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著者	中村　慶久
journal or publication title	IEEE Transactions on Magnetics
volume	35
number	5
page range	2697-2699
year	1999
URL	http://hdl.handle.net/10097/47868

doi: 10.1109/20.800956

Thermal Fluctuation of Magnetization in Nanocrystalline FePt Thin Films with High Coercivity

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***Abstract* — The effect of thermal fluctuations of magnetization on static and dynamic properties is discussed for nanocrystalline FePt films. The large magnetocrystalline anisotropy K_u of $L1_0$ type FePt results in large $K_uV/(kT)$ values of more than 70 even for very fine grain sizes of 7~8 nm, indicating the potential of this alloy film to resist thermal fluctuation of magnetization. It is successfully demonstrated that larger $K_uV/(kT)$ values of these films lead to lower magnetic viscosity. Annealing at higher temperature results in larger K_uV/kT values and smaller V_{act}. The remanent coercivity measured at high sweep rate by using pulsed magnetic fields indicates the high thermal stability of these alloy films at high frequencies. However, the results indicated that care should be taken to induce an adequate magnitude of K_u for the FePt alloy to be used as ultrahigh density recording media.**

***Index Terms* — FePt nanocrystalline films, magnetic viscosity, pulse magnetic field, remanent coercivity**

INTRODUCTION

$L1_0$ type FePt films are one of the candidates for ultrahigh density thin film media [1], [2]. The large magneto-crystalline anisotropy (K_u) value of these films is expected to result in high thermal stability even for fine grain structures. However, the large K_u value is likely to yield a large coercivity at the recording frequency, resulting in poor overwrite properties [3]. In the present study we fabricated nanocrystalline FePt films, and discussed the effect of the thermal fluctuation of magnetization on the static and dynamic magnetic properties.

EXPERIMENTAL PROCEDURE

Nanocrystalline FePt thin films were fabricated by thermal crystallization from an amorphous state. $(Fe_{50}Pt_{50})_{80}Ta_{20}$-N films having amorphous structure were fabricated by conventional r.f. sputtering with Ar + N_2 plasma under an N_2 gas flow ratio $F=N_2/(Ar+N_2)$ of 10% or 30%. Total gas pressure was fixed at 30 mTorr. Films were deposited on non-textured quartz substrates, and the film thickness was fixed at 10 nm. Films were crystallized by an annealing process in vacuum (10^{-6} mTorr) at various temperatures from 600 °C to 670 °C for 30min.

The time dependence of magnetization was measured using a VSM. In each measurement, the sample was initially saturated with a positive field of 30 kOe, and then a constant negative testing field was applied. The remanent magnetization curve was measured at two applied field sweep rates. One was at 100 Oe/s by the same VSM system, another was at ~10^8 Oe/s by a pulsed magnetometer (PMVSM01, Hayama Inc.). All magnetic properties were measured in the film plane.

Manuscript received February 25, 1999.
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RESULTS AND DISCUSSION

A. Structure and Remanent Magnetization Curves

Nanocrystalline FePt thin films, with very fine TaN at the grain boundaries, were successfully fabricated by the thermal crystallization process [4]. TEM diffraction patterns revealed that FePt has $L1_0$ tetragonal phase, and the (101) crystallographic plane was almost parallel to the film plane. The mean grain size of FePt, D, was found to increase with increasing annealing temperature T_A. Representative D values were ~5 nm (T_A=600 °C, F=30%) and ~8 nm (T_A=650 °C, F=30%).

Fig.1 shows the remanent magnetization curves for the films (F=30%) at various annealing temperatures. The sweep rate of the applied field was 100 Oe/s. The remanent coercivity, H_r, increased significantly with increasing T_A. The values of H_r for T_A higher than 640°C are more than 3 kOe, even though the mean grain size is only 7~8 nm in these films. Rotational hysteresis loss, W_r, analysis revealed that this increase of H_r is due to a significant increase of the magnetocrystalline anisotropy field of the grains, H_k [4]. The ratio of H_r to H_k of the present samples ranges from 0.05 to

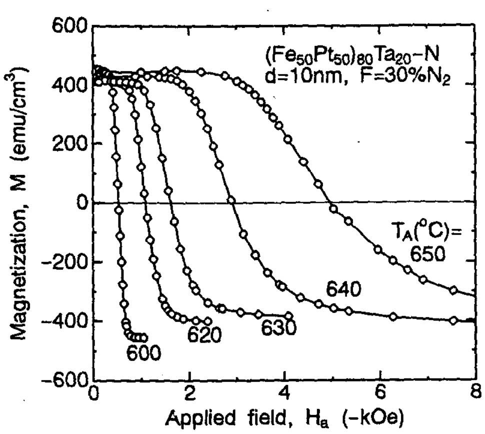

Fig.1 Remanent magnetization curves for films annealed at various temperatures, T_A.

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0.09, which indicates the presence of intergranular exchange coupling.

