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Slow magnetization relaxation and reversal in magnetic thin films

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Abstract

The slow magnetization dynamics in magnetic thin films has been investigated in this paper. It is shown that the experimental results of the time-dependent magnetization can be well described by the extended exponential function, $\exp(-(t/\tau)^{\beta})$ with $\beta > 0$, in a number of thin film systems. By investigating the characteristics of the magnetization process and examining the limitations of the present models, an explanation is provided based on the structural and dynamical properties of the magnetic domains. Meanwhile, some questions have been clarified in the study towards understanding the magnetization relaxation phenomenon in thin films.

1. Introduction

Magnetization processes in thin films are of great importance in information storage technologies such as magnetic recording and magneto-optical recording. Among all the magnetization processes, the magnetization reversal and relaxation in continuous thin film structures are of prime interest. A full understanding of the magnetization dynamics is crucial for applications in terms of media writeability and data retention. In addition, the investigation will help understand the underlying mechanisms. One may find reference [1] a particularly interesting and relevant reference on the subject of this paper.

Magnetization relaxation has been observed and investigated for several decades [2]. Research effort on magnetization relaxation in thin films has become intensive since 1990 when advanced experimental techniques and methods became available and capable for studying magnetic domain patterns and its time dependence [1] and new magnetic thin film materials were synthesized and fabricated. Through the numerous experimental studies that have been carried out, several issues remain in the interpretation of the results. First, there are several expressions to characterize the time dependence of the magnetization relaxation such as the Richter-type logarithmic relaxation and the stretched exponential relaxation. However, magnetization relaxations are characterized differently in similar types of materials, which do not have apparently different relaxation mechanisms. Second, the time dependence that fits well to a stretched exponential relaxation is occasionally analyzed and discussed by a normal exponential Debye relaxation. In this case, the energetic landscape of the magnetic system is oversimplified. Third, the characterization of the time dependence of magnetization relaxation and reversal in magnetic thin films has been merely qualitative based on the Fatuzzo model [1, 24]. In this paper, we start with a review and discussion of the existing expressions that have been widely used for describing magnetization relaxation. The experimental results of the time dependence of the magnetization reversal and relaxation in thin films are then revisited and reanalyzed. This work not only clarifies some key points in the understanding of magnetization relaxation in thin film structures, but also renders some interesting and useful results and sheds light on our future study that is related to the decay of magnetic recording media and other topics.

2. Review and discussion

Magnetic relaxation is a rather complicated phenomenon. Many models have been proposed based on phenomenological studies and first principles. We shall start with probably the best-known logarithmic relaxation in the magnetics research literature. The classification of slow magnetic relaxations will follow, which is mainly based on the time dependence of magnetization. A brief summary is provided at the end of this section.

2.1. Logarithmic relaxation

Consider a single magnetic Stoner–Wohlfarth particle. An external magnetic field opposite to the initial magnetization direction would cause the particle to overcome the energy barrier and reverse its field direction for the minimum energy state. The time dependence of the magnetization is written as

$$M(t) = M_0\,(2B(t) - 1) \tag{1}$$

and $B(t) = \exp(-t/\tau)$ describes the simple Debye relaxation that involves just a single energy barrier. At finite temperatures, the time constant τ for the magnetization reversal follows the Arrhenius–Néel law [3]:

$$\tau = \nu_0^{-1} \exp(\Delta E/k_{\mathrm{B}}T), \tag{2}$$

where the attempt frequency ν_0 in the range of 10^9–10^{11} Hz is related to the spin–lattice interaction. ΔE is the energy barrier that is a function of the applied field and $k_{\mathrm{B}}T$ is the thermal activation energy.

In an assembly of non-interacting magnetic particles, the time-dependent magnetization is determined by the statistical reversals of individual particles towards thermal equilibrium. Therefore, the magnetic relaxation by the system from measurement is

$$B(t) = \int_0^\infty \mathrm{e}^{-t/\tau(y)} g(y)\,\mathrm{d}y. \tag{3}$$

Here y denotes the energy barrier that determines the time constant of reversal, $\tau(y)$. $g(y)$ is the distribution function of energy barriers in the magnetic particle system. Under some strict and particular assumptions [4][1], the magnetization relaxation is obtained to be

$$M(t) \cong M_0 - S(H,T)\ln(t/t_0), \tag{4}$$

where $S(H,T)$ is known as the magnetic viscosity coefficient and is used to characterize the magnetic relaxation in the system. For a perpendicular recording media in the presence of magnetic fields [5], the magnetic decay can be written as

$$M(t) \cong M_0 - S(H,T)\ln(1 + t/t_0). \tag{5}$$

Then, both $S(H,T)$ and t_0 are functions of the applied field and temperature.

