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Field dependence of the activation volumes in Co/Pd multilayered films

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The field dependence of the activation volumes of nucleation and wall-motion processes in magnetization reversal of Co/Pd multilayers is presented, as observed using a magneto-optical Kerr-effect microscope capable of real-time domain imaging in a wide time range of $10^{-5}-10^{3}$ s. The analytic forms of the activation volumes are derived from a theoretical consideration of equilibrium conditions of the two typical domain evolution processes in a ferromagnetic film with uniaxial perpendicular anisotropy; the theory is shown to be in good agreement with the experimental results. It was found that the field dependence of the activation volumes can be characterized by the difference between the domain wall energy and the dipolar energy. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2776010]

I. INTRODUCTION

Magnetization reversal dynamics in ferromagnetic thin films is a challenging issue in magnetism as well as in spintronics.[1,2] Recently, understanding magnetization reversal dynamics has been tremendously enhanced by means of advanced domain imaging techniques that are capable of direct observation of the domain evolution patterns.[3,4] It has been found that magnetization reversal takes place by thermal activation overcoming the energy barrier when the applied field is smaller than the coercive field.[5–8] In thermal activation, the two processes of nucleation and domain wall motion are involved.[9–12] The field dependence of the energy barrier of each process enables measurement of the activation volume, which is the unit volume acting as a single domain during magnetization reversal.[13–15] In general, the two activation volumes are unequal and the inequality in the two activation volumes critically influences magnetization reversal behavior.[12]

Thus far, most experimental studies have reported that the activation volume is independent of the applied magnetic field, a condition ascribed to the linear analysis of the energy barrier.[9–12] However, the theoretical analytic form of the energy barrier was described in terms of the nonlinear dependence on the applied field, resulting in the field dependence of the activation volume.[14] Thus, the underlying physics as regards the activation volume in ferromagnetic films remains a question of debate and complete agreement between experimental results and related theory is lacking. Recently, although the authors have experimentally observed the field dependence of the activation volume in a Co/Pd multilayer using a magneto-optical Kerr-effect microscope, no analytical explanations were offered at that time.[6] In the present study, an analytic explanation is presented for the field dependence of the nucleation and wall-motion processes in the magnetization reversal of ferromagnetic films with uniaxial magnetic anisotropy. The field dependence of the activation volumes were investigated in ferromagnetic Co/Pd multilayers as they provide two contrasting domain evolution processes of the nucleation and domain wall-motion according to the Co-sublayer thickness. For an analytical explanation, a theoretical model was adopted that considers these two typical domain evolution processes in a ferromagnetic film with uniaxial perpendicular anisotropy, as originally proposed by Kirby *et al.*[19]

II. EXPERIMENTAL PROCEDURE

For this study, the magnetization reversal behavior was investigated by means of a magneto-optical microscope magnetometer (MOMM) capable of grabbing time-resolved domain evolution patterns under a constant applied field. Details of the MOMM are described elsewhere.[4] The magnetization reversal was triggered by applying a magnetic field in a field range of 0.41–1.40 H_c to an initially saturated sample. An electromagnet was used to apply low magnetic fields corresponding to a long time range ($10^{-2}<t<10^{3}$ s), while a small coil connected to a pulse generator circuit was used to apply high fields for a short time duration ($10^{-5}<t<10^{-1}$ s).

The $(t_{\mathrm{Co}}\text{-Co}/11\ \text{Å Pd})_{10}$ samples with Co-sublayer thicknesses of $t_{\mathrm{Co}}=2$ and 4 Å were prepared on glass substrates by alternatively exposing them to two electron-beam sources of Co and Pd under a base pressure of 2.0×10^{-7} Torr and at ambient temperatures. The thickness calibration and the existence of the multilayer structure were determined by low-angle x-ray diffraction using Cu $K\alpha$ radiation. A magneto-optical Kerr effect hysteresis loop mea-

