Influences of Process Condition of Magnetron Sputtering on Magnetostrictive Susceptibility of Fe_{2.2}Sm Alloy Film*¹

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Since morphological interface of iron-samarium alloy thin film prepared by direct current magnetron sputtering process is controlled by substrate temperature, sputtering argon gas pressure and residual gas pressure, influences of changes of morphology and its interface on compressive (negative) magnetostrictive susceptibility of Fe_{2.2}Sm alloy films are investigated. Decreasing the pressures of sputtering argon gas enhances the magnetostrictive susceptibility. The high susceptibility is also found under the low pressures of residual (impurity) gas at each substrate temperatures (T_s) from 423 to 523 K. The clear interface cannot be observed in the densely packed amorphous phase, when the high magnetostrictive susceptibility is obtained. Since the decreasing in impurity atoms at the unclear interface easily changes the domain direction and then doesn't prevent to move the magnetic domain wall in the amorphous phase, influences of the residual gas pressure on magnetostrictive susceptibility are explained by not only morphological interface but also its oxidation. [doi:10.2320/matertrans.47.2852]

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1. Introduction

The RFe₂ type cubic Laves phases of rare earth metals (R) and iron (Fe) show giant magnetostriction,¹⁾ as expected to apply for sensor and actuator.^{2,3)} The giant mover strain of Terfenol-D (Tb_{0.3}Dy_{0.7}Fe₂), which is larger than that of commercial piezo-ceramics such as PZT, has been found with low electronic potential driving, high power generation, high responsiveness and wireless operation by magnetic field.⁴⁻⁸⁾ In order to apply tiny acoustic devices, sensors and actuators driven by low intensity of magnetic field, giant magnetostrictive films have been expected.^{9,10)} The expansive and compressive magnetostriction values have been obtained for Fe-Tb and Fe-Sm films, respectively.^{11,12)}

Since the substrate temperature and sputtering pressure of direct current (DC-) magnetron sputtering process are important factors to control the morphology of thin film,¹³⁾ they may affect the magnetostrictive susceptibility. In addition, the residual gas pressure, which may easily controls the interface atomic structure of morphology, should be also one of basic factors to affect the susceptibility. To apply wireless acoustic sensor and tiny actuator with high responsiveness driven by low intensity of magnetic field and low electrical potential, influences of sputtering gas pressure, substrate temperature, residual gas pressure and their combinations of morphology on magnetostrictive susceptibility of Fe_{2.2}Sm alloys prepared by DC-magnetron sputtering process have been investigated.

2. Experimental Procedure

A unimorph structural mover device was driven by giant compressive magnetostrictive Fe_{2.2}Sm alloy film deposited on a (100) plane of silicon wafer. When an applied magnetic

field loaded the magnetostrictive thin film, a bending movement was obtained.

The Fe_{2.2}Sm alloy thin film was prepared by using DC-magnetron sputtering apparatus. The minimum vacuum pressure before argon sputtering and the sputtering pressure of argon gas (5N) were 5.0×10^{-4} Pa and 0.5 Pa, respectively. The mean leak rate of the chamber was about 1.64×10^{-5} Pa·m³/s.

The residual gas pressure, sputtering pressure of argon gas and substrate temperature were varied from 2.0×10^{-5} to 5.0×10^{-2} Pa, from 0.2 to 1.0 Pa and from 323 to 623 K, respectively. To evaluate the sputtering conditions in the present work, quantitatively, the standard sample was prepared under 0.5 Pa of argon sputtering pressure with 5.0×10^{-2} Pa of residual gas pressure at 323 K of substrate temperature.

The sputtering power, deposition time, sputtering distance between target and (100) plane substrate of single silicon crystal were 200 W, 3.6 ks and 90 mm, respectively.

The enhancement of substrate temperature just after sputtering was about 30 K. The film composition was controlled by sheet area ratio of samarium (99.9%) and iron (99.9%) on the Fe-35.0 at%Sm (Fe_{1.9}Sm) alloy plate. Coupling samarium and iron tips on the Fe-35.0 at%Sm plate with diameter of 75 and 1 mm thick assembled a sputtering target for formation of the Fe_{2.2}Sm film.

The atomic ratio of Fe-Sm film was varied with increasing the Ar gas pressure from 0.2 to 1 Pa.¹⁰⁾ Mean thickness of the Fe-Sm films deposited was about 2 μm.

