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Activation Volumes of Wall-Motion and Nucleation Processes in Co/Pd Multilayers

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The correlation between the activation volumes of wall-motion and nucleation processes in Co/Pd multilayers has been investigated. Each activation volume was estimated from the field dependence of the wall-motion speed and the nucleation rate, respectively, based on time-resolved domain patterns grabbed by a MOKE microscope system. Both the activation volumes are changed in the same manner around $0.2\sim1.1\times10^{-17}$ cm^3 with changes in the multilayered structure. Interestingly, the correlation between the activation volumes is sensitive to the multilayered structure; the wall-motion activation volume is smaller than the nucleation activation volume for a sample having a smaller number of repeats and a thinner Co-layer thickness, and *vice versa*. The correlation is closely related with the contrasting reversal modes; the process having the smaller activation volume dominates.

1. Introduction

Magnetization reversal dynamics in ferromagnetic thin films continues to be an important issue in achieving high performance in technological applications as well as in exploring fundamental magnetic behavior [1-3]. Most experimental studies have evidenced that magnetization reversal dynamics are governed by a thermally activated relaxation mechanism [4-9], which was theoretically predicted to take place via the thermal fluctuation energy k_BT overcoming the switching barrier energy E_B of the magnetization M_S of an activation volume V_A at a finite temperature T [10, 11];

$$\tau=\tau_0\exp(V_AE_B/k_BT) \quad (1)$$

where τ is the time needed to switch the magnetization direction and τ_0 is the characteristic switching time (or inverse of attempt frequency) when $E_B=0$. The activation volume is a key parameter in this phenomenon since it characterizes the basis volume acting as a single particle, and thus limits the minimum size of a stable magnetic volume for technological applications. The activation volume has been estimated from the field dependence of the reversal time, employing the fact that the switching energy barrier is linearly dependent on external magnetic field H near the coercive field H_C;

$$E_B(H)=E_B(H_C)+M_S\cdot(H_C-H) \quad (2)$$

It has been reported that the activation volume is in the range $10^{-18}\sim10^{-16}$ cm^3 for a number of ferromagnetic thin films, and is sensitively dependent on the composition, layered structure, and morphology of the films [4-9].

Recently, exploration of magnetization reversal dynamics has greatly advanced, largely motivated by direct observation of domain evolution patterns using magnetic imaging techniques [3, 12, 13], as well as by experimental observation of contrasting reversal modes in similar samples of many systems [2-5]. It has been revealed that the magnetization in ferromagnetic thin films reverses via two fundamental processes; 1) wall-motion of existing domains and 2) nucleation of new domains at random positions independent of existing domains. The wall-motion takes place either by a successive switching process of activation volumes adjacent to an existing domain via thermal activation energy overcoming finite wall-pinning energy when the applied field is smaller than the wall-pinning field, or by a viscous wall-motion process when the applied field is higher than the wall-pinning field [2, 9]. On the other hand, domain nucleation occurs by random switching of activation volumes at random places via thermal activation energy overcoming the nucleation barrier energy. Much effort has been devoted to measure the activation volumes of the wall-motion and nucleation processes in a number of systems [2, 4, 9, 14, 15]. Based on the experimental evidence that the activation volumes are not substantially different [1, 9], most theoretical studies so far have assumed that the activation volumes for wall-motion and for nucleation processes are identical [16, 17]. However, there is no clear physical reason that this should be the case and the correlation between the activation volumes still remains unclear.

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The present study was undertaken to characterize the correlation between the activation volumes for wall-motion and nucleation processes in ferromagnetic thin films. For this study, time-resolved domain reversal patterns of Co/Pd multilayers were grabbed in real time using a MOKE (Magneto-Optical Kerr Effect) microscope system [3]. The wall-motion speed and nucleation rate were quantitatively determined by analyzing the time-resolved domain reversal patterns [18]. The dependence of the wall-motion and nucleation processes on the applied field was characterized using a thermal activation relaxation mechanism. We report the correlation between the activation volumes for wall-motion and nucleation processes in connection with the magnetization reversal modes.

