ACTIVATION VOLUME OF FePt-TaN NANOCRYSTALLINE THIN FILMS WITH HIGH COERCIVITY

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Abstract --- Coercivity and magnetic aftereffect of nanocrystalline $L1_0$ type FePt films is discussed in connection with magnetocrystalline anisotropy and grain size. Magnetocrystalline anisotropy field shows very large value up to more than 40 kOe dependent on annealing temperature. This large magnetocrystalline anisotropy enabled to maintain large $K_u V/kT$ value more than 40 (10 years stability) even with very fine grain size of 7~8 nm, which described the potential of this alloy film to resist thermal fluctuation of magnetisation. Although intergranular exchange coupling enhances the thermal stability of the films, it is successfully described that the change in magnetic viscosity is qualitatively coincident with that of $K_u V/kT$ value. Activation volume V_{ac} of these films is much larger than grain volume, which is likely to be attributable to the existence of intergranular exchange coupling.

Key words: FePt nanocrystalline films, magnetocrystalline anisotropy, coercivity, magnetic aftereffect, activation volume

I. INTRODUCTION

The reduction of grain size is essentially required for thin film recording media to achieve ultrahigh density recording [1]. However, the size reduction degrades magnetic viscosity owing to thermal agitation of magnetic moment [2]. Therefore experimental analysis of magnetic viscosity phenomena is now very important to understand magnetisation reversal and coercive force mechanism of thin magnetic films. In the present study, we fabricated nanocrystalline FePt films, and discuss coercivity and magnetic aftereffect of the films in connection with magnetocrystalline anisotropy and grain size.

II. EXPERIMENTAL PROCEDURE

Nanocrystalline FePt thin films were fabricated by a thermal crystallization from amorphous structure. $(Fe_{50}Pt_{50})_{80}Ta_{20}$ -N films were fabricated by conventional r.f. sputtering with Ar + N₂ plasma under an N₂ gas flow ratio $F=N_2/(Ar+N_2)$ of 30%. Total gas pressure was fixed at 30 mTorr. Films were deposited on non-textured quartz substrates, and film thickness was fixed at 10 nm. Films were crystallized by an annealing process in vacuum (10^{-6} mbar) at various temperatures from 600 to 670 °C for about 30min. In each measurement of the time dependence of magnetization, the sample was initially saturated with a positive field of 30 kOe, and then a constant negative testing field was applied. Coercivity was measured at various sweeping rates of 12.5~200 Oe/sec.

III. RESULT AND DISCUSSION

Fig.1 shows TEM bright field images of the samples respectively annealed at 600 and 650 °C. It is clearly seen that fabricated films successfully consist of nanocrystalline grains less than 10 nm. TEM diffraction patterns revealed that these grains are fct-FePt ($L1_0$ tetragonal phase), and (101) crystallographic plane are almost parallel to the film plane. Although the formation of TaN was expected at the grain boundary of FePt because of strong bond between Ta and nitrogen, clear diffraction coming from TaN phase were not observed in the diffraction patterns. With increasing annealing temperature, T_A , the mean grain size of FePt was found to increase almost linearly from about 5 nm ($T_A=600$ °C) to about 8 nm (650 °C).

Fig.2 shows a T_A dependence of coercivity H_c . The films show isotropic magnetic property in the film plane. The given H_c values are those measured at 50 Oe/sec in sweeping rate, dH/dt . The value of H_c increases with increasing T_A , and goes beyond 3 kOe at 650 °C. This temperature is about 150 °C higher than that of pure FePt alloy films prepared in Ar + 4%H₂ atmosphere [3]. Saturation magnetisation M_s shows almost a constant value of about 550 emu/cm³ at higher T_A than 620 °C. It should be noted here that the squareness of magnetization curve, M_r/M_s , kept almost a constant value of about 0.75 against T_A , even though the H_c drastically increases.

Magnetocrystalline anisotropy field, H_k^{grain} , was estimated by a measurement of rotational hysteresis loss, W_r , as a function of applied field, H . The value of H_k^{grain} was defined as a field where W_r vanishes by using extrapolation method in the $W_r \sim 1/H$ plot [4].

Fig.1 TEM bright field images of the samples.

Fig.2 Annealing temperature T_A dependence of H_c .

Fig.3 The values of magnetocrystalline anisotropy field $H_{k\text{grain}}$ and calculated $K_u V/kT$ as a function of T_A . Close marks mean that these values were obtained under an assumption that the H_k^{grain} value linearly increases within this T_A range.

