$L1_0$型構造を持つFePt合金薄膜のc面配向精密制御

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Accurate Control of *c*-Plane Orientation of FePt Alloy Thin Film with $L1_0$ Structure Parallel to the Surface

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はじめに　$L1_0$型構造を持つFePt規則合金は6.6×10^7 erg/cm^{3}の高いK_uを示す．そのため，この合金薄膜は高密度磁気記録媒体やMRAMなどへの応用に向けて盛んに研究されている．デバイス応用のためには，磁化容易軸である[001]方位（c軸）を面直一方向に制御する必要がある．しかしながら，(001)配向の多結晶下地層，もしくは，単結晶基板上に膜形成を行うと，c軸が面直に向いた$L1_0$(001)結晶に加え，c軸が面内に存在する$L1_0$(100)結晶が膜中に混在する可能性がある[1,2]．これまで，我々は，高基板温度製膜[3]，もしくは，低温製膜後に熱処理[4]を施すことにより，MgO(001)基板上に$L1_0$構造を持つ40 nm厚のFePt膜を形成してきた．いずれの方法で形成した膜においても，$L1_0$(100)結晶が混在した．c軸方位制御には，面直方向の格子圧縮，もしくは，面内方向の膨張が有効であると考えられる．本研究では，MgO(001)基板上に低基板温度（200 °C）でエピタキシャル成長させた2～40 nm厚の不規則構造の単結晶FePt膜上に，2 nm厚のMgOキャップ層を形成した．その後，600 ℃の熱処理を施すことにより$L1_0$構造への規則化を促進させた．FePt膜に対して約10%の格子ミスマッチを持つ基板およびキャップ層で挟み込んだ状態で規則化させることにより，面内方向の引っ張り応力を促進させ，c軸方向の精密制御を試みた．

実験方法　膜形成には超高真空RFマグネトロンスパッタリング装置を用いた．構造評価にはRHEEDおよびXRD（$2\theta/\omega$スキャン面外および$2\theta\chi/\varphi$スキャン面内測定）を用いた．表面形態観察にはAFM，磁化曲線測定にはVSMを用いた．なお，$L1_0$（規則）と$A1$（不規則）構造の基本結晶軸の方位は異なるが，本研究では，$A1$構造の表記法を用いて，$L1_0$構造を示している．

実験結果　$L1_0$構造の(001)および(100)面の構造因子は，それぞれ，$S(f_{Pt}-f_{Fe})$および0で表せる（S：規則度，f：原子散乱因子）．そのため，面外XRDパターンにおいて，(001)超格子反射が観察されている場合，$L1_0$(001)結晶が形成されていることを示し，面内パターンにおいても超格子反射が確認される場合，$L1_0$(100)結晶も混在していることを意味する．熱処理後のキャップ層無しおよび有りの10 nm厚のFePt膜のXRDパターンを，それぞれ，Fig. 1(a)および(b)に示す．キャップ層無しの膜に対しては，面外パターンだけでなく，面内パターンにおいても，強度は弱いが(001)超格子反射が観察されており，膜中に$L1_0$(100)結晶が混在していることが分かる．一方，キャップ層有りの膜に対しては，面外パターンにおいてのみ，超格子反射が確認でき，$L1_0$(001)結晶のみから構成されていることが分かる．また，キャップ層無しおよび有りの膜の格子定数比および規則度（c/a, S）は，それぞれ，（0.9781, 0.58）および（0.9599, 0.82）であった．キャップ層を設けることにより，格子変形が促進され，規則度も向上した．これらの膜の磁気特性をFig. 2に示す．いずれの膜も，垂直磁気異方性を示しているが，キャップ層有りの膜の面内磁化曲線の保磁力は，非常に小さいことが分かる．これは，$L1_0$(001)結晶からのみ構成され，更により高い規則度を持つFePt膜の磁気特性を反映した結果であると考えられる．当日は，FePt膜厚を変化させた場合の格子歪，c軸方位，規則度，および，磁気特性の関係について，詳細に議論する．