The film composition was analyzed by Energy dispersive X-ray spectroscopy (EDS). The iron and samarium elements in the sample were homogeneously distributed. The film thickness was measured by using a cross sectional image of scanning electron microscope (SEM). Crystalline structures of the prepared film were determined by α -2 θ method of thin film X-ray diffraction (Cu-K α ; X'Part-MRD, PHILIPS), where irradiation angle to sample plane was 1.5 deg.

The magnetostriction ($\Delta\lambda_{||}$) of film was measured by a

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bending cantilever method using a He-Ne LASER under magnetic fields from -1200 to 1200 kA/m and was estimated by a following equation,^{8–10)}

$$\lambda_{\parallel} = d \cdot t_s^2 \cdot E_s(1 + \nu_s)/3t_f \cdot l^2 \cdot E_f(1 - \nu_f). \quad (1)$$

Here, t_s and t_f were the thickness values of silicon substrate and Fe_{2.2}Sm films, respectively. d and l were the bending displacement of the sample and distance of clamp and spot of LASER on film, respectively. The E_s and E_f values, the Young's modulus of silicon substrate and Fe_{2.2}Sm film, were assumed to be 130 GPa¹⁴⁾ and 40 GPa.¹⁴⁾

3. Results

3.1 Influences of residual gas pressure, substrate temperature and sputtering gas pressure on sample structure

The residual gas pressure, which depends on the partial pressure of water gas,⁶⁾ is one of basic factors to prepare films. Figure 1 shows X-ray diffraction patterns of Fe_{2.2}Sm film prepared at 323 K of substrate temperature under 0.5 Pa of sputtering argon gas pressure under each residual gas pressure from 5×10^{-2} to 2×10^{-5} Pa, together with standard X-ray references of ICDD cards and patterns at different substrate temperatures and under each sputtering pressure.

A preferred oriented remarkable crystalline peak of samarium oxide (Sm₂O₃) and/or samarium alloy at 28.5° is observed in Fe_{2.2}Sm alloy films under each residual gas pressure from 5.0×10^{-2} to 5.0×10^{-4} Pa under each sputtering gas pressure (P_s) from 0.5 to 0.8 Pa at each substrate temperature (T_s) from 323 to 623 K. On the other hand, the residual (impurity) gas is mainly composed with the water gas molecules.⁶⁾ Since excess P_s value enhances the formation rate of film, it prevents to grow up the preferred oriented oxide crystalline. As a result, excess P_s value forms the hydroxide, as well as oxide. Thus, the broad peak is found of the sample prepared at 1.0 Pa of P_s (see Fig. 1). Since the volume fraction of samarium oxidation mainly depends on the base (residual gas) pressure, the formations of samarium oxide (Sm₂O₃) crystalline and amorphous hydroxide are explained.

The remarkable peak is not observed in the sample prepared under the lowest residual gas pressure of 2.0×10^{-5} Pa and sputtering gas pressure (P_s) of 0.5 Pa at 323 K of T_s , under the lowest sputtering gas pressure of 0.2 Pa and residual gas pressure (P_r) of 5.0×10^{-4} Pa at 323 K of T_s , and at 523 K of T_s under 5.0×10^{-4} Pa of P_r and 0.5 Pa of P_s , as shown in Fig. 1. Although discontinuous X-ray profile with tiny peaks can be observed, crystalline phases of Fe₂Sm and Fe₃Sm are not remarkably observed in Fe_{2.2}Sm alloy films. Since an amorphous phase, glassy structure with and without nano-crystalline, is formed under the lowest P_r value of 2.0×10^{-5} Pa and the standard P_s value of 0.5 Pa at 323 K of T_s , under the lowest P_s value of 0.2 Pa and the standard P_r value of 5.0×10^{-4} Pa at 323 K of T_s , and under 5.0×10^{-4} Pa of the standard P_r value and 0.5 Pa of the standard P_s value at 523 K of T_s , the broad profiles from 30 to 50° of X-ray diffraction are obtained in Fe_{2.2}Sm alloy thin film (see Fig. 1). X-ray diffraction crystalline peaks are not remarkably detected in the amorphous phase. When crystal

Fig. 1 X-ray diffraction patterns of SmFe_{2.2} film prepared at different substrate temperatures under each sputtering pressure at each residual gas pressure, together with standard X-ray references of ICDD cards.

