Evolution of Growth Mode in Fe Thin Films with Different Grain Sizes under Strong Magnetic Fields and Its Influence on Magnetic Properties (Postprint)

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Abstract

Using physical vapor deposition under a strong magnetic field, nanocrystalline Fe films with progressively decreasing grain sizes were obtained by increasing the evaporation source temperature, and the influence of the strong magnetic field on the growth and magnetic properties of Fe films with different grain sizes was investigated. The results show that when the evaporation source temperature was 1440 °C, the Fe film exhibited fine grains, and the strong magnetic field transformed the film growth from a layered mode to a columnar mode, effectively reducing film defects. When the evaporation source temperatures were 1400 and 1350 °C, the Fe films had relatively coarse grains; the strong magnetic field could not alter their columnar growth mode, but increased the column width. The strong magnetic field increased the average grain size of the Fe films as well as the particle size (composed of grains), and reduced the surface roughness of the films. With decreasing grain size, the enhancing effect of the strong magnetic field on the coercivity, saturation magnetization, and remanence ratio of the Fe films became more pronounced.

Full Text

Growth Mode Evolution and Subsequent Magnetic Properties of Fe Films with Different Grain Sizes Under a High Magnetic Field

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Abstract

In order to increase the magnetic properties and realize essential applications in magnetic recording and spintronics devices, controlling the growth mode and grain size of Fe films is of significant importance. In this work, the effects of a high magnetic field (HMF) on the growth and magnetic properties of Fe thin films with different grain sizes prepared by physical vapor deposition were investigated. Decreased grain sizes were obtained by increasing the evaporation source temperatures. It was found that when the evaporation source temperature was 1440 ℃, the film grains were fine and the HMF changed the growth mode from layered to columnar, effectively reducing film defects. When the evaporation source temperatures were 1400 and 1350 ℃, the film grains were relatively coarse. The HMF did not alter their columnar growth mode but increased the column width. Additionally, the HMF increased the average grain size and particle size (composed of grains) of Fe films while reducing film surface roughness. As grain size decreased, the ability of the HMF to enhance the coercivity, saturation magnetization, and squareness ratio of Fe films was strengthened.

KEY WORDS high magnetic field, Fe film, nanocrystalline, columnar growth, magnetic properties

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Introduction

Iron-based nanocrystalline soft magnetic thin films exhibit high saturation magnetization and low coercivity, making them suitable for high-density magnetic recording head materials [**?**]. Additionally, Fe film/semiconductor systems are increasingly applied in magnetoelectronic and spintronic devices, serving as spin filters, spin injectors in semiconductor devices [**?**], and fundamental components for electrical and electro-optical heterostructure devices [**?**]. Consequently, Fe nanocrystalline thin films have attracted extensive attention in recent years [**?**, **?**, **?**].

The microstructure of thin films, including grain size and growth mode, significantly influences magnetic properties such as saturation magnetization, coer-

civity, and magnetic anisotropy. $YNi_xMn_{1-x}O_3$ films with large and uniformly distributed grain sizes exhibit higher ferromagnetic transition temperatures [?]. Size-driven phase transitions in $BaTiO_3$ films demonstrate a critical grain size for ferroelectric properties [?]. Moreover, conventionally grown carbon nanotubes show six times higher resistivity, thermal conductivity, and tensile strength than needle-like grown ones [?]. For Fe films, researchers have improved film quality and performance by altering growth modes and grain sizes to meet application requirements. Kima et al. [?] transitioned from three-dimensional island growth to columnar growth by increasing Fe film thickness. He et al. [?] achieved a transition from perpendicular to parallel magnetic anisotropy by changing growth modes through thickness variation. Wang et al. [?] enhanced Fe film grain size through annealing, significantly affecting film coercivity. Previous results indicate that changing Fe film growth modes can alter magnetic domain structures and achieve magnetic anisotropy transitions, while modifying grain size and distribution can change grain boundary density and intergranular exchange coupling, thereby tuning coercivity and saturation magnetization.

Therefore, controlling Fe film growth modes and grain sizes during deposition is crucial for enhancing magnetic properties and enabling practical applications. High magnetic fields possess non-contact characteristics, enhanced Lorentz force, magnetization force, and magnetic energy, playing significant roles in both bulk materials [?] and thin film preparation [?]. High magnetic fields can achieve grain alignment [?], oriented growth [?], phase transformation [?], and property improvement [?] without altering preparation conditions or environment, providing possibilities for controlling Fe film grain size and growth mode. Currently, high magnetic field applications in Fe film growth have focused on electrodeposition methods, primarily investigating effects on crystal structure and surface morphology [?, ?, ?, ?, ?]. Matsushima et al. [?, ?] applied a high magnetic field parallel to the substrate during Fe film electrodeposition, finding that surface morphology changed from angular to rectangular shapes aligned along the magnetic field direction. Koza et al. [?] reported that high magnetic fields affected nucleation processes and surface morphology during Fe film electrodeposition. Additionally, high magnetic fields controlled Fe film texture [?, ?], orienting the (110) plane along the field direction [?].

