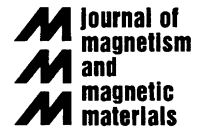




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Invited paper

Microscopic foundations of the Rayleigh law of hysteresis

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Abstract

The hysteresis properties of ferromagnetic materials at low fields are described by the Rayleigh law. We analyze the problem in the light of modern statistical mechanics models of hysteresis. In particular, we compute the demagnetization curve and derive the Rayleigh parameters a and b in the random-field Ising model and in a model of domain wall depinning. In the random-field Ising model, the Rayleigh law is obeyed only in the disorder dominated phase, while in the low disorder phase it is not possible to demagnetize the sample. This approach allows us to link a and b to microstructural parameters, such as the domain wall energy, the internal disorder or the exchange interactions. Finally, our results are compared with experiments. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Hysteresis; Domain wall dynamics

1. Introduction

In 1887, Lord Rayleigh analyzed the hysteresis properties of ferromagnetic materials at low fields, close to the demagnetized state [1–3]. When the field is cycled between $-H_m$ and H_m , the magnetization M is found to follow a simple quadratic law $M = (a + bH_m) \pm b(H_m^2 - H^2)/2$, where the signs \pm distinguish the upper and lower branch of the loop. The area of the loop can easily be computed and is given by $W = (\frac{4}{3})bH_m^3$. In addition, the response of the system to a small field change, starting from the demagnetized state is given by $M_m = aH_m \pm bH_m^2$. The Rayleigh law has been shown to hold in several ferromagnetic materials [4] and it has been also widely applied to describe ferroelectric materials [5,6]. A few papers have reported significant deviations from the simple quadratic law but no explanation has been provided [7].

In 1942, the Rayleigh law was interpreted by Néel in terms of the motion of a point (i.e. a rigid domain wall) in a random energy landscape, whose statistical properties determine the value of a and b [8]. In particular, a is associated to reversible motions inside one of the many

minima of the random potential, while b describes irreversible jumps between different valleys. Several generalizations of this approach have been proposed, considering more refined forms for the random energy landscape [9–13]. These approaches assume a single rigid domain wall and thus do not consider the effect of domain wall bowing and the interactions between different domains. When these effects are important, it is hard to link the Rayleigh law to the microstructural properties of the material, such as dimensionality, chemical composition or interaction types.

Here, we reconsider all these problems in light of novel statistical mechanics approaches to hysteresis. In particular, we compute the Rayleigh parameters a and b in the random-field Ising model (RFIM) [14] and in a model of domain wall depinning [15–21]. This approach allows us to link a and b to microstructural parameters, such as the domain wall energy, the internal disorder or the exchange interactions.

2. Models

A microscopic theory of hysteresis should, in principle, recover the Rayleigh law from the collective properties of interacting magnetic moments. The

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Rayleigh parameters would then be expressed in terms of micromagnetic parameters, that are available in the literature for a variety of ferromagnetic materials. Ferromagnets can in general be described by a locally varying magnetization $M_i(\vec{r})$ evolving under the action of the externally applied field. The evolution of M_i is ruled by appropriate relaxation equations that can be written in terms of the micromagnetic energy [2,3]. Considering for simplicity a uniaxial material, we can write the energy as

$$E = \sum_{i=1}^3 \int d^3r [A(\nabla \vec{M}_i)^2 + K(M_i n_i)^2 - \mu_0(H_i + H_{\text{dem}}^{(i)})M_i], \quad (1)$$

where A is the exchange interaction, K is the anisotropy constant, n_i is the anisotropy axis, H_i is the applied field, and $H_{\text{dem}}^{(i)}$ is the component i of the demagnetizing field and M_s is the saturation magnetization.