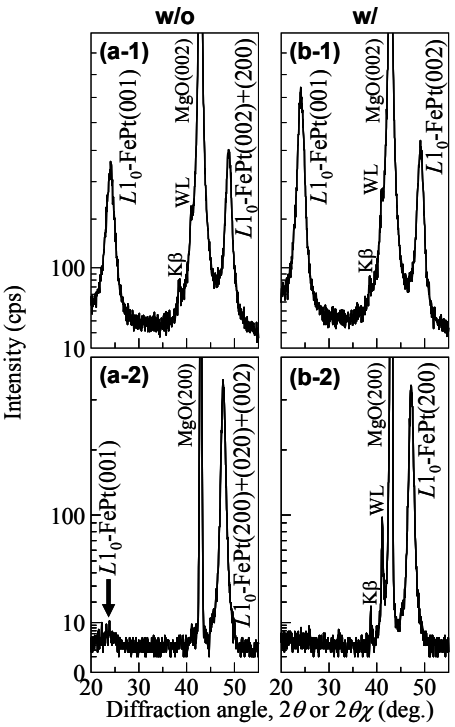

Fig. 1　(a-1, b-1) Out-of-plane and (a-2, b-2) in-plane XRD patterns of 10-nm-thick FePt films (a) without and (b) with cap layers after annealing at 600 °C. The scattering vector of in-plane XRD is parallel to MgO[200].

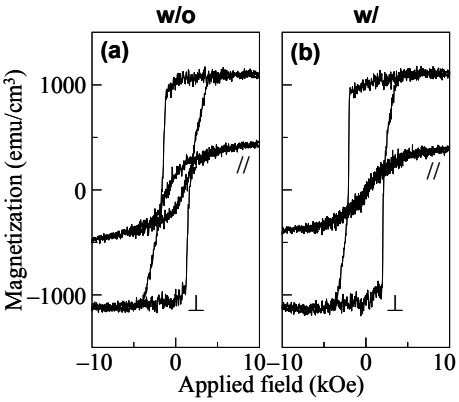

Fig. 2　Magnetization curves measured for 10-nm-thick FePt films (a) without and (b) with cap layers after annealing at 600 °C.

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Large grain size of Cr seed layer deposited on CrTi amorphous layer for future high K_u FePt-C granular medium

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Introduction Hard disk drive industry has been evolved through the increase of the areal density before the emergence of the trilemma. Thermally assisted magnetic recording has been introduced as one of the prospective technologies for future recording media by using the $L1_0$ ordered FePt with high magnetocrystalline anisotropy (K_u) of 7×10^7 erg/cc[1)]. In order to use the $L1_0$ ordered FePt as the recording media, it is necessary to fabricate the granular type such as FePt-C medium whose magnetic grains are completely separated each other by the C boundaries. However, it is reported that the reduction of the signal to noise ratio (SNR) arise from the magnetic anisotropy field variation, which is caused by the angular distribution of the (002) sheet texture in the FePt medium[2)]. Generally, the sheet texture of the crystalline film is known to be attributed to the epitaxial growth on crystalline underlayers. In this report, we proposed a new concept of the layered structure for reducing the angular distribution of sheet texture in FePt-C medium.