Saturation magnetization M_s showed an almost constant value of about 550 emu/cm^3 at T_A higher than 620 °C. It should be noted here that the squareness of the hysteresis loop, M_r/M_s, is approximately constant, with a value of ~0.75, against T_A, even though H_r increased significantly.

A series of films deposited at F=10% had slightly lower H_r values than those at F=30%, but showed almost the same T_A dependence. The value of M_s was ~600 emu/cm^3, which was larger than that at F=30%.

B. Long Term Stability

In the measurement of time dependence of magnetization within the time scale from 1 to 1500 s, an almost logarithmic time decay was observed in constant negative applied fields for all samples.

Fig.2 shows the viscosity coefficient S [5] as a function of applied field H_a. The estimated values of $K_uV/(kT)$ are also listed in the figure. Here, $K_u = H_kM_s/2$ (H_k was estimated by W_r analysis [4]), V is the volume of a grain by TEM, k is the Boltzmann constant, and T is the temperature (K).

The S of the T_A=600 °C sample shows a very large peak value of about 48 emu/cm^3, which corresponds to ~23% decay of S/M_r value with a factor of 10 increase in the observation time. The peak values of S at around H_a=-H_c show a significant decrease with increasing T_A, which is consistent with the change in $K_uV/(kT)$. In the samples annealed at T_A higher than 630°C, the large induced K_u results in large $K_uV/(kT)$ values of more than 70, even for a very fine grain size of 7~8 nm. This indicates the high potential of $L1_0$ type FePt for long term stability and resistance to thermal fluctuations.

C. Activation Volume

Experimentally observed magnetic viscosity enables an estimation of the activation volume, V_{act}, of these films. In principle the V_{act} gives the limit on recorded bit size.

Fig.3 shows the activation volume at H_a=-H_c as a function

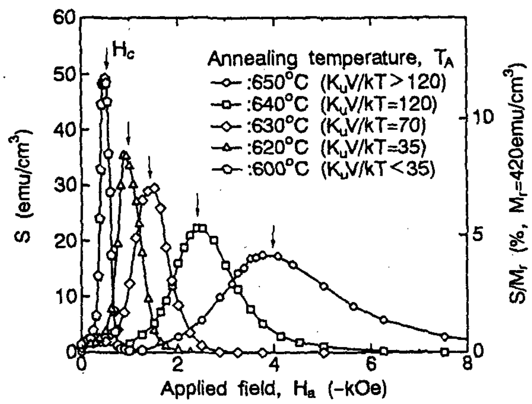

Fig.2 Applied field dependence of viscosity coefficient S. Estimated $K_uV/(kT)$ values are also shown.

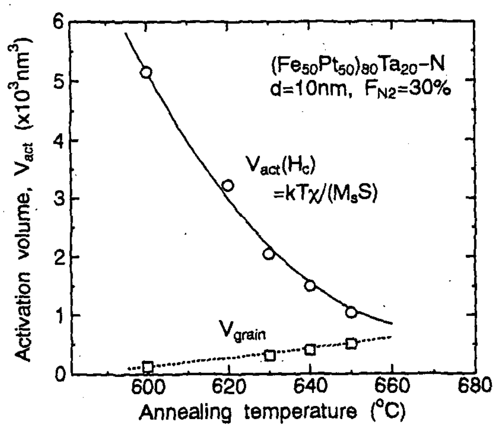

Fig.3 T_A dependence of the activation volume, V_{act}, and grain volume, V_{grain}.

of T_A, together with the grain volume, V_{grain}. The values of V_{act} were calculated using relations proposed by Street and Woolley [5] and Wohlfarth [6].

The values of V_{act} are much larger than V_{grain} values, particularly at lower T_A, which is different from the behavior observed in particulate films [7]. This is assumed to be caused by the intergranular exchange coupling, although the difference between V_{act} and V_{grain} is much greater than that computed theoretically [8]. It should be noted here that the value of V_{act} decreases significantly with increasing T_A, which suggests that a larger K_u (H_k) value reduces V_{act} through a reduction of the magnetic interacting volume.

D. Remanent Coercivity at High Frequency

A related dynamic property also due to thermal agitation is the field sweep rate dependence of H_r. The frequency dependence of the coercivity, H_c, has been studied mainly in particulate media [9], [10]. The following expression, based on coherent rotation, has been used successfully to represent this phenomenon [9], [10],

$$H_c(t') = H_0 \{ 1 - ((kT/K_uV) \ln (At'))^n \} \quad (1),$$

where A is a numerical factor, n is a value between 0.5 and 1, dependent on the anisotropy axes orientation and/or based on the assumptions of the calculation [9], [11], [12]. H_0 corresponds to the switching field at the frequency constant f_0 (=10^9~10^{10} s^{-1}), and t' is the time needed for a constant field equal to H_c to reduce the magnetization from saturation remanence to zero [10]. Intuitively, it is expected that this t' will have an approximately inverse relationship with the sweep rate of the applied field, dH/dt. This equation concerning H_c was originally derived from the switching of particle magnetization, therefore, (1) is principally applicable to H_r behavior.