However, these studies were limited to effects on crystal structure and surface morphology, with unclear impacts on growth modes and resulting magnetic properties. Moreover, the electrodeposition method used in these experiments caused high magnetic fields to act indirectly on film growth through magnetoconvection effects in the electrolyte, which is not conducive to finely observing high magnetic field effects on Fe film grain growth. Therefore, this work employs physical vapor deposition with direct high magnetic field action on deposited atoms. Furthermore, since the magnetic energy produced by high magnetic fields on materials is closely related to their volume and affects crystal orientation, arrangement, and motion states in the magnetic field, this work controls evaporation source temperature to prepare Fe films with different grain sizes, enabling high magnetic field control of Fe film growth modes. The study in-

vestigates high magnetic field effects on growth modes, grain sizes, and surface morphologies of Fe films with different grain sizes, as well as resulting changes in magnetic properties.

Experimental Methods

Fe thin films were prepared using a high magnetic field physical vapor deposition method [**?**]. The evaporation source material consisted of 99.99% pure Fe particles with 1–2 mm diameter. Si (100) substrates were placed at the maximum magnetic field intensity position, with the magnetic field direction perpendicular to the substrate surface. The substrate temperature was maintained at 200 ℃. The base pressure was better than 8.0$×10^{-5}$ Pa, and the working pressure was better than 1.0$×10^{-4}$ Pa. Different grain sizes of Fe films were obtained by varying the evaporation source temperatures of 1350, 1400, and 1440 ℃. To ensure identical film thickness, evaporation times were set to 90, 41, and 20 minutes, respectively. A 6 T magnetic field was applied during Fe film growth, with prepared films designated as 1350 ℃/6 T, 1400 ℃/6 T, and 1440 ℃/6 T films, and compared with those prepared without a magnetic field (designated as 1350 ℃/0 T, 1400 ℃/0 T, and 1440 ℃/0 T films).

Film thickness was analyzed using a Dektak 150 stylus profiler and JEOL JEM-2100F transmission electron microscopy (TEM). Cross-sectional TEM measurements showed Fe film thickness of approximately 60 nm, consistent with stylus profiler results. Crystal structure was analyzed using a DMAX2400 X-ray diffractometer (XRD) with 1° grazing incidence angle in –2 coupled scan mode, using Cu Kα radiation with wavelength $\lambda = 0.154056$ nm. Microstructure was examined by TEM. Surface morphology was analyzed using a Multimode IV atomic force microscope (AFM) and SUPRA-35 scanning electron microscope (SEM). Magnetic hysteresis loops were measured using a Lakeshore 7407 vibrating sample magnetometer (VSM).

Results and Discussion

Figure 1 [Figure 1: see original paper] shows XRD patterns of Fe films prepared under different conditions. The results demonstrate that high magnetic field application did not change the phase composition or preferred orientation of Fe films, which remained polycrystalline bcc structure with (110) preferred orientation. Grain sizes of Fe films under different conditions were calculated using the Scherrer formula [**?**], as shown in Figure 2 [Figure 2: see original paper]. Without magnetic field, increasing evaporation source temperature increased growth rate and gradually decreased film grain size, confirming that different grain sizes could be obtained by varying evaporation source temperature. Additionally, high magnetic field application increased the grain size of Fe films.