An essential contribution to the properties of the hysteresis loops is given by disorder, due to crystal imperfections, internal stresses, non-magnetic impurities, that are present in most magnetic materials. Néel recognized this fact and replaced the micromagnetic free energy by a random function of the magnetization (i.e. $E = E_R(M) - HM$) [8]. The approximation is rather drastic but makes the problem analytically tractable. In general, we can model the disorder by quenched local fluctuations of the micromagnetic parameters. Disordered interaction terms are conventionally denoted as random bonds ($A \rightarrow A(\vec{r})$), random anisotropies ($n_i \rightarrow n_i(\vec{r})$) and random fields ($H \rightarrow H_{\text{ext}} + h(\vec{r})$). A complete solution of micromagnetic equations including disorder is a very complicated task. One should then resort to some kind of approximation. In recent years, two main approaches have been undertaken to describe the magnetization properties of disordered materials.

The first approach takes into account the fact that in soft magnetic materials demagnetizing fields give rise to broad domains and the magnetization process is dominated by domain wall motion. One can thus reduce the problem to the motion of a flexible domain wall in a random potential. The domain wall contribution to the micromagnetic free energy can be expressed in terms of the domain wall coordinates $z(\vec{x})$, inserting in Eq. (1) $M_i(\vec{r}) = \delta_{i3} M_s g((r_3 - z(\vec{x}))/\delta_w)$ in Eq. (1), where $\delta_w \simeq \sqrt{A/K}$ is the domain wall width and $g(x) = \pm 1$ for $x \rightarrow \pm \infty$ [15,18,21]:

$$E_{\text{dw}} = \int d^2x [\gamma_w (\nabla z(\vec{x}))^2 - z(\vec{x})\mu_0 M_s (H + H_d(\{z(\vec{x})\}) + V(\vec{x}, z(\vec{x})))] \quad (2)$$

where the domain wall energy is given by $\gamma_w \simeq \sqrt{AK}$, the stray field is given by $H_d = M_s \int d^2x' \partial z / \partial x' (x - x') / |r - r'|^3$ and V is a random function taking into account all the disorder contributions. In the following we will

consider V as a superposition of pinning centers randomly distributed in space

$$V(\vec{x}, z) = \sum_p f_0 \exp - ((z - Z_p(\vec{x}))/\xi_p)^2, \quad (3)$$

where f_0 is the strength of each pinning center, of width ξ_p located in Z_p . The effective form of the pinning potential is not essential as long as the interaction range ξ_p is finite. This is not the case for pinning due to isolated dislocations, whose stress field decays as $1/r$. One should notice, however, that dislocations are typically arranged in patterns where the stress field is screened, providing us with an effective correlation length. In the following, we will restrict our attention to cases in which the pinning correlation length is smaller than the domain wall width, so that we can replace ξ_p by δ_w . This case corresponds mainly to the effect of non-magnetic impurities (see Ref. [16] for a discussion of this point).

Models based on Eq. (2) have been used in the past to compute the coercive field [15,17], analyze thermal relaxation [16], explain the statistical properties of the Barkhausen noise [19–22] and describe magnetization creep in thin films [23].

An approach based on domain wall motion is not adequate to describe the magnetization properties of hard ferromagnets, where the presence of strong random anisotropies prevents the formation of extended domains. For this class of materials, a description in terms of interacting spins seems more appropriate. Several disordered spin models have been proposed in the past to describe hysteresis. Among those, the simplest and most studied is the RFIM [14,24], where a spin $s_i = \pm 1$ is assigned to each site i of a d -dimensional lattice. The spins are coupled to their nearest-neighbors spins by a ferromagnetic interaction of strength J and to the external field H . In addition, to each site of the lattice is associated a random field h_i taken from a Gaussian probability distribution with variance R , $P(h) = \exp(-h^2/2R^2)/\sqrt{2\pi R}$. The Hamiltonian thus reads

$$E = - \sum_{\langle i,j \rangle} J s_i s_j - \sum_i (H + h_i) s_i, \quad (4)$$

where the first sum is restricted to nearest-neighbors pairs. The dynamics proposed in Ref. [14] is such that the spins align with the local field

$$s_i = \text{sign} \left(J \sum_j s_j + h_i + H \right). \quad (5)$$

In this way, a single spin flip can lead the neighboring spins to flip, eventually trigger an avalanche. Using this dynamics, it has been shown that the RFIM displays a phase transition in $d \geq 3$ as a function of R [14,24]. For $R < R_c$, the saturation loop has a discrete jump in the magnetization at $H = \pm H_c$. The jump disappears for

