangular region that is interpolated using the polynomial function $M_{FORC}^{\text{interpolated}}\left(H,H_r\right)=aH+bH_r+cH^2+dH_r^2+eHH_r$ , eliminating the need for the numerical derivation, while the value of the second mixed derivative of this polynomial is given by the coefficient $e$ of its mixed term. Beyond the mathematical procedure, another important contribution of the Pike's paper is the proposal to separate the FORC method from its roots, the Preisach Model, and to use it as a pure experimental method, called the FORC diagram method. Nevertheless, even if Pike's ideas started a new trend in hysteresis modeling – the FORC diagram analysis – his proposal was sometimes misunderstood by authors which disregarded the fact that when discussing the FORC diagrams in terms of coercive field and interaction distributions they are, in fact, relying on a Preisach-type model approach.

*This chapter collects results of the author aimed towards understanding the FORC diagram method starting from micromagnetic considerations[11]. In this framework we have proposed several new tools, like the second-order reversal curves[12] useful in analyzing both irreversible and reversible magnetization components, the 3D FORC diagram and the rotational FORC diagram[13] and we have proposed a new method of using directly the experimental FORC in simulations[14]. The method was also applied to ferroelectric hysteresis leading to understanding of the polarization phenomena[15].*

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# 5 Modeling multiple hysteresis in spin crossover compounds

## 5.1 General remarks on spin crossover compounds

Spin crossover complexes belong to the class of molecular magnets and exhibit thermal spin transition in from the low spin (LS) state, at low temperatures, to the high spin (HS) state at higher temperatures [107-109]. Depending on the strength of intermolecular interactions, the thermal transition can be gradual, or can be accompanied by a hysteresis, that makes these materials interesting as possible high-density information storage systems.

The spin transition molecules are composed by transition metals ions having four to seven electrons on their last electronic level $d$, situated in an octahedral ligand field that splits the $d$ orbitals into antibonding $e_g$ and weakly bonding $t_{2g}$ orbitals. Due to a higher occupancy of the $e_g$ orbitals in the case of HS molecules, their molecular volume is larger than the one of the LS molecules. The difference in molecular volume between the two possible spin states induces distortions of the sample lattice during the transition. These distortions are at the origin of intermolecular interactions that are treated here as connecting springs that can be either compressed or elongated relatively to their equilibrium length, depending on the relative positions of neighbor molecules. The macroscopic state of the switchable system is usually characterized by the proportion of spin crossover units in the HS state, that is called HS fraction and is denoted here as $n_{HS}$.

Intensive research has been carried out to establish the role of molecules interactions on the static and the dynamic properties of spin crossover solids. Using phenomenological interaction parameters, acting similarly for all molecules, the mean field models can reproduce several features of spin crossover compounds, such as the dependence of the thermal hysteresis width on intermolecular interactions, or the shift of the thermal transition towards higher temperatures [110], when applying higher external pressures. However, these simple models do not distinguish between short range and long range interactions and therefore do not allow an advanced analysis of the hysteresis loops, nor are able to explain how the individual switch of molecules spreads in the entire solid. In addition, these models cannot be applied for the study of size effects in nanoparticulate spin crossover systems. For these purposes Ising-like models with short and long range interactions have been proposed [111]. They led to the conclusion that while long range interactions act on the width of the hysteresis loop, the short range effect is essentially visible in the loop steepness. Another approach is based on the hypothesis that the spin transition occurs through domains of molecules in the same spin state; and allow the reproduction of minor hysteresis loops, by considering different interactions in the Preisach model [112].

*This chapter brings together contributions of the author in developing a new model for hysteresis in spin crossover compounds, the mechano-elastic model*[16].

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