Thermal fluctuations in perpendicular recording media: New methodology for estimation of activation moment

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Abstract. In nanoparticulate films with perpendicular magnetic anisotropy, a large demagnetizing field almost compensates for the experimentally designed change in the magnetic field applied parallel to the film normal. We propose a new method based on field-cycling to eliminate the uncertainty in the demagnetizing response from the analysis of the activation volume of the reversal or magnetic activation moment in such films. In this method, the applied field induced variation in the magnetic relaxation rate is measured before the effect of the demagnetizing field becomes dominant. We also discuss an analogical thermal-cycling method to clarify the temperature dependence of the barrier height for magnetization reversal in a magnetic field. We apply these methods to a Co₇₄Pt₁₆Cr₁₀-SiO₂ nanoparticulate film as an example. The results demonstrate that these methods are useful for studying thermal fluctuations in perpendicular recording media.

1. Introduction
Recently, thermal fluctuations in nanoparticulate films have been intensively studied, since thermally assisted switching of magnetization vectors in a desired region and the thermal stability in the direction of the vectors for the other recorded regions are central topics of current research on ultrahigh-density magnetic recording media. Therefore, a number of methods have been developed to clarify the effect of the thermal fluctuations [1-8]. For example, the typical energy barrier height for switching can be derived from the dependence of the M-H curve on the field sweep rate dH/dt [3]. Another example is the waiting time method [5,6], wherein the activation volume of the switching or magnetic activation moment μₐ is determined by the analysis of two relaxation curves at two different applied fields H_app. However, such analyses are complicated for perpendicular recording media, because a large demagnetizing field H_d perpendicular to the film plane compensates for the experimentally designed change in the applied field. In the former case, the variation in the actual field inside the film H = H_app - H_d abruptly slows down as the switching starts partially. Similarly, the latter method may be confusing if it is applied to perpendicular recording media, because at any measuring time during the observation, each nanoparticle is exposed to almost the same H between the two
relaxations with significantly different $H_{\text{appl}}$, as shown later. Thus, a new methodology is urgently required for the in-depth analysis of the thermal fluctuations in perpendicular recording media.

We propose a field-cycling method to simply clarify $\mu_A$ in perpendicular recording media, where $H_{\text{appl}}$ is temporarily changed to $H_{\text{appl}} + \Delta H_{\text{appl}}$ halfway through the relaxation. The new method was applied to a Co$_{74}$Pt$_{16}$Cr$_{10}$-SiO$_2$ nanoparticulate film provided by Hitachi Maxell, Ltd. [9]. The results demonstrate the proposed method’s simplicity and convenience.

2. Conventional methodology and its results
In nanoparticulate films, it is not easy to determine the magnitude of the magnetic moment $\mu$ for each nanoparticle. For this reason, we consider the fictitious magnetic moment $\mu_A$ given by $-\partial E_A/\partial H$ because the height of the potential energy barrier $E_A$ separating the direction of $\mu$ parallel to $H$ from the anti-parallel direction is lowered by the Zeeman energy $-\mu H$ in $H$. In general, variation in the total magnetization in $H$ is given by

$$\Delta M = (\partial M/\partial \ln t)|_t \Delta \ln t + (\partial M/\partial H)|_{\text{lin}} \Delta H = S \Delta \ln t + \chi \Delta H,$$

(1)

where $S$ is the magnetic viscosity and $\chi$ is the susceptibility. For perpendicular recording media, rotation of $\mu$ is almost negligible when $H$ is applied parallel to the film normal, because $H$ is parallel to the easy axis of the large perpendicular magnetic anisotropy. In other words, the reversal of $\mu$ due to thermally fluctuations is predominant. In this case, the relaxation time $\tau$ is expressed as $f_0^{-1} \exp(E_A(H)/k_B T)$, where $f_0$ is the attempt frequency and $k_B$ is the Boltzmann constant. If $E_A(H)$ is common in every nanoparticle at any $H$, magnetic relaxations may be scaled by the ratio $t/\tau$. $\Delta M = F(t/\tau)$, where $F(x)$ is a scaling function. This assumption leads to the relationship:

$$\mu_A = -\partial E_A/\partial H = k_B T \gamma / S = k_B T \langle \Delta \ln t/\Delta H \rangle |_M.$$

(2)

Therefore, $\mu_A$ or the activation volume of reversal $\mu_A/M$ is estimated by using the ratio $\gamma / S$ (Street-Woolley method [1]), where $M$ is the saturation magnetization. If $S$ significantly varies with $\ln t$ in the experimental time window, the waiting time method is also employed, where the times ($t_1, t_2$) required for the magnetization to decay to a constant level at slightly differing fields ($H_1, H_2$) are measured. $\mu_A$ is then estimated at $k_B T \ln(t_1/t_2)/(H_2 - H_1)$. [5,6].