$R > R_c$ and $R = R_c$ corresponds to a critical point and the model satisfies scaling laws [14,24].

3. Collective pinning effects in domain wall hysteresis

The hysteresis properties of interfaces in random media have been studied in the past in the context of frictional sliding [25]. Similar studies for ferromagnetic domain walls has only been restricted to the case of high driving frequencies and large fields [26]. It is interesting to note, however, that interfaces in random media obey return point memory, a typical properties of ferromagnetic hysteresis. Here, we use a model based on Eq. (2) to analyze the Rayleigh law at low frequency. For the sake of simplicity we do not consider here demagnetizing fields, which are essential, however, to account for the large scale behavior of the magnetization [21]. When considering small scale displacements of the domain wall, we expect the domain wall energy (scaling as q^2 in Fourier space for a deformation of wave vector \vec{q}) to be more relevant of the stray field contribution (scaling as q). The following arguments can nevertheless be generalized including the effects of demagnetizing fields [15].

The scaling properties of the Rayleigh parameters can be obtained using collective pinning theory [15,16,23]. The central concept is the identification of a coherence length L_c , defining a region of space where the domain wall moves freely from the pinning centers. The coherence length can be obtained comparing the domain wall energy with the disorder fluctuations over a region of length L_c , considering only small transverse wall deformations of the order of δ_w

$$\begin{aligned} \gamma_w \delta_w^2 &\sim f_0 (n_0 L_c^2 \delta_w)^{1/2}, \\ L_c &\sim (\gamma_w \delta_w^3)^{2/3} / (n_0^{1/2} f_0). \end{aligned} \quad (6)$$

The depinning field H_c , which can be identified with the coercive field, is obtained comparing the pinning energy with the magnetostatic energy over a region of length L_c [15,16,23]

$$\begin{aligned} \mu_0 M_s H_c L_c^2 \delta_w &\sim f_0 (n_0 L_c^2 \delta_w)^{1/2}, \\ H_c &= (n_0 f_0^2) / (\gamma_w \delta_w^2 \mu_0 M_s). \end{aligned} \quad (7)$$

This expression for the coercive field recovers the result obtained in Ref. [15] when two-dimensional domain vaulting is considered. In models based on a rigid domain wall [8,10,11], the coercive field is instead proportional to the standard deviation of the pinning field (i.e. $H_c \propto f_0 \sqrt{n_0}$).

Under the application of a small external field, the “unpinned” regions of the domain wall will bow slightly and contribute to the magnetization [16]. The expression for the susceptibility $a = dM/dH$ is similar to the one found in textbooks when discussing reversible susceptibility [2,3], with exception that the bowing length is now

given by L_c ,

$$a \sim \mu_0 M_s^2 L_c^2 / \gamma_w \sim (\gamma_w \delta_w^3) / (n_0 f_0^2). \quad (8)$$

This result differs from estimates based on generalizations of the Néel theory [8,10,11] where γ_w does not appear.

The first correction to the linear susceptibility, and hence the parameter b , can be obtained noticing that a small applied field can, in principle, lead to local depinning events, thus increasing the coherence length [25]. Using the arguments reported in Ref. [25], one can show that, to lowest order in H , $L_c(H) = L_c(0)(1 + c|H|/H_c)$ where c is a numerical factor. Inserting this expression into Eq. (8) and expanding for small H , we obtain $b \sim a/H_c$. A similar expression was already reported by Néel in 1942 [8].