Cross-sectional TEM was used to investigate high magnetic field effects on the growth modes of Fe films with different grain sizes, as shown in Figure 3 [Figure 3: see original paper]. All films consisted of Fe, Si, and SiO_2 layers, where the SiO_2 layer was the surface oxide layer of the Si substrate (Figure 3b2). Without magnetic field, increasing evaporation source temperature from 1350 to 1440 ℃ decreased the average grain size of Fe films and caused a distinct growth mode transition: from columnar grains penetrating the entire film thickness at 1350 ℃/0 T (Figures 3a1 and a2), to columnar growth with stacked elongated and spherical grains at 1400 ℃/0 T (Figures 3c1 and c2), and finally to layered growth with fine grains at 1440 ℃/0 T (Figures 3e1 and e2). After applying a high magnetic field, when evaporation source temperatures were 1350 and 1400 ℃, the HMF did not change the columnar growth mode of Fe films but significantly increased column width (Figures 3b1, b2, d1, and d2). Statistical analysis of column widths for each condition (200 columns counted) revealed that column width increased from 5–6 nm for 1400 ℃/0 T films to 7–11 nm for 1400 ℃/6 T films, and from 5–8 nm for 1350 ℃/0 T films to 6–11 nm for 1350 ℃/6 T films. Moreover, at 1400 ℃, the HMF transformed the growth mode from columnar growth with stacked elongated and spherical grains in 0 T films to columnar grains penetrating the entire film thickness. However, at 1440 ℃, the HMF changed the growth mode from layered growth in 0 T films to columnar growth (Figures 3f1 and f2), with column widths in 1440 ℃/6 T films larger than grain diameters in 1440 ℃/0 T films. As shown in the boxed region of Figure 3e1, the higher growth rate in 1440 ℃/0 T films resulted in numerous grain boundaries (white lines) with large spacing, indicating sparse growth regions, large intergranular gaps, and increased disordered structures and defects. After HMF application, grains arranged in a dense and orderly manner (Figures 3f1 and f2). Additionally, the HMF transformed grain shapes from spherical in 1440 ℃/0 T films to columnar in 1440 ℃/6 T films, reducing grain boundary density and thus decreasing defects and disordered structures. The trend of HMF effects on Fe film grain size determined by TEM was consistent with XRD calculations. Furthermore, TEM results indicated that the 200 ℃ substrate temperature created a transition layer in all films, which gradually thinned with increasing evaporation source temperature. When evaporation source temperature was higher (resulting in smaller grain sizes), the HMF could change Fe film growth modes, but as evaporation source temperature decreased (average grain size increased), the HMF effects on Fe film growth diminished.

SEM was used to examine high magnetic field effects on the surface morphology of Fe films with different grain sizes, as shown in Figure 4 [Figure 4: see original paper]. Without magnetic field, increasing evaporation source temperature increased the number of large surface particles and surface roughness (Figures 4a, c, e). This morphology resulted from two factors: first, higher evaporation source temperature increased film growth rate, reaching 3.16 nm/min at 1440 ℃, meaning Fe particles arrived before previous ones could fully relax, hindering surface vacancy and defect filling [**?**]; second, higher temperature caused concentrated atomic emission at the evaporation source, forming droplet emission

[?, ?], with higher temperatures producing larger and more numerous droplets that aggregated as large surface particles. After HMF application, large surface particles decreased significantly and surfaces became smoother at all evaporation source temperatures (Figures 4b, d, f), due to increased Fe particle energy from magnetic energy [?].

To determine HMF effects on Fe film particle size, Figure 5 [Figure 5: see original paper] presents AFM surface morphology and line roughness analysis results, where each peak represents a grain and peak height fluctuations indicate roughness variations. Without magnetic field (Figures 5a, c, e), increasing evaporation source temperature decreased peak numbers in line roughness profiles, indicating gradually increasing average particle size. After HMF application (Figures 5b, d, f), peak numbers decreased compared to 0 T films at different evaporation source temperatures, showing increased particle size (peak numbers decreased from 17 for 1350 ℃/0 T films to 13 for 1440 ℃/6 T films; from 13 for 1400 ℃/0 T films to 10 for 1400 ℃/6 T films; and from 12 for 1440 ℃/0 T films to 8 for 1440 ℃/6 T films). Additionally, after HMF application, peak height fluctuations decreased for all evaporation source temperatures, indicating reduced surface roughness, particularly at 1440 ℃ where large surface particles decreased dramatically and surface roughness dropped from 3.888 nm without magnetic field to 0.486 nm. However, after HMF application, surface roughness showed little variation with increasing evaporation source temperature. Since this study controlled deposition rate through evaporation source temperature to prepare films with different grain sizes (Figure 2), the effects of deposition rate were reflected through grain size examination. After applying a 6 T magnetic field, although evaporation source temperature (deposition rate) increased, all prepared Fe films exhibited columnar growth with similar grain sizes (Figures 2 and 3), meaning grains experienced similar magnetic energies [?], resulting in minimal HMF effects on surface roughness as evaporation source temperature increased.

Magnetic hysteresis loops of Fe films were measured perpendicular and parallel to the film plane using VSM, as shown in Figure 6 [Figure 6: see original paper]. The results show that films did not reach saturation even at perpendicular applied fields of 1.43$×10^6$ A/m, whereas saturation was achieved at parallel applied fields of 1.59$×10^5$ A/m. Perpendicular coercivity ($H_c^{\perp}$ = 1.11$×10^{4–3.69×10}4$ A/m) was significantly higher than parallel coercivity ($H_c^{//}$ = 5.49$×10^{2–2.33×10}3$ A/m), indicating that perpendicular magnetization was more difficult. This hard magnetization direction arises from film thickness effects: since film thickness is much smaller than surface length and width, shape anisotropy creates large demagnetizing factors and fields in the film normal direction, requiring larger applied fields for saturation compared to the parallel direction [?, ?]. As film applications primarily concern magnetic properties in the easy magnetization direction, this work focuses on properties with fields parallel to the film plane.

