Studies on FePt-based Hard Magnetic Thin Films and Their Application in Patterned Recording Media

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授　与　学　位	博士（工学）
学位授与年月日	平成２２年　３月２５日
学位授与の根拠法規	学位規則第４条第１項
研究科，専攻の名称	東北大学大学院工学研究科（博士課程）知能デバイス材料学専攻
学位論文題目	Studies on FePt-based Hard Magnetic Thin Films and Their Application in Patterned Recording Media　（鉄白金基硬質磁性薄膜とパターンドメディアへの応用に関する研究）
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論文内容要旨

Recently the ferromagnetic material *L1₀* FePt has gained significant attraction as a candidate for ultra high density magnetic recording due to its very high anisotropy constant ($K_u \sim 7.0\times10^7$ ergs/cm^3) and its high corrosion resistance. To control the magnetic properties of $L1_0$ FePt several aspects such as deposition conditions, substrate and buffer layer have to be taken into account. In order to apply FePt thin film for recording media, it needs to be grown along preferred orientation, and such films were usually obtained on a single crystal substrate. One of the major problems to use FePt in recording media is its extremely high coercivity that results in a write-ability problem, meaning with the present write head technologies it is hard to write bits on such a media. Additionally signal to noise ratio is also reported to be high. These problems can be solved by using a soft magnetic under layer and by patterning the individual bits of the media. Such a media is known as bit-patterned exchange coupled perpendicular recording media, and is expected to be thermally stable, have high signal to noise ratio. The mass-production of such a media in the recording density range of ~ Tbits/$inch^2$ is challenging because it requires a bit size of ~ 27 nm, which is extremely difficult and time consuming even with very modern lithographic techniques. Moreover growth of $L1_0$ FePt in preferred crystallographic orientation on a soft magnetic under layer is also challenging. In view of above problems, this thesis aim for finding a suitable combination of soft magnetic under layer on which $L1_0$ FePt can be grown in preferred orientation. Moreover such a material combination can allow pattern-ability to nanoscale dimensions at lower processing cost.

This thesis starts with the growth of FePt films on a single crystal substrate, and understands the magnetic properties which are usually reported in the literature. After that growth of FePt on non-epitaxial substrate is reported. In view of magnetic exchange coupling among the soft magnetic under layer and hard magnetic FePt, exchange coupling is studied by fabricating a nano-composite exchange coupled magnet in FePt-based thin films. Finally metallic glass thin films which have very good nano-fabrication abilities were used to grow preferred oriented thin films of *L1₀* FePt.

The results demonstrate, a high H_c ~ 33 kOe at 300K and 42 kOe at 5K for a ~ 20 nm thick $Fe_{51}Pt_{49}$ film deposited on MgO substrate at T_s ~550 °C. The surface morphology plays an important role for deciding the H_c. High H_c is only obtained for island like thin films in which islands are well separated from each other. The continuous films show very low H_c. The FePt films grown in the present study have a large value of magnetocrystalline anisotropy $K_u \sim 6.11 \times 10^7$ erg/cc, which is close to the theoretical value (7.0×10^7 erg/cc) reported for this system. The combination of H_c and K_u obtained in the present study are usually reported for the FePt thin films deposited at much higher temperature (~ 700 °C). Effects of two different crystallographic orientations i.e. (200) and (002) on the magnetic and structural properties of the films were studied. Anomalous temperature variation of H_c was observed at low temperatures, and the origin for this behavior was identified. The strong inter-particle interaction is the cause of anomalous temperature variation of H_c observed in the present case. The basic understanding of the magnetization behavior of single axis oriented (c-axis) and two axis (c- and a- axis) oriented FePt will be useful for understanding the magnetization behavior of poly-axis oriented film.