Fig.3 shows the values of H_k^{grain} and calculated $K_u V/kT$ as a function of T_A . Here, $K_u = H_k^{\text{grain}} M_s / 2$, V is the volume of a grain, k is the Boltzmann constant, and T is temperature (K). The values of $K_u V/kT$ which give thermal relaxation time $\tau = 1$ second and 10 years by using Arrhenius Neel law are respectively shown in the figure. The value of H_k^{grain} at 630 and 640 °C showed large values respectively of about 33 and 44 kOe. These values are several times larger than those of Co based thin film media [4]. The series of the $W_r \sim 1/H$ curves from $T_A = 600$ to 650 °C indicated that H_k^{grain} drastically increases with increasing T_A within this region. However the H_k^{grain} values at 600 and 620 °C were impossible to be estimated because of unstable magnetic torque in these films, which may be attributable to magnetic aftereffect during the measurement. On the other hand, the H_k^{grain} values at 650 °C was not possible to be estimated either, because the maximum applied field of the measurement system was not

enough to determine H_k^{grain} value by the extrapolation method in the $W_r \sim 1/H$ plot. Because of these reasons, the H_k^{grain} values given at 620 and 650 °C in the figure were estimated by using those at 630 and 640 °C under an assumption that the H_k^{grain} value linearly increases within this T_A range. The calculated $K_u V/kT$ value is found to increase drastically with increasing T_A due to the increases of V and K_u . It should be noted here that induced large magnetocrystalline anisotropy enables to maintain large $K_u V/kT$ value more than 40 (10 years stability) even with very fine grain size of 7~8 nm (in the T_A region more 630 °C), which obviously indicates the high potential of $L1_0$ type FePt to resist thermal fluctuation.

On the other hand, the values of normalised coercive force H_c/H_k^{grain} generally indicates the strength of intergranular exchange coupling [4,5]. The H_c/H_k^{grain} value of the present samples is almost a constant of 0.05~0.07. This suggests that the degree of intergranular magnetic coupling does not change remarkably against T_A . It should be mentioned here that these values of H_c/H_k^{grain} are a few times smaller than that of Co based thin film media [4], and several times smaller than the simulated value for the ideal media with no intergranular exchange coupling ($H_c/H_k^{\text{grain}} = \text{about } 0.45$) [2,5]. This result means that there is still a certain strength of intergranular exchange coupling.

In the measurement of time dependence of magnetization, almost logarithmic time decay have been observed under constant negative applied fields for all samples, and we show the representative behaviour in fig.4. Viscosity coefficient S [6] is determined as an slope of the $M(t) \sim \ln t$ as follows.

$$M(t) = M_0 - S \ln t \quad (1).$$

Here, M_0 is a constant magnetization. The values of M_0 and S are clearly dependent on applied field H_a .

Fig.5 shows S values as a function of H_a . The S value at $H_a = 0$ was very small less than a few erg/cm³ in every sample. Although the trend of $K_u V/kT \sim T_A$ plot in the fig.3 suggests that the τ value at 600 °C is much smaller than the measurement time scale, this sample is also thermally stable when $H_a = 0$. This is attributable to the existence of intergranular exchange coupling [7]. On the other hand, the S value is found to show the maximum at around $H_a = -H_c$ in all the samples. Particularly the S of the $T_A = 600$ °C sample shows a very large value of about 48 emu/cm³ which corresponds to about 23% decay of S/M_r value by the increment of observation time a factor of 10. The S value at $H_a = -H_c$ remarkably decreases with increasing T_A , which is well coincident with the change in $K_u V/kT$ value in fig.3.

Street and Woolley proposed [6] the following relationship between S and the irreversible susceptibility χ_{irr} , which defines fluctuation field H_f ,

$$S = \chi_{\text{irr}} H_f \quad (2).$$

Fig.4 Time dependence of magnetization $M(t)$ at various applied field H_a .

Fig.5 Applied field dependence of viscosity coefficient S .