Figure 7a [Figure 7: see original paper] presents quantitative values of coercivity

(H_c) and saturation magnetization (M_s) for Fe films with parallel applied fields. Without magnetic field, H_c gradually decreased with increasing evaporation source temperature. After HMF application, H_c increased to varying degrees for films with different grain sizes, with a particularly large 126% increase for 1440 ℃/6 T films compared to 1440 ℃/0 T films (from 5.57$×10^2$ to 1.26$×10^3$ A/m). For M_s, HMF enhancement became more pronounced with increasing evaporation source temperature.

Film coercivity is closely related to grain size, following the random anisotropy model [**?**]: H_c D^6, where D represents grain size and p_c, K_r, and A are constants. Although H_c is inversely proportional to M_s, grain size has a more significant influence on coercivity. Without magnetic field, increasing evaporation source temperature decreased Fe film grain size (Figures 2 and 3), thus reducing coercivity. After HMF application, grain sizes in 1350, 1400, and 1440 ℃ films increased compared to corresponding 0 T films, leading to increased coercivity. Particularly at 1440 ℃, HMF transformed grain shapes from fine spherical grains (Figures 3e1 and e2) to larger elongated grains (Figures 3f1 and f2). Additionally, the growth mode change from layered in 0 T films to columnar in 6 T films introduced higher strain in the films (strain perpendicular to film plane calculated from XRD [**?**], shown in Figure 7b). All films exhibited negative strain values, indicating compressive stress, with absolute strain values of 0.30% and 0.61% for 1440 ℃/0 T and 6 T films, respectively. This strain (stress) increased coercivity by enhancing magnetoelastic anisotropy energy, resulting in a substantial H_c increase for 1440 ℃/6 T films compared to 1440 ℃/0 T films.

Saturation magnetization is related to total spin electron number and film microstructure. HMF-induced grain growth reduced grain boundary density and enhanced intergranular exchange coupling, increasing M_s [**?**]. SEM surface morphology (Figure 4) showed that HMF eliminated large surface particles and smoothed surfaces, improving film density and benefiting M_s enhancement. Particularly at 1440 ℃, cross-sectional TEM (Figure 3) revealed that HMF significantly reduced disordered structures in 0 T films and improved film density, leading to pronounced M_s enhancement in 1440 ℃/6 T films.

Figure 7b shows the remanence ratio (M_r/M_s, where M_r is remanent magnetization) for Fe films with parallel applied fields. Without magnetic field, the remanence ratio increased with evaporation source temperature, but increased slowly at 1440 ℃. Stress between substrate and film (thermal and lattice mismatch) affects remanence ratio, with stress reduction improving remanence ratio. As evaporation temperature increased from 1350 to 1440 ℃, film growth mode transitioned from columnar to layered, which should gradually decrease strain (stress) magnitude. However, at 1440 ℃, numerous disordered structures and defects in Fe films (Figures 3e1 and e2) increased strain (stress) (originating from different magnetostriction produced by defects and ordered structures during magnetization [**?**]), resulting in slow remanence ratio increase at high evaporation temperature. After HMF application, remanence ratios increased

for all evaporation source temperatures. Since all films exhibited (110) preferred orientation while Fe' s easy magnetization axis is (100), preferred orientation effects on remanence ratio were excluded. Although HMF changed growth modes for 1440 ℃/0 T films (from layered to columnar grains penetrating the entire film thickness) and grain shapes for 1400 ℃/0 T films (from stacked elongated and spherical grains to columnar grains penetrating the entire film thickness), increasing strain compared to 0 T films, HMF-induced grain size increase in all films enhanced intergranular exchange coupling interactions and reduced random grain orientation, leading to improved remanence ratios.

Conclusions

(1) Without magnetic field, increasing evaporation source temperature produced Fe films with decreasing grain size, gradually transitioning the growth mode from columnar grains penetrating the entire film thickness to layered growth composed of nanoparticles. After HMF application, when Fe film grains were fine (high evaporation temperature: 1440 ℃), the HMF changed growth from layered to columnar, reducing film defects and disordered structures. For coarse-grained Fe films (low evaporation temperatures: 1350 and 1400 ℃), the HMF could not change the growth mode (remaining columnar) but effectively increased column width.

(2) The HMF significantly increased the average grain size and particle size (composed of grains) of Fe films with different grain sizes while reducing film surface roughness, but did not affect phase composition or preferred orientation.

(3) Columnar growth did not change the easy magnetization direction of Fe films (film plane direction). The HMF effectively increased coercivity, saturation magnetization, and remanence ratio of Fe films. As grain size decreased, the HMF' s ability to increase grain size, coercivity, saturation magnetization, and remanence ratio while reducing surface roughness was enhanced.

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