$Fe_{51}Pt_{49}$ thin films were deposited on SiO_2/Si substrate by dc magnetron co-sputtering technique. Effects of processing conditions on the structural and magnetic properties were studied. Polycrystalline *$L1_0$* FePt films with high degree of chemical ordering (0.98) were obtained at substrate temperature < 550°C. A maximum H_c ~ 15kOe was obtained at T_s = 500°C for a 20 nm thick polycrystalline *$L1_0$* FePt film deposited on SiO_2/Si substrate. Hard magnetic *$L1_0$* - $Fe_{50}Pt_{50}$ thin films (H_c ~ 8.2 kOe) can also be deposited on a silicon substrate with a very thin native SiO_2 layer at T_s up to 400 °C. Increasing T_s > 400 °C can result in soft magnetism (H_c < 200 Oe), by formation of various iron and platinum silicide-phases triggered by the diffusion of Fe and Pt with Si. If the surface of the substrate has some dimples, they can act as a nucleation sites for the growth of submicron-sized flower like patterns. Flowers are mainly made up of antiferromagnetic $FePt_3$ phase, and are surrounded by the ferromagnetic iron-silicides and non-magnetic platinum-silicides. Flower like nanostructures are not reported for FePt thin films. The ability to grow site specific antiferromagnetic micro/nano-scale structures in a ferromagnetic matrix on silicon substrate seems to be promising for understanding the basic physics of FM/AF interface and also for the development of future spintronics devices.

Exchange-coupling behavior was studied by fabricating a nano-composite FePtB thick film magnet. A mixture of soft Fe_xB (x = 2, 3), and $L1_0$-FePt hard magnetic phases with grain sizes ranging in the nano-meters was obtained in the FePtB films deposited at higher power and/or at $T_s > 300$ °C. The hard magnetic properties of these nano-composite films are dependent on the grain sizes and the crystalline quality of the hard and soft magnetic phases, which can be changed by the processing conditions. Under optimized sputtering conditions (Power = 200 W, Pressure = 0.5 Pa, $T_s = 370$ °C), a 1.4 μm thick FePtB film exhibits an in-plane $H_c \sim 7.6$ kOe and $(BH)_{max} \sim 12$ MGOe. In out of plane the $H_c = 7.1$ kOe and $(BH)_{max} = 16.7$ MGOe. The high H_c and the high $(BH)_{max}$ of thick FePtB films reported in the present study are due to the existence of strong exchange coupling among the nano-sized soft and hard magnetic phases, which are uniformly distributed throughout the film. These films also exhibit a very good spring back action, with a high recoil rate. Fe-Pt-B thin films were also deposited by PLD technique on silicon and glass substrates. The maximum T_s for deposition of FePtB films on silicon substrate was 450 °C. This was limited by the diffusion of film with the substrate. At this temperature a high value of inplane $H_c \sim 6.0$ kOe was obtained. The hysteresis loop shape was similar to a two phase-like magnet and is due to the weak exchange coupling among the soft (iron-boride) and hard ($L1_0$-FePt) magnetic phases. FePtB films deposited on glass substrate at $T_s \sim 525$ °C exhibit good hard magnetic properties [$H_c \sim 7.7$ kOe; $M_r/M_s \sim 0.9$].

The good hard magnetic properties of thick FePtB films, as demonstrated in the present study using sputtering as well as PLD are promising for the fabrication of micro or nano-sized permanent magnets and the various magnetic MEMS components. The basic understanding gained about the nature of exchange-coupling in soft and hard magnetic phase is valuable for tailoring the magnetic properties of soft and hard magnetic multilayered structure useful for the fabrication exchange-couple bit patterned media.