Concept of the layered structure The concept is to realize a highly oriented sheet texture in seed layer by promoting Frank-van der Merwe growth mode (layer by layer growth)[3)]. According to the initial state of the sputtering process when the sputtered atoms adhere to the surface of the crystalline film in liquid state, growth mechanism is determined by the wettability of the atoms. If the wettability is large enough, the atoms maximize the contact surface on the film that tremendously induces the epitaxial growth. Consequently, formation of the layer by layer fashion on the crystalline film namely, large grain, leads to the highly oriented sheet texture. Figure 1 shows a schematic of the new concept of the layered structure. The structure is consists of the magnetic layer (ML)/barrier layer (BL)/seed layer (SL)/texture inducing layer (TIL). Main function of each layer is as follows: TIL as determining the grain size, SL as contributing the highly oriented (002) sheet texture, BF as preventing the atomic diffusion between ML and SL by using MgO, and ML as FePt-C magnetic recording medium with the highly (002) sheet texture. The main issue in here, is to find out suitable materials for TIL and SL. M. Mikami reported that the grain size of the recording medium is controlled by Ni-based amorphous layers under the oxygen process[4)], suggesting that the grain size of seed layer would be changed depending on the amorphous material. Since the Cr alloy material has small lattice misft, the material can be used as seed layer. Accordingly, both amorphous and Cr-alloy can be applied as TIL and SL. To obtain (002) texture of SL, it is necessary to change its surface energy. In addition Cr alloy material is widely used in FePt medium because of the small lattice misft with the MgO. To obtain (002) texture of SL, it is necessary to change its surface energy. Here, we tried to investigate the various samples. We applied substrate heating process to fabricate the large grain seed layer by using above TIL and SL materials.

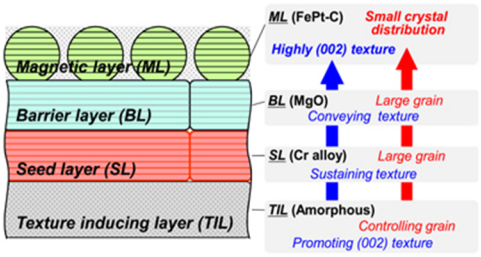

Fig. 1 Concept of the layered structure for reducing the (002) texture distribution of FePt-C medium.

Experimental results In order to fabricate the large grain seed layer, one example structure, Cr (SL) and CrTi (TIL). Stacking structure is following Cr(10) / CrTi(50) / Sub. Figure 2 shows the grain diameter (GD) and integrated intensity of sheet texture for Cr seed layers (I_{002}) as a function of the substrate temperature (T_{sub}) represented as red circle and black square. Here, GD was evaluated from Scherrer equation with (110) diffraction appeared in-plane XRD. As shown in the graph GD increases from 8 to 12 nm as increasing T_{sub} from 200 to 475 °C, and then decreases 12 to 11.5 nm as further increasing T_{sub}. Similar tendency was observed in I_{002} graph. Increase of I_{002} from 10 to 80 cps as increasing T_{sub} from 200 to 520 °C, and then decreases 80 to 25 cps as further increasing T_{sub}. These results suggested that the I_{002} is affected by the grain size of seed layer. In summary, we demonstrated increase of the grain size of Cr seed layer deposited on CrTi amorphous layer under the heat treatment.

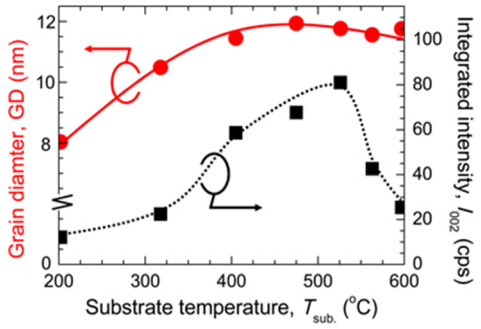

Fig. 2 Grain diameter (GD) and integrated intensity of sheet texture for Cr seed layers (I_{002}) with respect to the substrate temperature (T_{sub}).

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多結晶 MgO 下地層による FePt-C の微細構造制御

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Microstructure control of FePt-C by poly crystalline MgO underlayer