Equation (4) or equation (5) have long been used to characterize magnetic relaxation in granular magnetic recording thin film media and ferromagnetic nanoparticle assemblies [6]. However, one can quickly notice that equation (4) diverges at both ends, $t \to 0$ and $t \to \infty$. In fact, equation (4) is argued [4] (see footnote 1) to be valid in a limited range $\tau_1 \ll t \ll \tau_2$. For $t \ll \tau_1$, the magnetization decay $\Delta M(t)$ is a linear function of time. For $t \gg \tau_2$, $\Delta M(t) \propto t^{-1}\mathrm{e}^{-t/\tau_2}$. The two time parameters, τ_1 and τ_2, are associated with the distribution function of the system. Because the assumed distribution is not realistic, the physical meaning of the time parameters remains unclear.

[1] In this book, the change of magnetization over time is described as $\int_0^\infty \mathrm{e}^{-t/\tau} g(\tau)/\tau\,\mathrm{d}\tau$, where the distribution function $g(\tau)$ is assumed constant in a time period ($\tau_1 \leqslant \tau \leqslant \tau_2$) and zero outside.

2.2. Kohlrausch–Williams–Watts relaxation

Chantrell *et al* [7] reconsidered magnetic relaxation in particulate and thin film materials based on the reversal mechanism and the formulation of equation (3). Energy barriers were assumed to have a lognormal distribution for the non-interacting particles and grains. It can be seen that magnetization relaxation deviates remarkably from logarithmic behavior when the energy barrier distribution becomes narrow. Furthermore, we find that, for all the distribution widths shown in [7], the time-dependent magnetization can be well fitted to the stretched exponential function

$$B(t) = \exp\left(-(t/\tau)^{\beta}\right), \qquad 0 < \beta < 1, \tag{6}$$

in a long time span. The relaxation time constant τ is sensitive to the energy barrier while the exponent β changes little.

The relaxation described by equation (6) is also termed Kohlrausch–Williams–Watts (KWW) relaxation. It has been broadly observed in a variety of complex materials and systems such as supercooled liquids, spin glasses, amorphous solids, molecular systems, glassy soft matter, etc [8–12]. Although it is a universal phenomenon, understanding the relaxation dynamics has relied on the distinct characteristics and properties of the systems. Nevertheless, the stretched exponential time dependence is believed to appear in any strongly interacting disordered system. The KWW relaxation ($0 < \beta < 1$) is slower than the conventional Debye relaxation ($\beta = 1$) since the system goes through metastable states during the process towards the ultimate thermal equilibrium.

The stretched exponential relaxation has been observed and reported in a number of spin-glass and cluster spin-glass systems such as Au:Mn [13], γ-FeNiCr alloys [14], polycrystalline $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ [15] and amorphous Fe_2O_3 [16] by measuring the thermoremanent magnetization. Simulations have been carried out by Ogielski [9] to understand the relaxation dynamics in Ising spin glasses, followed by some phenomenological models [17–19]. The slow relaxation dynamics in spin glasses is much different from the relaxation dynamics in magnetic thin films. We shall return to the magnetic thin films that are the focus of this study. The treatment above of thin films as non-interacting grains is fairly crude and unrealistic. By including the dipole–dipole interaction in thin films with strong perpendicular anisotropy which resembles a 2D Ising spin system, Lottis *et al* [20] simulated the magnetization dynamics under a mean-field approximation and concluded that the relaxation can be better described by a stretched exponential rather than a logarithmic function. However, the magnetization structure of the thin film and its temporal evolution were not provided. We will come back to this point later.

2.3. Kolmogorov–Avrami–Fatuzzo relaxation

There exists a fundamental difference between the aforementioned models for spin glasses and the magnetic thin film systems that are the main subject of this study. In these models, some clusters, droplets or domains are presumed formed at the very beginning of the relaxation process. The magnetization

relaxation is the total effect of relaxing clusters or fluctuating droplets, etc [17–19], as the system approaches thermal equilibrium. In the thin films we consider, the magnetization is initially saturated in an easy axis direction. Then an external magnetic field is applied in the opposite direction. As a consequence, the magnetization relaxes and reverses field direction. For imperfect films with defects such as voids and pinning sites, the Stoner–Wohlfarth-like coherent reversal is not likely since the process is not favored energetically. The real picture is that first some reversed spins or grains appear in the films. Via the short-ranged exchange coupling, the adjacent spins or grains are more likely to reverse than the others. Therefore, magnetic domains are formed and grow over time from the reversed spins or grains, which are considered as nuclei. The magnetization reversal is achieved when the domains expand to cover the whole film. This so-called Kolmogorov–Avrami (KA) model was originally used to describe crystal growth [21, 22]. It has also been successfully applied to ferroelectric domain reversals [23].