In order to test these considerations, we perform numerical simulations based on Eq. (2). The coordinates of the domain walls evolve according to an overdamped equation $\Gamma dz/dt = -\delta E/\delta z$, where Γ is an effective viscosity. The equation of motion is discretized on a grid of size 50×50 and solved by an adaptive-stepsize Runge–Kutta method. We use quasistatic driving condition, applying a field H and integrating the motion until the domain wall comes to rest. The system is first demagnetized, with the successive application of positive and negative fields of decreasing amplitude, and then we cycle the field between $-H_m$ and H_m . The Rayleigh parameters are extracted from the scaling properties of the hysteresis loops (see Fig. 1) which are averaged over several realizations of the disorder. The procedure is

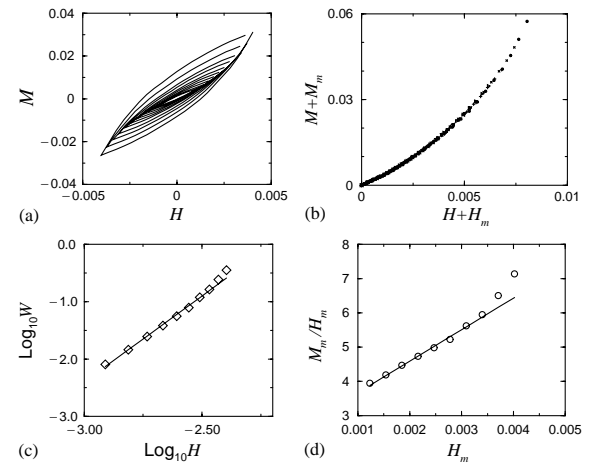


Fig. 1. The Rayleigh law computed in flexible domain wall in a random medium. (a) Hysteresis loop for different values of H_m . (b) The lower branch of the loops can be rescaled according to the Rayleigh law. (c) The area of the loop scales as $W \sim H_m^3$ (the line has a slope of 3). (d) The Rayleigh parameter can be also obtained by a linear fit of M_m/H_m vs. H_m . All units are arbitrary.

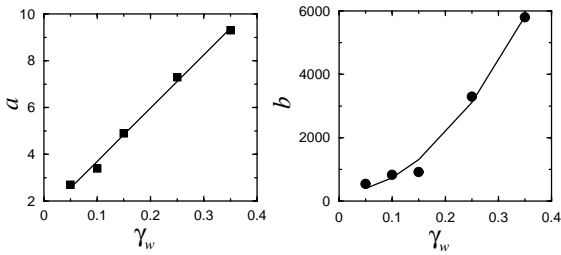


Fig. 2. The Rayleigh parameter a and b are computed for different values of the domain wall energy γ_w . The results satisfy the relation $a \propto \gamma_w$ and $b \propto \gamma_w^2$. All units are arbitrary.

repeated for different values of the domain wall energy γ_w and in Fig. 2, we show that simulations are in agreement with the scaling theory.

4. Random-field Ising model

The RFIM is probably the simplest model showing the combined effect of disorder and exchange interaction on the shape of the hysteresis loop. The model can be solved exactly in one dimension and minor loops can also be computed [27]. Recently, we have been able to obtain exact results for the entire demagnetization process in $d = 1$, including the Rayleigh laws. Here, we present numerical results for the Rayleigh laws in $d = 2$ and 3.

In $d = 2$, we perform a *perfect demagnetization* and thus obtain unambiguously the demagnetized state for a given realization of the disorder. This is done in practice by changing the field by precisely the amount necessary to flip the first unstable spin [24]. In this way, the field is cycled between $-H_m$ and H_m and H_m is then decreased at the next cycle by precisely the amount necessary to have one avalanche less than in the previous cycle. This corresponds to decrease H_m at each cycle by an amount ΔH , with $\Delta H \rightarrow 0^+$. We thus obtain the demagnetizing curve and extract the Rayleigh parameters close to the demagnetized state for a system of linear size $L = 50$ (see Fig. 3) for different values of R . During demagnetization, in higher order loops we observe that the same set of spins flips at precisely the same field, despite the fact that the demagnetization proceeds and the initial state is different.

