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

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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 tc and the bilayer number N by extraordinary Hall effect measurements. The perpendicular coercive field $H_c$ decreases with the increase of $t_c$, excepting a very thin thickness of 1 Å. While a threshold temperature ($T_o$) of $H_c$ vanishing linearly increases with the increase of $t_c$. The $H_c$ and $T_o$ 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 $a$ 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°C, that is, the $H_c$ was increased from 1.3 kOe to 2.9 kOe, while $T_o$ reduced from 210°C to 190°C.

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)†. 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‡, 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§, the stress induced anisotropy in Co and/or Co-Pd alloy¶, hybridization of Co 3d and Pd 4d©. 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) measurements⁵ 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 $5 \times 10^{-7}$ 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(t₀ Å)/Pd(8.0 Å)]ₙ₀, with $t_0$ ranged from 1.0 Å to 3.0 Å.

(2) [Co(1.7 Å)/Pd(8.0 Å)]ₙ₀, 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 ( $\lambda = 1.54$ Å). Magnetic properties of as-grown films were measured with a vibrating sample magnetometer (VSM). The extraordinary Hall Resistance ($R_h$) in the micro-structured samples, fabricated with the following process, was measured by DC 4-terminal methods. The Co/Pd
multilayer film was structured into $600 \times 600 \, \mu m^2$ 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)]$_{120}$ 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 tolerance 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°C to 150°C. The square hysteresis is sustained up to 100°C, 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°C, 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 $(K_v)$ including the shape anisotropy term. The vanishing of coercivity at $T_{th}$ can be explained by the compensation of opposite contributions from the $K_u$ 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 $K_u$ for different $t_{co}$ values. 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. The X-ray diffraction (XRD) patterns for samples with different $t_{co}$ are compared in Fig. 3(b).
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 $H_{c}$ 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\textsuperscript{11}, where the $H_{c}(R)$ is expressed as a square root of $\ln(1/R)$. The values of thermal coefficient $a = K V/k_B T$ ($V$: 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 $f_0 = 10^{12}$ are assumed in the fitting.

![Fig. 3(a) Dependence of $H_{c}$ (by VSM @ RT) and $T_{th}$ (by Extra Hall Resistance) on Co layer thickness $t_{Co}$.](image1.png)

![Fig. 4(a) Dependence of $H_{c}$ (by VSM @ RT) and $T_{th}$ (by Extra Hall Resistance) on layer repetition number $N$.](image2.png)

![Fig. 5 Field sweep rate dependence of $H_{c}$ for $[\text{Co}(1.7 \text{ Å})/\text{Pd}(8.0 \text{ Å})]_N$, with $N = 10$ and 30. Broken lines present the least square fitting to a theoretical prediction.](image3.png)
Another drastic enhancement of $H_c$ was realized by an air annealing at a relatively low temperature of 220°C, as shown in Fig. 6. The $H_c$ (at R.T.) for $[\text{Co}(1.7 \text{ Å})/\text{Pd}(8.0 \text{ Å})]_n$ was increased from 1.3 kOe to 2.9 kOe by annealing, while $T_m$ reduced from 210°C to 190°C. 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 $[\text{Co}(1.7 \text{ Å})/\text{Pd}(8.0 \text{ Å})]_n$ from 800 Oe to 1.6 kOe, by inserting a Pd seed layer with the thickness of 50 Å. In this case, $T_m$ also increases from 150°C to 200°C. 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 $T_m$ was decreased. The increase of both $H_c$ and $T_m$ 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)/$[\text{Co}(t_c \text{ Å})/\text{Pd}(8.0 \text{ Å})]_n$/Co(3.0 Å)/Cu(40.0 Å)/Co(3.0 Å)/Pd(8.0 Å)/Co(1.6 Å)]_n, where $t_c$ = 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°C. 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°C. The results demonstrate a potential performance of the composite Co/Pd multi layer as a candidate of heat assisted MRAM memory cell.
4. Conclusions

In conclusion, the structural control of magnetic properties in Co/Pd multilayer has been studied systematically with the Co thickness $t_c$ 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 $190^\circ C$ were realized by an air annealing of $[\text{Co}(1.7 \text{ Å})/\text{Pd}(8.0 \text{ Å})]_{13}$. 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.

References