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はじめに

次世代超高密度磁気記録方式として提案されている熱アシスト方式用記録媒体として、MgO 下地上に $L1_0$ 規則構造をもつ FePt-X ナノグラニュラー膜の研究が盛んに行われている。以前、我々はガラス基板に成膜した多結晶 MgO 下地上に成膜した FePt-C 層が、粒子サイズとサイズ分布が小さく、磁気特性に優れた良好なグラニュラー膜を形成することを報告した [1,2]。FePt-C 膜の粒子サイズは、成膜後のアニール温度 [1]や C の体積分率 [2]によって制御可能であるが、この系では膜厚が 6nm を超えると膜成長方向にも分離してしまい [3]、柱状成長が難しいという欠点がある。我々は、FePt-C 層厚 10nm 以上で、10nm 以下の FePt 粒子径とアスペクト比 1.5 以上の柱状成長の両立を目指し、成膜条件の検討を行ってきた。今回、 MgO 下地の成膜条件が FePt-C の柱状成長に寄与することを新たに見出したので、その検討内容について報告する。

実験方法

製膜は超高真空マグネトロンスパッタ装置を用いて行った。まず、ガラス基板上に NiTa(100nm)を製膜し、続いて、MgO(10nm)を 5～39mTorr Ar ガス雰囲気化、室温にて製膜した。さらに、それらの MgO 下地上に FePt-C 膜を、3.6mTorr Ar ガス雰囲気下、基板温度 600℃、Fe、Pt、C の 3 元同時スパッタによって製膜した。試料の構造は、X 線回折にて、平面及び断面構造形態は透過型高分解能電子顕微鏡により評価した。また、磁気特性は超伝導量子干渉振動試料型磁力計にて行った。

実験結果

図 1 に MgO(Ar 5mTorr)/FePt-C(12nm) (a)と MgO(Ar 39mTorr)/FePt-C(12nm) (b)の微細構造を示す。FePt-C 層厚が 5nm 以下の場合は、MgO 成膜時の Ar ガス圧による微細構造に差は見られなかったが、FePt-C 層厚が 12nm と厚くなると、MgO を 39mTorr と高圧下で成膜した場合に、柱状成長し易いことが分かった。多結晶 MgO 膜の粒子サイズを調査したところ、Ar 5mTorr では約 13nm、Ar 39mTorr では約 10nm と大幅に小さくなっていることが判明した。Ar 39mTorr 雰囲気化で成膜した MgO 上では、MgO の粒界がより多く存在することで、FePt 粒子の面内方向への成長や粒子の合体をより抑制しているものと考えている。

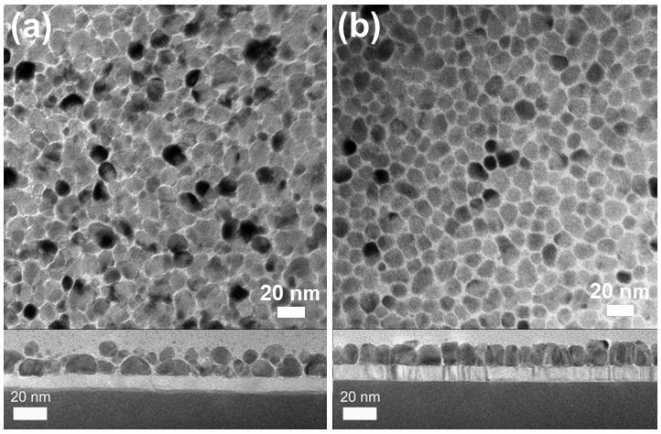

Fig.1　The in-plane and cross sectional TEM bright field image of FePt-C(12nm) on MgO deposited at Ar 5mTorr(a) and 39mTorr(b)

講演では、FePt のさらなる小粒径化検討や、垂直方向への FePt 粒子の配向性についても議論する。

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Effect of MgO seed layer misorientation on the texture and magnetic property of FePt-C granular film

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FePt-C based granular films with $L1_0$-ordered FePt nanoparticles have been considered as the most promising candidate for heat-assisted magnetic recording (HAMR) media for the recording density exceeding 1 Tbit/in^2. For the practical application of $L1_0$-FePt films as HAMR media, the thin-film structure has to be optimized with excellent alignment of the c-axis normal to the film plane and small grain size of less than 6 nm with less than 10% size distribution. In our previous work [1)], we demonstrated well-isolated uniform microstructure with high $\mu_0 H_c$ in FePt-C granular film on polycrystalline MgO underlayer. However, there are some remaining issues for the practical application, i.e. large switching field distribution and large in-plane hysteresis in the magnetization curve which could be an origin of poor SNR[2)]. In this work, we investigated the origin of the large in-plane component in the magnetization curve by comparing the FePt-C granular films deposited on a MgO single-crystalline substrate and a poly-crystalline seed layer.