Under the framework of the KA model, the time dependence of magnetization reversal is expressed as

$$B(t) = \exp\left(-\int_0^t n(\tau)s(t-\tau)\,\mathrm{d}\tau\right). \tag{7}$$

$n(\tau)$ is the nucleation rate at time τ. $s(t-\tau)$ is the domain expanding from a nucleus, which appears at time τ, for a given time t. The change of magnetization at time t is then the sum of the growth of the reversed domains from the nucleation that occurs from the beginning. We let the domain growth be a power-law function

$$s(t-\tau) = c(t-\tau)^{\gamma}, \tag{8}$$

where c is the domain growth speed and $\gamma > 0$, and consider two limiting cases. Supposing nucleation occurs at $\tau = 0$ immediately after the reverse field is applied, i.e. $n(\tau) = \delta(\tau)$:

$$B(t) = \exp(-ct^{\gamma}). \tag{9}$$

When nuclei appear at a constant rate, i.e. $n(\tau) = n_0$, the magnetization reversal is

$$B(t) = \exp\left(-\frac{cn_0}{\gamma+1}t^{\gamma+1}\right). \tag{10}$$

Both equations look similar to that of the KWW relaxation but, in the second case, the exponent, $\gamma + 1$, is known for sure to be greater than one.

Fatuzzo [24] has elaborated the thin film relaxation associated with nucleation and domain growth based on the KA model with the assumptions that the nucleation rate decreases exponentially in time:

$$n(t) = \exp(-Rt), \tag{11}$$

and circular domains grow with a speed v:

$$s(t-\tau) = \pi v^2(t-\tau)^2. \tag{12}$$

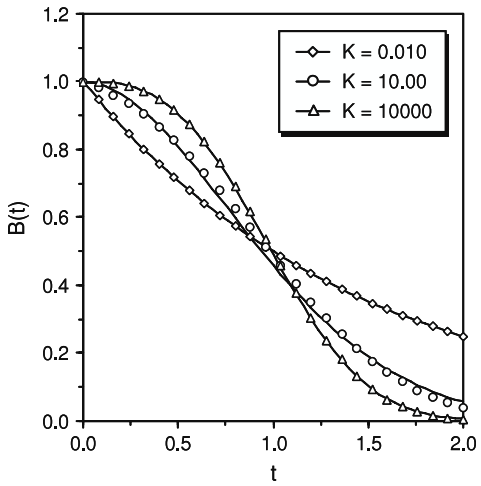

Figure 1. Equation (13) represented by the open dots fitted by compressed exponential functions for some typical cases, $k = 0.01$, 10 and 10 000. Time t is normalized to the characteristic time constant $t_{1/2}$, where $B(t)$ reduces to 1/2.

Thus, the relaxation (reversal) is obtained[2] as

$$B(t') = \exp\big(-k^2(2 - 2(t' + k^{-1}) + (t' + k^{-1})^2 - 2e^{-t'}(1 - k^{-1})) + (1 - t')\big), \tag{13}$$

where $t' = Rt$ and the parameter k is

$$k = v/Rr_c. \tag{14}$$

Here r_c is the radius of the nuclei.

The Fatuzzo model was originally intended to interpret the relaxation in ferroelectrics. It was borrowed from magnetic thin films [25] and has since been widely used [1]. However, the study based on the model has been qualitative partly because of the complexity of equation (13). We notice that the magnetization reversal as a function of t' is solely determined by the parameter k. Furthermore, the time dependence described by equation (13) can be approximated by

$$B(t') = \exp\left(-(t'/\tau(k))^{\beta(k)}\right),$$

or

$$B(t) = \exp\left(-(Rt/\tau(k))^{\beta(k)}\right), \tag{15}$$

in a significantly long time range. $\tau(k)$ and $\beta(k)$ are only dependent on k. Figure 1 shows excellent consistency between equations (13) and (15) for several typical k values. This transformation technique has been used by Orihara *et al* in their study of ferroelectrics [23]. It not only facilitates understanding of the experimental data, as will be seen in the next section, but also reveals some important characteristics of the relaxation.

The relaxation is characterized by a time constant, $t_{1/2}$, at which the magnetization becomes zero and $B(t) = 1/2$.

[2] Equation (7) was considered to be part of the reversal arising from domain expansion. Fatuzzo had an additional term, equation (7) in [24], to account for that of the nuclei in deriving equation (13).

