Effects of magnetic flux density and substrate temperature on Ni films prepared by means of unbalanced magnetron sputtering assisted by inductively coupled plasma

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Abstract. We prepared Ni films on glass substrates by means of Unbalanced Magnetron Sputtering Assisted by Inductively Coupled Plasma (UM-ICP). The effects of the magnetic flux density $B_C$ and the substrate temperature $T_S$ on their structures were investigated. We found that the surface grain size observed with an atomic force microscope became large as $B_C$ and $T_S$ increase. We also found that the (111) plane measured by X-ray diffraction was preferentially oriented on the films under $B_C=5$ mT at $T_S=60^\circ $C. These indicate that the energetic particle bombardment under $B_C$ and the increase of $T_S$ promote the surface diffusion on the Ni films. The ferromagnetic domain patterns observed with a magnetic force microscope showed the stripe domain structure having the domain width increasing with increasing $T_S$ and the fractal dimension of 1.88.

1. Introduction
Magnetron sputtering is an essential technology to deposit films in electronic devices. The sputtering enables us to deposit films of high melting point materials and compounds with complex compositions. However, highly ferromagnetic materials can be difficult to sputter, because a considerable portion of magnetic flux is shunted by ferromagnetic target [1]. Kawabata et al. developed the sputtering system called Multipolar Magnetic Plasma Confinement (MMPC) [2], which made up for the shunted magnetic flux by externally arranged multipolar magnets. MMPC enables the high-speed deposition of Ni films with the rate twenty times larger than the other sputtering methods [3-5]. MMPC combined with a high-frequency coil, which is called MMPC assisted by Inductively Coupled Plasma (MMPC-ICP) [6], promotes the ionization of sputtering gas and the preferred orientation of the (111) plane in the Ni films. The replacement of the multipolar magnets by a solenoid in MMPC-ICP enables to vary the magnetic flux toward a substrate $B_C$. This system is called Unbalanced Magnetron Sputtering Assisted by Inductively Coupled Plasma (UM-ICP) and able to increase the number of the charged particles incident on the substrate and enhance the energetic particle bombardment, by increasing $B_C$ [7].

The energetic particle bombardment promotes the surface diffusion on the substrate and affects the adhesion, the density and the grain size of films [8, 9]. The bombardment of the particles with suitable energies increases the grain size [10]. It is known that the surface diffusion is also promoted by thermal energy and makes the film highly dense in the temperature region of $0.3<T_S/T_M<0.5$ [11], where $T_S$ and $T_M$ are the substrate and the melting temperatures, respectively. This indicates that Ni
films with $T_M = 1455°C$ become highly dense in the region of $245°C < T_S < 591°C$, which includes the ferromagnetic phase transition temperature $T_C = 358°C$. The purpose of the present study is to clarify the effects of the magnetic flux density $B_C$ and the substrate temperature $T_S$ on the structure of the Ni films prepared by UM-ICP. The surface morphology and the magnetic domain structure were observed as their macroscopic structures and the lattice constant was measured as their microscopic structure at room temperature.

2. Experimental

The sputtering system UM-ICP used to deposit the Ni films is schematized in Figure 1. The pumping system is composed of a liquid nitrogen baffle, a turbo molecular pump and a rotary pump. The disposition of the magnets on the reverse of the target is symmetrical as shown in Figure 1. The solenoid was set up in the outside of the chamber. In order to enhance the ionization of sputtering gas and sputtered atoms, the high-frequency coil made of SUS304 was set up 60 mm above the target. Radio frequency power was applied to the high-frequency coil through a matching circuit. DC power was applied to the target through a low-pass filter. The heater was installed in the substrate holder. The target to substrate distance was fixed at 150 mm.

![Figure 1. Schematic diagram of the present sputtering system UM-ICP.](image)

The Ni target with the purity of 99.95%, the thickness of 5 mm and the diameter of 200 mm and the Ar gas with the purity of 99.9999% were used. The pressure and the flow rate of the Ar gas were maintained at $1.33 \times 10^{-1}$ Pa and 1 sccm, respectively. The high-frequency power and the target DC power were maintained at 30 W and 600 W, respectively. The magnetic flux density at the center of the external solenoid $B_C$ was maintained at 0 mT, 3 mT and 5 mT. Figure 2 shows the magnitude of the magnetic flux density measured at the position distant from the center of the solenoid $B_\perp$. When $B_C$ is maintained at 0 mT, 3 mT and 5 mT, respectively, $B_\perp$ has the averaged values of 1.1 mT, 2.3 mT and 3.3 mT on the wide area of the substrate.