Figure 1(a) shows a Co$_{74}$Pt$_{16}$Cr$_{10}$-SiO$_2$ nanoparticulate film with a thickness of 16 nm as an example; the film was completely saturated in a high magnetic field of $-20$ kOe at 300 K. The field was then isothermally reversed to $H_{\text{appl}} = 4.1$ kOe or $H_{\text{appl}} + \Delta H_{\text{appl}} = 4.15$ kOe at $t = 0$, and the magnetic relaxation of the film was measured at each field. $M$ was easily found to reach zero at $t_1 = 2$ ks in $H_{\text{appl}}$ and $t_2 = 0.6$ ks in $H_{\text{appl}} + \Delta H_{\text{appl}}$. Thus, $\ln(t_1/t_2)$ is 1.2. In contrast, consideration of $H_d$ is required to estimate the actual fields $H_1$ and $H_2$ at the applied fields. Generally, the self-demagnetizing field $H_d$ of each nanoparticle contributes to the shape anisotropy. Therefore, $H_d$ is expressed as $4\pi M - H_{gs}$. If we assume that the nanoparticles are disks with a diameter of roughly 50 nm [9], $H_{gs}$ is approximately $3\pi M$. Figure 1(b) shows the actual fields: in this case, $H_1 = H_{\text{appl}} - H_d$ and $H_2 = H_{\text{appl}} + \Delta H_{\text{appl}} - H_d$. Note that $H_d(t)$ is almost identical to $H_d(t)$ at any measuring time $t$, despite $\Delta H_{\text{appl}} = 0.05$ kOe. On the other hand, the difference under concern ($H_2(t_2) - H_1(t_1)$) is certainly equal to $\Delta H_{\text{appl}}$. As a result, $\mu_A$ was calculated to be $1.1 \times 10^5 \mu_B$. This non-intuitive estimation is confusing although the formalism is valid. The question arises as to why the two relaxation curves in Figure 1(a) are different from one another in the same actual field. One interpretation is that the difference is generated by a field-induced-reversal of $\mu$ in the very early stage of the relaxations, where $H_d(t) \neq H_d(t)$. However, the details are unclear since it occurs out of the experimental time window. Although the accuracy of the estimated $H_d$ is debatable, this issue is crucial as long as $H_d$ is large enough.
3. New methodology and its results

Now, we consider that the magnetic field is changed to \( H_{\text{appl}} + \Delta H_{\text{appl}} \) at \( t_0 \) after measurement starts at \( H_{\text{appl}} \) at \( t = 0 \). In this case, all of the states just before the changes are identical each other. Hence, \( H_d \) is uniform at \( t_0 \). After the changes, the relaxation of total magnetization \( M \) accelerates or slows down according to the variation in the barrier height \( E_A(H) \). Consequently, \( H_d \) starts to branch off in response to the divergence of \( M \) due to the acceleration/deceleration. Point is that, in the initial stage of the period, the difference of \( H_d \) is still small in comparison with \( \Delta H_{\text{appl}} \), i.e. \( \Delta H \sim \Delta H_{\text{appl}} \). If the field returns to \( H_{\text{appl}} \) at short time \( \Delta t \) later, we can estimate the extent of the acceleration or deceleration since contribution from field-reversible components such as the canting of \( \mu \) is almost the same between the periods before and after field-cycling. For this reason, the field-cycling method allows us to directly compare the two relaxations that start from the same initial state and proceed in different actual fields.

Figure 2(a) shows the same \( \text{Co}_74\text{Pt}_{16}\text{Cr}_{10}\text{-SiO}_2 \) nanoparticulate film being used as an example. First, the film was completely saturated in \( H_{\text{appl}} = -20 \) kOe, applied parallel to the film normal, at 300 K. The field was then isothermally reversed to \( H_{\text{appl}} = 4.1 \) kOe at \( t = 0 \). (b) Estimated variation in actual field \( H \) inside film during magnetic relaxation.

![Figure 1](image_url)

**Figure 1.** (a) Magnetic relaxation curves of \( \text{Co}_74\text{Pt}_{16}\text{Cr}_{10}\text{-SiO}_2 \) nanoparticulate film at applied fields of \( H_{\text{appl}} = 4.1 \) kOe and of \( H_{\text{appl}} + \Delta H_{\text{appl}} = 4.15 \) kOe. The film was completely saturated in a field of \(-20 \) kOe, applied parallel to the film normal, at 300 K. The field was then isothermally reversed at \( t = 0 \). (b) Estimated variation in actual field \( H \) inside film during magnetic relaxation.

The obtained exponential relationship \( \Delta t_{\text{eff}} \sim \Delta t \) was consistent with equation (2). More
simply, this relationship is explained by the ratio \( \tau(H+\Delta H)/\tau(H) = \exp[(E_A(H+\Delta H) - E_A(H))/k_B T] \sim \exp[\Delta H(\partial E_A/\partial H)/k_B T] \). Therefore, \( \mu_A \sim 0.7 \times 10^5 \) \( \mu_B \) can be derived from the gradient of the relationship. This value is comparable to \( \mu_A \) of 1.1 \( \times 10^5 \) \( \mu_B \) estimated by the waiting time method (see above), and \( \mu_A \) of 1.0 \( \times 10^5 \mu_B \) by the Street-Woolly method, where \( S \equiv 13 \) emu cm\(^{-3}\) and \( \chi_{\text{eff}} \equiv 0.3 \) emu Oe\(^{-1}\) cm\(^{-3}\) obtained from dc demagnetization curve. As discussed here, the field-cycling method enabled us to achieve a reasonable \( \mu_A \) in the straightforward framework.