Fig. 2 Applied magnetic field dependent magnetostriction of $\text{Fe}_{2.2}\text{Sm}$ alloy film prepared under each sputtering pressure at each substrate temperature.

anisotropy and grain boundary prevent to move the domain wall, the magnetostrictive susceptibility of the amorphous phase is probably higher than that of crystalline phase. Since a single phase of amorphous structure is remarkably found in $\text{Fe}_{2.2}\text{Sm}$ alloy films, no barrier prevents to move the domain wall. Therefore, the high magnetostrictive susceptibility is expected for the isotropic ferromagnetic $\text{Fe}_{2.2}\text{Sm}$ alloy.

3.2 Influences of substrate temperature, sputtering gas pressure and residual gas pressure, on magnetostriction

Figure 2 shows applied magnetic field dependent magnetostriction (λ) at each magnetic field (H) of $\text{Fe}_{2.2}\text{Sm}$ alloy film prepared at different substrate temperatures under each sputtering pressure and standard residual pressure ($P_r = 5 \times 10^{-4}$ Pa). Here, the standard residual pressure is conventionally achieved by three days evacuation in our magnetron sputtering process.

The decrease in argon sputtering gas pressure enhances the λ value at each magnetic field under the standard residual gas pressure at the constant substrate temperature ($T_s = 323$ K). The largest value of compressive magnetostriction is obtained under the lowest sputtering gas pressure of 0.2 Pa.

Furthermore, the largest value of compressive magnetostriction is obtained at 573 K of substrate temperatures (T_s) under the standard sputtering gas pressure ($P_s = 0.5$ Pa) and the standard residual gas pressure ($P_r = 5 \times 10^{-4}$ Pa).

To evaluate influences of the residual pressure on magnetostriction, the limited residual pressure ($P_r = 2 \times 10^{-5}$ Pa) is often achieved by two weeks evacuation in our magnetron sputtering process. Figure 3 shows applied magnetic field dependent magnetostriction (λ) at every magnetic field (H) of $\text{Fe}_{2.2}\text{Sm}$ alloy film prepared under each residual gas pressure from 2.0×10^{-5} to 5.0×10^{-2} Pa.

Applied magnetic field magnetizes the $\text{Fe}_{2.2}\text{Sm}$ alloy film and also generates the compressive magnetostriction (λ). Increasing magnetic field decreases the magnetostriction.

Fig. 3 Applied magnetic field dependent magnetostriction of $\text{Fe}_{2.2}\text{Sm}$ alloy film prepared under different residual gas pressures.

Decreasing residual gas pressure enhances the λ value at each magnetic field. The largest value of compressive magnetostriction is 1200 ppm under the lowest residual gas pressure of 2.0×10^{-5} Pa.

4. Discussion

4.1 Morphological discussion

To clarify the morphological discussion, SEM micrograph is observed in Fig. 4, together with schematic diagram of Zone I, Zone T and Zone II of Thornton model.¹⁵⁾ Porous tapered grains separated by voids in Zone I, densely packed fibrous grain in Zone T and columnar grains in Zone II have been explained by the Thornton model.¹⁵⁾ $\text{SmFe}_{2.2}$ alloy thin films have been prepared by DC-magnetron sputtering process under different sputtering argon gas pressures from 0.2 to 1.0 Pa at elevated reduced substrate temperatures (T_s/T_m) from 0.2 to 0.4, where T_s and T_m are substrate temperature and liquidus, respectively.

The porous tapered grain of Zone I, generated at low substrate temperature, is observed in SEM micrograph of Fe-Sm film. Since increasing P_s and P_r decreases the kinetic energy of sputtering particles on collision, the short mean free pass of samarium and iron atoms on depositing surface probably explains the porous grain. In addition, active residual gas atoms often form the molecules of oxide and nitride. Both phenomena prevent to migrate the samarium and iron atoms on depositing surface, resulting in the porous grains in Zone I.

Columnar grain with remarkable interface is found in Zone II ($P_s = 0.5$ Pa, $T_s/T_m = 0.4$) of Thornton model in Fig. 4. Since decreasing P_s increases the kinetic energy of sputtering particles on collision, the long mean free pass of samarium and iron atoms on depositing surface probably corresponds to the formation of the large size columnar grains in Zone II.