The Ni films were deposited on the glass substrates with the area of 15 mm × 15 mm (Corning 1737), which were supersonic-cleaned by acetone and ethanol. The film thickness $d$ was about 500 nm. The deposition rate varied between 26.5-32.5 nm/min. $T_S$ was maintained at 90°C, 200°C, 300°C, 380°C under 0 mT, 75°C, 200°C, 300°C, 380°C under 3 mT and 60°C, 200°C, 300°C, 380°C under 5 mT. The lattice constants $a$ of the Ni films were measured by X-ray diffraction (XRD) with the CuK$\alpha_1$ radiation. The averaged value of the grain size $D_G$, that is, the diameter of the grain, and the RMS roughness $R_q$ were obtained from the surface morphology observed with an atomic force microscope (AFM). The ferromagnetic domain patterns were observed with a magnetic force microscope (MFM). The average domain width $l$ was obtained from the Fourier transformed figures of the domain patterns.
3. Results and Discussion

3.1. Surface morphology of Ni films
The AFM images of the Ni films are shown in Figure 3. The figure shows that the surface morphology strongly depends on the magnetic flux density $B_C$ and the substrate temperature $T_S$. The average surface grain size $D_G$ increases as $B_C$ and $T_S$ increases.

$B_C=5\text{ mT}$

$T_S=60^\circ\text{C}$

$T_S=200^\circ\text{C}$

$T_S=300^\circ\text{C}$

$T_S=380^\circ\text{C}$

$B_C=3\text{ mT}$

$T_S=75^\circ\text{C}$

$T_S=200^\circ\text{C}$

$T_S=300^\circ\text{C}$

$T_S=380^\circ\text{C}$

$B_C=0\text{ mT}$

$T_S=90^\circ\text{C}$

$T_S=200^\circ\text{C}$

$T_S=300^\circ\text{C}$

$T_S=380^\circ\text{C}$

**Figure 3.** AFM images of Ni films with the scan size of $1\mu\text{m} \times 1\mu\text{m}$.

Figure 4(a) shows that $D_G$ under $B_C=5\text{ mT}$ at $T_S=380^\circ\text{C}$ is 4.5 times larger than $D_G$ under $B_C=0\text{ mT}$ at $T_S=90^\circ\text{C}$. The RMS roughness $R_q$ becomes large according to the increase in $D_G$, as shown in Figure 4(b). These show that the grains well grow under strong $B_C$ and at high $T_S$. Therefore, the surface diffusion on the substrate was promoted by the energetic particle bombardment under strong $B_C$ and by the heat distributed at high $T_S$. 

**Figure 2.** Magnetic flux density at the position distant from the center of the substrate $B_\perp$. 

3. Results and Discussion
3.2. X-ray diffraction patterns of Ni films

The XRD patterns of the Ni films are shown in Figure 5. The clear peaks of the (111) and the (200) reflections show that the Ni films have the fcc structure and are composed of polycrystals. The increase in the peak intensity with the increase of $B_C$ from 0 to 5 mT below $T_S=200^\circ$C in Figure 5 coincides with the increase in the grain size below $D_G=80$ nm with increase of $B_C$ in Figure 4(a).

![X-ray diffraction patterns of Ni films](image)

**Figure 5.** X-ray diffraction patterns of Ni films.

The lattice constants $a$ of the Ni films deposited on the glass substrate were obtained from the XRD patterns shown in Figure 5. All values of $a$ were smaller than the bulk lattice constant $a_{\text{bulk}}=3.5241$ Å [12], as shown in Figure 6. This shows the compressive stresses along the direction perpendicular to the Ni films. The substrate temperature dependence of the ratio between the integrated intensities $I(200)/I(111)$ is shown in Figure 7. The comparison of $I(200)/I(111)$ between the Ni films and the Ni powder brings us the information on the preferred orientation of the lattice plane in the Ni films deposited on the glass substrate. The preferred orientation of the (111) plane is shown under $B_C=5$ mT at $T_S=60^\circ$C, indicating that the surface diffusion is promoted under $B_C$. This is because the preferred orientation of the (111) plane, of which the lattice spacing is smaller than that of the (100) plane, is enhanced by the energetic particle bombardment [6]. It is expected, therefore, that the surface diffusion would be highly promoted under $B_C$ larger than $B_C=5$ mT. The preferred orientation of the (100) plane in the Ni films deposited at high temperatures would be due to a thermal activation process additional to the effect of $B_C$. The lowering of the preferred orientation of the (100) plane at $T_S=380^\circ$C could be attributed to the lowering of the compressive stress due to the absence of magnetostriction above $T_C=358^\circ$C.
3.3. Ferromagnetic domain patterns in Ni films

The MFM images of the Ni films are shown in Figure 8. The bright and the dark regions correspond to the north and the south poles of the ferromagnetic domains, respectively. The domain patterns show that the lamellar domain structure, which is called “stripe domain structure” [13], formed in the Ni films.