2. Experiments

A novel analysis model has recently been developed to quantitatively determine the wall-motion speed and nucleation rate based on time-dependent domain patterns [18]. In the model, domains expand at all domain boundaries with wall-motion speed V and simultaneously new domains are formed at a nucleation rate R per unit time and unit area during magnetization reversal from the initially saturated state. The changes in reversed domain area a and domain boundary length l in time dt are given by

$$da=lVdt+\pi r_0^2R(s-a)dt$$
$$dl=2\pi Vdt+2\pi r_0R(s-a)dt, \quad (3)$$

where r_0 is the characteristic radius of nucleation and s is the total area under examination. The first terms in the equations are ascribed to wall-motion, while the second terms are caused by the domain nucleation process. Inverting Eq. (3), the wall-motion speed V and the nucleation rate R are explicitly given by

$$V=(a'-r_0l'/2)/(l-\pi r_0)$$
$$R=(ll'/2\pi-a')/(l-\pi r_0)r_0(s-a), \quad (4)$$

where a' and l' denote the time derivatives of the reversed domain area and the domain boundary length, respectively. Using Eq. (4), one can simultaneously determine the wall-motion speed and the nucleation rate by measuring the domain area and the domain boundary length.

A MOKE microscope system has been used to measure the reversed domain area and domain boundary length during the magnetization reversal. The system is capable of grabbing domain images in real time with an advance video processing technique, and is equipped with an objective of 0.9 N.A. for 1,000× magnification [3]. The sample was first saturated by applying a magnetic field normal to the film plane and then, time-resolved domain images of 128 frames were grabbed at 10 frames per second under a reversing applied field. The image was composed of 200 × 160 pixels with the unit pixel size of 164 × 164 nm. The image was initially obtained in 256 gray levels and then intensified by noise filtering and black-and-white image extraction processes. The domain area and domain boundary length of each image were determined by counting black and white cells and by use of an edge-determining algorithm, respectively [18].

Figure 1 illustrates the typical time variation of the domain area and domain boundary length measured from the domain images as illustrated in the insets of the figure. The unreversed domain area is equivalent to the magnetization viscosity curve, which is the time variation of the magnetization under a constant reversing field [3]. The curve exhibits initially a slow decay and then, more rapid relaxation with time, which indicates the typical behavior of a low nucleation rate and fast wall-motion, since the initial decay rate is limited by the nucleation rate [19]. The domain boundary length initially increases with increasing domain area, but it decreases after domain coalescence [20].

The measuring system has been applied to characterize the activation volumes of Co/Pd multilayers. Co/Pd multilayer thin films were chosen because of their wide range of activation volumes due to the various magnetic and/or structural properties that result from varying the multilayered structure, as well as their large polar Kerr effect for clear domain imaging. A number of $(t_{Co}$-Co/11-Å Pd$)_n$ samples varying either the Co-layer thickness t_{Co} or the number of repeats n were prepared on glass substrates by alternatively exposing the substrate to e-beam sources of Co and Pd under a base pressure of 2.0×10^{-7} Torr at ambient temperature [3]. Low-angle x-ray diffraction studies using Cu K_α radiation revealed that all samples had distinct peaks indicating an existence of the multilayered structure and that the layer thickness was controlled within 4% accuracy. High-angle x-ray diffraction studies showed that the samples grew with the <111> direction normal to the film