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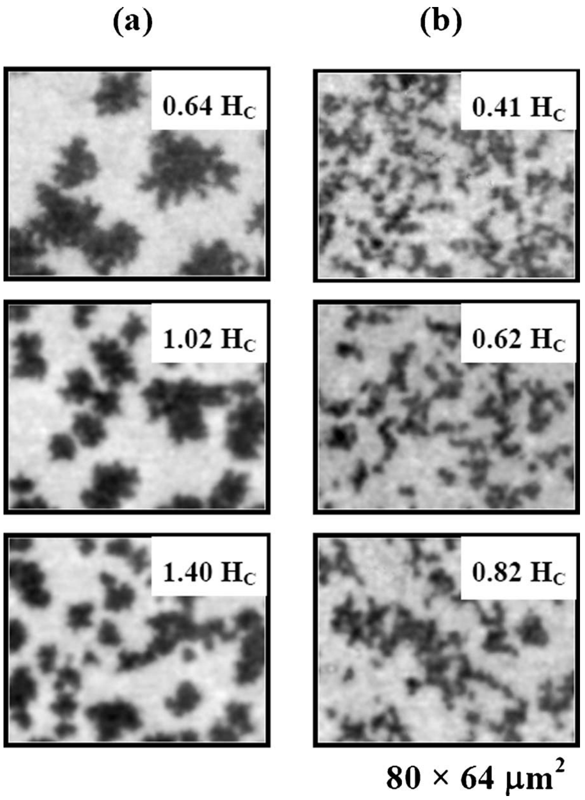

FIG. 1. Typical domain patterns of 30% magnetization reversal with respect to the applied field in the (a) $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ and (b) $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ samples, respectively, observed on a sample area of $80\times 64\ \mu\text{m}^2$ using MOMM. Here, H_c corresponds to the coercivity of each sample.

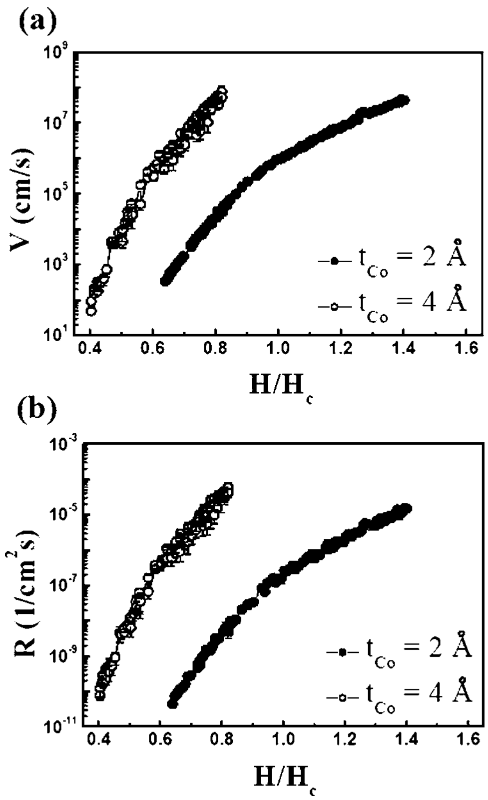

FIG. 2. (a) The wall-motion speed V and (b) the nucleation rate R with respect to the applied field in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ and $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ samples, respectively.

surement with a field sweeping rate of 30 Oe/s revealed that the samples had perpendicular magnetic anisotropy with a coercivity H_c of 609 and 203 Oe, respectively. It is well known that the Co/Pd multilayer system exhibits an increase of saturation magnetization and a decrease of the anisotropy constant as t_{Co} increases in this thickness region.[16] The saturation magnetization M_s, measured using a vibrating sample magnetometer increases from 266 to 462 emu/cm^3 as t_{Co} increases from 2 to 4 Å.

III. RESULTS AND DISCUSSION

Figure 1 shows typical domain patterns at 30% magnetization reversal with respect to each applied field in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ and $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ samples, respectively, observed on a sample area of $80\times 64\ \mu\text{m}^2$ using the MOMM, in which H_c corresponds to the coercivity of each sample. It is clearly seen that the magnetization reversal in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ sample exhibits a wall-motion dominant process at $H=0.64\ H_c$, whereas that of the $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ sample reveals a nucleation-dominant process at $H=0.41\ H_c$. The major origin of the different reversal behavior between the two samples is believed to be due to an increase in the dipolar energy, induced by an increase of M_s with the increase in t_{Co}.[17] Interestingly, the magnetization reversal behavior in each sample varies as the strength of the applied field increases. To be precise, as the applied field increases, the reversal behavior in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ sample changes from a wall-motion dominant to a nucleation dominant process, whereas in the $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ sample, it changes from a nucleation dominant to a wall-motion dominant process, as clearly seen in Fig. 1. The origin of these phenomena can be understood from a combination of a quantitative analysis of the domain evolution pattern and a theoretical approach, as explained later.

