Loss Separation for Dynamic Hysteresis in Ferromagnetic Thin Films

Francesca Colaiori,[1] Gianfranco Durin,[2] and Stefano Zapperi[1]

[1]*CNR-INFM, SMC, Dipartimento di Fisica, Università "La Sapienza", P.le A. Moro 2, 00185 Roma, Italy*

[2]*Istituto Nazionale di Ricerca Metrologica, strada delle Cacce 91, 10135 Torino, Italy*

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We develop a theory for dynamic hysteresis in ferromagnetic thin films, on the basis of the phenomenological principle of loss separation. We observe that, remarkably, the theory of loss separation, originally derived for bulk metallic materials, is applicable to disordered magnetic systems under fairly general conditions regardless of the particular damping mechanism. We confirm our theory both by numerical simulations of a driven random-field Ising model, and by reexamining several experimental data reported in the literature on dynamic hysteresis in thin films. All the experiments examined and the simulations find a natural interpretation in terms of loss separation. The power losses' dependence on the driving field rate predicted by our theory fits satisfactorily all the data in the entire frequency range, thus reconciling the apparent lack of universality observed in different materials.

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Power losses in ferromagnetic materials generally depend on the frequency of the applied field, a phenomenon referred to as dynamic hysteresis [1,2]. The problem has important implications for applications to high frequency devices, and, from a purely theoretical point of view, for the understanding of the dynamics of disordered magnetic systems, which represents a central issue in nonequilibrium statistical mechanics. While dynamic hysteresis in metallic bulk three-dimensional systems is well understood in terms of eddy current dissipation, a satisfying theory for thin films, where the effect of eddy current is expected to become negligible [3], is still lacking. Hence, in recent years a great attention, both experimental [2,4–17] and theoretical [1,18–20], has been devoted to magnetic reversal in thin and ultrathin ferromagnetic films.

An accurate interpretation of the experimental data requires a detailed understanding of the magnetization reversal properties on a microscopic scale. Based on the analogy with Ising type models, experimental data are often analyzed in terms of universal scaling laws, such as the one relating the dynamical hysteresis loop area (A) to external parameters such temperature (T), amplitude (H_0), and frequency (ω) of the applied magnetic field: $A \propto H_0^{\alpha}\omega^{\beta}T^{-\gamma}$, where α, β, and γ are scaling exponents [1]. The experimental estimates of these exponents, often based on a very limited scaling regime, display a huge variability [4–12], so that the validity of a simple universal scaling law is still under question. Some authors also interpret the lack of good scaling of $A(\omega)$ as a crossover between two distinct dynamical regimes, dominated, at low frequencies by domain wall (DW) propagation, and at high frequencies by the nucleation of new domains [2,10]. On the other hand, several authors recognized the presence of a static hysteretic term, yielding a nonvanishing loop area in the limit $\omega \to 0$ [16–18]. While this surely represents an important step forward for a better description of the data, we still need a more comprehensive theory to understand the magnetization reversal in thin films.

For bulk ferromagnetic materials, the conventional theory of power losses takes as a starting point the phenomenological principle of *loss separation* according to which the average power loss can be decomposed into the sum of static (hysteretic), classical, and excess contributions: $P = P_{\mathrm{hyst}} + P_{\mathrm{cl}} + P_{\mathrm{ex}}$ [3]. The hysteretic contribution P_{hyst} is due to the presence of quenched disorder, and gives rise to a nonvanishing area of the quasistatic hysteresis loops. The classical term P_{cl} is obtained computing the eddy currents present in an equivalent homogeneously magnetized sample [21]. Finally, excess losses P_{ex} take into account the dissipation associated to the presence of multiple moving DWs [22]. The theory of loss separation is usually considered inadequate for thin films, since eddy currents dissipation disappears as the sample thickness goes to zero. While this is certainly correct for what concerns the contribution from classical losses, the hysteretic and excess loss contributions have been derived under very general conditions which are in principle valid also for thin films [3].

In this Letter, we show that once the excess loss contribution is included, the theory of loss separation describes very accurately the properties of dynamic hysteresis in thin films. Instead of directly considering power losses, we focus on the behavior of the coercive field H_c as a function of the driving field rate $\dot{H} = dH/dt$, since these are the quantities used in published experimental data. Analyzing the behavior of the coercive field is particularly convenient also because it is less sensitive than the loop area to the particular driving conditions employed in the experiments (i.e., sinusoidal or triangular field, applied field or flux rate, etc.). Moreover, for fully saturated loops, the two quantities are proportional.

We can estimate the coercive field in analogy with the total losses, considering the average contributions obtained by dividing each loss term by the average magnetization rate, obtaining

$$H_c = H_p + H_{\mathrm{cl}} + H_{\mathrm{ex}}, \tag{1}$$

where H_p is the static (hysteretic) contribution of the pinning field, $H_{\rm cl}$ is the classical eddy current contribution, and $H_{\rm ex}$ is the excess field.