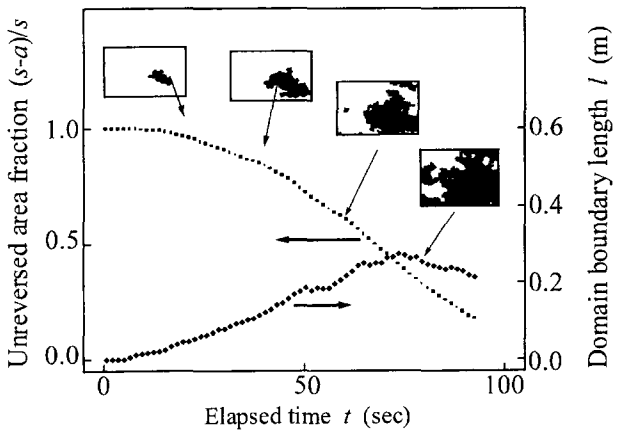

Fig. 1. The unreversed domain area and domain boundary length vs. elapsed time after a reversing field is applied. The domain area and domain boundary length of each image as shown by the insets were determined by counting black and white cells and from an edge determining algorithm, respectively.

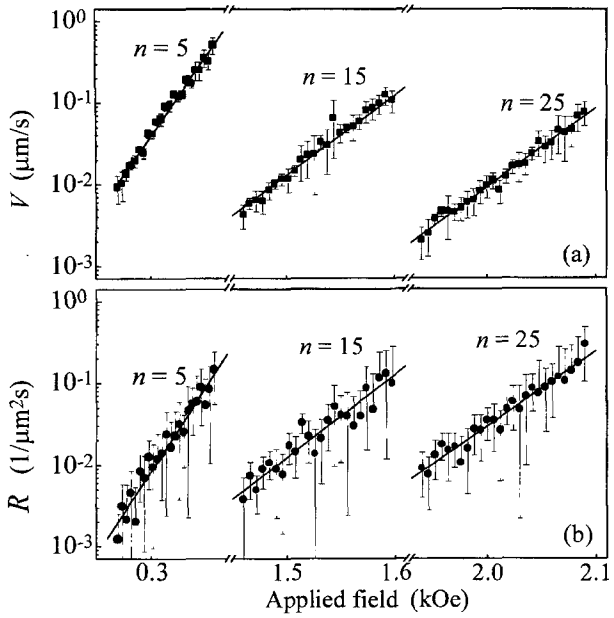

Fig. 2. The dependence of (a) the wall-motion speed V and (b) nucleation rate R with respect to the applied field for the (2-Å Co/11-Å Pd)$_n$ samples.

plane. All samples in this study had perpendicular magnetic anisotropy and showed square Kerr hysteresis loops.

3. Results and Discussion

The magnetic field dependence of the wall-motion speed V and nucleation rate R of Co/Pd multilayers has been determined from the time-resolved domain patterns during the magnetization reversal under various applied fields. In Figure 2, we plot (a) the wall-motion speed and (b) nucleation rate of the (2-Å Co/11-Å Pd)$_n$ sample with respect to the applied field H for three volumes of n Note that the wall-motion speed is very well defined, as shown in Figure 2(a), while the nucleation rate data are rather scattered, as shown in Figure 2(b). This is expected because the wall-motion is a successive process at every domain boundary, while the nucleation is a random process statistically governed by the switching probability.

The field dependence of the reversal processes has been analyzed within the context of a thermally activated relaxation process. Figure 2 vividly shows that both the reversal parameters are exponentially dependent on the strength of the reversing applied field. The solid line in each of the figures is the best fit of the correlated distribution using the fitting functions

$$\log V(H)=\alpha_W+\beta_W H$$
$$\log R(H)=\alpha_W+\beta_W H, \quad (5)$$

where α and β are the fitting parameters for the wall-motion and nucleation processes with subscripts W and N, respectively. The exponential dependence shows that both