For a quantitative analysis, the wall-motion speed and the nucleation rate in each sample were measured from the time-dependent domain evolution patterns at a given applied field using the quantitative model described in Ref. 18. Figure 2 shows the wall-motion speed V and the nucleation rate R with respect to the applied field in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ and $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ samples. The applied field was normalized by the coercivity of each sample. In Fig. 2, the exponential dependence on the applied field smaller than the critical field ($\sim 1.87\ H_C$) is clearly seen, which implies the reversal behavior governed by the thermal activation process.[6] Generally, the critical field is defined as the applied field at which the reversal behavior takes a transition from the thermal activation process to the viscous process when the applied field is increased. In addition, it should be noted that the energy barriers of V and R in the thermal activation process are closely related with the values of $\ln V$ and $\ln R$ according to the Arrhenius law, respectively.[20] Hence, as expected from the field dependences of V and R, it was found that the energy barriers of V and R in the two samples are nonlinearly dependent on the applied field. The nonlinear field dependence of the energy

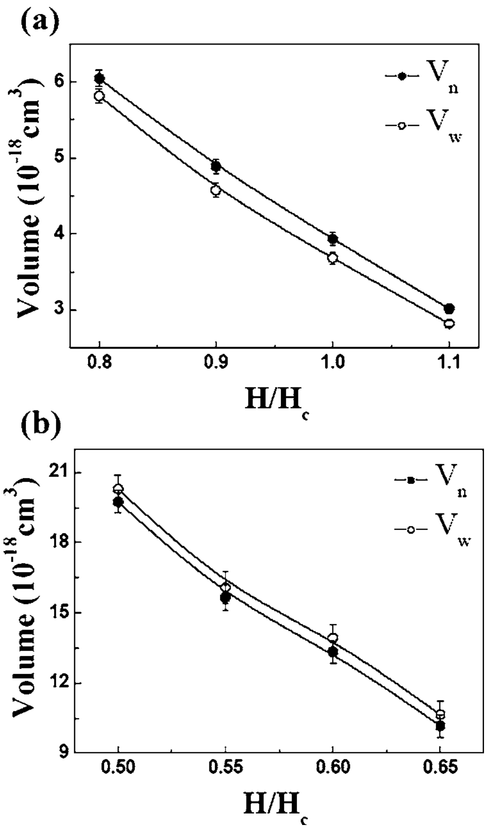

FIG. 3. The wall-motion activation volume V_w and the nucleation activation volume V_n with respect to the applied field in the (a) $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ and (b) $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ samples, respectively.

barrier can be witnessed via the systematic measurements of the thermal activation in a wide time range from 10^{-5} to 10^3 s.

Most strikingly, it was found that both the wall-motion and nucleation activation volumes vary with respect to the applied field. Each activation volume is determined from the field dependence of V and R using Gaunt's definitions.[13,14] Figure 3 shows the wall-motion activation volume V_w and the nucleation activation volume V_n with respect to the applied field in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ and $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ samples. As seen in Fig. 3, V_w and V_n no longer have a constant value and their values decrease with respect to the applied field. In addition, V_w and V_n are noticeably different from each other. The difference $\Delta V = V_w - V_n$ between the two activation volumes is estimated to be $-2.5 \pm 0.5 \times 10^{-19}$ cm^3 in the $(2\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ sample and $5.1 \pm 0.8 \times 10^{-19}$ cm^3 in the $(4\ \text{Å}\ \text{Co}/11\ \text{Å}\ \text{Pd})_{10}$ sample. The sign of ΔV was found to change from negative to positive with an increase in t_{Co}, the reason for which will be discussed later.

The experimental results of this study can be explained using a theoretical model considering a ferromagnetic system with perpendicular magnetic anisotropy, as originally proposed by Kirby *et al.*[19] In this model, the ferromagnetic thin film consists of identical single domain cells with the critical volume V_c on a hexagonal lattice with periodic boundary conditions, where V_c corresponds to the volume between the pinning centers of the domain wall. It is closely related to the Barkhausen volume during magnetization reversal. Each cell has a saturation magnetization M_s and a uniaxial perpendicular magnetic anisotropy K_u; its boundary has a wall energy density σ_w. The magnetization of each cell is aligned along the $+z$ direction, initially in the $-z$ direction. As described in Ref. 17, the magnetic energy E of a cell with the angle of magnetization direction θ from the $+z$ axis is expressed by