To perform a perfect demagnetization, however, it is not possible to go to very large system sizes. We have thus resorted to a different algorithm which performs an approximate demagnetization: instead of cycling the field between $-H_m$ and H_m we just flip the field between these two values. The no-passing rule [14] ensures that the system will go into the same states that would be obtained decreasing and increasing adiabatically H between $-H_m$ and H_m . The only difference is that, after

each cycle, we have to decrease H_m by an arbitrary value ΔH (i.e. we cannot perform the limit $\Delta H \rightarrow 0^+$ exactly). We have confirmed by numerical simulations that the results obtained with an approximate demagnetization with $\Delta H < 10^{-3}$ are in good agreement with the ones obtained under a *perfect demagnetization*.

An approximate demagnetization is then used in $d = 3$, in order to analyze the effect of the phase transition on demagnetization and the Rayleigh law (the transition is not present in $d = 1$, while in $d = 2$ the issue is controversial). We find that demagnetization is possible only for $R > R_c$, where $R_c = 2.16$ for $J = 1$ [24]. For $R < R_c$ the demagnetization curve coincide with the saturation loop and it is thus not possible to define the Rayleigh parameters. We thus measure a and b for different values of $R > R_c$ and linear system sizes ranging from $L = 25$ to 100. The results show that a and b vanish for $R \rightarrow R_c$ and follow a scaling law $a \sim (R - R_c)^{\beta_a}$ and $b \sim (R - R_c)^{\beta_b}$, with $\beta_a \simeq \beta_b \simeq 0.5$. This result suggests that the demagnetization curve scales simply as $M = (R - R_c)^{1/2} m(H)$ for $H \rightarrow 0$. It is interesting to compare this result with the behavior expected for $H = H_c$ on the saturation loop: in that case, the magnetization scales with an exponent $\beta \simeq 0.04$, while $\beta = \frac{1}{2}$ is valid only in mean-field theory (see Fig. 4).

5. Experimental comparison and conclusions

In order to test the result obtained above, we perform a set of experiments on magnetostrictive ribbons of $\text{Fe}_{64}\text{Co}_{21}\text{B}_{15}$ amorphous alloy under moderate tensile stress [28,29]. This material is characterized by extended domain walls and the statistical properties of its Barkhausen noise have been recently shown to be well described by models of domain wall pinning [22,29]. We thus expect that its hysteretic behavior could also be described by collective pinning theory.

The sample is first demagnetized and hysteresis loops are measured at low fields. We find that the Rayleigh law is not perfectly verified at very low fields, but the hysteresis cycle becomes parabolic at higher fields. At very low fields, the magnetization is completely reversible, then for higher fields a definite and *reproducible* magnetization jump appears. Finally, at still higher fields several jumps combined together give rise to what can be well approximated by a parabolic cycle. This behavior is summarized in Fig. 5a where we report the behavior of M_m/H_m as a function of H_m . The linear behavior at high field is described the Rayleigh law and the parameters a and b are consistent with estimates obtained from the loop shape.

In order to understand this behavior, we have simulated the domain wall model for a *single realization* of the disorder using a grid of linear size $L = 100$. Also in this case, we observe at low fields a reversible response