Table 1. Experimental magnetic properties, activation volumes, and reversal ratio of Co/Pd multilayers

n	t_{Co} (Å)	M_S (emu/cm^3)	H_C (kOe)	V_W (10^{-18} cm^3)	V_N (10^{-18} cm^3)	V/R (μm^3)
5	2.0	250	0.4	7.4 ± 0.1	7.8 ± 0.3	3.4 ± 1.8
10	2.0	265	1.1	4.3 ± 0.1	4.8 ± 0.2	2.4 ± 1.1
15	2.0	270	1.7	3.4 ± 0.1	3.5 ± 0.2	1.5 ± 0.3
20	2.0	290	2.1	3.3 ± 0.1	3.1 ± 0.1	0.5 ± 0.1
25	2.0	310	2.2	3.0 ± 0.1	2.7 ± 0.1	0.4 ± 0.1
10	2.5	370	1.0	4.8 ± 0.2	5.0 ± 0.3	2.1 ± 1.2
10	3.0	400	0.7	6.4 ± 0.2	6.3 ± 0.4	0.9 ± 0.4
10	3.5	460	0.4	10.7 ± 0.6	9.4 ± 0.2	0.4 ± 0.1

reversal processes are governed by thermally activated relaxation. The fitting parameter α is closely related to the activation energy barrier for each process, preliminarily defining the characteristic switching rate (or attempt frequency) of an activation volume [4, 6, 9]. On the other hand, the fitting parameter β is related to the magnetic properties by

$$\beta_W=V_W M_S/k_B T$$
$$\beta_N=V_N M_S/k_B T, \quad (6)$$

where V_W and V_N are the activation volumes for the two reversal processes, respectively.

The activation volumes for wall-motion and nucleation processes have been determined from the fitting parameters and the saturation magnetization M_S for each sample. The values of the saturation magnetization were measured using a vibrating-sample magnetometer as listed in Table 1. Figure 3 shows the activation volumes for the wall-motion and nucleation processes for (a) the (2-Å Co/11-Å Pd)$_n$ samples with varying number of repeats n and (b) the (t_{Co}-Co/11-Å Pd)$_{10}$ samples with varying Co-layer thickness t_{Co}. With increasing number of repeats, both activation volumes decrease as shown in Figure 3(a). This may result from the increasing accumulation of lattice misfits, residual stress,

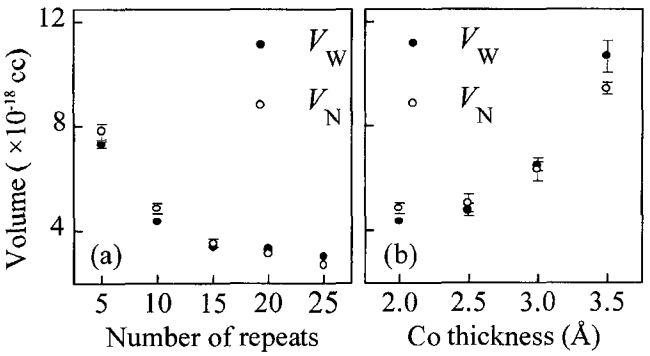

Fig. 3. The wall-motion activation volume V_W and the nucleation activation volume V_N of (a) the (2-Å Co/11-Å Pd)$_n$ samples with respect to the number of repeats n and (b) the (t_{Co}-Co/11-Å Pd)$_{10}$ samples with respect to the Co-layer thickness t_{Co}.

and other defects as the number of interfaces increases. On the other hand, with increasing Co-layer thickness, both the activation volumes increase, as illustrated in Figure 3(b). This can be understood since the lateral exchange length in a Co atomic monolayer is expected to be smaller than that in a few Co atomic layers if the sample preparation process is fully epitaxial. Note that both the activation volumes change together with changes in multilayer structure, which implies that both processes are influenced in the same manner by the structural and magnetic properties.