$$\begin{aligned} E = {} & K_u V_c \sin^2\theta - M_s V_c (H + \hat{H}) \cos\theta \\ & + 2(1 - \zeta \cos\theta) V_c \sigma_w / d_c, \end{aligned} \tag{1}$$

where H and $\hat{H}$ are the applied field and the demagnetizing field along the $+z$ direction, respectively. The third term of Eq. (1) corresponds to the domain wall energy of a cell, where the parameter ζ given by $\Sigma_k \cos\theta_k/6$ is the fraction of the magnetization state summed over the nearest neighboring cells k having a magnetization direction of θ_k with d_c the distance between the centers of the nearest neighboring cells. The energy barrier E_b of a cell for reversal, given by the difference in the magnetic energy between the initial value and the maximum, is then estimated in a simplified form expressed by

$$E_b = K_u V_c [1 - (h + m\hat{h} + \zeta w)]^2, \tag{2}$$

where $m = 2\pi M_s^2/K_u$, $w = \sigma_w/d_c K_u$, and $h = M_s H/2K_u$ are the ratios of the dipolar energy, the domain wall energy, and the Zeeman energy relative to the anisotropy energy, respectively. The parameter $\hat{h}$ given by $\hat{H}/4\pi M_s$ is the ratio of the demagnetizing field to its saturation value. Thus, the detailed energy barrier of a cell is determined according to the magnetization state of the neighboring cells, which is characterized by the two situation parameters of ζ and $\hat{h}$.

The two fundamental domain evolution processes of the wall motion and nucleation can be approximately characterized by the situation parameters ζ and $\hat{h}$, respectively, as described in Ref. 17: the wall motion takes place by switching a cell at the boundary of an existing domain, i.e., $(\zeta \approx 0, \hat{h} \approx 0)$, and the nucleation takes place at an isolated cell, i.e., $(\zeta \approx -1, \hat{h} \approx 1)$, where the self-demagnetizing energy of a cell is ignored. Assuming the values of the situation parameters as described earlier and applying the Arrhenius law[20] for each process, the wall-motion speed V and the nucleation rate R are expressed as

$$\begin{aligned} V &= V_0 \exp[-\beta'(1-h)^2], \\ R &= R_0 \exp[-\beta'(1-h+\alpha)^2], \end{aligned} \tag{3}$$

where V_0 and R_0 are the constants, $\beta' = K_u V_c / k_B T$ is the ratio of the magnetic anisotropy energy of a cell over the thermal activation energy, and $\alpha = w - m$ is a parameter that characterizes the difference between the domain wall and dipolar energies. From Eq. (3), it is clear that the energy barriers of V and R are nonlinearly dependent on the applied field, which is consistent with the experimental results here.

The analytic expressions of V_w and V_n derived from Eq. (3) using Gaunt's definitions[13,14] are as follows:

$$V_w = V_c(1-h),$$

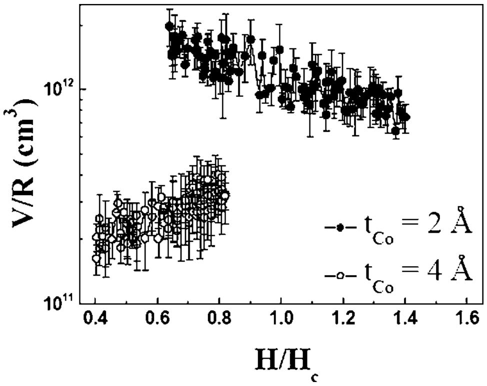

FIG. 4. The reversal ratio V/R values with respect to the applied field for the (2 Å Co/11 Å Pd)$_{10}$ and (4 Å Co/11 Å Pd)$_{10}$ samples.

$$V_n = V_c(1 - h + \alpha). \tag{4}$$

Similarly, the decrease of both V_w and V_n with respect to an applied field can be well explained by Eq. (4). Furthermore, it was found that the difference between V_w and V_n is determined by the parameter α, representing the difference between the domain wall energy and the dipolar energy, as given by $\Delta V = V_w - V_n = -\alpha V_c$. Accordingly, in the results, the change in the sign of ΔV with the increase in t_{Co} is due to the change in the sign of α, which can be ascribed to an increase in the dipolar energy induced by an increase of M_s with an increase in t_{Co}.