On the other hand, the amorphous phase of fine densely packed fibrous grain observed in Zone T of Thornton's Model (see Fig. 4) is found under the sputtering gas pressure from 0.2 to 1.0 Pa at different reduced substrate temperatures

Fig. 4 Schematic diagram of Thornton model¹⁵⁾ applied for magnetron sputtering process and SEM micrograph of Zone I, Zone T and Zone II.

(T_s/T_m) from 0.30 to 0.37 at the standard base (residual gas) pressure of 5.0×10^{-4} Pa. When the residual and argon gas don't impact to sputtering particles under their low pressures, the collision factor is not so large to decrease the kinetic energy of sputtering atoms. Namely, the low pressures of argon and residual gas don't largely decrease the kinetic energy of sputtering particles. Thus, it enhances the mean free pass of the particles, which migrate from landing on the sample surface to meta-stable site of amorphous structure. The atom migration with long mean free pass fills into the meta-stable site, resulting in the amorphous phase of fine densely packed fibrous grain.

In addition, amorphous phase is formed by rapidly depositing rate of DC-magnetron sputtering process, because the subsequent deposited particles rapidly deposit. Its rapid deposition prevents that the sputtering particles migrate the stable sites of the potential curve to form the crystalline. Namely, the densely amorphous formation is explained by the rapid deposition rate as well as the long mean free pass.

Based on the results of Fe-Sm film sample prepared under 0.5 Pa of sputtering argon gas pressure, the morphological boundary between Zone T and Zone II is found at about 0.4 of T_s/T_m , whereas the morphological boundary between Zone I and Zone T is found at 0.3 of T_s/T_m . These results correspond to the Thornton morphological model.¹⁵⁾

When the decreasing argon gas pressure decreases the kinetic energy of sputtering particles because of low probability of collisions between sputtering particles and argon gas, the fine densely packed fibrous grain can be obtained. Namely, we suggest that the P_s decreasing decreases the T_s/T_m value at the morphological boundary between Zone I and Zone T.

4.2 Sputtering gas pressure dependent $|d\lambda/dH|$

To discuss the physical meaning, precisely, the magnetostrictive susceptibility ($|d\lambda/dH|$) is defined as the slope of λ - H curve at each H value in Fig. 2. Figure 5 shows applied magnetic field dependent magnetostrictive susceptibility ($|d\lambda/dH|$) of Fe_{2.2}Sm alloy film prepared under each sputtering gas pressure (P_s) from 0.2 to 1.0 Pa under 5.0×10^{-4} Pa of P_r value at 323 K of T_s . The magnetization decreases the $|d\lambda/dH|$ value. The increase in P_s value also decreases the $|d\lambda/dH|$ value at each magnetic field.

Figure 6 shows compressive magnetostrictive susceptibility ($|d\lambda/dH|_{0.1}$) at 0.1 MA/m against sputtering argon gas pressure (P_s) for Fe_{2.2}Sm alloy film. The decrease in sputtering gas pressure (P_s) increases the $|d\lambda/dH|_{0.1}$ value and also decreases the experimental errors of $|d\lambda/dH|_{0.1}$ values.

When the substrate temperature and residual gas pressure are constants, a linear relationship between logarithmic

Fig. 5 Applied magnetic field dependent magnetostrictive susceptibility of $\text{Fe}_{2.2}\text{Sm}$ alloy film prepared under each sputtering gas pressure from 0.2 to 1.0 Pa at 5.0×10^{-4} Pa of standard residual gas pressure at 323 K of substrate temperature.

Fig. 6 Magnetostrictive susceptibility at 0.1 MA/m $|d\lambda/dH|_{0.1}$ against sputtering argon gas pressure (P_{Ar}) for $\text{Fe}_{2.2}\text{Sm}$ alloy film.

sputtering argon gas pressure ($\log(P_s/\text{Pa})$) and logarithmic magnetostrictive susceptibility ($\log |d\lambda/dH|_{0.1}/10^{-9} \text{ m/A}$) is expressed by a following equation.

$$(\log |d\lambda/dH|_{0.1}/10^{-9} \text{ m/A}) = -0.44 \log(P_s/\text{Pa}) + 0.35 \quad (2)$$