More detailed investigation revealed that the activation volumes for the wall-motion and nucleation processes are slightly different, and the difference changes systematically with changes in the multilayer structure. The nucleation activation volume is larger than the wall-motion activation volume for samples having fewer number of repeats, but the difference gradually decreases and finally reverses with increasing number of repeats, as seen in Figure 3(a). A similar trend was observed with changing the Co-layer thickness as seen in Figure 3(b); the nucleation activation volume is larger for the samples having thinner Co-layer thickness, but the difference is reversed and then, negatively increased with increases in the Co-layer thickness. We took the volume ratio V_W/V_N as a characteristic parameter, and this volume ratio was found to change from 0.9 to 1.1 with increasing either the number of repeats or the Co-layer thickness.

The activation volume ratio could be further analyzed with respect to the magnetization reversal modes. The magnetization reversal mode in Co/Pd multilayers have been reported to change from wall-motion dominated to nucleation dominated with increases in either the number of repeats or the Co-layer thickness [3, 18]. The contrasting reversal modes have been quantitatively characterized by the ratio of the wall-motion speed to the nucleation rate; wall-motion dominant reversal for a sample having a ratio larger than 1 μm^3, and nucleation dominant reversal for a sample having a ratio smaller than 1 μm^3. The reversal ratio changed slightly with changing applied field, and the field dependence was found to be in agreement with the difference between the activation volumes for the wall-motion and nucleation processes. In Table 1, we summarize the reversal ratio obtained from the reversal experiments under a reversing field of the coercivity for each sample determined by MOKE hysteresis loop measurements with the field changing at 60 Oe/sec.

It is very interesting to note that the activation volume ratio is closely related with the magnetization reversal modes [21]. Figure 4 clearly shows the correlation between the reversal ratio V/R and the activation volume ratio V_W/V_N. A wall-motion dominant reversal with larger V/R occurred for samples with $V_W < V_N$, while a nucleation dominant reversal with smaller V/R occurred for samples with $V_W > V_N$. Thus, it can be concluded that the correlation between the activation volumes of the wall-motion and

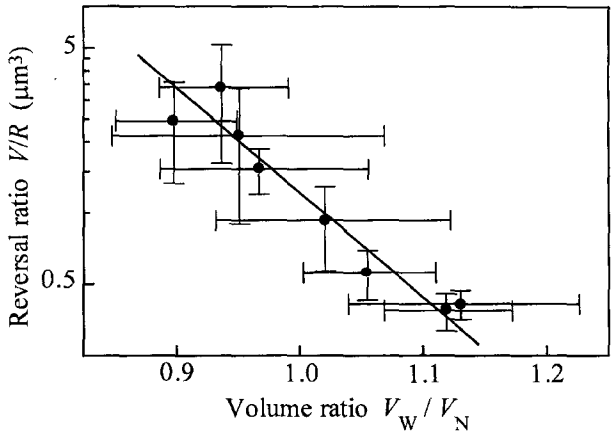

Fig. 4. The correlation between the reversal ratio of the wall-motion speed V to the nucleation rate R, and the volume ratio of the wall-motion activation volume V_W to the nucleation activation volume V_N.

nucleation processes has a crucial role in determining the magnetization reversal mode; the reversal mode is determined by the each balance between the switching rates of the wall-motion and nucleation processes, having an activation volume, where a smaller activation volume is easier to reverse than a larger one since the activation energy is proportional to the activation volume, as given in Eq. (1).

4. Conclusion

We have measured the activation volumes for the wall-motion and nucleation processes in Co/Pd multilayers by analyzing the field dependence of the magnetization reversal observed using a MOKE microscope system. Both activation volumes change in a similar way in the range $0.2\sim1.1\times10^{-17}$ cm^3 with changes in the multilayer structure. Interestingly, the difference between the activation volumes is systematically dependent on the multilayer structure; the wall-motion activation volume is smaller than the nucleation activation volume for a sample having a smaller number of repeats and a thinner Co-layer, and *vice versa*. The difference in the two activation volumes is closely related to the magnetization reversal mode; wall-motion dominant reversal occurs when the wall-motion activation volume is smaller than the nucleation activation volume, otherwise a nucleation dominant reversal occurs.

Acknowledgments

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