The parameter α also has an influence on the contrasting reversal process. The reversal ratio V/R is known to be an important value characterizing the contrasting reversal process: the domain reversal process changes from nucleation dominant to wall-motion dominant with an increase in V/R.[21] The analytic form of V/R determined from Eq. (3) is given by

$$V/R = V_0/R_0 \exp[-2\alpha\beta h + f(\alpha)]. \tag{5}$$

From these equations, $\ln(V/R)$ is clearly linearly proportional to an applied field and its slope depends on the value of α. For example, $\ln(V/R)$ in the sample with $0 < \alpha < 1$ decreases with the applied field, whereas $\ln(V/R)$ in the sample with $-1 < \alpha < 0$ increases with the applied field. The same trend was experimentally observed in the thermal activation reversal process of Co/Pd multilayers. Figure 4 shows the reversal ratio V/R with respect to the applied field in the (2 Å Co/11 Å Pd)$_{10}$ and (4 Å Co/11 Å Pd)$_{10}$ samples. As the applied field increases, $\ln(V/R)$ in the (2 Å Co/11 Å Pd)$_{10}$ sample decreases, whereas $\ln(V/R)$ in the (4 Å Co/11 Å Pd)$_{10}$ sample increases, as clearly seen in Fig. 4. These results are also consistent with the variation of the domain patterns with respect to the applied field, as shown in Fig. 1. Hence, the field dependence of V/R in the Co/Pd multilayers is well explained by the sign of α, induced by an increase of the dipolar energy with an increase in t_{Co}. Moreover, the variation of $\ln(V/R)$ in low fields with respect to α is characterized by $f(\alpha)$ which is an increasing function of α in the range of $-1 < \alpha < 1$. Specifically, the value of $\ln(V/R)$ in low fields is increased as α increases. Given that both $\ln(V/R)$ in low fields and ΔV strongly depends on the parameter α, it is natural for there to be a strong correlation between them.[9] Consequently, the reversal ratio V/R as well as the activation volumes with an applied field in ferromagnetic thin films can be understood in terms of the parameter α, that is, the difference between the domain-wall energy and dipolar energy.

An interesting quantity is the critical volume V_c as it represents the volume between the pinning sites in which the individual atomic moments are strongly correlated by the exchange interaction.[14,19] The critical volume V_c of each sample can be approximately estimated as $1.4 \pm 0.1 \times 10^{-17}$ cm^3 in the (2 Å Co/11 Å Pd)$_{10}$ sample and $5.1 \pm 0.3 \times 10^{-17}$ cm^3 in the (4 Å Co/11 Å Pd)$_{10}$ sample, by linear extrapolation from the field dependence of V_w to $H=0$ according to Eq. (4). Considering that the shape of the critical volume is hexagonal, the critical distance t_c defined as the width of the hexagon is estimated to be 35 nm in the (2 Å Co/11 Å Pd)$_{10}$ sample and 63 nm in the (4 Å Co/11 Å Pd)$_{10}$ sample, all of which are smaller than the domain wall thickness (~90 nm) measured via magnetic force microscope. These results reveal that the critical volumes in the two samples are largely influenced by the pinning sites rather than by the exchange interaction. In addition, the governing parameter α can be estimated as 0.02 in the (2 Å Co/11 Å Pd)$_{10}$ sample and −0.01 in the (4 Å Co/11 Å Pd)$_{10}$ sample using the relationship given by $\alpha = -\Delta V / V_c$. It was impossible to directly match the experimental value of α with the theoretical value of α estimated from the relationship $\alpha = w - m$ using the measured values of the macroscopic magnetic properties, which is to be expected considering the fact that the detailed microscopic magnetic properties during the domain reversal differ from macroscopic magnetic properties of a film. However, it was confirmed experimentally that the change of the sign of α with t_{Co}, induced by the increased M_s with t_{Co}, is a significant factor that determines the nature of the thermally activated domain reversal process in a wide field range.

IV. CONCLUSION

In this study, an experiment and analytic explanation is presented for the field dependence of the wall motion and nucleation activation volumes in the Co/Pd multilayers showing two contrasting domain evolution processes of the wall motion and nucleation. In an analytic study, a theoretical approach was carried out considering the two typical domain evolution processes in a ferromagnetic film with uniaxial perpendicular anisotropy. It was found that the parameter α, being the difference between the domain wall energy and dipolar energy, is essentially responsible for the change in the domain reversal and the difference between two activation volumes with the applied field.

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