As stated in the introduction, thermally assisted switching of \( \mu \) in nanoparticulate films is a topical issue for high-density magnetic recording. Thus, we applied the cycling method to the temperature dependence of \( E_A(H, T) \), i.e.,

\[
\partial E_A(H, T)/\partial T - E_A/T = k_B T (\Delta \ln T/\Delta T)_{E_A}. \tag{3}
\]

Figure 3(a) shows the magnetic relaxation curves of the same nanoparticulate film. After the film was completely saturated by \(-20\) kOe at \( T = 300\) K, the field was isothermally reversed to \( H_{\text{appl}} = 4.1\) kOe at \( t = 0 \), and the measurement was started. Halfway through the relaxation, the temperature was temporarily lowered to \( T + \Delta T = 297\) K, as shown in the inset. The relaxation was readily shown to slow down after cooling at \( t_0 = 1.3\) ks. The relaxation rate then recovered the original extent after reheating at \( t_0 + \Delta t = 10\) ks. When the relaxation curve obtained after thermal-cycling is shifted leftward by 5.2 ks, it can be superimposed on the isothermal relaxation (represented by open circles on the solid curve). In other words, the variation \( \Delta M \) in the period \( \Delta t = 8.7\) ks at \( T + \Delta T \) is almost equivalent to \( \Delta M \) in the period \( \Delta t_{\text{eff}} = 3.5\) ks at \( T \). Figure 3(b) shows the ratio of \( \Delta t_{\text{eff}} \) to \( \Delta t \) obtained for various \( \Delta T \). We are certain the relationship \( \ln(\Delta t_{\text{eff}}/\Delta t) \propto \Delta T/T \) for the weak thermal perturbations \( \Delta T \).

On the basis of the equation (3), the gradient of the relationship indicates that \( E_A(H, T) \sim \partial E_A(H, T)/\partial T \sim -0.005 \) (K\(^{-1}\)) at \( H \approx 4.1\) kOe, where \( E_A(H, T)/k_B T \) was assumed to be 30 in our experimental time window. This temperature dependence was significantly large in comparison with that of the anisotropy constant reported previously [10]. Although further information on \( E_A(H, T) \) cannot be derived from the results obtained here, this example demonstrates the possible applicability of the thermal-cycling method.

Figure 2. (a) Magnetic relaxation curves of Co\(_{74}\)Pt\(_{16}\)Cr\(_{10}\)-SiO\(_2\) nanoparticulate film with a field-cycling. The film was completely saturated in a field of \(-20\) kOe, applied parallel to the film normal, at 300 K. The field was then isothermally reversed to \( H_{\text{appl}} = 4.1\) kOe at \( t = 0 \). Halfway through the relaxation, the actual field \( H \) was temporarily changed, as shown in the inset. The open circles indicate time-shifted curves. (b) Ratio of \( \Delta t_{\text{eff}} \) to \( \Delta t \) as a function of amplitude of the field-cycling \( \Delta H \).
4. Summary
In nanoparticulate films with perpendicular magnetic anisotropy, a large demagnetizing field, whose amplitude is often unknown, compensates for the designed change in the applied field. Therefore, experimental results obtained by changing the applied field are always controversial as long as uncertainty remains in the demagnetizing response. Therefore, new methodology based on field cycling is proposed to eliminate the influence of the large demagnetizing field from the analysis of the activation volume of the reversal or magnetic activation moment in the nanoparticulate films. Additionally, an analogous argument shows that the relaxometry with thermal-cycling is also helpful in discussing the temperature dependence of the barrier height for magnetization reversal in a magnetic field. Thus, thermal/field cycling is found to be a promising methodology for investigating thermal fluctuations in perpendicular recording media.

References
[1] Street R and Woolley J C 1949 Proc. Phys. Soc. A62 562
[2] Wohlfarth E P 1984 J. Phys. F: Met. Phys. 14 L155
[3] Sharrock M P 1994 J. Appl. Phys. 76 6413
[4] Lyberatos A, Chantrell R W 1997 J. Phys.: Condens. Matter 9 2623
[5] O’Grady K, Dova P and Laidler H 1998 Mater. Res. Soc. Symp. Proc. 517 231
[6] El-Hilo M, O’Grady K and Chantrell R W 2002 J. Magn. Magn. Mater. 248 360
[7] Lauhoff G and Suzuki T 2000 J. Appl. Phys. 87 5702
[8] Dutson J D, Litvinov D, Gibbs M R J, Inaba Y, Muraoka H and O’Grady K 2006 J. Magn. Magn. Mater. 304 51
[9] Hagiya H, 2010 Master Thesis, Ibaraki Univ.
[10] Inaba N, Uesaka Y and Futamoto M 2000 IEEE Trans. Magn. 36 54

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