The classical term $H_{\rm cl} = C_{\rm cl}\dot{H}$ is linear in the driving field rate $\dot{H}$ through a coefficient $C_{\rm cl} \propto d^2\sigma\mu$ [3], where σ is the conductivity, μ is the permeability, and d is the sample thickness. Since this term goes rapidly to zero with d, it can be safely disregarded in thin films, and in the following we set $C_{\rm cl} = 0$.

The excess field has no explicit dependence on the thickness, thus in principle it should be taken into account also in thin films. Originally, it has been derived under two basic hypothesis: (i) eddy currents provide the dominant dissipation mechanism of magnetization reversal, and (ii) a number of correlated regions between DWs are progressively activated by the increase of the magnetization rate $\dot{I}$. As a matter of fact, these hypotheses can be made more general. Let us consider a single DW reversing all the sample magnetization. We can assume that the total excess field H_{dw} is proportional to the magnetization rate $\dot{I}$, as $H_{dw} = \Gamma\dot{I}$, where Γ is an effective damping coefficient. This coefficient was originally calculated for the eddy current dissipation, but can be generalized to *any* general dissipation mechanism (spin relaxation, etc.) which produces the excess field H_{dw}. Because in thin films the control parameter is $\dot{H}$, we also assume $\dot{I} = \mu\dot{H}$, where μ is the permeability. Given that, the factor $\Gamma\mu$ turns out to be an effective relaxation time relating the applied field rate to the excess field for a single DW, so that $H_{dw} = \Gamma\mu\dot{H}$.

In the case of n DWs, the excess field $H_{\rm ex}$ is proportionally reduced so that $H_{\rm ex} = H_{dw}/n$, as now each DW contributes on average to the total magnetization by I/n, and thus $H_{\rm ex} = \Gamma\dot{I}/n$. At the same time, the number of active DWs itself is a function of the excess field, and, as in the original model [22], we assume a simple linear relation $n \simeq n_0 + H_{\rm exc}/V_0$, where n_0 is the number of active DWs in the quasistatic limit, and V_0 is a characteristic field which controls the increase of the number of active DWs due to the excess field.

With these hypothesis, the excess field is calculated as

$$H_{\rm ex} = C_{\rm ex}[(1 + r\dot{H})^{1/2} - 1], \tag{2}$$

where $C_{\rm ex} = n_0V_0/2$, and $r = 4\Gamma\mu/(n_0^2V_0)$. The excess field thus only depends on the typical relaxation time $\Gamma\mu$, the number of DWs in the quasistatic limit n_0, and the characteristic field V_0. Remarkably, as reported in [3], when $r\dot{H} \ll 1$, we get $H_{\rm ex} = \Gamma\mu\dot{H}/n_0$, that is, for a large number of DWs ($n \sim n_0 \gg 1$) the excess field is a linear function of the applied field rate. On the other hand, for $r\dot{H} \gg 1$, $H_{\rm ex} = \sqrt{\Gamma\mu V_0\dot{H}}$, i.e., the excess field has a square-root dependence on the rate when the number of DWs is pretty small ($n_0 \sim 1$).

At this point, one can argue about the equivalence of performing dynamic experiments by controlling the driving field $\dot{H}$ instead of the magnetization rate $\dot{I}$. Clearly, these quantities are equivalent only when we consider quasirectangular hysteresis loops (constant μ), corresponding to a magnetization process which does not change along the loop, and holds for all the frequencies of the measurement [22]. While for bulk materials this is often a rough approximation, most magnetic thin films show rectangular hysteresis loops, especially when recorded with magneto-optical techniques, as frequently reported in the literature. Strong deviations occur for complex microstructures and/or for multilayers. In this Letter, we have selected only those films having quasirectangular loops, leaving to further studies the analysis of losses for more complicated loop shapes.

In order to test the generality of the loss separation mechanisms for hysteresis we consider the driven random-field Ising model (RFIM), which has been proposed in the past as a paradigmatic model to understand the effect of disorder in ferromagnetic hysteresis [23–25]. In the RFIM, a spin $s_i = \pm 1$ is assigned to each site of a two-dimensional square 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 is associated a random field h_i taken from a Gaussian probability density $\rho(h) = \exp(-h^2/2R^2)/\sqrt{2\pi}R$, with width R, which measures the strength of the disorder. The Hamiltonian thus reads

$$\mathcal{H} = -\sum_{\langle i,j\rangle} Js_is_j - \sum_i (H + h_i)s_i, \tag{3}$$

where $\sum_{\langle i,j\rangle}$ is restricted to nearest-neighbors pairs. We consider a simple relaxation dynamics obtained in the limit $T \to 0$ of the Glauber dynamics [23–25]: at each time step