10 nm thick FePt-*28vol.%* C were deposited by co-sputtering Fe, Pt and C at 600°C under 0.48Pa Ar on MgO (100) substrate (Sample A) and glass/ NiTa(100nm)/ MgO(10nm) stacking (Sample B), respectively. MgO seed layer was RF sputter deposited on the amorphous NiTa layer under an Ar pressure of 5.2 Pa at room temperature (RT) using a MgO target. The orientation and phase mapping experiments were conducted on a FEI Tecnai F20 TEM with a field emission gun and an accelerating voltage of 200 kV using the ASTAR™ (NanoMEGAS, Brussels, Belgium) system.

Figure 1 shows the in-plane and out-of-plane magnetization curves of Sample A and Sample B. $\mu_0 H_c$ of Sample A and Sample B are 4.3 and 3.7 T, respectively. Although both of the films show strong perpendicular anisotropy, compared with Sample A, Sample B presents a loop with smaller coercivity, broaden of switching field distribution and in-plane minor loop. By comparison of the orientation maps in Fig.2, one can see that the MgO seed layer introduces significant misorientation of the (001) texture along the normal direction and it should mainly responsible to the decay of magnetic properties in Sample B. With further ASTAR analysis, we found that about 23% of FePt grains in Sample B have 45° or even 90° miorientation from the [001] direction. They are mainly originate from misorientated MgO seed layer grains, MgO surface roughness and MgO grain boundaries which were confirmed by cross-sectional HRTEM observation.

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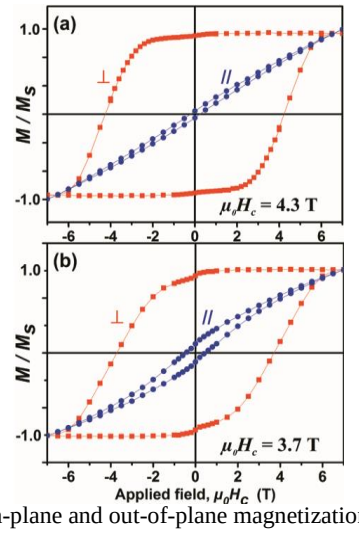

Fig.1 In-plane and out-of-plane magnetization curves of (a) Sample A and (b) Sample B.

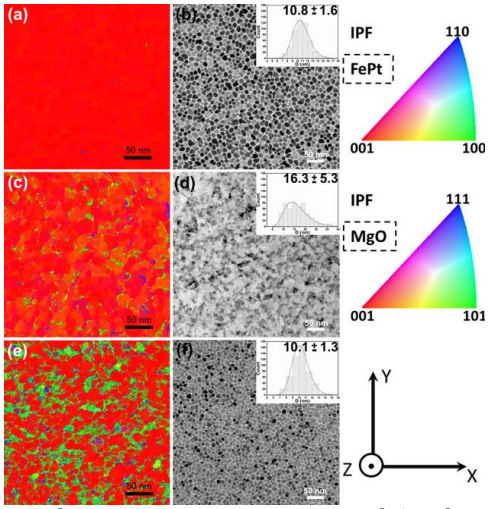

Fig.2 In plane orientation mapping (a, c and e) and virtual bright field TEM images (b, d and f) of Sample A (a & b); polycrystalline MgO (c & d) and Sample B (e & f).