Fig.4 shows representative pulse magnetic fields used for the present measurement. Two pulse magnetic fields with pulse heights of 5.37 kOe and 2.70 kOe are shown in the figure. The sweep rate of the field was defined as the slope of the line drawn between two points at H=0 and the maximum field, as shown in the figure. This sweep rate changes as a function of the pulse height, but its value is of the order of 10^8 Oe/s, except for pulse fields with very low heights.

Fig.5 shows remanent magnetization curves measured at 100 Oe/s and 10^8 Oe/s for a representative sample. The value of H_r at 10^8 Oe/s is found to be approximately 1.5 times larger than that at 100 Oe/s. The value of H_r at 100 Oe/s, measured by the VSM, is obviously reduced by thermal agitation.

Table I shows H_r values measured at 100 Oe/s (H_r^V) and at 10^8 Oe/s (H_r^P) for samples with various T_A. The values of $K_uV/(kT)$ and H_0 calculated using (1) are also shown in the table. Here, n=0.5 was assumed in the calculations.

It is clear that H_r^P of all samples are much larger than H_r^V. The samples annealed at higher T_A shows larger H_r^P values. But it should be noted that the ratio of H_r^P to H_r^V decreases significantly with increasing T_A, which indicates the high potential of $L1_0$ type FePt to resist thermal fluctuations.

The calculated $K_uV/(kT)$ show larger values at higher T_A, which is a similar dependence upon T_A to that obtained from torque analysis, as shown in Fig.2. It should be noted here that the H_0 values, which correspond to H_r values at high frequencies, close to f_0, are about 1.4~3 times larger than H_r^V values, depending on thermal stability of the sample. For recording media application the present films need to have a much smaller intergranular exchange coupling. However, it is obvious that the reduction of intergranular exchange coupling results in much larger H_0 [13], which would lead to poor overwrite performance. To avoid this, care should be taken to induce an adequate magnitude of K_u in the FePt alloys.

Table I The values of H_r measured at 100 Oe/s (H_r^V) and at 10^8 Oe/s (H_r^P) for representative samples with various T_A. The calculated values of $K_uV/(kT)$ and H_0 by using eq. (1) with H_r^V and H_r^P are also shown.

T_A (°C)	F (%)	H_r^V (Oe)	H_r^P (Oe)	H_r^P/H_r^V	$K_uV/(kT)$	H_0 (Oe)	H_0/H_r^V
600	30	530	960	1.8	48.7	1620	3.06
620	30	1090	1800	1.65	58.9	2850	2.61
640	10	2270	3290	1.45	84.8	4700	2.07
650	10	3440	4040	1.18	270.7	4820	1.40

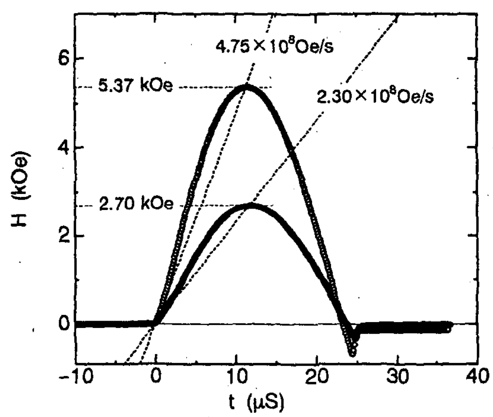

Fig.4 Two representative pulse magnetic fields used for the high sweep rate measurements.

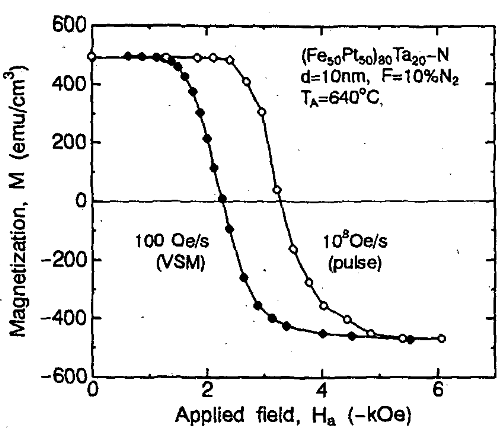

Fig.5 The remanent magnetization curves measured at 100 Oe/s and 10^8 Oe/s for a representative sample.

Conclusion

We demonstrated experimentally the potential of $L1_0$ type FePt alloys to resist thermal fluctuations of magnetization. It was successfully demonstrated that the large $K_uV/(kT)$ values of these films result in low magnetic viscosity (the long term stability). Annealing at higher temperature results in larger K_uV/kT values and smaller V_{act}. The remanent coercivity measured at high sweep rate by using pulse magnetic fields indicates the high thermal stability of these alloy films. However, the results indicate that, in order to be used as an ultra high density recording media, we must take care to induce an adequate magnitude of K_u in the FePt films.

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