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Structural Control of Magnetic Properties in Co/Pd Multilayer for Heat Assisted Perpendicular MRAM Application

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Abstract: Temperature dependence of magnetic properties in $[Co/Pd]_N$ multilayer have been systematically studied for lithographically patterned samples with different Co thicknesses t_{Co} and the bi-layer number N by extraordinary Hall effect measurements. The perpendicular coercive field H_c decreases with the increase of t_{Co}, excepting a very thin thickness of 1 Å. While a threshold temperature (T_{th}) of H_c vanishing linearly increases with the increase of t_{Co}. The H_c and T_{th} monotonously increases with the increase of N, ranged from 5 to 50. The superior crystalline structure for samples with the larger N was confirmed from enhanced Co/Pd(111) peak height in the XRD pattern. A practical thermal activation coefficient α of 174 was evaluated from a sweep rate dependence of H_c. A marked improvements for the heat assisted MRAM application was attained by air annealing of $[Co(1.7\ Å)Pd(8\ Å)]_{30}$ at 220℃, that is, the H_c was increased from 1.3 kOe to 2.9 kOe, while T_{th} reduced from 210℃ to 190℃.

Keywords: Magnetic multilayer, Perpendicular magnetic anisotropy, Magnetic random access memory, Thermally assisted magnetization reversal, Extra ordinary Hall Resistance

1. Introduction

Recently, heat assisted magnetization reversal of fine magnetic dots with high perpendicular magnetic anisotropy (PMA) has been believed to be a promising technique to realize ultra high-density magnetic recording. The concept can be introduced to a magnetic random access memory (MRAM)[1]. Among various PMA materials, a multilayer of Co/Pd is a promising candidate for a future MRAM application owing its extremely high PMA, insuring the thermal stability of magnetic dots with an order of 10 nm. Since the magnetization direction in MRAM have to be switched with a current induced magnetic field or a spin transfer torque[2], the heat assisted temporal reduction of the energy barrier between the bi-stable states is crucial to realize a tolerable power consumption in a high-density MRAM. Hence, deep understanding for a thermo-magnetic performance of Co/Pd and its artificial controlling are important. Various mechanisms for the PMA have been reported, that is, the atomic scale broken symmetry at the interface[3], the stress induced anisotropy in Co and/or Co-Pd alloy[4,5], hybridization of Co $3d$ and Pd $4d$[6]. In the present study, the temperature dependence of coercivity and its structural controlling have been investigated for Co/Pd thin films with various layer structures. The extraordinary Hall Effect (EHE) measurements[7] were used for characterizing lithographically patterned samples considering the application for micro-fabricated devices.

2. Experimentals

The structured multilayer films were fabricated on glass substrates by a tandem type DC magnetron sputtering with a multi-cathode system (Anelva SPC-350). The background pressure before deposition was below 5×10^{-7} Torr and the Ar pressure during the deposition was 20 m Torr. The substrate mounted on a sample holder was rotated at 50 rpm around the Co and Pd targets with a PC controlled shutter system. The sputtering rates for Co and Pd are 1.0 Å/sec and 0.36 Å/sec, respectively. After optimizing the Pd thickness as 8.0 Å from preliminary characterization, the following two series of samples were mainly studied.

(1) $[Co(t_{Co}\ Å)/Pd(8.0\ Å)]_{20}$, with t_{Co} ranged from 1.0 Å to 3.0 Å.
(2) $[Co(1.7\ Å)/Pd(8.0\ Å)]_N$, with N = 5, 7, 10, 15, 20, 25, 30 and 40.

The crystalline structure were characterized with a X-ray diffractometer of Cu $K\alpha$ radiation (λ=1.54 Å). Magnetic properties of as-grown films were measured with a vibrating sample magneto meter (VSM). The extra ordinary Hall Resistance (R_H) in the micro-structured samples, fabricated with the following process, was measured by DC 4-terminal methods. The Co/Pd

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multilayer film was structured into 600 × 600 μm^2 pattern by the photolithography and lift of process. Additional 4-terminal lead electrodes, consisted of Ti/Au, were fabricated with the lift off process. Composite multi layer samples with vertically integrated two different Co/Pd multi layers were also fabricated to confirm the fundamental MRAM memory operation. The magnetization switching behavior for the individual Co/Pd multi layers were studied with a current-in-plane magneto resistance (CIP-MR) measurement.