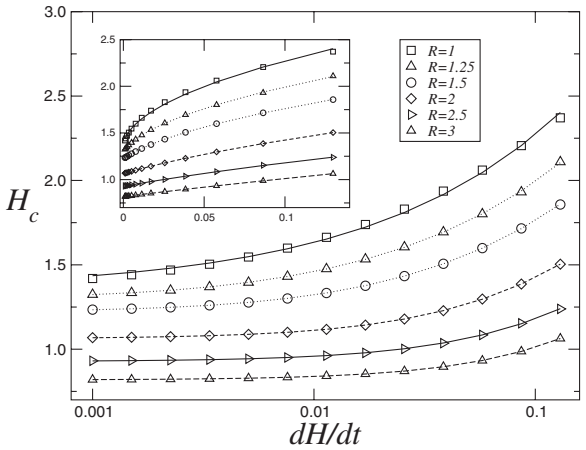

FIG. 1. The dynamic coercive field as a function of the applied field rate from simulations of the driven RFIM with different values of the variance R of the disorder distribution. The curves are fitted according to Eq. (2). In the inset: data in lin-lin scale. Note the linear behavior at large R, and the square-root dependence at low R.

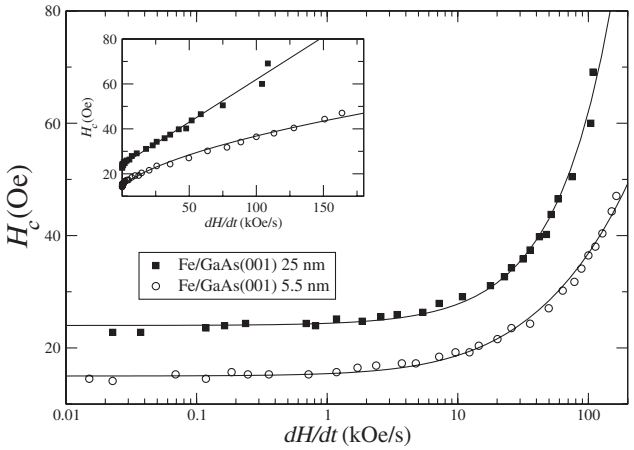

FIG. 2. Analysis of experimental data using Eqs. (1) and (2) on epitaxial Fe/GaAs(001) thin films, as reported in Ref. [10]. Data for the 25 nm film (■) are approximately described by a linear function, as better seen in the inset.

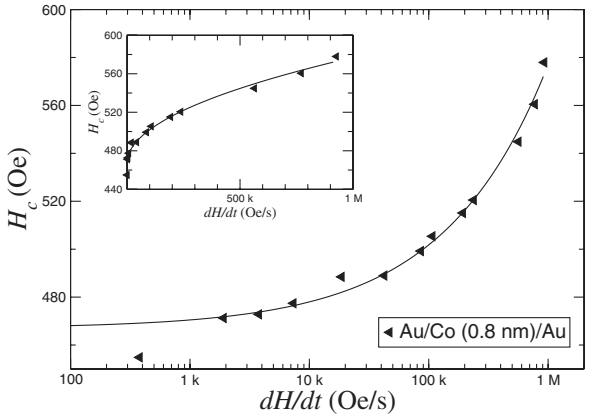

FIG. 3. Analysis of experimental data using Eqs. (1) and (2) on a MoS_2/Au/Co/Au sandwich taken at 300 K, as reported in Ref. [7]. The excess field has approximately a square-root dependence on the applied field rate.

the spins align with the local effective field $s_i = \mathrm{sgn}(J\sum_j s_j + h_i + H)$.

This model was originally studied in the quasistatic limit, however, dynamic effects can easily be incorporated assigning a time scale τ to spin relaxation and increasing the magnetic field in steps Δ, allowing the avalanches to overlap [26]. This is equivalent to an effective field rate $\dot{H} \equiv \Delta/\tau$. We numerically compute hysteresis loops for different values of the disorder strength R and obtain the coercive field averaging over different realizations of the disorder. In Fig. 1, we show that the data are described well by Eqs. (1) and (2). The resulting fit indicates that C_{ex} grows with the disorder, while r decreases. This fact can be understood by noting that the growth of disorder naturally leads to an increase in the number of the domains. In fact, when $R \sim 1$, n_0 is pretty small so that $r\dot{H} \gg 1$ and the excess field has a square-root dependence on the rate $\dot{H}$. At $R \sim 3$, n_0 is large ($r\dot{H} \lesssim 1$) and the dependence is linear (see the inset of Fig. 1).