The average domain width $l=\lambda/2$ was obtained from the wave number $2\pi/\lambda$ which characterized the Fourier transformed figure of each domain pattern. The substrate temperature dependence of $l$ is shown in Figure 9. The width $l$ increases as $T_s$ increases below $T_s=200^\circ C$. This indicates that the spontaneous magnetization $M_s$ decreased as $T_s$ increased. This is because $M_s$ of the stripe domain structure is proportional to $\sqrt{\Delta I/\lambda}$ [13]. The width $l$ of the Ni film deposited at $B_C=5$ mT and $T_s=60^\circ C$ is the smallest in Figure 9, showing that the value of $M_s$ is the largest in the domain patterns in Figure 9.
8. This is due to that the magnetization easy axis normal to the (111) plane is nearly perpendicular to the Ni film having the preferred orientation of the (111) plane in Figure 7. The decrease of \( l \) under low \( B_C \) and high \( T_S \) corresponds to the lowering of the preferred orientation of the (100) plane in Figure 7. All domain patterns showed the fractal dimension of 1.88, which is nearly equal to that of the percolation cluster 1.89 \cite{bib14}. Thus the ferromagnetic domain structures observed at room temperature conserves the characteristics of the structures formed at \( T_S \), though \( l \) should become large while the films are aged at room temperature.

![Figure 9. Effects of the magnetic flux density \( B_C \) and the substrate temperature \( T_S \) on the average domain width \( l \) in Ni films.](image)

4. Conclusions
We prepared the Ni films on the glass substrates by means of the sputtering system UM-ICP and investigated the effects of the magnetic flux density \( B_C \) and the substrate temperature \( T_S \) on the structures of the films. We found that the surface grain size became large as \( B_C \) and \( T_S \) increased and the preferred orientation of the (111) plane occurred under \( B_C=5 \) mT at \( T_S=60^\circ C \). This indicates that the increases of \( B_C \) and \( T_S \) promote the surface diffusion on the Ni films. We also found that the characteristics of the ferromagnetic domain structures formed at \( T_S \) were conserved after cooling to room temperature. Thus we conclude that the structure of the Ni films prepared by the sputtering system UM-ICP is controllable by \( B_C \) and \( T_S \).

References
\[ \text{[1] Zhang H, Poole J, Eller R and Keefe M 1999 } \textit{J. Vac. Sci. Technol. A} 17 1904 \]
\[ \text{[2] Kawabata K, Tanaka T, Kitabatake A, Yamada K, Mikami Y, Kajioka H and Toiyama K 2001 } \textit{J. Vac. Sci. Technol. A} 19 1438 \]
\[ \text{[3] Ghebouli B, Layadi A and Kerkaiche L 1998 } \textit{Eur. Phys. J. AP} 3 35 \]
\[ \text{[4] Howell R S, Sarcona G, Saha S K and Hatalis M K 2000 } \textit{J. Vac. Sci. Technol. A} 18 87 \]
\[ \text{[5] Yu M, Qiu H, Chen X, Wu P and Tian Y 2008 } \textit{Thin Solid Films} 516 7903 \]
\[ \text{[6] Takesako K, Okayama Y and Kawabata K 2006 } \textit{J. Vac. Soc. Jpn.} 49 156 \]
\[ \text{[7] Eboshi R, Muraoka Y, Kurokawa S, Tachibana A, Hasui Y and Kawabata K 2008 } \textit{J. Vac. Soc. Jpn.} 51 182 \]
\[ \text{[8] Mattox D M 1989 } \textit{J. Vac. Sci. Technol. A} 7 1105 \]
\[ \text{[9] Rossnagel S M and Cuomo J J 1989 } \textit{Thin Solid Films} 171 143 \]
\[ \text{[10] Asahara H, Inokuchi A, Watanuki K, Hirayama M, Teramoto A and Ohmi T 2009 } \textit{Jpn. J. Appl. Phys.} 48 04C135 \]
\[ \text{[11] Thornton J A 1986 } \textit{J. Vac. Sci. Technol. A} 4 3059 \]
\[ \text{[12] Ha K, Ciria M, O’Hndley R C, Stephens P W and Pagola S 1999 } \textit{Phys. Rev. B} 60 13780 \]
\[ \text{[13] Saito N, Fujiwara H and SugitaY 1964 } \textit{J. Phys. Soc. Jpn.} 19 421, 1116 \]
\[ \text{[14] Stauffer D 1979 } \textit{Phys. Rep.} 54 1 \]