3. Results and Discussion

MH hysteresis for an as-grown film and an R_H hysteresis for a micro-structured sample of [Co (1.7 Å)/Pd (8.0 Å)]$_{20}$ are compared, as shown in **Fig. 1**.

It should be noticed that any significant difference is not observed for the squareness and coercive field H_c, confirming the torrelance of magnetic properties for a standard micro fabrication process. The following magnetic properties were measured by the EHE measurements.

Figure 2(a) shows a series of R_H hysteresis curves measured at various temperatures ranged from 23℃ to 150℃. The square hysteresis is sustained up to 100℃, suggesting a nucleation type magnetization reversal at the raised temperature. The H_c linearly decreases with the increase of temperature and extrapolates to zero value at 150℃, defined as a threshold temperature T_{th}, as shown in **Fig. 2(b)**. The net uniaxial perpendicular anisotropy K_u can be a summation of the surface anisotropy ($2K_s/t_{Co}$) and the volume anisotropy (K_v)

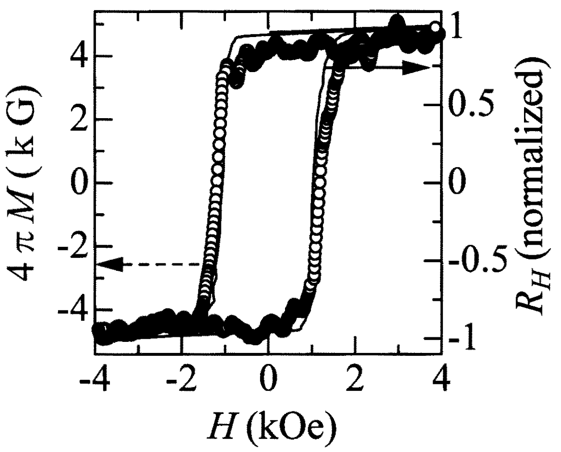

Fig. 1 Hysteresis curves of magnetization M and extraordinary Hall resistance R_H for both as-grown and micro-structured samples of [Co(1.7 Å)/Pd(8.0Å)]$_{20}$.

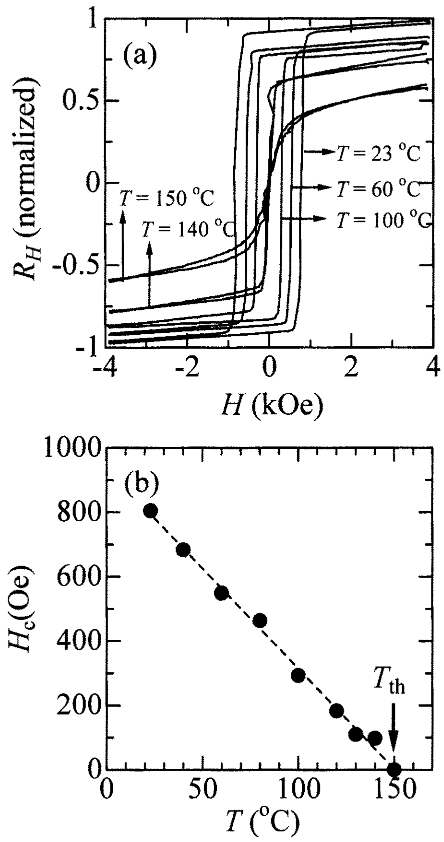

Fig. 2(a) R_H hysteresis measured at various temperatures. **(b)** Temperature dependence of H_c. Threshold temperature T_{th}, at which H_c vanishes, is noted in the figure.

including the shape anisotropy term[8]. The vanishing of coercivity at T_{th} can be explained by the compensation of opposite contributions from the K_s and K_v with different temperature dependences. The thermally activated magnetization reversal is another cause for the H_c reduction, as discussed later.