It is interesting to remark that in Ref. [27] similar results for the loop area in the RFIM have been fitted by a scaling law $A = A_0 + A_1\omega^{\beta}$, obtaining a disorder dependent β. While it is difficult to discriminate between different fitting functions, we notice that an exponent depending on microscopic parameters is difficult to justify theoretically. In this respect, Eq. (1) interpolates between $\beta = 1$ for $r\dot{H} \ll 1$ and $\beta = 1/2$ for $r\dot{H} \gg 1$, as said. We also note that $\beta = 1/2$ was obtained from the solution of a model for the dynamics of a single DW [16,28], fully compatible with the present results.

As Eqs. (1) and (2) provide a good description of dynamic hysteresis under fairly general conditions, we can try to compare its predictions with some of the experimental measurements reported in the literature. In particular, we reexamine the measurements of dynamic coercivity in magnetic thin films and multilayers of various thicknesses, as reported in Figs. 2–5 (from Refs. [7,10,12,14], respectively). Significantly, all the data are well fitted by Eqs. (1) and (2) over the entire range of the applied field rate. Both limiting cases are present ($r\dot{H} \ll 1$, and $r\dot{H} \gg 1$), with an approximately linear (black squares) and a square-root (black triangles) dependence on $\dot{H}$. Despite the large variability of compositions, structures, and static coercive fields (which span 2 orders of magnitude), the fitting curves are compatible with a limited range of the model parameters: $n_0 = 1$–100, $V_0 = 0.1$–10 Oe, and $\Gamma\mu = 10^{-2}$–10^{-4} s. In particular, the time constant lays within the expected range for the dynamics of DWs in thin films [2]. This observation strongly supports the validity and the large applicability of this model. This also seriously puts in doubt the existence of a dynamic transition between a regime dominated by propagation and one dominated by nucleation of new domains. As matter of fact, the model

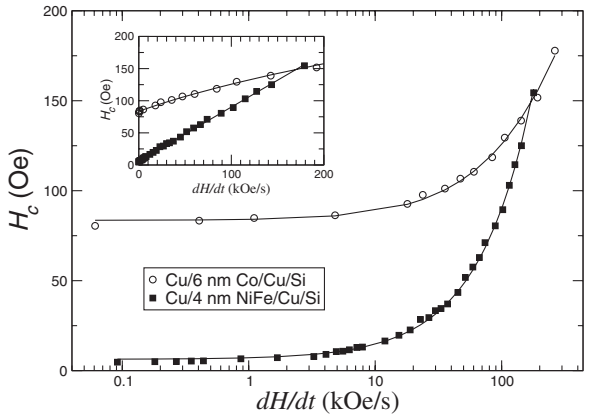

FIG. 4. Analysis of experimental data using Eqs. (1) and (2) on NiFe and Co single magnetic layer films, as reported in Ref. [12]. Data for the NiFe (■) are approximately described by a linear function, as clearly seen in the inset.

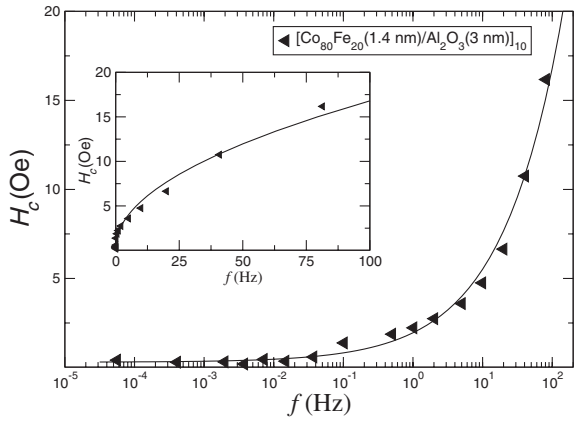

FIG. 5. Analysis of experimental data using Eqs. (1) and (2) on a superparamagnetic multilayer $[Co_{80}Fe_{20}(1.4\ nm)/Al_2O_3(3\ nm)]_{10}$, as reported in Ref. [14]. The excess field has approximately a square-root dependence on the applied field rate.

does not exclude the nucleation of new domains, as an increasing value of n can imply both the activation of existing DWs and the creations of new ones. Nevertheless, DW activation and propagation are inextricably linked in the evolution of the system.

In summary we have shown that the properties of dynamic hysteresis in ferromagnetic thin films can be explained in terms of the theory of loss separation, originally derived for bulk metallic materials. This conclusion is supported by numerical simulations and by the analysis of experimental data reported in the literature. We have selected those films which have been measured up to saturation and having quasirectangular hysteresis loops in the whole range of frequencies. In all the cases the model fits with high accuracy the measured data. It will be interesting to analyze films or multilayers whose hysteretic properties are much more complex, in order to evaluate the general validity of this approach to describe the magnetization dynamic in low-dimensional structures.

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