Mechanism of coercivity enhancement by Ag addition in FePt-C granular media for heat assisted magnetic recording

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FePt granular thin films are considered to be one of the suitable candidates for ultrahigh density perpendicular recording media beyond 1 Tbits/in^2 because of the high magnetocrystalline anisotropy of the $L1_0$-FePt phase (~7×10^7 erg/cc). We previously reported highly $L1_0$-ordered FePtAg–C nanogranular film as a potential high-density storage medium for heat assisted magnetic recording (HAMR) [1,2]. Although the addition of Ag is known to increase the H_c, the mechanism of H_c enhancement is not clarified yet. In this paper we investigated the Ag distribution in FePtAg-C granular films by aberration-corrected scanning transmission electron microscope-energy dispersive X-ray spectroscopy (STEM-EDS) and X-ray absorption fine structure (XAFS).

$(FePt)_{0.9}Ag_{0.1}$-28vol%C (FePtAg-C)and FePt-28vol%C (FePt-C) granular films were deposited by co-sputtering Fe, Pt, Ag and C targets on a pre-deposited glass/*a*-NiTa/MgO substrates at 600°C. In this work, we employed a new alternating layer deposition technique to control the grain growth in the perpendicular direction suppressing the growth of the randomly oriented spherical particles on the [001] textured FePt granular layer. The film stack was glass/*a*-NiTa(60nm)/MgO(15nm)/[$(FePt)_{0.9}Ag_{0.1}$-48vol%C or FePt- 48vol%C(0.25)/FePt(0.15)]$_{25}$ deposited on a heat resistant glass substrate [3]. Figure 1 shows the in-plane and out-of-plane magnetization curves of (a) FePt-C and (b) FePtAg-C films. Both of the films show strong perpendicular anisotropy due to the strong c-axis texture. Coercivity H_c of FePt-C and FePtAg-C films are 3.0 and 3.9 T, respectively. TEM bright-field images (not shown here) indicated that FePt-C and FePtAg-C show well-isolated uniform microstructure with average particle sizes are 10.5 nm and 10.0 nm, respectively. The higher H_c in the FePtAg-C film in spite of the similar microstructure is attributed to the higher degree of $L1_0$ ordering. Figure 2 shows STEM-EDS elemental maps of (a) Fe, (b) Pt, (c) Ag and (d) a combined map of Fe and Ag in the FePtAg-C film. The elemental mapping shows that FePt particles are enveloped by Ag-rich shells. EXAFS results showed that Ag shells have fcc-like structure. From these details analysis, we can conclude that Ag is rejected from the core of FePt grains during the deposition, forming Ag-enriched shell surrounding $L1_0$-ordered FePt grains. Since Ag has no solubility in both Fe and Pt, the rejection of Ag induces atomic diffusions thereby enhancing the kinetics of the $L1_0$-order in the FePt grains [3].

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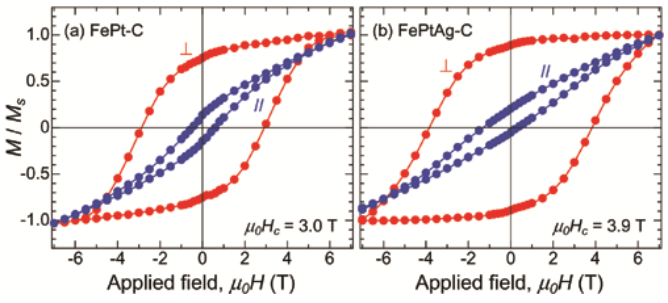

Figure1. In-plane and out-of-plane magnetization curves of (a) FePt-C and (b) FePtAg-C granular films.

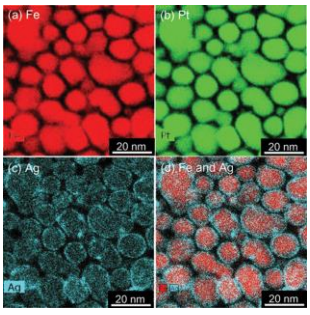

Figure 2. EDS mappings of (a) Fe, (b) Pt, (c) Ag and (d) Fe and Ag of FePtAg-C granular film.