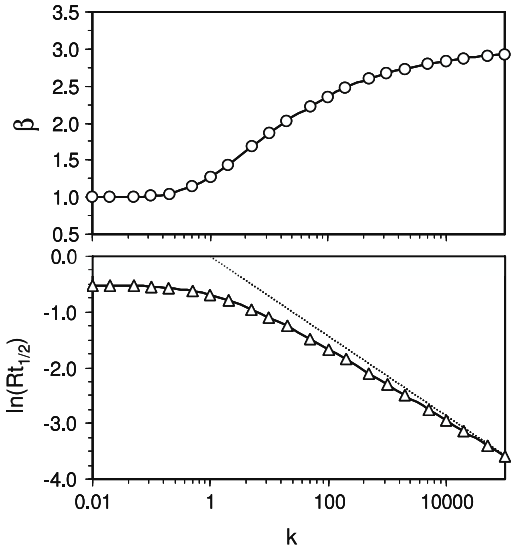

Figure 2. Exponent β and time constant $t_{1/2}$ as functions of k. The dotted line represents equation (20).

Because of the property of equation (13), $t_{1/2}$ can be written as $t_{1/2} = h(k)/R$. Specifically, from equation (15),

$$t_{1/2} = (\ln 2)^{1/\beta(k)}\tau(k)/R. \tag{16}$$

For the limiting case that the nuclei do not grow, i.e. $k \to 0$:

$$B(t) = \exp(-Rt), \tag{17}$$

and

$$t_{1/2} = \ln 2/R. \tag{18}$$

For the other limiting case when domains grow extremely fast, i.e. $k \to \infty$:

$$B(t) = \exp\left(-k^2R^3t^3/3\right), \tag{19}$$

and

$$t_{1/2} = (3\ln 2)^{1/3}/Rk^{2/3}. \tag{20}$$

Thus, $\beta(k) = 3$ and this is equation (10) with $\gamma = 2$ for the growth of circular domains, since the domains grow so fast that the change of nucleation rate over time is negligible. Figure 2 shows the exponent, β, and the time constant, $t_{1/2}$, as functions of k. β increases gradually from 1 to 3 when k goes from 0 to $+\infty$. $t_{1/2}$ is close to $\ln 2/R$ for low k and follows equation (20) for high k. The characteristics of nucleation and domain growth can be readily obtained by fitting experimental data to equation (15) and then converting the fitting values for β and $t_{1/2}$ to R and k from this figure.

2.4. Power-law relaxation

Another class of magnetization relaxation is with the power-law time dependence

$$M(t) = M_0t^{-\zeta}, \qquad \zeta > 0. \tag{21}$$

This has been observed in spin glasses such as the γ-FeNiCr alloys [14]. This behavior can be understood in the context of the flipping of spin clusters [26]. Recently, Monte Carlo simulations were carried out to study the magnetic relaxation in single-domain ferromagnetic nanoparticles and showed that for all the particle densities the magnetization approaches a finite remanent value slowly by a power law [27]. Dispersivity and dipolar interaction of the nanoparticles are found to play significant roles in the slow relaxation.

To summarize the section, several typical kinds of magnetic relaxation behavior have been reviewed. First, one should be careful when trying to fit experimental measurements to any of the above functions that all describe a monotonic magnetization decay. In particular, the logarithmic relaxation is valid in a limited range based on the property of the function itself and the strict assumption from which it is derived. Its popularity in magnetization decay lies partially in the history, that it was the very first one used to describe the decay phenomenon in magnetics [2]. Similarly, the power law is not suitable for the study where we investigate the process of magnetization relaxation and reversal in thin films from $+M_0$ and $-M_0$. Therefore, an 'extended' exponential function:

$$B(t) = \exp\left(-(t/\tau)^{\beta}\right), \tag{22}$$

is chosen for the purpose. Note that $0 < \beta < 1$ describes the stretched exponential relaxation, i.e. the Kohlrausch–Williams–Watts relaxation and $\beta = 1$ is the conventional Debye relaxation. $1 < \beta \leqslant 3$ is the regime for the Fatuzzo model and then equation (22) is a compressed exponential function. This classification of magnetization relaxation seems too crude and might be inappropriate without knowing the mechanisms and dynamics. It will be clarified with experimental data in the next section. Another clarification is that the slow relaxation refers to timescales far longer than 10^{-9}–10^{-11} s for spin–lattice interactions and 10^{-11}–10^{-13} s for spin–spin interactions. As will be shown, the measurements we considered were done in the time range of 10^{-2}–10^{4} s. The dynamics within 10^{-2} s of the onset of the magnetization process were not detected due to the measurement capability. Associated mechanisms will not be discussed.