A magnetostrictive susceptibility at 0.1 MA/m of magnetic field of $\text{Fe}_{2.2}\text{Sm}$ alloy film increases with decreasing in an argon gas pressure as sputtering gas. The mainly affected impurity is the water molecules rather than the oxygen molecules in the sputtering gas.⁶⁾ Adsorbed water molecules at inner surface of vacuum chamber mainly convert to the water gas. In order to decrease the sputtering pressure (P_s) of our magnetron sputtering apparatus, opening the exhausted gas valve to evacuate enhances the conductance of argon gas

flow. Since the increasing the exhausting rate of gas decreases not only the sputtering pressure (P_s), but also the partial pressure of water gas because of the constant generating rate of water gas from adsorbed water molecules on inner surface of vacuum system. To obtain the high magnetostrictive susceptibility in our sputtering apparatus, the sample should be prepared at the lowest value of argon gas pressure of 0.2 Pa.

When the space distance between target and substrate is about 90 nm, the decreasing P_s decreases the volume fractions of crystal samarium alloy and oxide¹⁶⁾ and then decreases the collision probability of sputtering particles to argon gas atom. As a result, decreasing P_s doesn't prevent to deposit. Namely, the deposition rate becomes high, because decreasing P_s does not largely induce the particle scattering.

Since the residual gas pressure (P_r) is also strongly related to the partial pressure of water gas generated from adsorbed water molecules in vacuum system, the formation of samarium oxide crystalline is not easily formed under the low P_r value. In addition, the decreasing P_r is not easy to form samarium alloy crystalline from amorphous phase, although the oxides act as a catalyze to form the crystalline with low $|d\lambda/dH|$ value. Namely, when the volume fraction of amorphous phase with high $|d\lambda/dH|$ value is high, the magnetostrictive susceptibility of Fe-Sm alloy film becomes high.

In addition to the absorptions of water and oxygen molecules at morphological interface and solid solution of oxygen atom, oxidation occurs at surface and/or morphological interface. Oxygen atoms generally act as obstacles of domain wall at morphological interface, resulting in the low $|d\lambda/dH|$ value. When oxygen atoms don't act as domain wall obstacles, the $|d\lambda/dH|$ value probably becomes high.

The experimental error probably depends on the oxidation volume and its sites at morphological interface. When they can be controlled at low P_s value, the small experimental error is also found (see Fig. 6). Namely, the high magnetostrictive susceptibility with small experimental error is obtained at the low P_s value (0.2 Pa).

4.3 Residual gas pressure dependent $|d\lambda/dH|$

The residual gas pressure, which mainly depends on the partial pressure of water gas and easily controls the interface atomic structure, is one of basic factors to determine the magnetostrictive susceptibility ($|d\lambda/dH|$). Figure 7 shows the magnetic field dependent $|d\lambda/dH|$ value under each residual gas pressure for $\text{Fe}_{2.2}\text{Sm}$ alloy film. The magnetization from 0.1 to 1.2 MA/m decreases the $|d\lambda/dH|$ value under each residual gas pressure. Although increase in residual gas pressure also decreases the $|d\lambda/dH|$ value at each magnetic field, decreasing residual gas pressure from 5×10^{-4} to 2×10^{-5} Pa enhances the compressive magnetostrictive susceptibility at each magnetic field from 0.02 to 0.15 MA/m.

Figure 8 shows the magnetostrictive susceptibility ($|d\lambda/dH|_{0.1}$) at 0.1 MA/m against residual gas pressure for $\text{Fe}_{2.2}\text{Sm}$ alloy film. Decreasing residual gas pressure enhances the compressive magnetostrictive susceptibility at each magnetic field. When the substrate temperature and sputtering gas pressure are constants, a linear relationship between

Fig. 7 Magnetic field dependence of magnetostrictive susceptibility under each residual gas pressure for Fe_{2.2}Sm alloy film.

Fig. 9 Applied magnetic field dependent magnetostrictive susceptibility of Fe_{2.2}Sm alloy film prepared at different substrate temperatures.