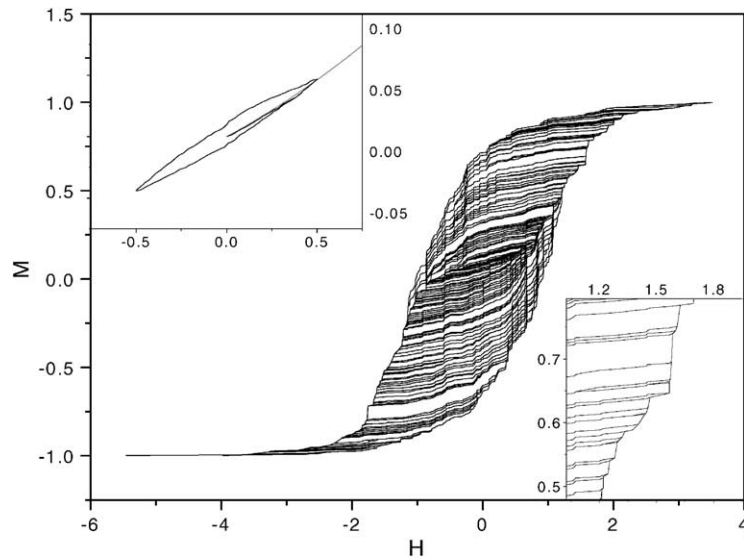


Fig. 3. An example of exact demagnetization process of the RFIM in $d = 2$ for $R = 1.64$. In the upper inset, we show the demagnetization curve and a small loop around the demagnetized state, both averaged over 30 realizations of the disorder. In the lower inset, we show a detail of the main figure. All units are arbitrary.

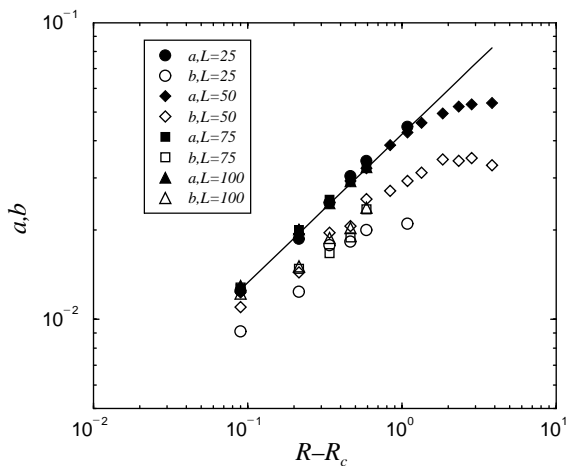


Fig. 4. The Rayleigh parameters a and b are computed for different values of the disorder R in the RFIM in $d = 3$, the results are averaged over N realizations of the disorder: $N = 100$ for $L = 25$; $N = 50$ for $L = 50$; $N = 20$ for $L = 75$ and $N = 5$ for $L = 100$. The Rayleigh law is only found for $R > R_c$ and the parameters scale to zero as power law when $R \rightarrow R_c$. All units are arbitrary.

and a jump at higher fields. After the jump the hysteresis loop is well described by the Rayleigh law (see Fig. 5b). Notice that when we average the cycles over different disorder configurations, the jump disappears. Clearly, disorder averaging cannot be performed directly for experimental data, unless different samples are used.

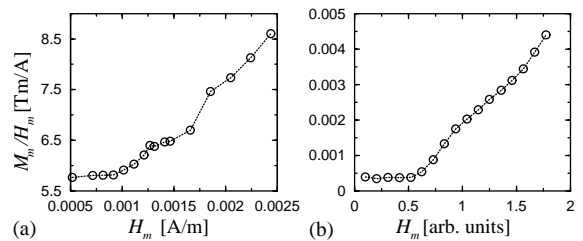


Fig. 5. The peak susceptibility M_m/H_m as a function of H_m : (a) for a single realization of the disorder in the domain wall model and (b) for a $\text{Fe}_{64}\text{Co}_{21}\text{B}_{15}$ amorphous ribbon.

One should thus keep this in mind when Rayleigh parameters are estimated.

In conclusions, we have discussed the occurrence of the Rayleigh law of hysteresis, analyzing two classes of models: disordered spin models and domain wall depinning models. This is a step towards the ambitious goal to recover hysteresis loops properties from the magnetic microstructure.

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