3. Results and analysis

3.1. Extended exponential relaxation

Rare earth–transition metal (RE–TM) alloy thin films with perpendicular anisotropy [25] were the first systems used for the study of slow magnetization reversal and relaxation. Since then slow magnetization dynamics has been investigated in many other thin films [28–41], multi-domain particles [42], and 1D magnetic nanowires [43, 44] and similar behavior has been observed. Figure 3 shows the magnetization change versus time for a GdTbFe sample in [25]. It fits excellently to a compressed exponential decay with $\beta = 1.63$, which corresponds to $k = 4.8$, referring to figure 2. It is different from the value, $k = 1$, reported in [19], where the estimate was made in the limit, $Rt \ll 1$. Before proceeding to the following analysis and discussion, it is necessary to point out that Wernsdorfer *et al* [42, 43] have applied the extended exponential function to characterize magnetization reversal in

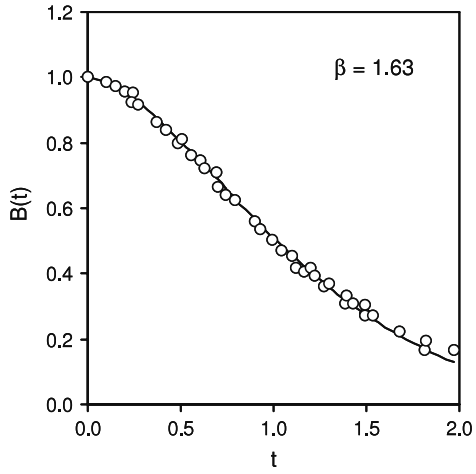

Figure 3. Experimental data taken from [25] fitted to equation (22). Time t is normalized to $t_{1/2}$. Note the data points are combined with the measurements at different magnetic fields.

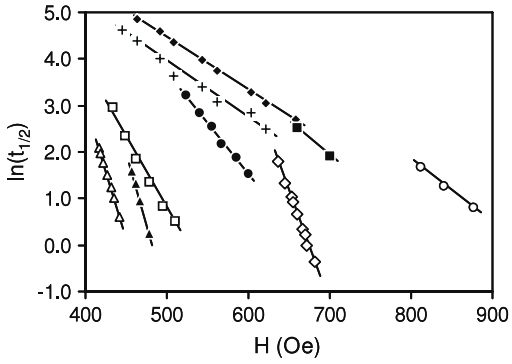

Figure 4. Time constant $t_{1/2}$ as a function of time in the presence of reverse magnetic fields. Data is taken from [25] (●) for the sample GdTbFe; [28] (□) for sample I of 5 ML Au/Co/Au and (Δ) for sample II of 5.5 ML Au/Co/Au; [30] (♦) for the 485 Å TbFeCo film and (+) for the 1550 Å TbFeCo film; [31] (■) for an Au/Co/Au sandwich film; [32] (○) for an Au/Co(3 ML)/Au sample and (▲) for an Au/Co(5 ML)/Au sample; and [36] (◊) for an Au/Co(10 Å)/Au sample.

magnetic particles and nanowires. In both kinds of magnetic structures, the exponent β can be either less than or greater than unity. In particular, the magnetization switching in the Co particles was concluded to be associated with the domain wall nucleation and annihilation processes in the particles [42]. In this paper, we will limit our discussion to ferromagnetic thin films.

One of the important findings in [25] is the field dependence of the magnetization relaxation. It is easy to understand that the smaller the reverse magnetic field, the slower the magnetization reversal and relaxation. However, when normalized to the time constant $t_{1/2}$, the magnetization relaxation fits in a unique curve shown by figure 3 for all the reverse fields. In other words, the exponent β is independent of the reverse field strength. This phenomenon has also been reported in other thin films as well [28, 34, 41]. We know from the previous analysis that β is a monotonic function of k (see equation (15) and figure 2). k is then independent of the applied field.

Figure 4 shows some of the relaxation data collected from several magnetic thin films. An exponential relationship between the time constant $t_{1/2}$ and the applied reverse field H can be established. According to equation (16) and the field-independent k, it suggests that the nucleation rate is related to the field by

$$R = R_0 \exp\left(\alpha(H - H_N)\right), \tag{23}$$

where R_0 is the nucleation rate at a reverse field of H_N. R_0 and H_N cannot be determined independently in data fitting. In [25] where the nucleation is believed to be thermally activated, the parameter α is written as $\alpha = 2M_sV_N/k_BT$, where M_s and V_N are the magnetization and the effective activation volume of the nucleus.