Fig. 8 Magnetostrictive susceptibility at 0.1 MA/m $(d\lambda/dH)_{0.1}$ against residual gas pressure for Fe_{2.2}Sm alloy film.

logarithmic residual gas pressure ($\log(P_r/\text{Pa})$) and logarithmic magnetostrictive susceptibility ($\log |d\lambda/dH|_{0.1}/10^{-9} \text{ m/A}$) obtained at 0.1 MA/m is expressed by a following equation.

$$(\log |d\lambda/dH|_{0.1}/10^{-9} \text{ m/A}) = -0.19 \log(P_r/\text{Pa}) - 0.15 \quad (3)$$

The highest magnetostrictive susceptibility measured is $8.5 \times 10^9 \text{ m/A}$ under the lowest value of residual gas pressure of $2 \times 10^{-5} \text{ Pa}$.

4.4 Substrate temperature dependent $|d\lambda/dH|$

The Fe-Sm alloy thin films are prepared by DC-magnetron sputtering process at elevated substrate temperatures (T_s : substrate temperature) from room temperature to 673 K under the constant P_s and P_r values. Figure 9 shows applied magnetic field dependent magnetostrictive susceptibility of

Fe_{2.2}Sm alloy film prepared at different substrate temperatures. The magnetization decreases the compressive magnetostrictive susceptibility ($|d\lambda/dH|$) at different substrate temperature (T_s). High $|d\lambda/dH|$ values are obtained for Fe_{2.2}Sm thin films heated from 474 to 523 K of substrate temperature.

Figure 10 shows magnetostrictive susceptibility ($|d\lambda/dH|_{0.1}$) at 0.1 MA/m against reduced substrate temperature (T_s/T_m). Here, T_m is melting point = 1553 K¹⁷⁾ under the standard sputtering (0.5 Pa) and standard residual ($5.0 \times 10^{-4} \text{ Pa}$) gas pressures. The high $|d\lambda/dH|_{0.1}$ values are obtained for Fe_{2.2}Sm thin films heated from 0.30 to 0.37 of (T_s/T_m), although low susceptibility values are obtained for films heated below 0.27 or above 0.4 of T_s/T_m . When the T_s is 323 K ($T_s/T_m = 0.20$) and 523 K ($T_s/T_m = 0.34$), the volume fractions of crystal samarium alloy and oxides become low, resulting in high volume fraction of amorphous phase with high magnetostrictive susceptibility.¹⁸⁾

On the other hand, the high volume fraction of the crystalline phases with low magnetostriction and the low volume fraction of amorphous phase with high magnetostriction are observed in Zone II of Thornton model in Fig. 4, when the substrate temperature is 623 K ($T_s/T_m = 0.4$).¹⁸⁾

4.5 Effects of morphological interface on $|d\lambda/dH|$ in Zone T

The high magnetostrictive susceptibility ($|d\lambda/dH|_{0.1}$) is obtained at the optimum substrate temperature from 0.30 to 0.37 of T_s/T_m (see Fig. 10) under the lowest sputtering pressure of 0.2 Pa (see Fig. 6) in Zone T (see Fig. 4). In order to explain the high magnetostrictive susceptibility, Thornton morphological model¹⁵⁾ and SEM micrograph are convenient tools. The influences of substrate temperature and sputtering argon gas pressure related to kinetic energy of sputtering (depositing) particles on morphological texture can be explained for DC-magnetron sputtering process. Namely, the model exhibits influences of both thermal energy of

Fig. 10 Magnetostrictive susceptibility at 0.1 MA/m $|d\lambda/dH|_{0.1}$ against reduced substrate temperature (T_s/T_m , T_m is melting point (1553 K¹⁷⁾) under standard sputtering pressure (0.5 Pa) and standard residual gas pressure (5.0×10^{-4} Pa).

substrate and kinetic energy of sputtering (depositing) particles on morphological structure as follows.

The densely packed fibrous amorphous phase with unclear interface is observed in Zone T ($0.2 \text{ Pa} < P_s < 0.5 \text{ Pa}$, $0.2 < T_s/T_m < 0.34$) in Fig. 4. The clear interface cannot be observed by SEM morphological observation. Since the unclear morphological interface in fine densely packed fibrous amorphous phase in Zone T probably exists, the cooperative phenomena among domains probably act to move the domain wall through unclear interface. Since the decreasing in impurity atoms at the unclear interface easily changes the domain direction and then doesn't prevent to move the magnetic domain wall in the amorphous phase, the high magnetostrictive susceptibility (see Fig. 8) of the Sm-Fe film deposited under the lower residual gas pressure can be explained.