Non-magnetic and magnetic metallic glass thin films were deposited by magnetron sputtering. Our results demonstrated very good surface and magnetic properties of metallic glass thin films. Metallic glass thin films showed excellent micro/nano-fabrication abilities on a large area using a low cost mass-production technique such as imprinting. A feature dimension of ~ 34 nm can be obtained easily on a large area. This is not the limit for these materials but it is limited by our experimental technique. Excellent nano-fabrication ability of metallic glass thin films gives the possibility of mass production of bits at lower cost. $L1_0$ FePt thin films can be grown preferentially along (111) crystallographic direction on a soft ferromagnetic metallic glass thin film at lower substrate temperature (~ 400 °C). The switching field of FePt can be easily tailored to any desirable value below 8 kOe, just by changing the thickness of soft magnetic metallic glass under layer. The magnetic easy axis of preferred oriented $L1_0$ FePt grown on soft magnetic metallic glass is tilted about 25 ^{0} away from the out of plane direction. At this angle exchange coupling among the soft magnetic metallic glass under layer and $L1_0$ FePt hard magnetic layer is very strong. This is believed to be very useful for the fabrication of tilted bit-patterned exchange coupled perpendicular recording media.

Results of this thesis suggest the possibility of fabrication of tilted bit-patterned exchange coupled perpendicular recording media at lower cost.

論文審査結果の要旨

$L1_0$-FePt 規則構造は極めて高い結晶磁気異方性を有することから、超高記録密度媒体の素材として有望視されている。しかしながら、FePt 薄膜の応用を見据えた場合、FePt 相を強配向させることが必須であり、従来のほとんどの研究では単結晶基板へ製膜する高コストな手法が用いられている。また、FePt 相の極めて高い保磁力によって、現行技術では FePt 基記録媒体への磁気ヘッドによる書き込みが困難であることが報告されている。さらに、テラビット級の高記録密度を達成するためにはビットサイズを 27nm 以下に制限する必要があり、現行のリソグラフィー技術では極めて困難であると同時に莫大な時間がかかるばかりか、軟磁性下地層への $L1_0$ 相の配向制御についても課題が残されている。

本研究では、垂直磁気記録用パターンドメディアの作製技術で現在直面している諸問題を解決するため、FePt 相の配向制御を可能とする軟磁性下地層の組み合わせの検証と、作製した薄膜における低コストでのナノパターニングの可能性の検証を目的としたもので全 7 章からなる。

第 1 章は諸言であり、背景と研究目的について述べている。

第 2 章では実験方法について解説している。

第 3 章ではスパッタ法で MgO(111)基板上に作製した島状構造を有する FePt 薄膜における硬磁気特性と試料厚さの関係について調査し、FePt 薄膜の保磁力は試料厚さと表面性状に強く依存することを見出した。特に、厚さ 20nm の薄膜は室温で 33kOe の高保磁力を示すこと、ならびに本試料の結晶磁気異方性定数は約 6.1×10^7erg/cm^3 であり理論値の 90%に近い値が得られることを見出した。

第 4 章では SiO_2/Si 基板への FePt 相の製膜傾向について調査し、基板表面層の SiO_2 がガラス質であるにも関わらず、厚さ 20nm の薄膜において 0.98 の高い規則度が得られ、15kOe の高保磁力が得られることを見出した。

第 5 章では FePt 硬磁性相と Fe-B 軟磁性相の複相薄膜の作製を行い、厚さ 1.4 μm の薄膜では、均一に分散したナノサイズの軟磁性相と硬磁性相間の強い交換結合によって、面内方向と面垂直方向のそれぞれで高保磁力と高磁気エネルギー積を示すことを見出した。

第 6 章では軟磁性金属ガラスを下地層とするためのパターニングを行い、使用装置の性能限界である 34nm 幅のドット加工が容易に行えることを示し、量産化プロセスで課題となる軟磁性下地層を低コストで作製できる可能性を示した。また、微細加工の施された軟磁性金属ガラスに FePt 相を製膜し、面垂直方向から傾斜した $L1_0$ 配向組織が得られることと、軟磁性下地層の厚さを調節することで 8kOe 以下の保磁力を任意に制御できることを見出した。

第 7 章は総括である。

以上要するに、本研究は FePt 硬磁性薄膜の学術的発展とパターンドメディアへの応用に際する基礎に寄与するところが少なくない。

よって、本論文は博士（工学）の学位論文として合格と認める。