According to the Fatuzzo model, we know from equation (14) that the domain growth $v = kr_cR$. The nucleation radius r_c is associated with the intrinsic defects of the films and then should be independent of the external field. When domain growth speed, similar to the nucleation rate, is written as [25]

$$v = v_0 \exp\left(\alpha'(H - H_D)\right), \tag{24}$$

where $\alpha' = 2M_sV_D/k_BT$, we come to a strong conclusion that the domain growth speed and the nucleation rate have the *same* field dependence, i.e. $\alpha' = \alpha$. It implies that the nucleation and domain growth arise from the same mechanism and it might be true that at relatively low fields the domains grow by pushing the domain wall through the inhomogeneities or defects in the films [45, 33]. Nevertheless, it looks odd since $\alpha' = \alpha$ means that the activation volume of nuclei V_N and the activation volume for domain wall motion V_D should be equal. At higher fields in the viscous regime, domains grow at a speed proportional to the field strength. The field dependence of domain growth also relies on film quality. In nearly homogeneous Ising films the speed of the domain wall motion is [46] measured to be $v \propto \exp(\varepsilon/H^\mu)$, where $\mu = 1/4$, and understood by creep theory. Mangin *et al* [41] found that μ was larger than unity, e.g. 3/2 or higher, at low temperatures in their GdFe/TdFe samples where the GdFe domain walls propagated in the film plane through the energy barriers, including those arising from the coupling with the TdFe layer.

Table 1 summarizes the relevant properties of the magnetic thin films and the slow magnetization relaxation found from the films. Film material ranges from TM metals, RE–TM alloys, to noble metal–TM alloys in structures of single layer films, bilayer films, sandwich films and multilayers. The films were prepared by a variety of deposition methods and the magnetic films can be amorphous, granular, textured or epitaxial. The magnetic layer thickness ranges from 2 Å to more than 1000 Å. Except for the last two in the table, Ag/Fe/Ag sandwiches and Co/Cu bilayer films, all the films show magnetization perpendicular to the film sustained by the perpendicular magnetic anisotropy (PMA). The values quoted in the table for coercivity for reference were obtained from hysteresis loop measurements that were usually completed in

Table 1. Exponent β and coefficient α extracted from the magnetization relaxation experiments conducted and reported on a number of magnetic thin films. The table also shows the measurement temperature T, thickness of a single magnetic layer t_{FM} and coercivity H_c for reference. RT stands for room temperature and ML for monolayer.

Material	T	t_{FM}	H_c (Oe)	b	a (Oe^{-1})	Ref.
GdTbFe	RT	∼200 A	640	1.63	0.022	[25]
TbFeCo	RT	485 A	∼8800	0.59	0.012	[30]
		1550 A	∼15600	0.26	0.011	
GdFe/TbFe/GdFe	4 K	1000 A	∼45	1.76		[41]
CoPt	RT	100 A	∼1000	2.01		[33]
		300 A	∼2250	1.09		
Au/Co/Au	RT	5.0 ML	510	0.39	0.032	[28]
		5.5 ML	450	3.36	0.057	
Au/Co/Au	RT	8.0 A	770	1.92	0.016	[31]
Au/Co/Au	RT	3 ML	∼1000	2.13	0.013	[32]
		5 ML	∼500	1.69	0.064	
		7 ML	∼400	0.16		
Au/Co/Au	RT	10.0 A	∼700	0.53	0.048	[36]
Fe/Cu	120 K	3 ML	∼30	1.04	0.356	[40]
	270 K			0.44	1.415	
$[Fe/Dy]_m$	RT	5.6 A	∼4300	2.08		[29]
		8.1 A	∼3900	0.36		
$[Co/Pd]_{10}$	RT	2.0 A	1200	1.91		[34]
		3.0 A	600	1.35		
		4.0 A	400	0.72		
$[Co/Pd]_5$	RT	2.5 A	149		0.111	[37]
$[Co/Pd]_{10}$			368		0.063	
$[Co/Pd]_{15}$			916		0.030	
$[Co/Pt]_9$	RT	3.6 A	820	1.32		[39]
Ag/Fe/Ag	RT	10 ML	3	0.35		[35]
Co/Cu	RT	30 ML	∼70	0.33		[38]

less than several seconds. We find that the magnetization reversal and relaxation in these films can be fitted to the extended exponential function, equation (22), fairly well. The exponent β varies in the range from 0.16 to 3.36. The coefficient α for equation (23) is commonly below 0.12 Oe^{-1}, except for that measured at low temperatures on the Fe/Cu films [40] with a low coercivity around 30 Oe.