A magnetostrictive susceptibility at 0.1 MA/m of magnetic field of Fe_{2.2}Sm alloy film also decreases with increasing in residual gas pressure. When the barrier (obstacles), controlled by residual gas pressure, at interface of morphology is formed and then prevents to move the magnetic domain wall, the oxidation at the interface of the grains in the films probably decreases the $|d\lambda/dH|$ value, as shown in Fig. 8. Therefore, influences of residual gas pressure on magnetostrictive susceptibility are mainly explained by the oxidation at morphological interface as well as the morphological interface formation.

4.6 Influence of morphological change on $|d\lambda/dH|$ in Zones I and II

Porous tapered crystalline grains separated by voids with remarkable interface have been found in Zone I (see Fig. 4).¹⁵⁾ Since the remarkable interface among grains probably acts as obstacles (barrier) to prevent to move the domain wall, the low magnetostrictive susceptibility ($0.5 \text{ Pa} < P_s < 1.0 \text{ Pa}$ [see Fig. 6], $T_s/T_m = 0.27$ [see Fig. 10]) of Fe-Sm alloy film in Zone I is explained.

The drop of $|d\lambda/dH|_{0.1}$ from 0.20 to 0.27 of T_s/T_m is large, as shown in Fig. 8. Since the reaction rate between impurity and samarium atoms at clear interface of porous tapered grains in Zone I increases at elevated temperatures, the formed barrier (obstacles) prevents to move the domain wall. Thus, the $|d\lambda/dH|_{0.1}$ tremendously decreases at elevated temperatures from 0.20 to 0.27 of (T_s/T_m) in ZONE I (see Fig. 10).

On the contrary, the low substrate temperature generally decreases the rate of oxidation at clear interface of porous tapered grains in Zone I. Since the clear interface, which doesn't largely trap the impurity atoms, is not effective barrier to prevent to move the domain wall, decreasing the substrate temperature from 0.27 to 0.20 in ZONE I in Fig. 2 enhances the $|d\lambda/dH|_{0.1}$ value (see Fig. 10).

On the other hand, columnar phase with remarkable interface is also found in Zone II ($P_s = 0.5 \text{ Pa}$, $T_s/T_m = 0.4$) in Fig. 4. Since the remarkable interface among columnar grains probably act as the obstacles (barrier) to prevent to move the domain wall, the low magnetostrictive susceptibility (see Fig. 10) is explained in Zone II of SmFe_{2.2} alloy thin films.

5. Conclusion

Influences of residual gas pressure, substrate temperature and argon gas pressure on negative (compressive) magnetostrictive susceptibility of SmFe_{2.2} alloy thin films prepared by direct current magnetron sputtering process are investigated.

- (1) The magnetostrictive susceptibility ($|d\lambda/dH|_{0.1}$), sensitive to morphological structure change in the densely packed amorphous phase, increases with decreasing the pressure of argon gas (P_s /Pa) and is expressed by a following equation.

$$(\log |d\lambda/dH|_{0.1}/10^{-9} \text{ m/A}) = -0.44 \log(P_s/\text{Pa}) + 0.35$$

- (2) The magnetostrictive susceptibility ($d\lambda/dH$)_{0.1}/10⁻⁹ m/A obtained at 0.1 MA/m of the densely packed amorphous phase decreases with increasing the pressure of residual gas (P_r /Pa) and is expressed by a following equation.

$$(\log |d\lambda/dH|_{0.1}/10^{-9} \text{ m/A}) = -0.19 \log(P_r/\text{Pa}) - 0.15$$

- (3) High magnetostrictive susceptibility of fine densely packed amorphous phase is found at each substrate temperatures (T_s) from 423 to 523 K.
- (4) When the residual gas pressure affects the density of impurity atoms at the unclear morphological interface in densely packed fibrous amorphous phase, the high magnetostrictive susceptibility can be explained that the decreasing in impurity atoms at the unclear interface doesn't probably prevent to move the magnetic domain wall in the amorphous phase.
- (5) The low substrate temperature generally decreases the rate of oxidation at clear interface of porous tapered grains in Zone I. When the clear interface is not effective barrier to prevent to move the domain wall, decreasing the substrate temperature from 0.27 to 0.20 in ZONE I enhances the magnetostrictive susceptibility.

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