Fig.6 shows a representative H_a dependence of H_f determined by using eq.(2). Here, χ_{irr} is the differential of the remanent magnetisation curve. The H_a dependences of S and χ_{irr} are shown in the inserted figure. With increasing H_a , the value of H_f drastically drops, and shows almost a constant in the range of field values larger than H_c for which it can accurately be measured. Here, the dotted line in the figure corresponds to H_a dependence of H_f values theoretically calculated for a uniaxial non-interacting fine particles system with randomly oriented anisotropy axes [8], with using H_k^{grain} value experimentally obtained. The value of H_f in the range of field values larger than H_c is found to be much smaller than that theoretically obtained, and this may be attributable to the enhancement of thermal stability due to the intergranular exchange coupling in these films. On the contrary, in the range of field values close to zero, S shows certain values even though χ_{irr} is almost zero, resulting in the large H_f values. It should be noted here that H_f in this field range is larger than that of non-interacting system. It is assumed in this field range that local

Fig.6 Applied field dependences of H_f , χ_{irr} , and S of the sample with $T_A=640^\circ\text{C}$.

magnetizations still remain almost parallel to each other in the direction of plus field initially saturated. It is likely that the large values of H_f in this field range are associated with strong intergranular exchange interaction which promotes the magnetized state.

Another representative magnetic aftereffect is the frequency dependence of the coercivity and this has been studied mainly in particulate media [9-11]. A following expression [9-11] based on coherent rotation has been successfully used to represent this phenomena.

$$H_c(t') = H_k \{ 1 - ((kT/K_u V) \ln(A t'))^n \} \quad (3),$$

where A is a numerical factor, n is the value from 0.5 to 1 dependent on anisotropy axes orientation and/or based assumption for calculation [10,12], and t' is the time needed for a constant field equal to H_c to reduce the magnetisation from saturation remanence to zero. Intuitively, it is expected that this t' will have an approximately inverse relationship with the sweep rate of applied field dH/dt .

Fig. 7 shows the values of $\Delta H_c/H_{c0}$ as a function of logarithmic scale of dH/dt . Here, H_{c0} is coercivity at $dH/dt = 1$ Oe/second, and ΔH_c is the difference of coercivity at certain dH/dt value and H_{c0} . The value of $\Delta H_c/H_{c0}$ is found to increase linearly with increasing $\ln(dH/dt)$ in the present observation range of $\ln(dH/dt)$ values. It is obvious that the slope in the $\Delta H_c/H_{c0} \sim \ln(dH/dt)$ plot decreases with increasing T_A , which is well coincident with the change in $K_u V/kT$ mentioned above.

These magnetic aftereffect experimentally observed enables an estimation of activation volume V_{act} of these films. Wohlfarth [13] proposed the following relationship between H_f and V_{act} .

$$V_{act} = kT / (H_f M_s) \quad (4).$$

Fig.7 The values of $\Delta H_c / H_{c0}$ as a function of logarithmic scale of dH/dt .

Fig.8 T_A dependence of activation volume V_{act} with that of grain volume V .

The value of V_{act} is obviously field dependent. On the other hand, Bruno et al. proposed [12] the following relationship between V_{act} and the sweeping rate dependent coercivity.

$$V_{act} = kT / (qM_s),$$

where

$$q = dH_c / d(\ln(dH/dt)) \quad (5).$$

Fig.8 shows the values of V_{act} at $H_a = -H_c$, calculated by using eq.(4), as a function of T_A . In the figure, the values of V_{act} obtained by using eq.(5), and grain volume V are also shown in comparison. The values of V_{act} obtained by using eqs.(4) and (5) are quantitatively well agreed with each other. The value of V_{act} drastically decreases with increasing T_A . It should be noted here that V_{act} values are much larger than V values particularly at lower T_A , which is completely different from the behaviours observed in particulate films or systems [14]. This characteristic property is assumed to be caused by the intergranular

exchange coupling although the difference between V_{act} and V is much more enhanced than that theoretically computed [15].

IV. SUMMARY

We experimentally described the potential of $L1_0$ type FePt alloy to resist thermal fluctuation of magnetisation. Magnetocrystalline anisotropy field shows very large value up to more than 40 kOe dependent on annealing temperature. This large magnetocrystalline anisotropy enabled to maintain large $K_u V / kT$ value more than 40 (10 years stability) even with very fine grain size of 7~8 nm. Although intergranular exchange coupling enhances the stability of the films to resist thermal fluctuation, it is successfully described that the change in magnetic viscosity is qualitatively coincident with that of $K_u V / kT$ value. Activation volume V_{act} of these films is much larger than grain volume, which is likely to be attributable to the existence of intergranular exchange coupling.

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