It can be seen from the table that generally the exponent β decreases with increasing magnetic layer thickness in the same thin film system. The magnetization relaxation in [Co/Pd] multilayers is redrawn in figures 5(a)–(c) and β is shown by figure 5(d) to change from 1.91 to 0.72 with the Co sublayer thickness. Coercivity may be lower for films with a thicker magnetic layer, which agrees with the theoretical modeling and experimental observation [45] that the activation energy of domain wall motion decreases with increasing magnetic layer thickness. In addition, there exists a correlation between the coercivity and the coefficient α. The sensitivity of the nucleation rate and domain growth speed on the magnetic reversal field appears to be high in low coercivity films.

3.2. Analysis and discussion

In the Fatuzzo model, the magnetization reversal and relaxation is described by a compressed exponential function ($1 \leqslant \beta \leqslant 3$). In the case when the nucleation dominates in the reversal process, i.e. $k \ll 1$, the relaxation is the normal exponential function ($\beta = 1$). The fact that the observed slow relaxation can be either stretched exponential ($\beta < 1$) or compressed exponential ($\beta > 1$) behavior indicates that the model is no longer complete in describing this magnetization dynamics. We shall carefully examine the two assumptions, nucleation rate and domain structure/expansion, on which the model is based.

First, the exponential time dependence of the nucleation rate, equation (11), has not been confirmed experimentally. As mentioned in the previous section, the magnetization relaxation of dispersive non-interacting ferromagnetic nanoparticles could follow a stretched exponential. It implies that the nucleation would be similarly a non-exponential function of time as well. It is generally believed that nucleation is strongly associated with the defects in the films. Nevertheless, its properties, including time dependence, are yet to be characterized until the mechanism is well understood. The previous study [28–40] just simply related the stretched exponential decay ($\beta < 1$) to nucleation-dominated magnetization relaxation. However, an explanation is needed for understanding the thickness dependence, when the magnetization relaxation crosses over from compressed exponential behavior to stretched exponential behavior with increasing magnetic layer thickness.

Second, circular domains are assumed in the Fatuzzo model. In fact, it is not true in inhomogeneous films. Magnetic domains have been observed to grow and evolve in a dendritic structure [34, 45, 48] and can be generated from the random field Ising model [49] for perpendicularly magnetized films. Furthermore, the dependence of the domain structure on the thin film thickness is evident [34]. In the general case, the domain and domain wall/boundary are characterized differently [42, 50] with their own fractal dimensionalities. Since domain growth relies on the local characteristics and property of the film at the domain boundary, the specific expression of equation (12) is no longer valid. We should go back to the generic equation (8), where both the domain growth speed c and the index γ are variables of the magnetization dynamics.

Now that the index γ is related to the domain structure, it is completely determined by the magnetic film structural quality such as the morphology and imperfections. It is known that defects play a critical and fundamental role in domain structure. Defects are more likely to be located at magnetic film surfaces as forms of loose spins due to film roughness and reduced surface anisotropy [47]. They can also be formed as voids and pinning sites in the bulk of the films during deposition. On the other side, the domain growth speed c is related to the activation energy of domain wall motion, which is dependent upon the intrinsic magnetic property of the film, the film structural and magnetic defects, and the extrinsic factors, such as applied fields, as well. When we go back to consider the Kolmogorov–Avrami mechanism in the case where nucleation occurs at time $t = 0$ and equation (9) is applied to the slow relaxation, questions associated with the experimental observations can be clarified. First, the puzzle around the awkward relationship, $\alpha' = \alpha$, does not exist any more. The domain growth is a function of the applied