Figure 3(a) presents the T_{th} and H_c as a function of t_{Co}. The H_c increases with the decrease of t_{Co} (1.7 ~ 3.0 Å), reflecting the property of the surface anisotropy enhanced at the thinner thickness. The rapid reduction of H_c at t_{Co} = 1 Å can be attributable to an imperfect interface between Co and Pd layers. The results can be related to the previously reported temperature dependence of K_u[4,9]. The mostly linear decrease of T_{th} with decreasing t_{Co} can be explained by the reduction of the exchange interaction.

Resultantly, complementary properties for H_c and T_{th} can be realized by adjusting the t_{Co}, which can be utilized for an exchange coupled bi-layer in heat assisted MRAM architecture[10]. The X-ray diffraction (XRD) patterns for samples with different t_{Co} are compared in **Fig. 3(b)**. The

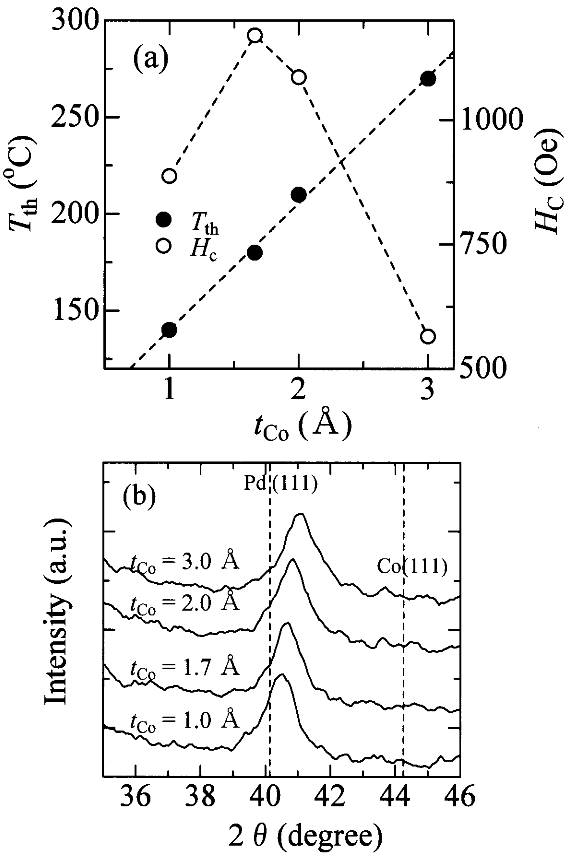

Fig. 3(a) Dependence of H_c (by VSM @ *RT*) and T_{th} (by Extra Hall Resistance) on Co layer thickness t_{Co}.
(b) XRD patterns for samples with different t_{Co}.

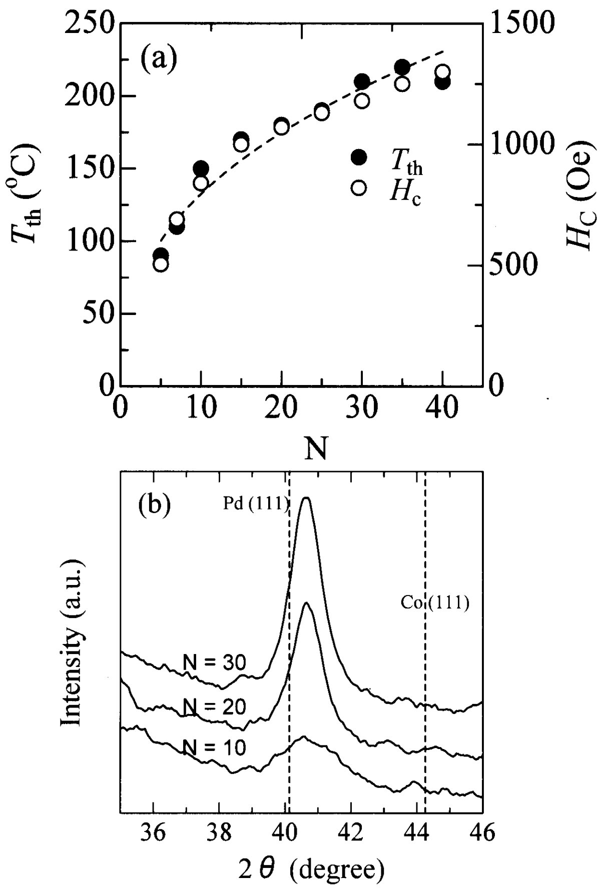

Fig. 4(a) Dependence of H_c (by VSM @ *RT*) and T_{th} (by Extra Hall Resistance) on layer repetition number *N*.
(b) XRD patterns for samples with different *N*.

main peak is continuously shifted from that of Pd(111) to Co(111) with the increase of t_{Co}, which can be ascribed to the increase of Co composition in the Co-Pd alloy layer formed at the interface.

The magnetic properties can also be controlled with the repetition number *N*, as shown in **Fig. 4(a)**. A correlative increase of the H_c and T_{th} is observed for the *N* values ranged from 5 to 40. The notable increase of H_c and T_{th} at N <10 can be attributable to the deterioration of crystalline structure as demonstrated by the significant decrease of the XRD peak height in **Fig. 4(b)**.

The sweep rate (R) dependence of the H_c for samples with N = 10 and 30 are plotted, in **Fig. 5**. The experimental results reasonably fit with the theoretical prediction[11], where the $H_c(R)$ is expressed as a square root of ln(1/*R*). The values of thermal coefficient α ($\fallingdotseq K_uV/k_BT$, *V*: activation volume) are evaluated from the least square fitting as 47 (N = 10) and 174 (N = 30), respectively. The perpendicular anisotropy field of 4.7 kOe (N = 10), 8.0 kOe (N = 30), from VSM measurements, and an attempt frequency $f_0 = 10^{10}$ are assumed in the fitting.

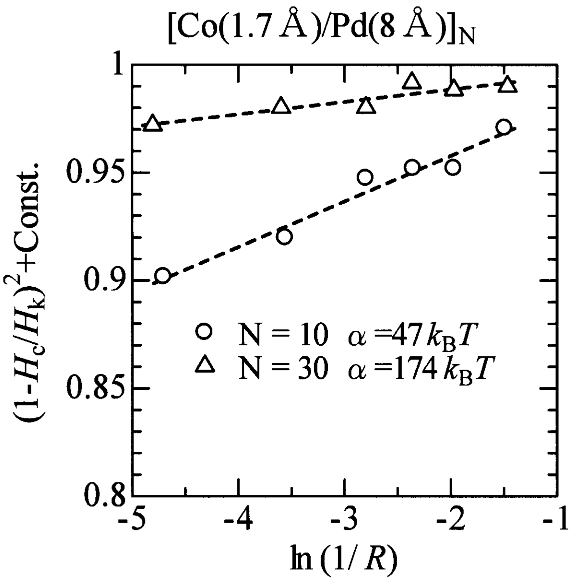

Fig. 5 Field sweep rate dependence of H_c for [Co(1.7 Å)/Pd(8.0 Å)]$_N$, with N = 10 and 30. Broken lines present the least square fitting to a theoretical prediction.

Another drastic enhancement of H_c was realized by an air annealing at a relatively low temperature of 220℃, as shown in **Fig. 6**. The H_c (@ R.T.) for [Co(1.7 Å)/Pd(8.0 Å)]$_{30}$ was increased from 1.3 kOe to 2.9 kOe by annealing, while T_{th} reduced from 210℃ to 190℃. An exchange decoupling of the grains would be a probable cause for the enhanced H_c.