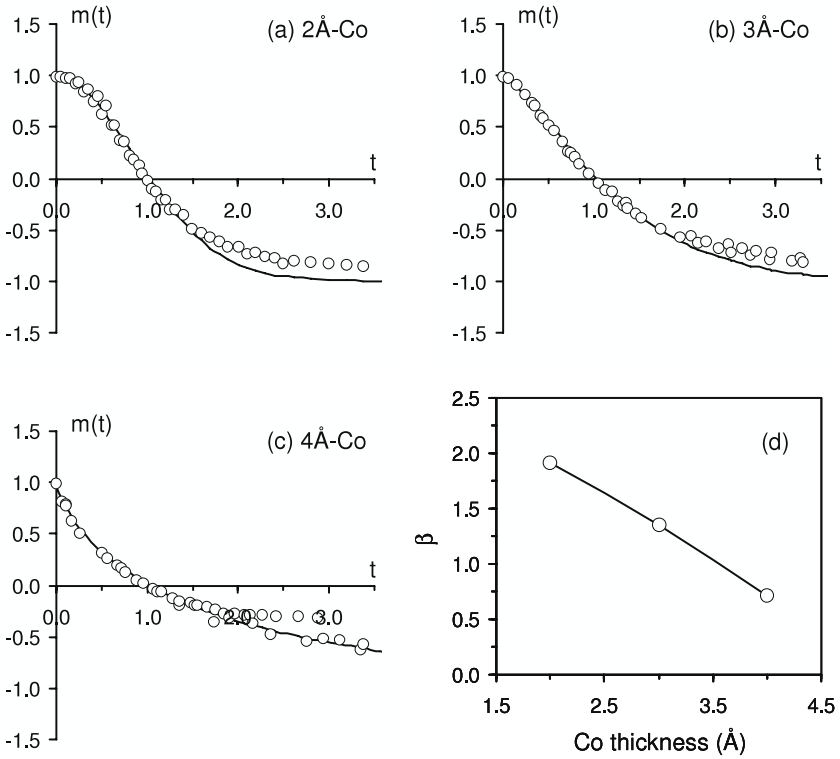

Figure 5. Time dependence of the magnetization relaxation in $[Co/Pd]_{10}$ multilayer films with a single Co layer thickness of (a) 2 Å, (b) 3 Å and (c) 4 Å. Data is taken from [34]. The exponent β extracted from the relaxation curves as a function of the Co layer thickness is shown by (d).

field by equation (24) with v replaced by c while the index γ (or β) in equation (22) is independent of the field. Second, the magnetization relaxation can be easily modified in the exponent β by an Au cap layer to smooth out the surface [32]. The thickness dependence of β observed in a number of thin film systems can be understood by the addition of defects in thicker magnetic films. Weir *et al*'s Monte Carlo simulations clearly show slower relaxation in the films with higher pinning site densities [48]. Unfortunately the effect of pinning site density broken down for c and γ is not given. Last, sharp jumps in the relaxation curve are observed in some thin films [35, 38], indicating the magnetization overcomes some major pinning sites in the reversal process.

We notice in our data fitting that the relaxation becomes slower than expected when the magnetization approaches the opposite direction. This behavior is shown in figure 5 and is more pronounced for compressed exponential decay. An explanation is given as follows: the moving front of a domain is curved or jagged during expansion. Shown in figure 6, non-reversed magnetic 'bubbles' are formed when two domains coalesce and when a domain passes around a pinning site or void [49]. The bubbles can be stabilized by the demagnetizing field generated by the surface magnetic charge against the applied field. The magnetization relaxation associated with the bubbles is similar to that described by the droplet and domain models [18, 19] for spin glasses by thermal flipping of the bubbles. Therefore, both domain growth and bubble reversal contribute to the relaxation. The magnetization relaxation can be clearly seen to cross over from compressed exponential

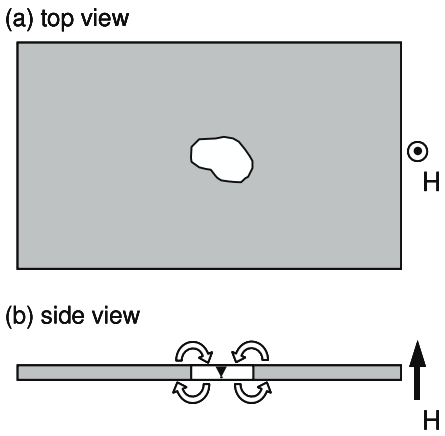

Figure 6. (a) Side view and (b) top view of a magnetic bubble formed in the background of reversed magnetization.

decay to stretched exponential decay when domains stop growing and bubble reversal becomes dominant in the late phase of the process. Technically it is not as easy to identify the bubble contribution in data analysis for the case where the relaxation associated with domain growth is also a stretched exponential.

4. Summary and conclusions

In summary, the experimentally measured time dependence of the magnetization processes in magnetic thin films that we

can find is fitted to the extended exponential function. The relaxation and reversal processes in thin films associated with the domain formation and growth described by the general Kolmogorov–Avrami model can be either the compressed exponential behavior ($\beta > 1$) or the stretched exponential behavior ($0 < \beta < 1$). The Fatuzzo model is not adequate in understanding the phenomenon. This conclusion is drawn from the study of the field dependence and the magnetic layer thickness dependence of the magnetization process.

The magnetization reversal and relaxation in thin films are directly related to the structural and dynamical properties of the magnetic domains. These processes are ultimately influenced by the structural properties such as morphology, dispersivity, defects, and by the magnetic properties including anisotropies and exchange coupling. Aside from the nucleation and domain growth mechanism, thermal flipping of the magnetic bubbles resulting from domain expansion and coalescence also contributes to magnetization relaxation and becomes significant in the late phase of the relaxation. Like the relaxation process in disordered systems, the magnetization processes associated with the bubble mechanism can be characterized by a stretched exponential function.

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