It was also found that an addition of Pd seed layer is effective to improve the PMA. **Figure 7** shows the enhancement of H_c in [Co(1.7 Å)/Pd(8.0 Å)]$_{10}$, from 800 Oe to 1.6 kOe, by inserting a Pd seed layer with the thickness of 50 Å. In this case, T_{th} also increases from 150℃ to 200 ℃. Thus, the physical origin of the enhancement of H_c appears to be different from that for the annealing effect as shown in **Fig. 6**, where T_{th} was decreased. The increase of both H_c and T_{th} can be reasoned to be the intrinsic enhancement of PMA realized with the well defined *fcc* (111) crystalline structure formed by the template effect of Pd seed layer.

Based on the above mentioned results, composite multi layer samples, consisted from two different Co/Pd multi layers, were fabricated. The field induced magnetization switching behavior was investigated by CIP-MR measurements, which reflect the relative magnetization orientation in the two Co/Pd multi layers. **Figure 8** shows the MR change caused by an external field sweep from -4.0 kOe to +4.0 kOe, measured for two samples of Ta(50.0 Å)/[Co(t_{Co} Å)/Pd(8.0 Å)]$_{10}$/Co(3.0 Å)/Cu(40.0 Å)/Co(3.0 Å)[Pd(8.0 Å)/Co(1.6 Å)]$_{20}$, where t_{Co} = 2.0 Å and 3.0 Å. Selective switching of the two Co/Pd multi layers was confirmed from the observed plateau like resistance change. That is, the high and low resistance states correspond to the anti-parallel and parallel magnetization orientation in the two Co/Pd multi layers, respectively.

A temperature dependence of CIP-MR profiles for the composite multi layer sample was shown in **Fig. 9(a)**. The plateau like MR behavior was successfully held for a practical temperature range from 23 to 200℃. As can be seen in **Fig. 9(b)**, about 50 % reduction of H_c for the top and bottom Co/Pd were realized by increasing the temperature to 250℃. The results demonstrate a potential performance of the composite Co/Pd multi layer as a candidate of heat assisted MRAM memory cell.

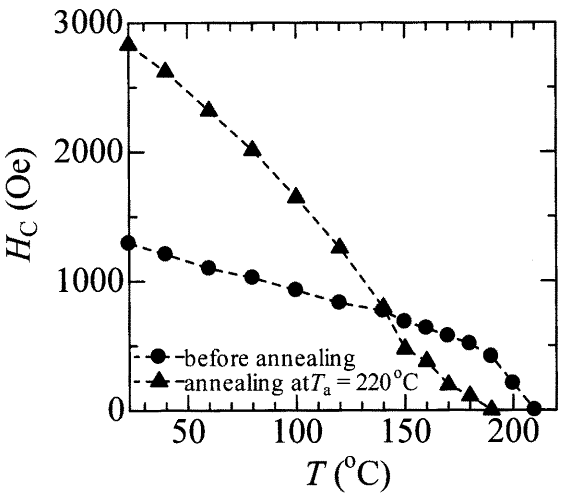

Fig. 6 Comparison of temperature dependence of H_c for [Co(1.7 Å)/Pd(8.0 Å)]$_{30}$ before and after annealing.

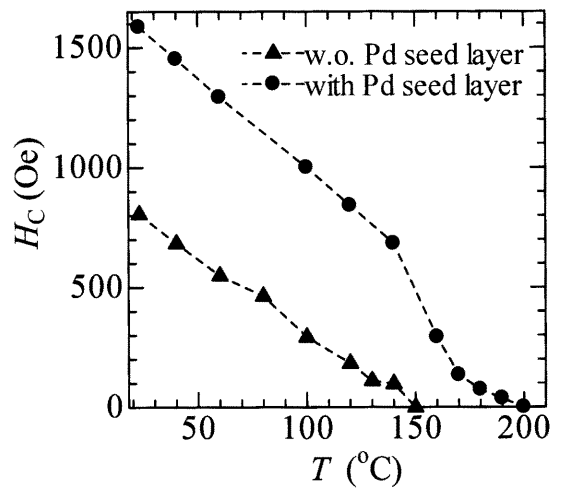

Fig. 7 Comparison of temperature dependence of H_c for [Co(1.7 Å)/Pd(8.0 Å)]$_{10}$ with and without Pd(50 Å) seed layer.

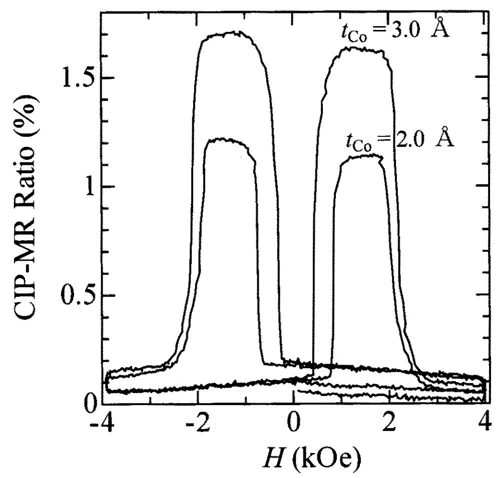

Fig. 8 Selective magnetization layer switching of Ta(50.0 Å)/[Co(t_{Co} Å)/Pd(8.0 Å)]$_{10}$/Co(3.0 Å)/Cu(40.0 Å)/Co (3.0 Å)[Pd(8.0 Å)/Co(1.6 Å)]$_{20}$ with t_{Co}=2.0 Å and 3.0 Å.

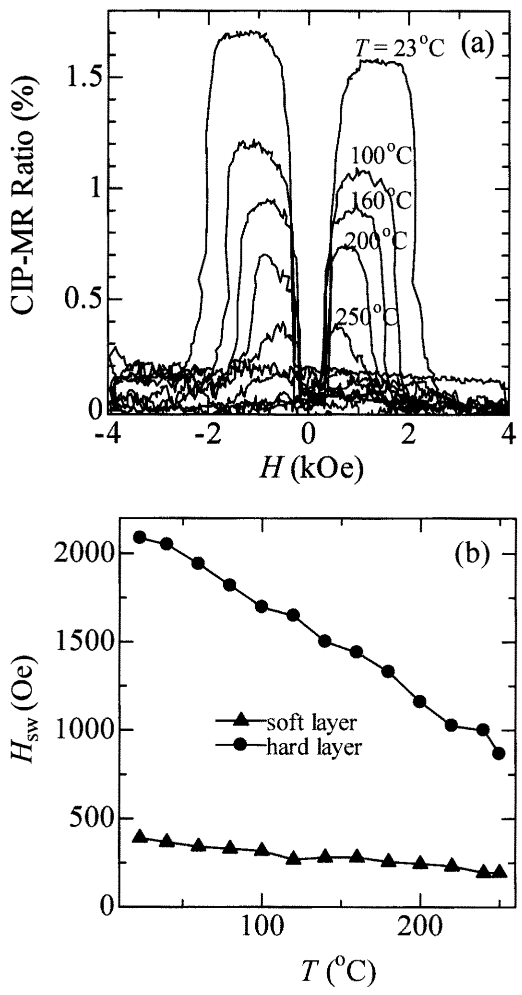

Fig. 9(a) Temperature dependence of CIP-MR behavior for Ta(50.0 Å)/[Co(3.0 Å)/Pd(8.0 Å)]$_{10}$/Co(3.0 Å)/Cu(40.0 Å)/Co(3.0 Å)[Pd(8.0 Å)/Co(1.6 Å)]$_{20}$.
(b) Temperature dependence of H_c for the top and bottom Co/Pd layers.

4. Conclusions

In conclusion, the structural control of magnetic properties in Co/Pd multilayer has been studied systematically with the Co thickness t_{Co} and the total layer repetition number N as parameters. The practical thermal stability of 174 k_BT was evaluated from the sweep rate dependence of H_c. A high H_c value of 2.9 kOe at an ambient temperature and its low temperature diminishing at 190 ℃ were realized by an air annealing of [Co(1.7 Å)/Pd(8.0 Å)]$_{30}$. The property is suitable for the heat assisted MRAM operation. The wide range of structurally controlled magnetic properties can be applied for a vertically integrated multi-bit MRAM cell architecture.

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